THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Environmental Assessment of Battery Systems:

Critical Issues for Established and Emerging Technologies

CARL JOHAN RYDH

Environmental Systems Analysis CHALMERS UNIVERSITY OF TECHNOLOGY Göteborg, Sweden 2003 Environmental Assessment of Battery Systems: Critical Issues for Established and Emerging Technologies

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Abstract

Energy and material flows associated with portable and industrial rechargeable batteries have been quantified in a life-cycle perspective, as guidance for development of battery systems. The study included portable batteries based on nickel-cadmium, nickel-metal hydride and lithium-ion. Energy return factors and overall energy efficiencies were calculated for a stand-alone photovoltaic (PV)-battery system under different operating conditions. Eight different battery technologies for stationary energy storage were evaluated: lithium-ion (Ni), sodium-sulphur, nickel-cadmium, nickel-metal hydride, lead-acid, polysulphide-bromide, vanadium redox and zinc-bromine.

In applications where batteries are difficult to collect at the end of their life, dissipative losses of toxic metals from incineration and landfills are of main concern. Indicators of global metal flows were used to assess the potential environmental impact of metals used in portable batteries. Lithium-ion and nickel-metal hydride batteries have lower impact based on indicators of anthropogenic and natural metal flows than nickel-cadmium batteries.

Energy requirements during production and usage are important for battery systems where the metal losses throughout the battery life cycle are low. For a PV-battery system with a battery capacity three times higher than the daily energy output, the energy return factor is 0.64-12, depending on the battery technology and operating conditions. With a service life of 30 years, the energy payback time is 1.6-3.0 years for the PV-array and 0.55-43 years for the battery, which highlights the energy related significance of batteries and the large variation between different technologies. Some of the emerging technologies studied, e.g. lithium-ion and sodium-sulphur, show favourable performance for use in PV-battery systems, resulting in higher energy return factors and higher overall battery efficiencies than for established battery technologies.

The environmental impact can be reduced by matching operating conditions and battery characteristics in a life-cycle perspective. To decrease the environmental impact of battery systems, the development of battery technologies should aim at the recycling of materials, increased service lives and higher energy densities. To decrease the environmental impact arising from the use of metals in battery systems, metals with high natural occurrence should be used and regulations implemented to decrease the need for virgin metals. To increase the overall energy efficiencies of battery systems, the development of battery technologies should aim at higher charge-discharge efficiencies and more efficient production and transport of batteries.

Keywords: rechargeable, battery, energy efficiency, environmental indicators, energy analysis, life cycle assessment, substance flow analysis, photovoltaic cells, renewable energy, materials management, metal recycling

Miljöbedömning av batterisystem: Kritiska aspekter för etablerade och framväxande teknologier

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Sammanfattning

Den snabba tillväxten av bärbar elektronik, behov av säker strömförsörjning och lagring av förnybar energi har lett till att användningen av batterier ökar. För att undvika att utvecklingen av etablerade och framväxande batteriteknologier begränsas krävs att deras kritiska miljöaspekter identifieras och kontrolleras.

För att ge vägledning i utveckling av batterisystem har energi- och materialflöden för uppladdningsbara batterier kvantifierats ur ett livscykelperspektiv. Fallstudier har genomförts på småbatterier baserade på nickel-kadmium, nickel-metallhydrid och litiumjon. Energiåterbetalningsfaktorer och totala energiverkningsgrader för ett solcellsystem med olika typer av batterier har beräknats för olika driftsförhållanden. De åtta batteriteknologierna för stationär energilagring som studerats är: litium-jon (Ni), natriumsvavel, nickel-kadmium, nickel-metallhydrid, bly-svavelsyra samt redox flow batterier baserade på polysulfid-bromid, vanadin-vanadin och zink-brom.

I applikationer där batterier är svåra att samla in efter att de nått sin användbara livslängd uppstår största miljöpåverkan genom spridning av giftiga metaller från avfallsförbränning och deponering. Indikatorer för globala metallflöden användes för att bedöma den potentiella miljöpåverkan för olika marknadsvolymer av olika teknologier av småbatterier. Baserat på indikatorer för antropogena och naturliga metallflöden bedömdes framväxande teknologier som litium-jon och nickel-metallhydrid batterier ha lägre miljöpåverkan än nickel-kadmium batterier.

Energianvändning vid batteritillverkning och under drift är framförallt viktig för batterisystem där metallutsläppen under batteriets livscykel är små. För ett solcellbatterisystem med en batterikapacitet tre gånger högre än den utmatade energin beräknades energiåterbetalningsfaktorn till 0.64-12 för olika batteriteknologier och driftförhållanden. Med en livslängd av 30 år är energiåterbetalningstiden 1.6-3.0 år för solcellerna och 0.55-43 år för batteriet vilket visar den stora betydelsen av batterier ur energiperspektiv och den stora variationen mellan olika teknologier och driftsförhållanden. Flera av de framväxande batteriteknologierna, exempelvis litium-jon och natrium-svavel, har fördelaktiga egenskaper för användning i solcell-batterisystem, som ger kortare energiåterbetalningstid och högre total verkningsgrad än etablerade batteriteknologier.

Miljöpåverkan av batterier kan reduceras genom att välja batteriteknologier med tanke på användningsområde och driftsförhållanden ur ett livscykelperspektiv. För att minska miljöpåverkan av batterisystem bör utvecklingen av olika batteriteknologier sikta på höga återvinningsnivåer av material, lång batterilivslängd och hög energidensitet. För att minska miljöpåverkan från metallflöden som uppstår genom användning av batterier bör metaller med hög naturlig förekomst användas och regleringar införas för att minska behovet av att bryta jungfruliga metaller. Energibehovet för batterisystem reduceras effektivast genom ökad energiverkningsgrad vid laddning och urladdning av batterier följt av effektivare produktion och transport av batterier.

Nyckelord: uppladdningsbara batterier, metallåtervinning, energiverkningsgrad, miljöindikatorer, materialflödesanalys, energianalys, livscykelanalys, förnybar energi

List of appended papers

This thesis is based on the work described in the following papers, which are referred to by their Roman numerals in the text:

- I Rydh, C. J. and Karlström, M. (2002) Life Cycle Inventory of Recycling Portable Nickel-Cadmium Batteries, *Resources, Conservation and Recycling* Vol. 34, No. 4, p. 289-309
- II Rydh, C. J. and Svärd, B. (2003) Impact on Global Metal Flows Arising from the Use of Portable Rechargeable Batteries, *The Science of the Total Environment* Vol. 302, No. 1-3, p. 167-184
- III Rydh, C. J. (1999) Environmental Assessment of Vanadium Redox and Lead-acid Batteries for Stationary Energy Storage, *Journal of Power Sources*, Vol. 80, No. 1-2, p. 21-29
- IV Rydh, C. J. and Sun, M. (2003) Life Cycle Inventory Data for Materials Grouped According to Environmental and Mechanical Properties, Accepted for publication in *Journal of Cleaner Production*
- V Rydh, C. J. and Sandén B. A. (2003) Energy Analysis of Batteries in Photovoltaic Systems, Submitted for publication in *Energy Conversion and Management*

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Preface

"...with every advance the difficulty of the task is increased" *Planck's principle of increasing effort*

By putting things in perspective and relating them to each other, their relative importance can be understood. For example, in the year 0 the global population was estimated to be 0.30 billion. The world population reached 1 billion in 1804, 2 billion in 1927 and 6 billion in 1999 (UN 1999). The major underlying reason for this extraordinary increase of the human population is the utilisation of energy in fossil fuels, i.e. coal, oil and natural gas, which have been created during millions of years. During the past century, the global primary energy supply increased from $30 \cdot 10^{18}$ J/year to $403 \cdot 10^{18}$ J/year (Grübler 1998). In 1997, fossil fuels provided 86% of the global primary energy supply (WRI 2000). Extraction of minerals from the earth's crust has provided us with materials that have enabled technological development that was hard to imagine many years ago. Human developments have become so far-reaching that they influence material and energy flows on a global scale, which will eventually lead to irreparable disturbances in the functioning of the ecosystems, which support human life. Since physical flows of materials and energy are essential for any organism (or product system), knowledge and management of these flows are fundamental for survival. During the past century, material flows in the technosphere have become more complex than ever, making it difficult to understand how they are connected.

Before man took the step into the agricultural society, only a few different professions and skills were needed in hunter-gatherer societies to make a living. The development of specialised agricultural methods created surplus resources and it was no longer necessary for the whole community to be involved in providing food. Specialised professions evolved in different fields, for example agricultural technology, administration and military defence. The industrial society relies on an innumerable number of highly specialised professions (e.g. battery experts!). Specialisation of human resources has led to a more complex and effective society. However, at a certain level of complexity¹, Tainter (1995) proposes that increased specialisation will require more input resources than the benefits created. Increased administration and complexity of society may result in inefficient use of resources and will eventually result in collapse to a lower level of energy use. Tainter (1995) also suggests that increasing complexity was the underlying reason for the collapse of earlier advanced civilisations, for example, the Roman Empire and the Lowland Classic Maya.

¹ Complexity refers to things as the size of society, the number and distinctiveness of its parts, the variety of specialised social roles that it incorporates, the number of distinct social personalities present and the variety of mechanisms for organising these into a coherent, functioning whole (Tainter 1995).

The complex industrial society works since it is subsidised by fossil fuel. Our capital of fossil fuels can be used to give short term benefits or be invested in long term sustainable energy systems. The use of fossil fuels may be constrained by unacceptable environmental effects or resource scarcity. It is therefore important to understand how energy and material flows can be managed as efficiently as possible.

The choice of batteries for this study can be traced back to my hobby during the 1980s-1990s, which was competitive racing with radio-controlled model cars. At that time, rechargeable lithium-ion and nickel-metal hydride batteries had not yet been commercialised. Many other things have changed rapidly during my short journey in life. In the beginning of the 1990s it was very popular to travel around in Europe with an Interrail pass. Today, inter-flying around world has become common practice. When I wrote my Master's Dissertation in 1997, I read physical journals and books, and references were "saved" by using photocopying machines. Only a few years later, information has become more accessible than ever via the Internet, and references are now saved as downloadable text files.

Human induced changes to the earth are occurring at a faster rate than ever in human history. It is fascinating to have the opportunity to experience this revolutionary era. At the same time, it is worrisome since we have no historical references to help us see what can be expected in the future. During my studies, my perspectives have broadened, and I am particularly grateful for having had the opportunity to learn how to assess, process and present information critically.

This thesis provides quantitative data on energy and material flows associated with battery systems as they can be described today. We do not know much about the future, but this study provides a starting point or a historical footprint of the situation at the beginning of 21st century. I hope that this work will contribute to improved environmental management of battery systems and the application of methods for environmental assessment of all types of product systems.

During the course of this work, I have come into contact with many people who have influenced and inspired my work. I would like to especially thank the following people:

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Valuable inspiration for the work on this thesis has been found in a variety of activities in contrast to paper reading and writing. Restaurant Rice and Wine has given me lots of energy. Apart from the activity of eating there has been some time left for outdoor and indoor activities. The time spent kayaking, ice-skating, biking and hiking, especially with Henrik Engström, Kerstin Kahl, Erik Landmark, Åse Möller, Magnus Nicklasson, Mattias Persson and Olof Pettersson, has charged my batteries and given me the energy to complete the research. Thanks also to Linus and "the Gang", for giving me bruises, practise in fighting and cheap tricks during the compulsory "Tuesday-innebandy". It gave me the necessary skills to master my computer.

The last word goes to my five-year-old computer, which is totally exhausted after writing this thesis and running all new software.

Tail Johan God

Carl Johan Rydh Kalmar, December 2003

Nomenclature

AC	air conditioning		
battery efficiency	Charge-discharge (direct current) energy efficiency of a battery. See also <i>overall battery efficiency</i> .		
battery system	The technical system of a rechargeable battery, in some cases also including a charger, and life-cycle activities for materials production, battery production, use and end-of-life treatment.		
calendar life	Service life in years of a battery under normal operation.		
CERA	cumulative energy requirement analysis		
CHP	combined heat and power		
СР	condensing power		
DOD	depth of discharge		
energy density (gravimetric)	The ratio between the energy capacity and the mass of a battery (Wh/kg).		
energy payback time, <i>t</i> *	The ratio between the primary fossil energy that is required to produce the PV-battery system and the average annual gross primary fossil energy use of a diesel generator. The time it takes to pay back the energy that was used to produce a PV-battery system, by not using the diesel generator (or another source of electricity that is replaced).		
energy return factor, f	(1) The ratio between the service life of the PV-battery system and the energy payback time.		
	(2) The ratio between the fossil energy replaced and the fossil energy required to produce the PV-battery system.		
environmental aspect	Element of an organisation's activities, products or services that can interact with the environment.		
ERA	environmental risk assessment		
float service life	Service life in years of a battery due to corrosion processes. Assumed to be equivalent to <i>calendar life</i> for redox flow batteries.		
GWp	giga (10 ⁹) watt peak		
industrial battery	Battery with a mass greater than 1 kg. See also stationary battery.		
LCA	life cycle assessment		
LCI	life cycle inventory		
LEI, i_E	lithospheric extraction indicator. The ratio between anthropogenic and natural metal flows.		

Li ion	lithium ion
MFA	material flow analysis
MSW	municipal solid waste
NiCd	nickel-cadmium
NiMH	nickel-metal hydride
NaS	sodium-sulphur
overall battery efficiency, $\eta^{*}{}_{B}$	The ratio between the electricity output of the battery system and the total input in equivalent electricity.
PbA	lead-acid
portable battery	Battery with a mass less than 1 kg.
primary battery	Battery intended not to be recharged.
primary material	Material produced from matter extracted for the first time. Virgin material.
PSB	polysulphide-bromide
PV	(solar) photovoltaics
SD	standard deviation
secondary battery	Battery intended to be recharged.
secondary material	Material produced from raw material that has been used in processes or products before. Recycled material.
SFA	substance flow analysis
SHS	Solar Home System. A small stand-alone electricity system based on a PV array and a rechargeable battery.
SLI	Starting, Lighting and Ignition. Starter battery for cars with internal combustion engine.
stationary battery	Industrial battery used for UPS, load-levelling, power quality or energy storage.
technosphere	Material and energy in the human sphere (constructions, machines, tools, etc.)
ton	metric ton
traction battery	Battery used for propulsion of an electric vehicle or onboard power. Commonly used for continuous deep cycling.
UPS	uninterruptible power supply. Energy supply with back-up batteries providing energy in the case of a power failure of the grid.
VRB	vanadium-redox battery
ZnBr	zinc-bromine

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APPENDED PAPERS

1 Introduction

1.1 Background

The development of portable electronic products² has increased the demand for highperformance batteries. Figure 1 shows that the market for portable rechargeable batteries has quadrupled between 1989 and 2000. The demand for batteries with higher energy density has led to the development and commercialisation of new electrochemical systems, other than the established lead-acid and nickel-cadmium batteries. The introduction of new technological standards for wireless communication is likely to contribute to the growth of battery use in the future.



Fig. 1. Number of rechargeable portable battery cells produced globally per year from 1989 to 2000 (portable lead-acid batteries not included) (NRI 1999, Paper I). (NiCd = nickel-cadmium, NiMH = nickel-metal hydride, Li-ion = lithium-ion)

A battery application of growing importance is large-scale and cheap storage of renewable energy (Butler *et al.* 1999). The use of photovoltaic (PV) cells in off-grid applications, which may include batteries, grew 25% per year during the past three years (1999-2002) on a world basis (Maycock 2003). The annual growth rate of the peak power of photovoltaic cell production was 35% during the period 1997-2002 (Fig. 2). The cumulated produced peak power globally was 1 GWp in 2001. About half of the installed capacity is used in stand-alone applications where many systems are equipped with energy storage to increase the usefulness of the PV system (Fig. 2).

About 1.6 billion of the world's 6.1 billion people have no access to any form of electricity (IEA 2002). To enable widespread access and large-scale diffusion of distributed electricity including energy storage in batteries, economic, technological and environmental factors ought to be optimised. To give future generations the benefit of the capital of fossil fuels, non-renewable resources can be used to create renewable substitutes. If electricity from PV-

² 3C products: camcorders, cellular phones and computers.

battery systems is to contribute to a renewable energy supply, they must give net energy yield throughout their lifetime.³ It should be noted that net energy generation is less important in energy systems with low energy turnover. For example, solar home systems (SHSs) can be used improve the standard of living in poor environments by providing a means of moving energy from industrialised areas to rural areas.⁴



Fig. 2. Annual world photovoltaic production by application (Maycock 2003).

The chemical and physical properties of some metals make them attractive for use as electrode materials in batteries (Linden and Reddy 2002). For a few metals, batteries are the major end use. Seventy percent of the cadmium mined world wide each year is used in NiCd batteries (Papers I & II) and 70% of lead (virgin and recycled lead) is used in lead-acid batteries (Paper III). Some new types of electrochemical systems use metals, e.g. cobalt, neodymium, praseodymium and vanadium, whose eco-toxicological characteristics are not well known. Throughout the product life cycle of batteries, metals may give rise to

³ In stand-alone photovoltaic systems, batteries are used to accumulate electricity generated by PV modules, since the supply of solar energy is irregular. Batteries are not an energy source but a means of concentrating low-power solar energy into high-power electricity. Energy is indirectly required to produce batteries. The net energy output for rechargeable batteries, i.e. the energy output minus the energy required for battery production, depends on the energy turnover in the battery. For stationary batteries in a PV-battery system, 0.08-1.6 times the energy output may be used for battery production (Paper V). For portable batteries, the ratio between primary energy requirement for battery production and electrical energy output is 5-116 times for rechargeable batteries and 39-755 times for non-rechargeable batteries (Barlinn 1997, Scholl and Baumann 1997). If the ratio is higher than one, more energy is required to produce the battery than the useful energy output from the battery.

⁴ If one billion households had a SHS with a 100 Wp PV panel and assuming an annual yield of 150 kWh/year, the total electricity generated would be 150 TWh/year. This represents only 1% of the global electricity generation in 2001 (15 476 TWh/year, (IEA 2003)).

elevated background concentrations of metals in the environment and eco-toxicological effects.

Within the European Community, environmental concern has led to prohibition and regulation of batteries containing mercury (EC 1998). In the ongoing work of modifying the battery directive 91/157/EEC (EC 1991), a ban on cadmium in batteries has been discussed (EC 2003, EC 2003b). The Swedish battery ordinance (SFS 1997) regulates the market and the levies on batteries depending on their content of heavy metals. The Swedish Parliament has adopted Government Bill 1997-98:145 (SOU 1997) with the aim of phasing out the use of mercury, cadmium and lead in the long run, mainly through voluntary measures.

The long-term competitiveness of a battery system⁵ in the society is determined by complex interactions between different factors, e.g. usefulness, cost and risks. Environmental aspects and management of materials are important parameters requiring consideration in the strategic development of systems that include batteries (Ainley 1995, Robertsson *et al.* 1997). To make a battery system sustainable from a long-term perspective, the distance to constraining limits has to be as great as possible. Material flows of human society have to be incorporated into (e.g. phosphate) or isolated from (e.g. radioactive material) the natural turnover of energy and material. Three classes of constraints on industrial metabolism can be identified (Karlsson and Holmberg 1996): (1) limits on extraction due to limited resources, (2) limits on emissions due to nature's limited assimilation capacity and, (3) limits on the manipulation of nature that are possible without disrupting nature's productivity, functions and biodiversity.

These constraints can be applied to structure problems and guide the direction of development. Battery technologies requiring scarce materials or having low overall energy efficiency may be constrained by limited availability of concentrated materials and energy resources in deposits and natural energy flows and funds. The use of batteries with low energy efficiency or high energy requirements for their production may also be restricted by measures taken to mitigate climatic change and acidification while the use of battery systems with high dissipative losses of toxic metals may be restricted by heavy-metal pollution. Manipulation of nature includes soil erosion and extinction of species, which may for example be caused by open pit mining.

To avoid undesired effects of the use of a substance or a product, appropriate information and assessment methods are needed. If too narrow or short-term perspective is considered, there is a risk of sub-optimisation. Sub-optimisation is experienced when the efficiency of a unit process is increased, but at the same time, the overall efficiency is decreased. To improve batteries from an environmental perspective, greater knowledge concerning the environmental impact of battery systems must be gained. Emerging battery technologies, which may be implemented on the same scale as established technologies, should be assessed before possible environmental effects occur. Important factors and constraints related to battery technologies should be identified and managed from an environmental

⁵ The term battery system is used to define a technical system consisting of a rechargeable battery, in some cases also including a charger, and life cycle activities for materials production, battery production, use and end-of-life treatment.

perspective. Environmental information can be used to set up development goals and to evaluate steering effects (SOU 1996a).

Based on this background, the following questions ought to be answered in order to evaluate and manage batteries from an environmental perspective:

- Which methods are appropriate for assessing the environmental aspects of battery systems?
- Which are the most important environmental aspects of different battery systems?
- How do different parameters influence the environmental impact and energy flows of battery systems?

The environmental performance of portable batteries has been assessed in several studies (Hofstetter and Häne 1990, Törnblom 1996, Lankey 1998, Staal-Jensen and Petersen 1999) as well as batteries for traction purposes (Koonts *et al.* 1993, Acurex 1995, Gaines and Singh 1995, Kertes 1996, Garcia and Schlüter 1996, Socolow and Thomas 1997a, 1997b, Lave *et al.* 1995, 1996, 1997, Patyk and Reinhardt 1998, Steele and Allen 1998, Almemark et al. 1999, Karlsson 1999). The main objectives of these studies were to characterise environmental performance and compare different battery types in one type of application.

Only one study has been found dealing with environmental aspects of batteries in industrial applications and renewable energy systems. Alsema (2000) concluded that the production of the batteries used in SHSs contributed significantly to the gross energy requirement of the solar home system.

Despite the large number of studies, none has summarised critical issues regarding the assessment of the environmental impact of battery systems in different applications. By identifying parameters that are relevant in describing the environmental performance of battery systems, it will be easier to conduct further assessments and comparisons of batteries. To facilitate the communication of environmental information on batteries to stakeholders throughout the battery life cycle, there is a need to summarise the environmental aspects of batteries. Identification of important parameters can be used to direct research and product improvements. By developing computer models, the effects of changes in performance can be easily updated and evaluated. A comparison of different battery technologies can be used as a guide to battery choice for specific conditions.

From the variety of methods available for environmental systems analysis (Wrisberg *et al.* 2000), appropriate methods have to be chosen to assess the environmental performance of batteries. The choice of method for analysis at different spatial and temporal levels has been discussed by Bouman *et al.* (2000). However, there seem to be no practical experience of applying and comparing different methods to the analysis of batteries.

1.2 Goal and scope

The objectives of this work were:

- to identify and quantify environmental aspects of established and emerging battery systems, and
- to assess how different parameters influence the environmental impact of and energy flows resulting from battery systems.

Based on experience of analysis, methods for environmental assessment of battery systems will be discussed. Energy and material flows are assessed using substance flow analysis, energy analysis and life cycle assessment. Safety, health and working environment aspects are not considered. Environmental consequences due to power failures of battery systems in applications are not included.

Case studies were performed for portable and industrial rechargeable⁶ batteries. The studies on portable batteries (Papers I & II) include the following technologies: nickel-cadmium (NiCd), nickel-metal hydride (NiMH) (AB₅, AB₂,⁷) and lithium-based batteries (Li-ion: Co, Ni, Mn; Li-polymer: V). Eight different battery technologies for stationary energy storage were analysed regarding use in stand-alone photovoltaic systems: lithium-ion: Ni (Li-ion), sodium-sulphur (NaS), nickel-cadmium (NiCd), nickel-metal hydride AB₅ (NiMH), lead-acid (PbA) and three types of redox flow batteries (regenerative fuel cells): polysulphide-bromide (PSB), vanadium-redox battery (VRB) and zinc-bromine (ZnBr) (Papers III & V).

Depending on the particular battery application, different battery technologies may not be practically interchangeable with each other due to financial aspects and/or differences in technical performance characteristics (e.g. robustness, temperature range, etc.). However, in Papers II and V different technologies were assumed to be interchangeable in order to enable comparisons. In Paper II, metal flows arising from portable batteries are presented for the battery market 1999 (Case 1). In Case 2, it is assumed that different battery technologies replace the market share of NiCd batteries, and Case 3 represent a projected battery market for 2009.

In Paper I, the functional unit is defined as "a battery with an energy storage capacity of 1.0 Wh of electrical energy". The functional unit in Papers III and V is defined as "an electricity storage system with a power rating of 50 kW, a storage capacity of 450 kWh and an output of 150 kWh electricity per day".

A method of obtaining estimates of life cycle assessment data on materials in mechanical design is proposed and LCI data are presented (Paper IV). The life cycle of battery systems

⁶ Although the amount of portable non-rechargeable batteries (zinc-carbon) is three times that of portable rechargeable batteries in Europe, by weight (BIO 2003), they were excluded since no other alternative technologies are emerging. Automotive starter (SLI) batteries, in which approx. 50% of the globally mined lead is used (Paper III), were excluded since no alternatives are currently on the market.

 $^{^{7}}$ AB₅ alloy contains mischmetal, which is a mixture of lanthanides (rare earths). In AB₂ alloys, A stands for a hydride forming element (e.g. zirconium or titanium) and B other metals, such as Ni, Co, V, Mn, Al, Cr or Fe.

is analysed from cradle to grave. The geographical scope is global in Paper II and Sweden in Papers I and III. Life cycle inventory data are representative for 1995-2002 (Papers III & V). The temporal scope is 1998 in Paper I.

1.3 Overview of the appended papers

The appended papers focus on portable and industrial batteries. The methods used in the papers are given in the left column in Fig. 3. In order to identify and quantify environmental aspects of portable batteries, a life cycle inventory of portable nickel-cadmium batteries in Sweden was made (Paper I). Activities with significant impact were identified by varying the recycling rate of batteries and the different time boundaries for emissions of landfilled metals.

The use of portable rechargeable battery cells and their effects on global metal flows were assessed for three cases (Paper II). Based on the lithospheric extraction indicator (LEI), which is the ratio of anthropogenic to natural metal flows, and the significance of battery production related to global metal mine production, the potential environmental impact of metals used in different types of battery was evaluated.



Fig. 3. Structure of the research presented in the appended papers. Arrows denote information flows. Portable batteries were studied in Papers I & II, industrial batteries were studied in Papers III & V. Paper IV provided data for the study in Paper V.

The environmental impact of using vanadium redox batteries and lead-acid batteries in PV applications was evaluated by using LCA (Paper III). LCI data from Paper III were used in Paper V where the energy analysis of eight different battery technologies was performed, from cradle to grave, of a PV-battery system. Energy return factors and overall battery system efficiencies were calculated for different operating conditions and assumptions. The energy return factor was calculated by dividing the fossil energy replaced by operating the PV-battery system with the fossil energy required for the production of the PV-battery system.

Paper IV describes how life cycle assessment data for materials can be combined to give groups which are represented by average environmental values. Life cycle inventory data

for material groups (Paper IV) were then used to estimate energy requirements for the production of batteries (Paper V).

1.4 Outline of the thesis

The choice of different types of batteries and their applications are briefly described and the methods used for environmental assessment of battery systems are presented in Chapter 2. The methods described are substance flow analysis, energy analysis and life cycle assessment. The history, purpose and use of the results, methodology, and limitations of the different methods are discussed.

In Chapter 3, quantitative data on material and energy flows are presented for portable and industrial batteries. The influence of different parameters is analysed.

In Chapter 4, uncertainties in data and methodological choices in environmental assessment of batteries are considered. The relevance and the implications of the results are discussed. Recommendations for future research are made. The conclusions of the work are summarised in Chapter 5.

2 Method

2.1 Selection of type of battery for different applications

Different types of batteries have characteristics that make them more or less suitable in particular applications. This section provides a short overview of different rechargeable battery technologies and the requirements on batteries in some applications. Batteries can be categorised as portable if their mass is below one kg, otherwise they are regarded as industrial. Automotive lead-acid starter batteries (SLI) and batteries for traction applications are categorised as industrial batteries but they are outside the scope of this work.

Different types of portable rechargeable batteries include nickel-cadmium, lead-acid, nickel metal-hydride, lithium-ion and lithium-polymer batteries. The market for portable batteries has been dominated by sealed NiCd batteries for many years (Fig. 1). In applications of portable electronic products (footnote 2, page 1) consumers are willing to pay for batteries with low weight and high energy storage capacity, which has led to the emergence of NiMH and Li-ion batteries. Sealed portable NiCd batteries are used particularly in applications with high discharge rates, temperature extremes and where long cycle life is required, e.g. in power tools and emergency lighting. The financial cost of portable NiCd batteries is lower than for NiMH and Li-batteries, which makes them attractive in portable consumer products. Portable sealed lead-acid batteries are used as back-up power supplies in electronic products.

In battery applications for load-levelling, power quality, UPS and stand-alone systems, the European market is dominated by lead-acid batteries (97%) and the rest is made up of NiCd batteries (BIO 2003). Industrial NiCd batteries have higher investment costs than PbA, and NiCd batteries are used in niche applications where the batteries are exposed to mechanical strain, temperature extremes, limited maintenance and in applications with high reliability requirements e.g. offshore oil rigs, lighthouses and aviation. Several battery technologies suitable for stationary energy storage are emerging, including high-temperature sodium-sulphur batteries and redox flow batteries based on redox couples of, e.g. sulphur-bromide, zinc-bromine and vanadium-vanadium. The PSB, VRB and ZnBr batteries are based on liquid electrolytes which are pumped into the battery stack. The size of the stack determines the power rating and the volume of the electrolyte determines the storage capacity. Large battery systems can be constructed from Li-ion and NiMH cells, but their relatively high production costs have led to their being seldom employed in applications for storage of several hundreds of kWh.

The choice of battery technology from an environmental perspective should be assessed for each specific application. In particular applications only one type of battery technology can meet the demands. The environmental impact arising from the use of a battery system must be related to the avoided environmental effects that could be the consequence of a power failure, e.g. interruption of industrial processes and traffic accidents. However, the use of electricity from the PV-battery system is outside the scope of this work. The influence of the performance characteristics of different types of batteries on energy use is analysed in Paper V. It is assumed that the different technologies can be designed to meet the battery requirements of a PV-battery system in Paper V.

2.2 Selection of method

The choice of method for environmental assessment is determined by the *information needs* and the *resources available* (Wrisberg et al 2000). The information needs related to the actions that will follow, e.g. strategic planning, capital investments, design and development, communication and marketing, and operational management. The information needs in this study are dictated by different decision situations depending on the user, which may range from policy decisions to practical technical changes of a battery design. Available resources include: data, time, knowledge available to apply the method, the number of people involved, software and data processing resources. Data availability is a crucial aspect when assessing emerging technologies for which limited amounts of data are available.

Substance flow analysis (Paper II), energy analysis (Paper V) and life cycle assessment (Paper I, III) were applied in this work to assess the potential environmental impact of battery systems. The methods were chosen in order to focus on different environmental aspects and geographical scales of battery applications (Bouman *et al.* 2000).

Substance flow analysis (SFA) was used in Paper II to assess whether or not a technical option could solve a problem in principle on a macro level. Energy analysis was applied in Paper V since battery systems require energy for production and in order to function, and the use of energy is coupled with resource use and emissions. Life cycle assessment was used in Papers I and III in order to identify significant environmental aspects of different battery technologies. Compared with other methods (e.g. material flow analysis and energy analysis) it includes potential environmental impact connected to both material and energy flows, and it can be used to determine whether decreased emissions or resource use are shifted to other environmental problems.

To allocate resources to areas where the greatest benefits can be gained, parameters or activities with major influence should be identified (the Pareto principle⁸). In environmental assessment, several different physical parameters have to be compared. In order to do this, physical flows can be compared with some kind of reference values. One way of doing this is to relate to assessments of social or political priorities for various forms of recognised problems. Reference values can also be related to natural science, e.g. by relating a substance to biogeochemical cycles, eco-toxicity, reserve-to-use-ratio etc. In LCA, environmental impact assessment seeks to identify and quantify the most important resource uses and emissions. Different characterisation and weighting methods can be used for this purpose. Weighting methods for environmental impact assessment can be based on, for example monetary values, expert panels, distance to political targets, or critical environmental load (Bengtsson 2000).

⁸ The Pareto principle, also known as the 80-20 rule, or "vital few and trivial many", states that a small number of causes (20%) is responsible for a large percentage (80%) of the effect (Chen *et al.* 1994). In 1906, Italian economist Vilfredo Pareto created a mathematical formula to describe the unequal distribution of wealth in his country, observing that twenty percent of the people owned eighty percent of the wealth. Pareto has also formulated the economic definition of Pareto efficiency "Resources are allocated (Pareto) efficiently when no person can be made better off without some other person being worse off" (Fischer *et al.* 1988).

Limited data availability on battery system performance and metal toxicity may force the use of a simplified method for environmental assessment. In Paper II, substance flow analysis was chosen for the evaluation of emerging technologies since the use of metals in batteries is of environmental concern. The ratio between anthropogenic and natural flows gives an early indication as to whether an activity may cause environmental impact. The method provides a rapid means of analysis with small data needs. The focus is early in the cause-effect chain and gives an early warning of the potential impact of a new technology.

Energy use in batteries is not significant in comparison with energy use in other sectors of society (Paper I). However, the use of energy is related to significant environmental impact throughout the battery life cycle (Paper III). Depending on the source of energy and conversion efficiency, emissions and resource use may differ significantly. Since the generation of net energy is an important purpose of large PV systems, it is important to consider the effects on energy flows when PV systems are used in combination with different battery technologies (Paper V).

In the study on recycling portable NiCd batteries (Paper I), a life cycle inventory was made since the potential environmental impact is strongly related to metal emissions and energy use. Metal use in batteries causes a significant addition to the overall societal and natural flows of cadmium and lead (Papers II and III, Rydh 1999a). According to impact assessment weighting with the ET and EPS methods, the potential environmental impact of these metals is greater than any other emissions or resource use (Paper III). The use of primary metal resources is strongly connected to energy use (Papers III & IV). LCA was applied in Paper III to provide a comprehensive overview of different environmental aspects related to the use of battery systems. In Paper IV, the weighting of material inventory data was used as one criterion for grouping.

The following sections give an overview of different battery types and applications and a description of the methods regarding (1) their historical development, (2) their purpose and how the results can be used, (3) the principles of the method, (4) the specific methods used for the assessment of batteries and grouping of materials, and (5) limitations of the method.

2.3 Substance flow analysis

2.3.1 History

The principle of substance flow analysis (SFA) is based on the Lavoisier's law of mass conservation which was formulated in 1789 (Lavoisier 1789). Early applications of substance flow analysis can be found in ecology and studies of nutrient budgets (Lotka 1924). Later examples are studies of biogeochemical cycles, the availability of resources and pollution problems. During the past decade there has been a certain development towards a general methodology of SFA (van der Voet 2002, ConAccount 2003).

2.3.2 Purpose and use of results

Material flow analysis aims at specifying the pathways of materials in, out and through the technosphere of a given region and over a given period of time in order to provide relevant information for overall management strategy. The method can be used to trace the direct causes and origin of certain emissions and the fates of accumulated stocks. Two main complementary approaches exist: material flow analysis (MFA) and substance flow analysis (SFA).

MFA includes flows of materials (chemical compounds e.g. coal, wood, plastics; and products e.g. batteries) to direct priorities for policy measures towards increased resource efficiency. MFA follows a cradle-to-grave approach for all processes connected with the material flow studied. Material flows may be traced beyond the system boundary in order to detect unexpected side effects. (Wrisberg et al 2000)

SFA includes the assessment of a single substance, or a group of substances, that is associated with specific environmental effects. The relationship between industrial metabolism and environmental effects allows quantitative cause-effect modelling. The results of SFA may be linked to environmental risk assessment methods. SFA only considers the flows and accumulation in the technosphere in as far as they are connected to the substance under study.

2.3.3 Method

Quantitative relationships for substance flows are established based on mass balance principles. No standardised method has been defined, but the general procedure includes the following steps (van der Voet 2002): (1) definition of system boundaries (space, function, time, materials), (2) quantification of stocks and flows, and (3) interpretation.

Different modelling techniques for material flow analysis include (van der Voet 1996) bookkeeping, and static and dynamic modelling. In bookkeeping, flows and stocks are registered in order to identify trends and evaluate steering effects. Static modelling is based on linear equations that are used to describe steady-state relations between stocks and flows. This modelling technique can be used to the trace origins of pollution problems and to estimate the effectiveness of abatement measures. Dynamic modelling includes time as a modelling parameter, which makes it possible to predict future situations and the effects of polices. Future situations can be modelled with scenarios where the fate of accumulated stocks is estimated based on leaching (emission factors) or delay in the technosphere (residence time).

2.3.4 Indicators for assessing impact on global metal flows arising from the use of portable rechargeable batteries

The aim of this method is to provide a simple assessment of technologies when only few data are available. It has its focus early in the cause-effect chain and detects potential problems rather than actual problems. An early indication regarding metals of concern can be used to direct research and further actions. Here the method is used to assess potential effects on global metal flows arising from different market cases of portable batteries.

The potential environmental impact resulting from the use of metals in batteries is assessed from the mobilisation of metals. Lithospheric extraction indicators (LEIs) are calculated for each metal as the ratio between the anthropogenic metal flow and natural metal flow (Lantzy and Mackenzie 1979, Benjamin and Honeyman 1992). LEIs and mine production for the year 1999 were used as the reference, and all changes due to battery use were relative to these values. The lithospheric extraction indicator, i_E , is calculated from

$$i_E = \frac{F_A}{F_N} \tag{1}$$

where F_A is the annual metal flow from mine production and the mobilisation of metals due to combustion of fossil fuels (kton/y) and F_N is the natural turnover of metal due to weathering and volcanic activity (kton/year). The calculated LEIs indicate potential problems⁹. They do not represent absolute values of environmental impact, but provide a simple system for environmental ranking of different metals.

By assuming that the use of metals in batteries influences the amount of metals mined annually, while other metal use is constant, the LEI at a certain time, t, and for a certain metal can be calculated from

$$i_{E}(t) = \frac{F_{A} + F_{B}(t) - F_{B}(t_{0})}{F_{N}}, \qquad F_{B}(t) \ge F_{B}(t_{0})$$
(2)

where $F_B(t)$ and $F_B(t_0)$ are the virgin metal flows due to battery production (kton/year) in a specific year, *t*, and a reference year (t_0 = 1999), respectively. The metal flow may end up in products or be emitted to air, soil, water or sediment. The fraction of recycled metal used in batteries, *R*, reduces the metal flow required for manufacturing of batteries, F_B gross (kton/year), resulting in the net metal flow

$$F_B = F_{B \text{ gross}} \cdot (1 - R) \,. \tag{3}$$

The metal demand for the manufacture of a specific battery, $F_{B \text{ gross}}$, is calculated from $F_{B \text{ gross}} = n_{Cells} \cdot m_{Cell} \cdot c_{Metal}$ (4)

where
$$n_{Cells}$$
 is the number of batteries manufactured annually (cells/year), m_{Cell} is the mass of a battery cell (g/cell), and c_{Metal} is the metal content of a battery (wt%).

To calculate the metal demand for an equivalent energy capacity (Case 2 in Paper II), different energy densities and cycle lives were considered for the various battery technologies in order to make the battery technologies comparable.

To compare the magnitude of battery metal flows to other anthropogenic and natural flows three complementary indicators where calculated: i_N compare battery metal flows to natural flows

$$i_N = \frac{F_B}{F_N} \tag{5}$$

 i_A is the contribution of batteries to anthropogenic flows

⁹ If LEIs should also be an indicator of actual future environmental impact the following general assumptions must apply: (1) all mined metals will be released to the environment in the future due to geological processes, (2) anthropogenic emissions of metals are distributed in the same way as naturally occurring elements and have the same degree of bio-availability, (3) organisms and ecosystems have adapted to natural background concentrations of metals and would be harmed by increased levels.

$$i_A = \frac{F_B}{F_A},\tag{6}$$

and i_M is the fraction of mine production (F_M) used for batteries

$$i_M = \frac{F_B}{F_M} \,. \tag{7}$$

With the equations above, the change in anthropogenic mobilisation of metals for the cases concerning different battery types was calculated. The values for 2009, $i_E 09$, (Eq. 2) were related to the values for reference year 1999, $i_E 99$, and the change compared with the reference year is given as $((i_E 09 - i_E 99)/i_E 99)$.

Metals of special interest were identified according to: (1) i_E or $i_N > 1$ and/or (2) i_A , or $i_M > 1\%$. The value of LEI was chosen as an arbitrary limit to distinguish between low and high impact on natural flows. A value of $i_E > 1$ indicates that the anthropogenic use of a metal exceeds the natural turnover and thus indicates a risk for substantially increased metal concentrations in the environment and thus increased environmental impact.

High values of i_E and i_N imply that the use of the metal in batteries may result in increased background concentrations. If i_E is high but i_N is low for a particular metal, the impact of batteries is low but some concern is warranted, since uses in other products may slander the metal, which may have consequences for its use in batteries.

If i_A is high, the use of the metal in batteries has the potential to significantly influence anthropogenic metals flows. A high value of i_M means that the demand for metals in batteries may have substantial influence on the mining of that metal.

2.3.5 Limitations

Indicators of metal flows can be used for the identification of metals that may warrant further investigation. Indicators of metal flows are only indicative, while actual emissions of metals are highly depended on the type of application. LEIs do not consider the chemical properties of different metals or their distribution between air, soil, water and sediment. LEIs do not assess actual toxicological effects or temporal aspects. Therefore, it can not be assumed that a high LEI for a certain metal will automatically result in high environmental impact, since some metals may have low toxicity or may be present as a chemical species that is not bio-available.

2.4 Energy analysis

2.4.1 History

During the 1960s, awareness of the risk of exhaustion of fossil fuels and the scarcity of metal resources, as well as increasing environmental pressure society increased. This resulted in need to describe the behaviour of industrial systems, resulting in energy analysis, resource analysis and environmental profile analysis. One of the earliest reports concerned some aspects of the chemical industry (Smith 1969). Energy and resource analysis were used in decision making which led to the implementation of legislation regarding the recycling of packaging materials.

At the height of the energy crisis during the 1970s, policies affecting the use of energy were implemented throughout the world (Spreng 1988). Net energy analysis (NEA) provides a method of assessing how much energy an energy system requires to maintain itself, and, was one of the methods that gained great prominence at that time. The US congress (1974) required every governmentally sponsored scheme for producing energy to be subjected to net energy analysis.

Net energy analysis is also known as energy analysis, full fuel cycle analysis (FFCA) and cumulative energy requirement analysis (CERA). The method of energy analysis has become standardised through the work of VDI (1997) and ISO 13 600 (1999).

2.4.2 Purpose and use of results

Energy analysis is used to quantify the primary energy requirement for products and services in a life cycle perspective. The primary energy requirement represents the sum of the energy required for production, use and disposal of a product. Energy analysis can be used to (Spreng 1988): (1) quantify the energy requirement of products, services and national economies, (2) analyse options for energy savings in industry, and (3) provide energy requirement data for base materials to support engineering and construction of products.

2.4.3 Method

An inventory of energy flows is conducted within the life cycle from cradle to grave for a product. Primary energy is defined as the energy content of energy carriers that have not yet been subjected to any conversion (Boustead and Hancock 1979). Converted energy can be divided into the categories thermal, electrical and feedstock energy. Feedstock energy accounts for the heating value of an input material if this material may also be applied as an energy carrier. Primary energy is assumed to be the heating value for fuels (gross calorific value i.e. the combustion enthalpy).

The primary energy requirements are made comparable and are aggregated into one number by ensuring homogeneity between different energy qualities. This is achieved by multiplying energy of different qualities with specific conversion efficiencies (see page 18). The amounts and types of primary fuels used for the production of secondary energy forms will be quite different for each secondary form, but may also differ between production sites. This means that not only conversion efficiencies but also environmental effects will show large variations. To improve the transparency of energy analysis, Fritsche *et al.* (1999) suggested that the total result should be presented for different energy sources: $E_{Non-renewable}$ (including nuclear and fossil fuels), $E_{Renewable}$, (renewable fuels), E_{Others} (energy of rest products) and E_{Total} , (sum of all components).

As for all methods described in this chapter, the choice of system boundaries and allocation principles is important to ensure a reliable assessment. The system boundaries can be defined for different orders, where the first order includes direct energy, the second order also includes energy for materials production, the third order also includes the energy required for capital equipment and the fourth order also includes the energy required for make the capital equipment (Boustead and Hancock 1979).

If a process has multiple outputs, the inflow of energy or material must be divided between them. Within the development of LCA, the following allocation principles have been proposed (ISO 14 041:1998): (1) regard only one main product, (2) expand the system to

avoid allocation, (3) allocate on mass basis and (4) allocate on the basis of economic value or other parameters describing the quality of each product.

2.4.4 Energy efficiency indicators for batteries in photovoltaic systems

Energy efficiency can be defined in many ways, but in all cases it is a measure of the amount of energy inputs that is needed to provide an energy service (output). Two complementary indicators for the calculation of energy efficiency are presented, the *energy return factor* and the *overall efficiency*.

Direct and indirect energy requirements

The gross energy requirement, E_G , of an energy conversion device with an energy output E_O can be decomposed into two parts (Fig. 4): the direct input, E_D , of energy during operation and the indirect energy requirement E_I , i.e. the energy required to produce the device and transport it to the site of operation.

$$E_G = E_D + E_I \tag{8}$$



Fig. 4. The general energy balance for an energy conversion device.

From these energy flows three measures of energy efficiency can be calculated: the direct (or normal) energy efficiency of the device

$$\eta = \frac{E_O}{E_D}, \qquad 0 < \eta < 1 \tag{9}$$

the overall energy efficiency,

$$\eta^* = \frac{E_O}{E_G}, \qquad 0 < \eta^* < 1 \tag{10}$$

and the energy return factor

$$f = \frac{E_o}{E_I}. \qquad f > 0 \tag{11}$$

Energy return factor

The most common way to describe the energy balance of a PV system (or any other energy flow conversion technology) is by calculating energy payback times, t^* (Alsema 1998). A diesel engine with a generator continuously converts fossil fuel into electricity. To produce a PV array and a battery that could replace the diesel generator requires energy (assumed to be fossil energy), while only solar energy will be used during operation (Fig. 5). After a certain time, the energy payback time, the energy that was used to produce the PV-battery system will have been paid back by not using the diesel generator.



Fig. 5. The energy flows of the PV-battery system and the reference system (the diesel generator).

Both systems have the same output, E_{use} (MJ_{el}/year). The average annual gross primary fossil energy use of the diesel system, E_{G0} (MJ_{pf}/year) is calculated from

$$E_{G0} = \frac{E_{use}}{\eta_0^*} \tag{12}$$

where, η^*_{0} , is the overall efficiency of the diesel generator. The energy payback time is then calculated from:

$$t^* = \frac{Q_{pf}}{E_{G0}}$$
(13)

where Q_{pf} (MJ_{pf}) is the energy (Q) given as primary fossil energy that is required to produce the PV-battery system.

The energy return factor, f, ¹⁰ is then the ratio between the service life of the PV-battery system t (year), and the energy payback time or, to put it differently, the ratio between the

¹⁰ The net energy (output) is defined in some studies as E_O-E_I , in relation to the indirect energy requirement (E_O-E_I)/ $E_I = f-1$ or to the energy output (E_O-E_I)/ $E_O = 1-1/f$. The indirect energy requirements can also be expressed as percentage of the energy output, $E_I/E_O = 1/f$.

replaced fossil energy (diesel) and the fossil energy required to produce the PV-battery system:

$$f = \frac{t}{t^*}, \qquad f = \frac{E_{G0} \cdot t}{Q_{pf}} = \frac{E_{G0}}{E_{I,pf}}$$
 (14a, 14b)

where $E_{I,pf}$ (MJ_{pf}/year) is the average annual energy required to produce and replace the PV-battery system. Eq. 14b is a version of Eq. 11 where the energy flows have been transformed into a common energy "currency". Since the service life of the components in the PV-battery system differs, the meaning of an energy payback time becomes ambiguous and the energy return factor is then a better measure.

Overall battery efficiency

The overall efficiency of the battery system, η_B^* , is the ratio between the output from the battery system, E_{use} (MJ_{el}/year), and the sum of the electricity input to the charger from the PV array and total inputs for production and transportation of the charger, the battery and inverter transformed into an electricity equivalent, E_G (MJ_{el}/year) (see Paper V, Fig 4).

$$\eta_B^* = \frac{E_{use}}{E_G} \tag{15}$$

When calculating the energy return factor the solar electricity was implicitly regarded as an abundant free resource. When calculating the overall efficiency the electricity input is seen as the scarce resource worth saving.¹¹ The overall efficiency could be a relevant measure of efficiency in a closed solar energy system. For the designer of a PV-battery system the direct efficiency of the battery system is of interest. A large direct efficiency would save resources (materials, energy, capital and labour) used to produce the PV-system and space taken up by the PV-arrays. In a world that relies largely on solar energy, the battery system must also be produced from solar energy (we can no longer borrow fossil fuels to build the system). Thus, more PV-systems (or other solar energy technologies) would have to be produced. The total electricity input can thus be interpreted as the output from PV arrays at the site and from PV arrays producing electricity that is used to produce and transport batteries. The closed solar energy system is just an example. The overall efficiency measure is valid for electricity produced from any energy source.

Energy quality and conversion factors

The fact that energy may take different forms poses a problem. The gross energy requirement and the indirect energy requirement are normally made up of many different kinds of energy inputs. To be able to define single measures for the overall efficiency or the

¹¹ The overall efficiency of the PV-battery system can be calculated by taking into account the direct efficiencies and indirect energy requirements of the PV array. This would then become a measure of how efficiently the solar irradiation is used. However, the focus is on battery performance in this work.

energy return factor, different energy forms need to be converted to a common energy currency.

In the calculation of the energy return factor, primary fossil energy was used as currency. The energy replaced by the PV-battery system (diesel) is derived from primary fossil energy and the available data for indirect energy requirement is given in terms of primary energy (implicitly assumed to be fossil). However, this is not the only possible choice. There are for example great differences between coal, oil and gas (e.g. with regard to CO_2 emissions or resource availability). However, in principle, the diesel oil saved could be used for the heat, electricity and motor fuel production required for the production and transport of the PV-battery system.

Since the direct energy input and the output of the battery system are electricity, electricity is used as the energy currency for the calculation of the overall efficiency. The energy requirements for production and transport given in the currency primary fossil must be converted into electricity equivalents. To do this, it is assumed that electricity and thermal energy are used for the production of the components and transport fuel is used for transport. The energy requirement for producing and transporting component *i* is calculated from the gravimetric energy requirement for production $q_{P,i,pf}$ (MJ/kg), and transport to the site of operation $q_{T,pf}$ (MJ/kg):

$$q_{P,i,el} = q_{P,i,pf} \cdot \left(\beta_i \frac{\alpha_{pf,el}}{\alpha_{el,el}} + (1 - \beta_i) \frac{\alpha_{pf,th}}{\alpha_{el,th}}\right)$$
(16)

$$q_{T,el} = q_{T,pf} \cdot \frac{\alpha_{pf,tr}}{\alpha_{el,tr}}$$
(17)

where the conversion factors α are given in Table 1. The factor β_i is the estimated proportion of primary fossil energy used to generate the electricity used in the production of component *i*.

Table 1 shows that the average conversion efficiency for electricity generation from fossil fuels is estimated to be 0.35. Losses in distribution and conversion of primary fossil fuel to thermal energy result in a conversion efficiency of 0.95 (NTM 2003). The efficiency for refining and distribution of primary fossil fuel to diesel for transportation is 0.88 (Ahlvik and Brandberg 2001).

When PV generated electricity is used to produce the PV-battery system, electricity has to be converted into thermal energy and fuels for transportation (diesel or hydrogen), since different forms of energy are required in the production processes. Two extreme cases are investigated for the conversion of solar electricity, the *Reference case* and the *All PV case*.

In the *Reference case*, the PV-battery system is open to other fuels and the electricity produced can be used to replace a certain amount of primary fossil energy, which in turn can be used to produce heat and transport fuel. When electricity replaces the need for the combustion of fossil fuels for electricity generation, the conversion factor for electricity to heat is 2.71 ($\alpha_{el, th}$ = 0.95/0.35). For transportation, the primary fossil fuel includes 12% losses for refining and distribution of diesel fuel (Ahlvik and Brandberg 2001), resulting in an energy conversion factor of 2.51.

The *All PV case* corresponds to a closed renewable energy system where solar electricity cannot be traded and has to be used directly to produce heat and a transport fuel. Electricity is then used directly for the generation of high temperature thermal energy (heat pumps not considered) with the conversion factor 1. Transportation in a renewable energy system may be based on vehicles powered by fuel cells and electrical motors where hydrogen is used as a motor fuel. The conversion efficiency of electricity to hydrogen by electrolysis of water is estimated to be 80% and the efficiency in the distribution of hydrogen fuel is 80%. A fuel cell vehicle could thus be 33% more energy efficient per ton km transported goods than a conventional diesel truck (Ahlvik and Brandberg 2001), resulting in a conversion factor of 0.85.

Tabla	1	Engran	o o mo o mai o m	fastona
Table I	1.1	energy	conversion	jaciors.
		· · · · ·		

	Conversion from primary fossil energy			Conversion from electricity		
				Reference case	All PV case	
Electricity	$\alpha_{pf, el}$	0.35	$\alpha_{el, el}$	1.0	1.0	
Thermal energy	$\alpha_{\rm pf, th}$	0.95	$\alpha_{el, th}$	2.71	1.0	
Transport fuel	$\alpha_{\rm pf, tr}$	0.88	$\alpha_{el, tr}$	2.51	0.85	
C						

Source: Paper V

2.4.5 Limitations

The energy requirement indicates a basic environmental pressure associated with the use of energy. As in the case with material flows, the energy requirement cannot be used to quantify specific environmental effects (e.g. ozone depletion) without characterisation of inventory data.

Energy analysis suffers from limitations, as do other environmental systems analysis methods, in that it is difficult to confirm that all significant impact is within the system boundary definition. The principles for allocation of energy between joint products should be justified. A serious problem lies in the aggregation of energies of different qualities into a single homogeneous entity (Chwalowski 1996). The effects of choice of system boundaries, allocation and energy qualities in the energy analysis of PV modules are discussed by Alsema (1998).

2.5 Life cycle assessment

2.5.1 History

Local environmental problems caused by human activities, e.g. smog, and acid rain, showed that international measures were also required to curb such problems. In the 1980s the potential threats of global warming and ozone depletion added to the need to consider emissions to air and water. Initially, end-of-pipe measures were employed to decrease environmental impact but the focus later shifted to the analysis and improvement of products. The methodology for evaluating emissions from a product system is similar to that for calculating energy requirements (Section 2.4.3). Energy analysis was therefore expanded to include more environmental data categories in the calculations. The method has been described as *ecobalance, ecoprofile, cradle to grave analysis, life cycle analysis* and *life cycle assessment*, all essentially describing the same type of approach. At a
workshop in 1990, SETAC (1991) recognised the need to describe the causal link between inputs and outputs of a product system and the potential environmental impact. The method was divided into three phases, namely inventory, interpretation and improvement. The method of life cycle assessment has become standardised within ISO 14 040-14 043 (ISO 1997, 1998, 2000).

2.5.2 Purpose and use of results

The goal of LCA is to present a comprehensive evaluation of flows of material and energy throughout the life cycle of a product. LCA can be applied to hot spot identification in product systems, product development, product comparisons, green procurement and market claims. It can also be used on the strategic level for business or policy development. LCA is a comprehensive method that covers the product chain from cradle to grave. LCA is normally used to assess the flows that cause global and regional environmental effects, such as global warming and acidification. In principle, it can include all material flows, and even potential toxicological impact can be assessed.

2.5.3 Method

LCA is a method that aims at analysing and evaluating the environmental impact of products or services. The whole chain of activities required for the production of a certain product or service is taken into consideration. Both emissions of potentially harmful substances from these activities and their consumption of natural resources are analysed. In this way, different technical systems producing a comparable utility (product or service) can be followed from cradle to grave (from extraction of natural resources, transport, production, and use, to recycling/disposal), and can be compared with regard to their impact on the environment.

LCA is described as a four-step procedure (ISO 14 040, 1997) including the following steps:

- *Goal and scope definition*, in which the purpose of the study is presented and the system boundaries are defined. The functional unit, i.e. the basis for comparison, is defined.
- *Life cycle inventory* is the phase in which information on the emissions and the resource consumption of the activities in the system is collected from various sources. Relationships between different activities are identified. Where reliable data are unavailable, assumptions may have to be made.
- *Life cycle impact assessment* is the phase in which the environmental consequences of the inventory data are assessed. Characterisation and/or weighting methods are used for aggregation of inventory data.
- *Interpretation* of data is carried out for all phases. Often, some kind of sensitivity analysis or discussion of uncertainties is included in this step.

A further description of the LCA methodology is given by Rydh *et al.* (2002), Guinée (2002) and Baumann and Tillman (2004).

2.5.4 Specific method - grouping of life cycle inventory data

Compilation of quantified inventory data for LCAs may be difficult since information about products may be confidential or unknown, particularly in the case of emerging

technologies. Average inventory data (Paper IV) can be used as estimates when specific data are unavailable (Paper V, Rydh 2003). The more specific the material data that can be used in an analysis, the more accurate the result will be. Little specific data on materials are available early in the design phase, and thus data representative of average materials can be used.

Grouping of materials and identification of the number of material groups that can be used to present average environmental data with low standard deviation (SD) have been presented in a previous study by Sun *et al.* (2003). The materials were grouped to increase data availability for product designers. The criteria for grouping were based firstly on environmental properties and, secondly, on the mechanical and physical properties of the materials. Cluster analysis and scatter plots of weighted environmental impact were used to identify groups of different materials. Data consistency for the LCIs of material was ensured regarding the system boundaries and allocation principles used (Sun *et al.* 2003). When a particular group of materials had a standard deviation greater than 30% of the group average, analysis of material properties (e.g. material composition or density) was conducted for further sub-division (Sun *et al.* 2003).

As a measure of overall environmental impact, LCI data for 407 different materials were weighted with the ECO'99 weighting method. They were initially arranged in 41 material groups according to the mechanical properties of the materials. The groups were based on classification and grouping by Ashby (1999). The further merging of material groups was based on similar environmental performance and affiliation to material classes.

The standard deviation (expressed as the coefficient of variance = SD/ average \cdot 100%) was calculated for ECO'99 scores for each group. The average standard deviation was calculated for each number of material groups. Fig. 6 shows that 17 groups have a relatively low standard deviation (22%), and further division into more specific material groups gives only a marginal improvement in accuracy. The five groups were composites/paper, metals, polymers, porous ceramics/glass and wood.



Fig. 6. Average standard deviation of ECO'99 weighted impact for different numbers of material groups. (Sun et al. 2003)

2.5.5 Limitations

An advantage of LCA is that it is a standardised method, which means that the method is known and accepted. A broad perspective is considered in LCA, which makes it possible to avoid sub-optimisation and problem shifting from one stage in the product life cycle to another.

The limitations and drawbacks of LCA are mainly due to its broad scope. Because all processes and environmental consequences have to be specified, LCA has extensive data requirements which, in practice, often cannot be fully met. The considerable need for data tends to require the involvement of many organisations. The high degree of detail and consideration of methodological issues requires expert knowledge and a great deal of time. To assess the influence of variations in data, uncertainties in LCA are dealt with by sensitivity analysis. Methodological choices such as allocation procedures and decisions on system boundaries may alter the outcome of the study.

3 Results

In this chapter, quantitative data on energy and material flows are presented for portable and industrial batteries, and the influence of various parameters is analysed.

3.1 Portable batteries: energy and material flows

3.1.1 Effects of battery recycling

The results of the life cycle inventory of portable NiCd batteries include primary energy, emissions of CO_2 , SO_x and NO_x , and metal flows of nickel and cadmium. When excluding the user phase, the greatest energy use is in battery manufacturing. Fifty-five per cent of the CO_2 emission originates from battery manufacturing, 44% from raw materials production and 0.8% from battery distribution, collection and sorting. Transportation in the materials production phase contributes 1% to the total CO_2 emission. Consumption of metal resources is ascribed to unit processes, which disperse concentrated metals and make them difficult to recover. Consequently, the greatest resource use is found in incineration and landfilling (Table 2).

Model simulations of the NiCd battery life cycle shows a minimum at 90% recycling rate for energy use and NO_X emissions (Table 2). This minimum is due to the fact that recycling materials and longer transportation distances have less impact than extraction and refining of virgin materials. At recycling rates greater than 90%, local transport resulting from emptying collection boxes and delivery of batteries to sorting plants is modelled to increase rapidly.

The use of energy decreases by 13% when increasing the recycling rate from 25% to 90%. The difference between 25% and 90% recycling corresponds to a decrease of approximately 87% in cadmium and nickel resource use and emissions. CO_2 and SO_x emissions decrease by 30% and 80%, respectively. Extraction and refining of virgin nickel give rise to high SO_x emission, which decreases significantly at higher recycling rates.

	Landfill. 100%	Incineration 60%	Recycling 90%	Recycling 100%
		Landfill. 40%	Incineration 6%	
			Landfill. 4%	
Renewable energy (MJ/Wh)	0.16	0.16	0.14	0.14
Non-renew. energy	5.18	5.15	4.29	4.32
(MJ/Wh)				
CO ₂ (kg/Wh)	0.41	0.41	0.26	0.26
$NO_x (g/Wh)$	0.56	0.56	0.34	0.47
SO_x (g/Wh)	5.45	5.45	0.83	0.32
Cd (resource) (g/Wh)	4.1	4.1	0.41	0
Ni (resource) (g/Wh)	5.1	5.1	0.51	0

Table 2. Selected inventory data for the NiCd battery life cycle (excluding user phase) for different end-of-life treatment methods.

Source: Paper I

As shown in Fig. 7, an increase in recycling rate from 0 to 90% decreases the total primary energy use by 17%. Expressed as a percentage of the total energy use, the energy required for collection and sorting increases from 0.6% to 5%, while energy use in raw materials production decreases from 36% to 15%. By using recycled metals, the energy required for the processing of raw materials used in batteries is reduced by 65% compared with virgin materials only. Energy use in battery manufacturing remains constant irrespective of the recycling rate.

Energy flows in the user phase of rechargeable batteries varies significantly depending on the way of use (Rydh 2001). For a battery and charger, the energy requirement may be 2-32 times higher during use than the energy for manufacturing of the battery. The energy efficiency of the charger and it no-load loss plays an important role in the overall energy use (Rydh 2001).



Fig. 7. Primary energy use in the NiCd battery life cycle (excluding user phase) at different recycling rates (Paper I).

The method of electricity generation may alter the absolute values of primary energy use. If all electricity is generated by hydropower, the total primary energy will be 2.1-2.8 MJ/Wh and NO_x emission 0.17-0.31 g/Wh. Corresponding values for coal power are 8.4-10 MJ/Wh primary energy and 91-107 g NO_x/Wh. Compared with the country-specific electricity mix, primary energy use is reduced by half or doubled, due to the different energy conversion efficiencies of the different power sources. Assuming that electricity is generated by hydropower, the contribution of energy required for transportation increases to 10% of the total energy use at 100% recycling. However, recycling is still energy efficient.

To quantify the energy savings made by using secondary materials, the total energy required for recycling should be allocated (Section 2.4.3) between the nickel and the cadmium recovered. Allocation on a mass basis is preferable to economic allocation since physical parameters are constant. Economic values of recovered metals fluctuate over time

and do not follow open market prices since the Swedish battery fund pays for the recycling of NiCd batteries. Boustead and Hancock (1998) used mass allocation when calculating the energy required to produce cadmium as by-product from zinc production. Considering the primary energy expended in the collection and recycling of 1 kg of cadmium (mass allocation Cd/Ni= 44/56), 19 MJ is needed for transportation and 19 MJ for the recycling process. Corresponding values for 1 kg nickel are 20 MJ for collection and 20 MJ for the recycling process. Compared with the extraction and refining of virgin metal, 54% and 75% less primary energy is needed to recycle cadmium and nickel, respectively. Economic allocation may be used if physical relationships cannot be established between co-products (ISO 14 041, 1998). If economic allocation is used, the calculated result show 90% and 59% lower primary energy requirement to recycle cadmium and nickel, respectively, than for virgin metal production (Cd/Ni= 7.6/92, metal prices USD 98/kg (USGS 1999, LME 2000)).

3.1.2 Impact on global metal flows arising from the use of portable rechargeable batteries

Case 1-3: Summary of metal flows of portable batteries

Based on the material composition of portable batteries and their mass introduced on the global market annually, the total amounts of metals used in batteries were calculated and related to anthropogenic and natural metal flows (Eqs 1-7). Table 3 shows that the average LEIs range from 0.07 to 5.6 for the different metals used in batteries. Bold numbers indicates values of particular interest.

Changes in the metal indicators for the different cases are exemplified for nickel and cadmium due to their high LEIs, and for cobalt because of batteries high contribution to global mining demand. All these three metals are used in the different battery technologies. Fig. 8 shows that the LEI for cadmium decrease 31% in Case 2, when assuming replacement of NiCd batteries with NiMH and Li-ion, respectively. At the same time, the LEIs increase 0.5% for nickel and 7% for cobalt. For the other metals studied, the LEIs of which increase by 0.1-7% compared with 1999, the LEIs do not exceed 0.7, which indicates low potential environmental impact of these metals.



Fig. 8. Lithospheric extraction indicators for Case 1-3 (Paper II).

Element	$i_E (F_A/F_N)$	$i_N (F_B/F_N)$	$i_A (F_B/F_A)$	$i_M (F_B/F_M)$	F_B (ktons/year)	F_A (ktons/year)	F_N (ktons/year)	F_M (ktons/year)
Ni	5.6	0.073	0.013	0.020	7.1 - 36 (22)	1670	300	1100
Cd	4.4	1.4	0.31	0.37	2.6 - 12 (7.3)	23	5	20
Li	0.65	0.0016	0.0025	0.038	0.14 - 1.0 (0.58)	235	360	15
V	0.33	0.0022	0.0066	0.065	1.4 - 3.7 (2.6)	390	1200	40
Zr	0.27	0.00002	0.00007	0.0001	0 - 0.13 (0.063)	960	3500	820
Co	0.18	0.0076	0.042	0.15	2.0 - 6.2 (4.1)	98	540	28
Nd	0.18	0.0013	0.0076	0.084	0.13 - 1.9 (1.0)	132	750	12
La	0.10	0.0030	0.031	0.095	0.19 - 3.0 (1.6)	52	540	17
Ce	0.083	0.0011	0.0013	0.044	0.11 - 2.5 (1.3)	100	1200	30
Pr	0.073	0.0019	0.026	0.094	0.042 - 0.60 (0.032)	12	170	3

Table 3. Indicators of metal use in portable batteries for 1999 (Case 1). (Average values)

Table 4. Indicators of metal use in portable batteries for 2009 (Case 3). (Average values)

Element	$i_{E09} (F_{A09}/F_N)$	$i_{N09} (F_{B09}/F_N)$	$i_{A09} (F_{B09}/F_{A09})$	$i_{M99} \left(F_{B09} / F_{M99} \right)$	F_{B09} (ktons/year)	F_{A09} (ktons/year)	F_{M09} (ktons/year)
Ni	5.7	0.18	0.031	0.048	19 - 88 (53)	1702	1132
Cd	3.4	0.37	0.11	0.097	0.70 - 3.2 (1.9)	18	15
Li	0.66	0.011	0.017	0.27	1.0 - 7.0 (4.0)	238	18
V	0.34	0.015	0.044	0.44	9.9 - 26 (18)	405	55
Zr	0.27	0.00002	0.00009	0.0001	0 - 0.17 (0.085)	960	820
Co	0.21	0.039	0.18	0.75	10 - 32 (21)	115	45
Nd	0.18	0.0041	0.023	0.25	0.38 - 5.7 (3.0)	134	14
La	0.10	0.0090	0.088	0.29	0.58 - 9.2 (4.9)	55	20
Ce	0.086	0.0033	0.039	0.13	0.34 - 7.6 (4.0)	103	33
Pr	0.077	0.0057	0.074	0.28	0.13 - 1.8 (1.0)	13	4.0

The use of cadmium in portable batteries only, results in an LEI of 1.4 (Fig. 9). Consequently, a change of the use of NiCd batteries has significant influence on the total LEI of cadmium. Nickel and cobalt have low impact only on the ratio and a replacement of NiCd batteries (Case 2) indicates low potential impact for the other metals studied. The use of nickel in batteries has a relatively small influence (2%) on the nickel demand since the largest fraction of mined nickel is used in stainless steel production.



Fig. 9. Lithospheric extraction indicators, i_N , arising from the use of metals in portable batteries in Case 1-3 (Paper II).

Case 1: Metal flows of portable batteries in 1999

The contribution of battery metals to the total anthropogenic flow is 31% for cadmium and 4.2% for cobalt, while it is below 3.1% for the other metals (Table 3). This indicates that particularly the use of cadmium in portable batteries influences anthropogenic metal flows.

The metal demand for batteries can be obtained by relating the mass of battery metals to annual mining production (Eq. 7). Fig. 10, shows that metals contributing more than 1% to the global mining production are: Cd, Co, La, Pr, Nd, V, Ce, Li and Ni.



Fig. 10. Case 1: Metal demand for the manufacture of portable batteries as a percentage of global mining production in 1999. The bars indicate uncertainties in the demand of each battery technology. The lithospheric extraction indicator and the 1999 mining production for each metal are also given (Paper II).

Fig. 10 shows that cadmium in portable NiCd batteries constitutes 13-60% of the total cadmium mining production, corresponding to 2.6-12 ktons cadmium per year. The broad ranges represent uncertainties due to variations in average cell weight, energy density and assumed recycling rate (5-50% for NiCd and NiMH). Literature data indicate that approximately 70% of the cadmium mined is used in the manufacture of NiCd batteries (Plachy 2000). Of the cadmium used in batteries, 75-80% is used in the production of portable NiCd batteries, while the rest is used in industrial NiCd batteries (Plachy 2000). This means that the calculated high estimate (60%) of mining demand for cadmium is likely to correspond to the actual demand.

The major end use of nickel is in the steel industry. Portable batteries account for 0.6-3.3% of the end use. Of this, NiMH batteries make up 53% and NiCd batteries 35%. Cobalt use in batteries constitutes 7.2-22% of global Co mining production. Fifty-eight percent of this cobalt is used in Li-ion(Co) and 33% in NiMH(AB₅) batteries.

Case 2: Different battery types replace NiCd batteries

Changes in LEIs were calculated for cases in which NiMH(AB₅), NiMH(AB₂) or Li-based batteries, respectively, were assumed to replace NiCd batteries¹². It was also assumed that changes in demand for metals on the battery market would be reflected in corresponding changes in global mining production.

Replacement of NiCd batteries by other battery technologies would result in a decrease in LEI for cadmium from 4.4 to 3.0. The replacement of NiCd batteries by NiMH(AB₅) batteries would lead to the greatest increases in LEIs for La (4.1%), Pr (3.4%), Ce (1.7%) and Co (1.4%). Metal flows resulting from the introduction of NiMH(AB₂) batteries would result in increases in LEI for Co (0.6%), V (0.3%) and Ni (0.2%). Replacing NiCd batteries by Li-based batteries would result in an increase in LEI for Co (7.1%) and V (1.9%). The small increases in LEIs of metals resulting from a replacement indicate that the potential increase in environmental concentrations is low compared with background concentrations of these metals.

Case 3: Projected battery market in 2009

Metal flows were calculated for the projected battery market in 2009 in which the total number of cells has increased 2.2 times (Paper II). The assumed market shares are 6% for NiCd, 53% for NiMH and 41% for Li-based batteries. The assumed market share of Li-based batteries (+6.1 times compared with 1999) may increase the demand for mining of several metals (Fig. 11).

The LEI for cobalt would increase by 15% from 0.18 to 0.21. Li-polymer (V) batteries contribute 78% to the battery-related cobalt flow. For the other metals studied, the relative changes in LEI are below 6.3%. The LEIs (i_{E09}) for other metals are below 0.66 and the ratios of battery metal flows to natural flows (i_{N09}) are below 0.06, which means that their material flows would be expected to have only a slight impact on the global level.

A growing battery market shows that portable batteries may be an important end-use for Co, Nd, La, Ce, Pr and Li. Increasing demand for these metals may result in higher metal prices, which may limit the growth of Li-ion(Co) and NiMH(AB₅) technologies. Higher prices for metals used in batteries may create incentives for battery collection and recycling.

¹² Small sealed lead-acid batteries may be the most practical alternative to NiCd batteries in many applications (BIO 2003). To estimate the potential impact of the use of lead in portable batteries, indicators of metal use were calculated based on the following estimates: $m_{Cell}=50$ g/cell, $c_{Pb}=65$ wt%, R=30%, $F_A=3$ 085 ktons/year, $F_N=290$ ktons/year (Paper II, Table 4). **Case 1:** $n_{Cells}=0.3 \times 10^9$ (NRI 1999), $F_B=2.9$ ktons/year. **Case 2:** No. of cells (NiCd replacement) = 1.7×10^9 - 3.0×10^9 , F_B case 2=17-30 ktons/year. The ratios for battery metal flows to natural flows were calculated and found to be $i_{N99}=0.01$ and $i_{N Case 2}=0.06-0.1$. Small lead-acid batteries have no significant influence on global lead flows, since the greatest lead use is in SLI-batteries. However, the LEI for lead is very high ($i_E=11$, Paper II, Table 4) and all dissipative uses of lead should be avoided.



Fig. 11. Case 3: Projected metal demand of portable batteries in 2009 as a percentage of global mining in 1999. The bars indicate the uncertainties in demand of each battery technology. The lithospheric extraction indicator and the 1999 mine production for each metal are also given (Paper II).

Parameters influencing metal indicators for portable batteries

Important parameters influencing metal flows arising from the use of portable batteries are service life, energy density and recycling of metals, while the natural occurrence of metals influences the relative contribution of anthropogenic metal flows.

Development of battery technologies should aim at high energy density and long service life. Only one third of the metal is required for storing electrical energy with rare earths than with cadmium. The metal intensity is 1.2 kg/kWh for rare earths, 0.14-0.52 kg/kWh for lithium and 3.4 kg/kWh for cadmium (Råde and Andersson 2001). The turnover of batteries and metals decreases with increasing service life.

Metals with high natural occurrence should be used to decrease their relative contribution to natural flows. For example, the natural occurrence of rare earths ¹³ in the earth's crust is about 100 times higher than that of cadmium.

Metals from spent batteries should be recovered and regulations implemented to decrease the need for mining of virgin metals. In order to control the mobilisation of certain metals, interactions with other metal use must be addressed. For example, the demand for zinc also leads to mining of cadmium, since it is a by-product of zinc¹⁴. A way of avoiding the

¹³ AB₅ alloy contains mischmetal, which is a mixture of rare earth metals. Rare earths include the following 17 elements: scandium, yttrium (the lanthanides): lanthanum, cerium, praseodymium, neodymium, promethium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium and lutetium.

¹⁴ 0.4 kg cadmium is recovered as by-product per 100 kg zinc produced from zinc concentrate (Boustead and Dove 1998).

mobilisation of metals is to deposit low-demand metals at the extraction mine to avoid dissipative losses. Problems associated with the collection of spent batteries and the small quantities of metals available for recycling make it difficult to realize commercial recycling of batteries. Replacing NiCd batteries would lead to a decrease in the demand for cadmium metal and consequently lower prices and costs for safe storage. Regulations would have to be implemented to ensure that cadmium in products and from mining were collected and stored safely. The use of cadmium in industrial NiCd batteries, a product application where high collection efficiencies are achievable, may give market incentives for continued collection of cadmium containing products.

3.1.3 Summary of environmental aspects for portable batteries

For portable batteries, the small size of each battery unit, the large number of battery users, the low concentration of economic value and type of application are examples of product characteristics that are related to losses of metals. Portable batteries also have a short effective service life, which increases the turnover of materials.

Dissipative losses of cadmium and nickel from incineration and landfills are of environmental concern. Emissions and resource consumption of metals are significant in the end-of-life treatment, regardless of whether short- or long-term emissions are considered. The collection of spent batteries is more important than the technical efficiency of recycling processes in decreasing losses of metals to the environment. NiCd battery recycling is energy efficient at transportation distances greater than 10 000 km at collection rates of 10-85%. Transportation for the collection of spent NiCd batteries has low environmental impact compared with the impact throughout the whole life cycle and thus NiCd batteries can be transported long distances for recycling and this would still be beneficial from an environmental perspective.

Energy losses arising from the standby mode of the charger and also the losses during charging of portable batteries contribute significantly to the total energy use due to the low efficiency of many commercially available chargers. Excluding the usage phase, primary energy use and the emission of CO_2 are most significant in battery manufacturing. Batteries manufactured with recycled cadmium and nickel have 16% lower primary energy requirements than if only virgin metals are used. Using recycled cadmium and nickel requires 46% and 75% less primary energy respectively, compared with extraction and refining of virgin metal, considering allocation on mass basis.

The assessment of global metal flows arising from the use of portable rechargeable batteries shows that cadmium and nickel are of concern due to their high LEIs (4.4-5.6). The ratios of battery metal flows to natural flows are 1.4 for cadmium, 0.07 for nickel and below 0.01 for other metals, indicating that cadmium use in batteries may have an environmental impact. Battery metal demand for cadmium and nickel constitute 37% and 2% of global mining production in 1999, respectively.

To decrease the impact on global metal flows arising from the use of portable batteries the following points should be considered.

- 1. The development of battery technologies should aim at high energy density and long service life.
- 2. Metals with high natural occurrence should be used.
- 3. Metals from spent batteries should be recovered and regulations implemented to decrease the need for virgin metals.

3.2 Industrial batteries: energy analysis of batteries in photovoltaic systems

This section presents the results of the energy analysis of eight different battery technologies used in a stand-alone PV-battery system (Paper V). The battery technologies analysed are: lithium-ion Ni (Li-ion), sodium-sulphur (NaS), nickel-cadmium (NiCd), nickel-metal hydride AB_5 (NiMH), lead-acid (PbA) and three types of redox flow batteries (regenerative fuel cells): polysulphide-bromide (PSB), vanadium-redox (VRB) and zincbromine (ZnBr). The uncertainties, contributing components and the influence of different parameters and conditions of use are assessed. The design of the PV-battery system and the input data are described in detail in Paper V.

3.2.1 Uncertainties and contributing components

To show the uncertainties and the contribution of different components to the gross energy requirement, the results for the reference case (Case 1) are presented when the battery service life is limited by cycle life and the temperature is 25° C (Paper V). It is assumed that the batteries are produced from 100% recycled materials and that the different components are transported 3 000 km by heavy truck. The solar irradiation is assumed to be 1 700 kWh/m² year.

Fig. 12 shows that the energy return factor for the PV-battery system ranges from 2.3 for NiMH batteries to 12 for the Li-ion batteries. The PV-array excluding batteries has an energy return factor of 10-19. This means that 8.1-44% of the energy output is required to produce the PV-battery system. The Li-ion battery has the highest average energy return factor (9.8), which means that the PV-Li-ion battery system will replace 9.8 times more energy throughout its life time than the energy required for production of the PV-battery system.

For a PV-battery system with a service life of 30 years and taking into account the different service lives of the components, the energy payback time is 2.4-13 years, depending on the battery technology used. With a service life of 30 years, the energy payback time is 1.6-3.0 years for the PV-array. The energy payback time is 0.55-10 years for the different battery technologies, showing the energy related significance of batteries in PV-battery systems.

The uncertainty in the average value of the energy return factor is +/- 14-53%. The greatest uncertainty in *f* is found for the PbA battery, due to its high variability in cycle life. Fig. 12 shows the importance of using specific data when comparing different battery technologies, due to the high variability.



Fig. 12. Energy return factors for the PV-battery systems. Case 1: T=25°C, 100% recycled battery materials, service life limited by cycle life and transportation by heavy truck. The variation in the average value is +/-14 to 53%. (Paper V).

Production and transportation of batteries contributes 25-70% to the total production energy of the PV-battery system, also underlining the energy related significance of batteries in PV systems (Fig. 13). The relative contribution due to the production of batteries is lowest for the ZnBr battery and highest for the NiMH battery.

The contribution of production and transport of the PV array is 26-70% (NiMH-ZnBr). The highest absolute energy requirement for PV array production is 80-87 GJ/year for the redox flow batteries due to their relatively low efficiency, resulting in the need for a larger PV array and charge regulator.

Production and transport of the charge regulator contribute 1-4% (NiMH-ZnBr) to the gross energy requirement. The corresponding figures for the inverter are 2-5%.

The contribution of transport of all the components to the gross energy requirement is low (1.0-9.2%) for 3 000 km transport by heavy truck. The lowest energy requirement for transport is for the ZnBr battery due to its high energy density and the possibility of recycling the electrolyte. The transport of PbA batteries contributes 9.2% to the gross energy requirement since these batteries have a relatively low energy density and cycle life, and therefore a larger mass of batteries has to be transported.



Fig. 13. Energy requirements for production and transport of various PV-battery systems. Case 1: T=25°C, 100% recycled battery materials, service life limited by cycle life and transportation by heavy truck. The uncertainty is +/- 14 to 53%. (Paper V).

3.2.2 Comparison of indicators of energy efficiency

Depending on the origin and availability of energy resources, different energy indicators can be used to evaluate PV-battery systems. In applications where both solar and non-renewable energy are used as input energy to the PV-battery system, the energy return factor and the overall battery efficiency can be combined in order to make trade-offs between competing interests.

In cases where the focus is on using fossil fuels efficiently, a high energy return factor is important. This measure may be important in the expansion phase of PV-battery systems. PV-battery systems with similar energy return factors (e.g. ZnBr and Li-ion, in Fig. 14) may have different overall battery efficiencies. If electricity generated by solar energy can be considered as a free energy source, this measure would be less important. In the case of battery systems being charged with electricity generated from fossil fuel, the direct and overall battery efficiencies are important measures.

The overall battery efficiency is an important measure of the efficiency of a closed renewable system, where renewable energy has to be used as efficiently as possible, for example due to limited area for energy production.

Fig. 14 shows that the most energy-efficient battery technologies are found in the upper right hand corner. The Li-ion and NaS battery have low energy requirements for production and high charge-discharge efficiency. The direct efficiency of the charger, battery and inverter is 0.50-0.85. When considering the charger-battery-inverter system including their production, the corresponding figure for the overall battery efficiency is 0.41-0.82, which is an average decrease of 8.7%. The average efficiency of the NiMH battery decreases by 18%, from 0.65 to 0.53, which shows the effect of high energy requirements for battery production on the overall battery efficiency.



Fig. 14. Relation between energy return factor and overall battery efficiency (Paper V).

Three battery technologies that represent different performance characteristics have been compared: Li-ion batteries have a high direct efficiency and low energy requirements for production, PbA batteries have low direct efficiency and high energy requirement for production, while PSB batteries have a low direct efficiency and low energy requirements for production. The output energy corresponds to the functional unit and differences between the batteries in gross electricity input are mainly determined by the battery efficiencies (Table 5). The relative contribution of output energy to gross electricity input is 71% for Li-ion 53% for PbA and 51% for PSB. The relative contribution to output energy decreases with increasing number of days of storage capacity, i.e. the size of the battery.

Low battery efficiency influences the power of the charger and the PV array, resulting in higher energy requirements for their production, which can be seen in the row "E Production PV, charger & inverter" in Table 5. The ranking of the battery technologies in Table 5 according to the energy return factor is Li-ion, PSB and PbA. When considering the overall efficiency, the batteries are ranked Li-ion, PbA and PSB in the Reference case.

In cases when specific output energy is required and the area for mounting of PV arrays is limited, for example to one roof only, batteries with low direct efficiency may not be practical to use. For example, a PV-battery system with a PSB battery requires a PV array with an area of 484 m², while a Li-ion battery requires 337 m² (Table 5).

The PbA battery uses the gross energy input more efficiently than the PSB battery in the Reference case. In the All PV case, solar electricity is used for production and transportation of the batteries resulting, in higher gross energy input than in the Reference case. Since the PbA battery requires more energy for its production than that for the PSB battery, its overall battery efficiency decreases more than for the PSB battery. The comparison of PSB and PbA batteries shows how the overall battery efficiency can be used as a measure that take into consideration both energy requirements for battery production and battery efficiency.

		Li-ion			PbA			PSB	
Exchange of fuels ^a	Reference	All PV	Reference	Reference	All PV	Reference	Reference	All PV	Reference
	(GJ _{el} / year)	(GJ _{el} / year)	(GJ _{pf} / year)	(GJ _{el} / year)	(GJ _{el} / year)	(GJ _{pf} / year)	(UJ _{el} / year)	(UJ _{el} / year)	(GJ _{pf} / year)
Euse	197	197	^D 986	197	197	[₽] 986	197	197	[□] 986
E losses ^c	59	59		104	104		171	171	
E production battery	21	32	50	68	122	186	19	36	44
E production PV, charger &			70			81			98
inverter									
Area PV array (m ²)	337	337	337	397	397	397	484	484	484
Direct efficiency, η^{c}	0.78	0.78		0.66	0.66		0.54	0.54	
Overall battery efficiency,									
$\eta^{*_{B}}$	0.71	0.68		0.53	0.47		0.51	0.49	
Energy return factor, f			8.2			3.7			7.0
Energy payback time, t^*			3.7			8.1			4.2
(years) ^d									
E production of E use (%) ^e			13			27			14

Table 5. Indicators of the energy efficiency of a PV-battery system (Case 1).

Source: Paper V

Note: Case 1: T=25°C, 100% recycled battery materials, service life limited by cycle life and transportation by heavy truck.

Note: Case 1: 1-25 °C, 100% recycled battery materials, service me innited by cycle me and transportation by neavy truck. ^a Reference case:= the electricity produced can replace a certain amount of fossil fuel. All PV case:= closed renewable energy system where electricity is used for production and transportation of the PV-battery system. See explanation on page 18. ^b Primary fossil fuel replaced by the PV-battery system, $E_{G0} = E_{use}/\eta^*_{0}$, where $\eta^*_{0} = 0.20$. ^c Efficiency of charger, battery and inverter. ^d PV-battery system service life= 30 years

e Ei/Eo = 1/f

3.2.3 Influence of service life

To evaluate the influence of depth of discharge and battery temperature on the battery service life, four different cases were analysed with the energy model of the PV-battery system. The effects of using air conditioning (AC) on the total energy efficiency were evaluated to assess for which batteries the installation of AC can be motivated.

Battery service life was limited by cycle life except in Case 2, where it was limited by float service life¹⁵. The battery temperature in the various cases was set to (1) 25° C, (2) 25° C, float, (3) from 40°C to 25° C by using active cooling with AC, and (4) 40°C. The other parameter settings are the same as in the previous section, i.e. it is assumed that the batteries are produced from 100% recycled materials and the different components are transported 3 000 km by heavy truck.

Considering one cycle per day, float service life is longer than cycle life according to Case 2 for NiCd, NiMH and PbA batteries. The energy return factors are in this way increased by 10% for NiMH, 23% for NiCd and 52% for PbA. For Li-ion batteries, the energy return factor decreases by 14% for float service life compared with cycle life. Float service life will be the life-limiting factor for NaS, PSB, VRB and ZnBr batteries.

Fig. 15 shows that the energy return factor for the PV-battery system ranges from 2.7 (PbA at 40°C) to 9.8 (Li-ion at 20°C) for Case 1, 3 and 4. The uncertainty is +/- 14 to 61%. The greatest uncertainty is for the PbA battery due to the great variability in cycle life.

Increasing the temperature from 25° C to 40° C, causes the energy return factor to decrease by 0-47% for the different battery technologies. For batteries whose service life is temperature dependent, cooling of the batteries by operation with AC improves the service life compared with operation at 40°C. The effect of increased battery service life has to be related to the energy losses due to operation with AC, to ascertain whether if active cooling results in improved efficiency.

Operation with AC increases the energy return factor for NiCd from 4.6 to 5.2 (+14%), for NiMH from 3.0 to 3.8 (+27), for Li-ion from 6.8 to 9.3 (+37%), and for PbA from 2.7 to 4.9 (+79%). AC is therefore beneficial for these technologies. NaS and PSB batteries are excluded from Case 3 with external AC, since these technologies have built-in thermal management systems. VRB and ZnBr batteries are excluded from Case 4 since these technologies require AC at temperatures above 30° C, and it is therefore not possible to evaluate the use of AC.

¹⁵ Service life of a battery due to corrosion processes.



Fig. 15. Energy return factors for the PV-battery systems at different temperatures. The uncertainty is +/-14 to 61% (Paper V).

The difference in overall battery efficiency between service life limited by cycle life and float life is -2 to 10%. Float service life is longer than cycle life for NiCd, NiMH and PbA batteries, resulting in 2-10% higher overall energy efficiency for these batteries.

Fig. 16 shows that the overall energy efficiency of the PV-battery system ranges from 0.46 to 0.73. The uncertainty is +/- 9 to 33%. When considering the energy return factor, operation with AC is motivated for NiCd, NiMH, Li-ion and PbA batteries, but this situation changes when evaluating the overall efficiency. A comparison with AC turned on with 40°C shows that AC results in higher efficiency for PbA batteries, while it decreases the overall efficiency for Li-ion, NiMH and NiCd batteries (Fig. 16). A high energy return factor is important in an expansion phase of PV-battery systems while the overall battery efficiency is important when considering the efficient use of solar energy in a long-term perspective.

The direct energy requirement for operating AC is $21-60 \text{ GJ}_{el}$ /year, corresponding to 98% of the direct and indirect energy requirements of the AC unit. To improve the overall efficiency, it is most effective to decrease the energy use in the usage phase of the AC system.



Fig. 16. Overall battery efficiencies at different operating temperatures. The uncertainty is +/-9 to 33% (Paper V).

3.2.4 Influence of material recycling and transport

The relative importance of using battery materials of recycled or virgin origin, and transport by plane instead of truck from the battery manufacturer to the site of operation, have also been analysed. The temperature assumed was 25°C and the transportation distance for the PV-system components 3 000 km.

Fig. 17 shows that batteries with low energy density and short cycle life are most influenced by air transport. Air transport decreases the energy return factor for PbA batteries by 70%. Although redox flow batteries have a low energy density, the energy requirements for their transport are reduced since distilled water can be supplied at the site of operation and the active materials in the electrolyte can be recycled on site. With no recycling, the energy return factor of the ZnBr battery is reduced by 35%.



Fig. 17. Relative changes in the energy return factor for different recycling rates and modes of transportation compared with Case 1 (Paper V).

The overall battery efficiency decreases by 1-6% when using virgin material production compared with recycled material production. The greatest change in the overall battery efficiency is seen for the ZnBr battery, for which the value decreases from 0.55 to 0.52 (Paper V, Table 10). Air transport decreases the overall battery efficiency by 3-33%. The overall battery efficiency of the PbA battery decreases from 0.56 to 0.38 (Paper V, Table 10).

3.2.5 Influence of battery parameters

Improvement in the battery efficiency has the greatest influence on the energy return factor for Li-ion, NaS, PSB, VRB and ZnBr batteries (Table 6). The battery efficiency is the second most important parameter for NiMH, NiCd and PbA batteries. An improvement in the battery efficiency by one percent results in an increase in the energy return factor of 0.40-0.73%. Since losses in the battery must be compensated by higher energy input, low battery efficiency results in a larger PV-array and charger, which means higher indirect energy requirements for their production.

For NiMH, NiCd and PbA batteries, the energy density and the service life have greatest influence (0.51-0.63 % f/ $\%\Delta\eta_3$). The substantial influence of these parameters is explained by the energy requirement for battery production, which is 56-70% of the gross energy requirement for NiMH, NiCd and PbA, compared with 25-35% for the other battery

technologies. Low energy density results in higher material intensity and energy requirements for the production of materials, as well as a higher battery mass to be transported. Short battery service life means that batteries have to be replaced more often, resulting in higher energy requirements for battery production and transport.

The energy requirement for battery manufacturing is an important parameter for NiCd and NiMH batteries. Energy requirements for battery production, q_{P3} , have a 4-16% smaller influence than *d* and $t_{cycle,3}$, since q_{P3} does not influence the energy requirement for battery transport.

Technology	η	d, t _{cycle}	q_P
Li-ion	0.42	0.28	0.28
NaS	0.65	0.26	0.25
NiCd	0.40	0.52	0.50
NiMH	0.30	0.63	0.61
PbA	0.41	0.51	0.43
PSB	0.66	0.26	0.25
VRB	0.62	0.29	0.28
ZnBr	0.69	0.23	0.23

Table 6. Percent change in the energy return factor when changing various battery parameters (% f / %\Delta \eta_3).

Source: Paper V

Note: Case 1: T=25°C, 100% recycled battery materials, service life limited by cycle life and transportation by heavy truck. t_{cycle} = cycle life, d= gravimetric energy density, η = battery efficiency, q_P = energy requirements for battery production. **Bold** face indicates the highest values.

The battery efficiency, η , has the greatest influence $(0.69-0.96\%/\%\eta)$ on the overall battery efficiency. This is due to the high energy turnover in the battery (50-72% of the total turnover). Changes in the efficiency of the Li-ion battery have a relatively small influence compared with the other battery technologies. This is because the losses in the Li-ion battery correspond to 9% of the total gross energy requirement, compared with 19-33% for the other technologies. This results in a smaller improvement than for the other battery technologies and a relatively small change compared with the already high efficiency of the Li-ion battery.

Improvement of energy requirements for production, service life and gravimetric energy density by one percent change the overall battery efficiency by 0.03-0.15%. All these parameters influence the indirect energy requirements for production and transport. Since the electricity output from the PV array is partly converted to other energy carriers (thermal energy and transport fuel), the output energy for production and transportation gives 2.5-2.7 times higher energy yield, which decreases the relative importance of production and transport of the batteries and charger.

3.2.6 Summary of environmental aspects for industrial batteries

Industrial batteries are generally made for high-power applications and have a high energy turnover compared with batteries in portable applications, and analysis of energy flows is therefore an important aspect when assessing industrial batteries. The energy return factor and the overall battery efficiency were estimated for eight different battery technologies used in a stand-alone PV-battery system. With a battery storage capacity three times higher than the daily energy output, the energy return factor for the PV-battery system ranges from 0.64 to 12 for the different cases. This means that 8.1-156% of the energy output is required to produce the PV-battery system. If the value of the energy return factor is less than one, the indirect energy used to produce and replace the device is greater than the direct energy output. In this case, the device works as a non-rechargeable battery moving energy from one place to another.

In the reference case, the production and transport of batteries contributed 25-70% to the indirect gross energy requirement. The contribution of production and transport of the PV array is 25-66% depending on the battery technology used. For a PV–battery system with a service life of 30 years, the energy payback time is 2.4-46 years depending on the battery technology. The energy payback time is 1.6-3.0 years for the PV-array and 0.55-43 years for the battery, showing the energy related significance of batteries in PV-battery systems. The Li-ion battery had the highest energy return factor due to its high energy efficiency and low energy requirements for battery production. The lowest value of the energy return factor was for the PbA battery when the battery is transported by air and produced from virgin materials only.

Conditions and battery parameters were varied in order to identify their relative influence on the energy flows of battery systems. For batteries in stand-by applications (UPS) which are discharged very seldom, low no-load losses and high energy efficiency of the charger are important in order to reduce energy losses throughout the battery life cycle (Papers III & V). In applications where batteries are cycled frequently, high charge-discharge efficiency of the battery is most important. The battery charge-discharge efficiency has considerable influence on the energy return factor for batteries with relatively low energy requirements for production and transportation (Li-ion, NaS, VRB, ZnBr and PSB). Service life, gravimetric energy density and battery production are equally important for NiCd, NiMH and PbA batteries. For NiCd, NiMH, Li-ion and PbA batteries, whose service life is temperature dependent, cooling of the batteries by operation with AC improves the energy return factor compared with operation at 40°C. The overall battery efficiency is significantly influenced by the charge-discharge efficiency.

When comparing battery technologies specific data has to be used corresponding to the particular application since the charge-discharge efficiency and service life of batteries depend on operating conditions. PV-battery systems can be made more energy efficient by matching operating conditions and battery characteristics in a life cycle perspective. The energy efficiency of the PV-battery system can be increased by: (1) optimised charging algorithms, (2) passive temperature regulation to $\sim 20^{\circ}$ C, (3) increased utilisation of the active battery material, (4) lower material requirements for battery production and, (5) efficient production and transport of PV-battery system components.

4 Discussion

4.1 Uncertainties in data and choice of method

In environmental assessment of battery systems, it is important to be aware of uncertainties in parameters that have significant influence on the results. In this section, methodological choices and critical parameters are discussed.

4.1.1 Choice of method

Application of LCA proved to be useful for the identification of activities associated with portable batteries that have environmental impact (Paper I). After identification of environmental hot spots, targeted methods may be applied. It is important to take into account the eco-toxicity of hazardous substances when evaluating battery systems, and this is difficult to interpret in LCA. To thoroughly address eco-toxicity aspects, LCA can be complemented with other methods, for example environmental risk assessment (ERA), in order to model the exposure to and response of organisms to different metals. ERA has extensive data requirements and the focus is to assess the risk of actual effects, implying that the cause-effect chain is largely known.

Assessment of new technologies is difficult since the conditions of use of the technology may be unknown, and data on manufacturing processes are usually not available. Approximate approaches can be useful in order to obtain an overview and to identify possible critical parameters. Application of SFA (Paper II) enables the assessment of limits on emissions based on nature's limited assimilation capacity early in the cause-effect chain. Depending on how actors select and interpret information, the concept of *frames* can be used to analyse the views of various actors (Schön and Rein 1994). Two major frames can be identified: the precautionary frame in which potentially harmful practices should be avoided if feasible alternatives are available, and a proof-first frame in which scientific evidence of proven harm is required before a practice can be stopped. The interpretation of the results from the SFA in Paper II (precautionary) and ERA (proof-first) depends on the view on what information that signifies a problem.

Emerging technologies may be difficult to assess with LCA since the method has comprehensive data requirements. In LCA of fuel cell technology, Pehnt (2003) applied forecasting methods to estimate data in order to allow comparisons with currently available technologies. By conducting LCAs continuously, environmental hot spots and bottlenecks in emerging technologies may be identified (Paper III). Life-cycle inventory data for groups of materials can be used as estimates in life-cycle assessments when specific data on materials are not available (Paper IV). In this way, the environmental significance of a material in a specific application can be identified.

In industrial battery applications with high energy turnover, for example, renewable energy systems or load levelling, energy is of significant environmental importance since material flows of large batteries are generally are easier to control than those of portable batteries (Section 4.2.1). The energy return factor and the overall energy efficiency can be used as

indicators in assessing the usefulness of energy storage from an energy perspective and in monitoring the performance of energy systems. The choice of conversion efficiencies for different energy qualities and system boundaries is crucial for a reliable assessment (Paper V).

The environmental impact assessment phase in LCA aggregates different kinds of environmental impacts, such as resource consumption, health effects and ecological effects. Assessment specifically focusing on limits on extraction can be made by comparing the material requirements of a technology with the available resources and mining production capacity. Recent examples of the application of this methodology are assessments of thin-film photovoltaics, batteries and fuel cells (Andersson 2001, Råde 2001).

4.1.2 Choice of system boundaries

The results of environmental systems studies depend on the choice of temporal, geographical and technological scope. Future risks and environmental impact arising from landfilled materials are difficult and most LCA studies of batteries do not consider long-term emissions. By expanding the temporal scope of analysis, the importance of such emissions increases (Paper I). Instead of using static modelling, as is commonly done in LCA, dynamic models may be used to more accurately describe metal emissions due to changing application patterns of products, utilisation of resources and changing landfill environments.

The geographical scope of an analysis influences the environmental impact of battery systems since electricity generation and end-of-life treatment may differ between different countries. End-of-life treatment of batteries in non-OECD countries with less developed infrastructure for battery collection and less controlled landfilling and incineration has been little described in the literature.

In many environmental assessments of rechargeable batteries, auxiliary components for charging are assumed to be identical and are not included. When assessing batteries used in applications with a high energy turnover, the charger, charging regimes and battery energy efficiency are of major importance (Paper I and V, Kirmayer 1995, Garica and Schlüter 1996, Barlinn 1997, Rantik 1999, Staal-Jensen and Petersen 1999). Small differences in battery energy efficiency may have a significant influence on the results, which means that it is important to use specific data for each battery type and application evaluated. In Paper V, it is shown how the battery charge-discharge efficiency influences the power ratings of several components (PV-array, charger and air conditioning) in the battery system. Staal-Jensen and Petersen (1999) concluded that manufacturing of electronic parts and the coil of a charger for a mobile phone causes greater environmental impact than the production of a battery for a mobile phone. In studies in which whole energy systems are compared with each other to identify significant effects, it is therefore important to include the charger in the evaluation.

4.1.3 Collection efficiency of spent batteries

Collection efficiencies and material recovery rates are important parameters in establishing the material balance of a battery system throughout its life cycle. Due to uncertainties in determining collection efficiencies, different cases may be modelled in order to illustrate the best and worst potential conditions (Papers I, II, III & V).

The collection efficiency can be calculated in different ways in order to consider hoarding effects, service life, imports/ exports and market growth. Four different methods for calculation have been proposed, and they all give different results (Lankey 1988, Wiaux 2002, BIO 2003).

The fraction of batteries collected in relation to battery sales, η_{Sales} , is calculated from:

$$\eta_{Sales} = \frac{Q_{Collected}}{Q_{Sales}} \tag{18}$$

where Q_{Sales} (ton/year) represents the battery sales in a specific year and $Q_{Collected}$ (ton/year) is the mass of batteries which have been collected in the same year. This ratio may be appropriate for products with a very short service life, but for batteries there is no relationship between actual collection efficiency and sales, since the service life may be several years and hoarding effects are not considered.

When considering the service life of batteries, the collection efficiency of waste batteries, $\eta_{Battery waste}$, is calculated from

$$\eta_{Battery \ waste} = \frac{Q_{Collected}}{Q_{Battery \ waste}} \tag{19}$$

where $Q_{Battery waste}$ (ton/year) is the market sales in the year corresponding to the average service life of the battery. To reach high collection efficiency with this ratio, hoarding of spent batteries has to be minimised. It is difficult to account for imports and exports of batteries, but these factors may be included specifically (Lankey 1998).

To include the effects of hoarding, $\eta_{Batteries in MSW}$, gives the fraction of batteries, that is collected relative to the spent batteries available for collection in municipal solid waste (MSW):

$$\eta_{Batteries in MSW} = \frac{Q_{Collected}}{Q_{Collected} + Q_{MSW}}$$
(20)

where Q_{MSW} (ton/year) is the mass of batteries found in municipal solid waste. Uncertainties in the calculation include time lags during which collected batteries may be stored before they are transported to recycling plants. Representative sampling of the municipal waste stream is difficult to achieve since batteries can be unevenly distributed and some may be disposed of in an uncontrolled manner. Regular sampling of batteries in MSW is carried out in a few countries only.

Another measure of collection efficiency, which is, however not useful for establishing material balances, is the actual collection efficiency, $q_{Inhabitant}$ (g/inhabitant year), which can be calculated from:

$$q_{Inhabitant} = \frac{Q_{Collected}}{n_{\text{Region}}}$$
(21)

where n_{Region} is the number of inhabitants in a region. This measure is independent of actual sales but it provides a quantitative goal.

In the proposal of the new battery directive (EC 2003b) quantified targets have been established for $\eta_{Battery in MSW}$ and $q_{Inhabitant}$.

In Paper I, the collection efficiency was calculated based on Eq. 18 due to a lack of data, which made it impossible to apply other approaches. The value of $\eta_{Battery waste}$ was calculated and found to be 22-31% in Sweden during the period 1996-1999. In a study by Wiaux (2002), 165 tons of portable NiCd batteries were collected and 25 tons were estimated to be discarded in MSW in Sweden in 2000. This results in a collection efficiency of 87%. The average collection efficiency of six European countries was found to be 72% (Wiaux 2002). Considering this new data, the actual collection efficiency in Paper I is underestimated.

4.1.4 Metal emissions from landfilling and incineration

It is difficult to estimate representative factors for metal emission to the environment from landfilling and incineration since local conditions vary considerably. During this work, emission factors for NiMH batteries were quantified in laboratory studies, but their temporal perspective and relevance to field data could not be determined (Eriksson 2001). Generalised emission factors may be used in order to estimate the magnitude of different flows and to identify life cycle activities of significance (Finnveden 1996).

In Paper I, generalised emission factors were assumed for metal emissions from landfills and incineration in order to estimate their relative contributions compared with other life cycle activities. It is assumed that the emission is proportional to the mass of the disposed batteries. However, this may lead to an overestimation of emissions since it has been claimed that no proportional relationship exists, and the cadmium concentration in leachate is independent of the amount of NiCd batteries in MSW (Wiaux 2003). Alkaline and reducing environments and the presence of complexing agents (organic material or sulphides) may result in very low mobility of metals.

In LCA, compounds containing the same metals are often aggregated to form one model compound since it is difficult to determine chemical species. However, the chemical species of metals determines their mobility and bio-availability, and thus their potential environmental effects (Wolterbeek and Verbrug 2001). Cadmium oxide, which is the form of cadmium used in NiCd batteries, is relatively insoluble, while cadmium chloride is highly soluble (Morrow 2001). Aggregation of different cadmium metal species may therefore lead to over- or underestimated environmental effects in LCA. To assess ecotoxicological effects of a particular substance, environmental risk assessment is a more appropriate method than LCA since it is specifically intended to model the exposure and response of different organisms.

A targeted risk assessment of cadmium oxide used in batteries within the European Union concluded that portable NiCd batteries contribute 0.1-1.1% to the total cadmium emission to different environmental compartments (TRAR 2003¹⁶). Industrial plants producing and using cadmium contribute 3.8% to the total cadmium emission to air and surface water, while the main emissions originate from phosphate fertilisers, fossil fuel combustion and

¹⁶ Belgium is rapporteur for the risk assessment which was initiated in 1999 in accordance with council regulation EEC/793/93. The final draft from May 2003 is currently being peer-reviewed by the Scientific Committee on Toxicity and the Environment (SCTEE).

the metal industry. At a global level, the use of portable NiCd batteries gives a small contribution to the total emission, which may have eco-toxicological effects. However, since portable NiCd batteries comprise approximately 50% of the end use of cadmium, the potential is high for long-term emissions (Paper II). Dissipative uses in other applications than batteries may slander cadmium, which may result in restrictions on the use of all cadmium-containing products.

Assessment of local risks indicates a possible effect of portable NiCd batteries in local sediment due to already high background concentrations, but actual bio-availability has not been considered (TRAR 2003). The ratio of Predicted Environmental Concentrations (PEC) to Predicted No Effect Concentrations (PNEC) in the worst-case local scenario was found to be 0.40-5.6 for cadmium (TRAR 2003). The study concluded that a risk on a local scale cannot be excluded due to the possibly of high background concentrations in sediment but there is at present no need for further risk reduction measures beyond those, that are already being applied (TRAR 2003).

Uncertainties in the TRAR (2003) lie in the fact that long-term diffuse cadmium emissions due to re-use, disposal and/or incineration residues have not been quantified due to a lack of appropriate methods. It is not possible to determine the future change in municipal solid waste composition or the composition of leachate from landfills. More information is needed to refine the regional background concentrations and release estimates for local scenarios since regional cadmium concentrations are already above the PNEC in sediment. No risk assessment has been conducted regarding air emissions, due to a lack of data on toxicity in the atmospheric compartment. Emissions of cadmium may be underestimated since it is difficult to obtain data for all plants handling cadmium. Environmental cadmium concentrations were not expressed as bioavailable concentrations since these vary with the properties of the compartment (soil, atmospheric, sediment, aquatic) (TRAR 2003). Most industrialised countries have implemented extensive control measures to immobilise metals treated in municipal solid waste, which means that predicted emissions are valid for these countries only. PNEC values have been estimated for certain organisms only, and synergetic effects with other substances have not been considered. Uncertainties, that are difficult to quantify arise from unexpected events such as flooding or fires. Flooding of landfills may result in oxidising conditions thereby mobilising metals. Uncontrolled landfill fires may affect as much as 0.2% of the annually landfilled waste of all municipal landfills in Sweden (RVF 2003a and b).

4.1.5 Uncertainties in energy analysis of PV-battery systems

Since the performance of a specific component or technology is dependent on battery system design and operating conditions, it is not possible to present general results without considerable uncertainties. In Paper V, the broad ranges of values indicate the uncertainties in the results as well as the improvement potential of different technologies. Input data with high influence and large uncertainty is the battery charge-discharge efficiency and the battery service life. The uncertainty in output results for different battery technologies varies between 8% and 61% (Paper V).

In Paper V, the difference between low and high values of input data is 1.1-2.2 times, where the highest variability is for NiMH and PbA. Since all battery technologies, except for PbA and NiCd, are immature for PV applications there are uncertainties about their performance. The absolute values of the energy return factors are dependent on the

conditions of use due to the high influence of the conversion efficiency to electricity. With higher efficiency of the replaced reference system for electricity generation ($\eta_0 = 0.35$ instead of $\eta_0 = 0.20$) the absolute values of the energy return factor decrease by 43%. Assuming that electricity generated by the PV array is used for the production of PV-battery systems in a closed renewable energy system (the *All PV case* in Table 1, Paper V), the overall battery efficiencies decrease by 3-15% to 0.35-0.80. This reduction in overall efficiency means that energy-efficient production of the PV-battery system becomes more important when considering a closed renewable energy system.

Energy requirements for the production of batteries were assessed from cradle to gate, including materials production and battery manufacturing. Comparisons of different LCA studies of batteries (Rydh 2003) are often difficult since important assumptions are not transparently reported. To enable a thorough evaluation of different energy analyses of battery systems, the following parameters should be reported: (1) battery mass, (2) battery capacity, (3) choice of system boundaries (geographical, temporal, technological) (4) the proportion of the gross primary energy requirement made up by electricity, the assumed efficiency of electricity production and method used to sum up different energy qualities, (5) recycling rate of spent batteries, (6) energy requirements for battery manufacturing processes and production of virgin and recycled materials, (8) battery design, (9) battery material composition, and (10) the allocation principles for multi-output processes.

Only a small number of demonstration units have so far been built using VRB, PSB, ZnBr and NaS batteries, and mass production is likely to improve the production efficiency of these batteries. Energy requirements for the production of batteries may vary considerably depending on material requirements, and where and how the batteries are manufactured. Uncertainties are due to the restricted availability of information since companies manufacturing batteries protect their technology from competitors. Material requirements for immature technologies can change rapidly in the course of development. Despite these factors, the analysis in Paper V indicates the technical potential of different technologies.

4.1.6 Environmental impact assessment

To rank different product alternatives, identify data categories and activities which are of high environmental priority from a diverse set of inventory data, environmental impact assessment can be applied by characterisation and/or weighting. Several LCA studies comparing battery technologies (Li-ion, NiCd, NiMH and PbA) do not rank the batteries based on environmental performance (Törnblom 1996, Kertes 1996, Almemark *et al.* 1999, Staal-Jensen and Petersen 1999). Unreliable or a lack of data for different batteries makes it difficult to draw conclusions based on inventory results only. Differences can be found between certain impact categories. For portable batteries, impact assessment shows that NiCd batteries have a higher impact on eco-toxicity than Li-ion and NiMH batteries (Törnblom 1996, Staal-Jensen and Petersen 1999). Morrow (2001) concluded that disposal is the most important factor in determining the total environmental impact of a battery system over its entire life cycle. The most effective way to reduce the environmental impact of battery systems is to increase the recycling rates, the battery performance, and, to lower the content of hazardous material (Morrow 2001).

Morrow (1997) compared four different battery systems using five different weighting methods, but the results were difficult to interpret since they pointed in different directions. Since the models for weighting are based on different subjective values, the user has to

decide the value of highest rank. To be useful for companies, LCA should be straightforward to use. As a basis for communication there is a desire for standardization of impact assessment methods. Companies and practitioners require off-the-shelf characterization and weighting factors. Hertwich *et al.* (2003) suggest that a best available practice and uniform method for weighting should be developed in order to increase the acceptance and credibility of the results. However, Bengtsson (2002) argues that weighting will have to be accepted for each decision maker or organisation, since different stakeholders have different priorities.

In Paper III, weighting of the inventory data with the EPS-96 method resulted in resource consumption of metals totally dominates over other factors. The environmental priority is therefore to ensure no or minimal losses of metals, or to use metals with less impact on resource consumption. All other measures to reduce environmental impact are insignificant compared with metal resource losses, which means that life cycle inventories of other data categories are unnecessary. However, by using various weighting methods, different significant environmental impacts can be identified giving a diverse picture of environmental effects.

4.1.7 Environmental systems analysis in decision making

Different methods of environmental systems analysis may be applied to evaluate the possible consequences of a decision. However, depending on the system boundaries and modelling techniques of a technical system, the results may be inconsistent. For example, the ability to generate positive net energy has recently been at the heart of a debate on the benefits of producing ethanol from corn (Shapouri *et al.* 2002, Kim and Dale 2002, Pimentel 2003).

The spread in results causes confusion and debate among decision makers, and it is possible that studies that support one or the other decision will be cited. It is therefore important to understand the reasons for the heterogeneity in the results obtained by different authors in order to help reach a consensus and to ensure that the decisions taken are based on sound assumptions.

Examples of how the results of environmental studies of the same object differ, and how they are interpreted, can be found in the debate regarding the revision of the EU directive 91/157/EEC (EC 1991) on batteries. The resulting proposal (EC 2003b) is based on different types of environmental assessments, and on social and economic considerations. In an online consultation, different stakeholders expressed their opinions on different policy options (EC 2003).

The government in the United Kingdom (UK 2003) states, "Collection of batteries, for all types, and through whatever means, will have significant environmental and economic impacts", which is in contrast to the conclusion of Paper I. It is also stated that there are no excess risks associated with the disposal of cadmium- or lead-containing batteries to landfills or by incineration. The first statement may find justification in a study commissioned by the UK Department of Trade and Industry, which concluded that separate collection of batteries causes higher environmental impact than the reduction achieved by recycling materials from batteries (ERM 2000). The second statement is related to the risk assessment of cadmium (TRAR 2003), which is discussed in Section 4.1.4.

The environmental consequences of collecting and recycling batteries in the study by ERM (2000) are in conflict with the results of other studies (Lankey 1998, Paper I). In recent environmental systems analysis studies of waste management (Finnveden *et al.* 2000, Sundkvist *et al.* 2002) it was concluded that the following prioritisation of waste treatment is valid, as a rule of thumb: (1) reduce, (2) reuse, (3) recycle materials, (4) incinerate with heat recovery, and (5) landfilling. Increased transportation distances arising from the collection of waste do not change the ranking of different waste treatment options, but passenger car transportation should be minimised. Also, for low-energy materials, like paper, it was found that it was more energy efficient to recycle or to incinerate them, than to landfill them (Finnveden *et al.* 2000).

ERM (2000) concluded that increased collection of batteries and recycling of battery materials would result in increased environmental impact regarding air emissions and depletion of oil resources. It is reported that system expansion is used to model the benefits of recycling. In other words, the system includes all effects associated with production of the materials from alternative sources (a mix of virgin and secondary), sufficient to match the composition of the total battery waste stream when these materials are not recovered from batteries themselves¹⁷ (ERM 2000, pages 83 and B11).

The authors of the ERM study make the assumption that all materials can be modelled as virgin or recycled steel¹⁸, resulting in a model that does not correspond to reality (see Appendix A, Fig. A2-A3). Unfortunately, no quantified sensitivity analysis is performed in the study to show if their assumption results in an over- or underestimate. Due to the lack of transparency in the report, it is not possible to identify activities contributing to different environmental effects since they have been aggregated for each scenario. Quantitative data on waste scenarios and alternative sources of production of materials in terms of environmental impact and financial costs are lacking, which makes evaluation of their relative contributions impossible.

According to the model developed by ERM (2000), revision of the battery directive would not result in any financial or environmental benefits. By changing the assumption regarding alternative sources for the production of materials, as exemplified above, it is likely that the results regarding environmental impact will be inconclusive. By presenting the figures from ERM for overall collection rates, BIO (2003, p. 84) concluded that the CO₂ emission resulting from battery material production, collection and recycling decreases with increasing overall recycling rates. The assumptions behind the financial aspects can not be evaluated since no background data were presented by ERM (2000) and a monetary evaluation is not within the scope of the present work.

¹⁷ However, the displaced need for alternative materials production resulting from materials recycling is not included in the input data requirements of the system boundaries (Fig 5.1 in ERM 2000).

¹⁸ (Page B11 in ERM 2000) "Hence, in order to maintain comparability between the baseline system and those scenarios developed for the recover of the various batteries, all such materials were modelled as either virgin steel or recycled steel as appropriate. The same assumption has also been made to model production of virgin and recycled plastics. This presents potentially either an over-or under-estimate of the burdens associated with these aspects of LCA systems modelled. However, the principal environmental impacts associated with the production and recycling of metals and plastics are predominately energy-related and are thus expected to be the same order of magnitude."

The revision of the battery directive is aimed at reducing the future risks of heavy-metal contamination. Remediation and treatment of contaminated soil and water involve the operation of heavy equipment and the mobilisation of large quantities of materials (US EPA 2003). It would, therefore, have been appropriate to quantify the possible environmental impact and financial costs of treatment of contaminated leachate and soil remediation, as indicated in Fig. A1, in a sensitivity analysis.

The conclusion drawn from this comparison of studies is that the complexity of systems analysis makes it extremely important to clearly specify and motivate the system boundaries and assumptions made. A study should identify the most important parameters and present the uncertainties in the input data. The meaning of the word *environmental impact* must also be specified, especially when unknown, long-term effects of heavy metals is the main concern, and their possible environmental effects must be compared with environmental effects related to energy use. The choice of system boundaries and different scenarios must be made so as to include the effects and consequences influenced by a policy or decision. Furthermore, all results should be transparently presented in order to allow assessment of the assumptions and the validity of the results. Finally, critical review by an external expert in accordance with ISO 14 040 (1997) increases the credibility of a study.

4.2 **Relevance and implications of the results**

Based on the results presented above, important aspects concerning the environmental impact of battery systems are discussed below in a broader context. The significance of battery application and strategies for managing material flows of toxic metals are reviewed. Furthermore, the opportunities and constraints on the use of batteries to contribute to a renewable energy system are examined, and strategies for improving the environmental performance of battery systems are presented.

4.2.1 Environmental impact determined by the battery application

The environmental impact of a battery system depends on its application, which in turn influences (1) the turnover of energy during use of the battery and, (2) the risk of uncontrolled disposal and dissipative losses at the end of life.

For batteries used in UPS applications, or in applications where the energy turnover in the battery is low, a low rate of self-discharge in the battery and low no-load losses of the charger are important parameters for the total energy requirements of the battery system. For batteries used in cycling applications with high energy turnover in the battery (e.g. renewable energy systems or load levelling) the battery charge-discharge efficiency is the most important parameter in reducing the gross energy requirement (Paper V).

Since the power used in industrial applications is high compared with portable applications and the former are designed for continuous use, their relative contribution to total energy flows of energy systems may be significant. The power and energy turnover in portable battery applications are low compared with other electrical appliances, and their relative contribution to electricity use in an average home in the Western world is low. However, in a global perspective it is important to consider the energy efficiency in portable applications since the accumulated use of portable batteries is significant. Energy losses at no-load¹⁹ of simple chargers for portable batteries are the most significant contributors to the life cycle gross energy requirement (Rydh 2001). Standby losses, including no-load losses, in power supplies contribute substantially to the electricity consumption of households in Europe (EC 2000). Due to an increase in the use of external power supplies, it has been estimated that energy use will increase from 8 TWh in 1996 to about 14 TWh in 2006 in a Business as Usual scenario (EC 2000).

For portable batteries, the small size of each battery unit, the large number of battery owners and low concentration of economic value are examples of characteristics that are associated with losses of metals (Rydh 2001). Portable batteries also have a short effective service life, which increases the turnover of materials (Paper II). In certain applications of portable non-rechargeable batteries, such as hearing aids and watch batteries, the appliance may be returned to a professional dealer when the battery has to be replaced, which may improve the control at the end of the battery's life.

Material flows of large industrial batteries are easier to control due to the limited number of owners, and the large size reduces the risk of loss and inappropriate disposal. Large battery systems are characterised by a high concentration of metal constituting an economic value, and thus owner responsibility (Rydh 2001). High collection efficiency is only likely to be achieved in countries with a well-developed infrastructure.

4.2.2 Managing material flows of toxic metals

In the use and development of batteries, there are uncertainties associated with different substances in batteries. Problems may appear when a substance has been used for a long time, and it may be difficult to identify the origin of the problem due to complex interactions. There are several substances in the history of industrialisation that proved to be environmentally problematic when used on a large scale. Examples of substances and applications are PCB (e.g. transformer oil), DDT (e.g. insecticide), chloroflourocarbons (e.g. coolant in freezers), chlorine (e.g. paper bleaching), Cd compounds (e.g. pigments), Pb compounds (e.g. gasoline) and Hg compounds (e.g. fungicide).

Critical issues facing battery technologies based on toxic metals are related to the management of these metals throughout the battery life cycle. The ban on button cell batteries containing mercury is a recent example of an environmentally constrained technology (EU 1998).

The use of cadmium in NiCd batteries can serve as an example of the importance of managing material flows to avoid environmental constraints. Since the boom in use of portable NiCd batteries in the late 1980s, the following instruments have been discussed or applied in an effort to decrease the input and control the flow of cadmium in batteries (Rentz *et al.* 2001): (1) banning the use of cadmium in batteries, (2) introducing collection programmes organised through the voluntary initiative of branch organisations, (3) subsidising safe deposition of cadmium when mined as a by-product, (4) taxes or levies on

¹⁹ The charger is connected to the power grid but no battery is connected.

NiCd batteries, (5) refunds on batteries, (6) consumer information campaigns on appropriate battery handling, and (7) the support of alternative types of batteries.

The revision of the EU battery directive 91/157/EEC (EC 1991) has been strongly debated (CollectNiCad 2001, EC 2003) since a ban on cadmium in batteries was initially proposed in 1997. Based on impact assessments of different policy options (BIO 2003, TRAR 2003), collection and recycling of all types of batteries is expected to provide the same environmental protection at a lower cost than a ban on portable NiCd batteries. A ban on portable NiCd batteries would not cover existing and hoarded batteries, which may result in continued landfilling of batteries. Shorter service life of batteries which can replace NiCd batteries may result in increased material flows of other metals (Paper II, BIO 2003). The trend seems to be that NiMH and Li-ion batteries in household appliances are taking market shares from NiCd batteries (EC 2003b). In other applications, sealed lead-acid batteries may provide an alternative to portable NiCd batteries, which may not lead to reduced environmental impact (Footnote 12, page 31). The current proposal (EC 2003b) sets the collection targets for waste batteries at 160 g batteries per inhabitant per year and 80% collection efficiency of portable NiCd batteries. The material recovery efficiency is set to 65 wt.% for lead-acid, 75wt.% for NiCd and 55% for all other types of batteries.

The industrial initiative CollectNiCad (2001) has been working since 1998 on establishing high collection efficiencies, currently with the target of collecting 15 g portable NiCd batteries per EU inhabitant per year. During the past five years public statistics and information regarding battery material flows have become available, indicating the increasing societal importance of managing material flows arising from batteries. The future success of NiCd batteries lies in proving that low dissipative losses of metals can be achieved, that the long-term risks are acceptable, and that the stock of accumulated metals can be managed. Modelling of accumulated cadmium at a hypothetical steady state in the Netherlands, showed that the level of acceptable daily intake in humans will never be exceeded (van der Voet *et al.* 2000).

In order to decrease the need for input of primary material through maintaining a stock of metals by recycling, the recovery efficiency must be high. Assuming a hypothetical collection rate of 100% portable NiCd batteries, 0.03% Cd would be lost from the NiCd product system during the life cycle of the NiCd battery (Paper I). NiCd battery recycling is energy efficient, even at very long transportation distances, at collection rates of 10-85% compared with battery production from virgin materials (Paper I). Several studies (Socolow and Thomas 1997a, 1997b, Lave *et al.* 1995, 1996, 1997, Karlsson 1999) have analysed and debated the technical efficiency of recycling lead in lead-acid batteries, as well as the level of metal emission. Karlsson (1999) has concluded that at steady state with a high collection rate of spent batteries the losses of lead from the system will be small in comparison with natural lead flows and historical losses during industrialisation.

The possibility of avoiding dissipative losses and to achieving closed technospheric flows is dependent on the type of battery application (see Section 4.2.1). Material flows of large battery systems with few battery owners may assist safe handling of spent batteries. The use of toxic or scarce metals may be appropriate for large battery systems, which prove to have low dissipative losses of metals. For metals where the incentives for collection and recycling are so far small, the need for metals in large battery systems may create market incentives for their collection and recycling (Paper II).

The application of batteries in solar home systems in less developed countries may give rise to problems if no scheme is established for recycling. At remote locations, batteries may be dumped at the end of their life if there are no incentives for recycling. Education on proper maintenance and handling of spent batteries must be provided.

4.2.3 Can PV-batteries contribute to a renewable energy system?

In many applications, batteries are part of an energy system that moves energy from one place to another (Paper V). Due to irregularities in the supply of renewable energy (e.g. solar and wind), batteries may be used to increase the usefulness and reliability of a renewable energy system. Depending on the performance of the batteries and energy requirements for the production of battery systems, the addition of a battery may influence the overall energy efficiency of the renewable energy system.

Assuming that the world's PV systems have to produce their own energy for manufacture, Lysen and Daey Ouwens (2002) estimated that the first world-wide net kWh on an annual basis was produced in 2002, and on a cumulative energy basis this will be produced in 2007. Since the use of batteries in PV-systems may add 5-15 years to the energy payback time of a PV-system (Paper V), it will take even longer for net energy to be produced from PV arrays. If PV and PV-battery systems are to contribute to a renewable energy system, it is important to improve the energy efficiency of all their components. Some of the emerging technologies studied (e.g. Li-ion and NaS) have a performance suitable for use in PV-battery systems, resulting in higher energy return factors and overall battery efficiencies than for established battery technologies. Calculations of the energy return factor and overall battery efficiency can be used to monitor the development of energy technologies and identify areas where improvement is necessary.

The energy payback time for wind turbines has been determined to be 2-3 months for an average modern turbine at an average site (Vindmølleindustrien 1997). With a service life of 20 years, the energy return factor is 80. The energy payback time is shorter for wind turbines than for PV since the conversion efficiency for wind is higher than for solar irradiation and the energy requirements for production are lower. The percentage of electricity generated, which was used to produce the plant and fuel, as well as decommissioning, has been estimated over the service life of different plants for electricity production to be as follows (Brännström-Norberg *et al.* 1996)²⁰: hydro 0.5%, wind 1.3%, nuclear 2.5%, biomass combined heat and power plant (CHP) 4%, natural gas CHP plant 4.5%, oil condensing power (CP) plant 13% and gas combined cycle CP plant 23%. The corresponding values for electricity from a PV system, as calculated in Paper V, are 6%-11% (*f*= 9 to 16) for the PV array and 8%-156% (*f*= 0.64 to 12) for the PV-battery.

However, comparisons between different generating technologies may be difficult since they can have different energy generation potential. As the solar energy flow is greater than wind energy, there is the potential to construct more PV systems than wind turbines. Other aspects that make comparisons difficult is that technologies with the same energy return factor can have different environmental impacts depending on requirement of scarce metals (PV) and whether the energy required for their production originates from fossil fuels or

 $^{^{20}}$ The indirect energy requirements can be expressed as a percentage of the energy output, $E_{l}/$ E_{O} = 1/f.
renewable sources. Regardless of the origin of the energy, the energy return factor provides a measure of efficiency, that can be used for benchmarking in the development of different PV and battery technologies.

4.2.4 Improving the environmental performance of battery systems

Improvement of battery systems should preferably be done in a systematic way, starting with the actions that have the greatest influence on the total system and the highest return on investment. The following strategies can be used to reduce the environmental impact of batteries (Modified from Karlsson *et al.* 1997): (1) reduce the flow of materials and energy by reducing weight and increasing efficiency, (2) slow down the flow of materials and energy by using lower power and increasing service life through maintenance, regeneration and ability to repair, (3) close the flow of materials and energy by re-using or recycling them in a closed or open loop, (4) replace the flow of materials and energy by using new solutions and less energy-intensive materials from a life cycle perspective. The consequences of an action need to be evaluated in a life cycle perspective in order to avoid sub-optimisation.

PV-battery systems can be made more energy efficient by matching operating conditions and battery characteristics in a life cycle perspective (Paper V). The energy efficiency of the PV-battery system can be increased by: (1) optimised charging algorithms, (2) passive temperature regulation to $\sim 20^{\circ}$ C, (3) increased utilisation of the active battery material, (4) lower material requirements for battery production and, (5) efficient production and transport of PV-battery system components.

One of the most efficient ways to reduce the environmental impact of battery systems is probably to educate the users of batteries. Instructions on the best way to handle batteries and how different actions influence the overall technical and environmental performance must be disseminated.

Organisations measure what they care about and want to develop. Environmental indicators can be used to inform different stakeholders of important parameters regarding the battery life cycle. They can be used to set up goals and monitor steering effects. The overall efficiency described in Paper V can be used as a complementary indicator to the energy return factor, which provides a measure of the total energy turnover of the system.

4.3 Recommendations for future research

Regarding the supply of environmental information and assessment of battery systems, research should be pursued in the following three areas.

- To reduce the uncertainties in substance flow analysis for batteries, further work could be done to monitor metal flows and estimate their fluctuations. Existing indicators of metal flows can be further developed and refined in order to allow assessment on different temporal and geographical scales. Metal flows arising from small sealed lead-acid batteries and different types of non-rechargeable (primary) batteries could be assessed. The bio-availability of metals and the exposure of organisms should be further explored.
- 2. Data on energy requirements for the production of batteries originate from different sources, which makes comparisons difficult as the system boundaries may be

inconsistent. Further work is therefore needed to improve the quality of data on material and energy requirements for batteries. For redox flow batteries, energy requirements should be determined for power and storage capacity, in order to enable evaluation of the potential benefits of independent sizing of power and capacity of redox flow batteries. The influence of temperature on battery service life should be further analysed. The effects of different battery charge-discharge efficiencies on the battery temperature and cooling requirements should also be further investigated. The energy model presented in Paper V can be developed to include functions for assessing the influence of the rate of battery capacity. The model can be extended to include the assessment of resource use, emissions and potential environmental impact of different technologies. PV systems with energy storage in flywheels and fuel cells with hydrogen storage should be evaluated and compared with batteries.

3. Uncertainties about future handling of products containing batteries make it important to assess leaching and mobilisation of metals from batteries. Despite difficulties in assessing the long-term behaviour of metals with leaching tests (Jones 1978, Oda 1989, Panero *et al.* 1995, Eriksson 2001), it is important to estimate emission factors for different metals and battery technologies.

In order to communicate environmental information efficiently between different actors research is needed in the following areas.

- 1. Battery systems involve several stakeholders throughout the life cycle their functions and demand for environmental information have to be assessed. Data on energy and material flows of battery systems are difficult to find, the data are not continuously updated and their use is restricted to a few stakeholders only. An information management system could be used to compile data on battery life cycles and make data more easily accessible. Procedures should be developed for co-operation between stakeholders, database accessibility and data security. Routines for measuring and updating data should be defined.
- 2. To motivate the interest of different stakeholders in managing energy and material flows of batteries, clear incentives must be introduced. The kind of incentives appropriate should be investigated, together with efficient ways of communicating feedback to stakeholders.

5 Conclusions

Critical environmental aspects have been identified and quantified for established and emerging battery systems. The environmental impact of a battery system is mainly influenced by its application and conditions of use and the choice of battery technology should be assessed for each specific application. In applications where batteries are difficult to collect at the end of their life, material flows and dissipative *losses of toxic metals* are of main concern. *Energy requirements* during production and usage are important for battery systems where the material losses throughout the battery life cycle are low.

For portable batteries, dissipative *losses of toxic metals* from incineration and landfills are of environmental concern. Indicators of global metal flows were used to assess the potential environmental impact of metals used in portable batteries. Metals of special concern were identified according to their lithospheric extraction indicators (LEIs), calculated for each metal as the ratio between the anthropogenic metal flow and natural metal flow. A LEI value greater than one, shows that the anthropogenic use of a metal exceeds the natural turnover and thus indicates a risk for substantially increased metal concentrations in the environment and thus increased environmental impact. The assessment of the battery market 1999 showed that cadmium and nickel are of concern due to their high LEIs (4.4-5.6). The ratios of battery metal flows to natural flows are 1.4 for cadmium, 0.07 for nickel and below 0.01 for other metals, indicating higher environmental concern for cadmium than for the other metals used in portable batteries.

For the projected market increase of NiMH and Li-based batteries assumed here, NiMH batteries may cause a net increase in nickel use compared with 1999. However, the use of nickel in batteries has a low ratio of battery metal flows to natural flows (0.06-0.3), indicating low potential environmental impact. For the other metals studied, the LEIs are below 0.7 and the ratios of battery metal flow to natural flow are below 0.06, which suggests that these material flows have a low impact on the global level. Emerging portable battery technologies (lithium-ion and nickel-metal hydride) have a lower impact, based on indicators of anthropogenic and natural metal flows, than the NiCd technology. A growing battery market indicates that portable batteries may be an important end-use of cobalt, neodymium, lanthanum, cerium, praseodymium and lithium. Increasing demand for these metals may result in higher metal prices, which may limit the growth of Li-ion(Co) and NiMH(AB₅) technologies. Higher prices of metals used in batteries may create incentives for battery collection and recycling.

Important parameters influencing the *energy use* throughout the life cycle of batteries were assessed through energy analysis of eight different battery technologies used in a standalone PV-battery system. Measures of the performance of the different battery technologies used in a PV-battery system were obtained by the energy return factor and the overall battery efficiency.

With a battery storage capacity three times higher than the daily energy output, the energy return factor for the PV-battery system ranged from 0.64 to 12 for the different cases. This means that 8.1-156% of the energy output is required to produce the PV-battery system. If the value of the energy return factor is less than one, the indirect energy used to produce

and replace the device is greater than the energy output. In this case the device works similar to that of a non-rechargeable battery, simply moving energy from one place to another. If PV-battery systems are to contribute to a renewable energy supply, it is important to improve the energy efficiency of all their components.

For a PV-battery system with a service life of 30 years, the energy payback time is 2.4-46 years, depending on the battery technology and operating conditions. The energy payback time is 1.6-3.0 years for the PV array and 0.55-43 years for the battery, showing the energy related significance of batteries in PV-battery systems. Some of the emerging technologies studied (e.g. Li-ion, NaS) exhibit performance suitable for use in PV-battery systems, resulting in higher energy return factors and overall battery efficiencies than for the established battery technologies.

The influence of different parameters on environmental impact and energy flows of battery systems have been assessed. Important parameters influencing *material flows* are material requirements for battery production, battery service life, collection efficiency and material recovery rate. Product characteristics of portable batteries, that are related to losses of metals, are the small size of each battery unit, the large number of battery owners, low concentration of economic value and type of application. Portable batteries also have a short effective service life, which increase the turnover of materials. Material flows of industrial batteries are easier to control due to the limited number of owners and the large size, which reduces the risk of loss and inappropriate disposal. To decrease losses of metals to the environment, collection of spent products is more important than the technical efficiency of recycling processes. NiCd battery recycling is energy efficient, even at very long transportation distances, at collection rates of 10-85%.

Important parameters affecting *energy flows* in battery systems are the battery chargedischarge efficiency, the type of cycling regime, the battery service life and the energy requirements for battery production.

In cases where the focus is on the efficient use of fossil fuels, and electricity generated by solar energy can be considered as a free energy source, a high energy return factor is important. This measure may be important in the expansion phase of PV-battery systems. Sensitivity analysis showed that the charge-discharge efficiency is the battery parameter with the highest influence on the energy return factor and is most important for lithium-ion, sodium-sulphur, polysulphide-bromide, vanadium-redox and zinc-bromine batteries. Service life, energy density and energy requirements for battery production are of equal importance for nickel-cadmium, nickel-metal hydride and lead-acid batteries.

The overall battery efficiency provides a measure of the efficiency of a closed renewable system, where renewable energy has to be used as efficiently as possible. The battery charge-discharge efficiency has the greatest influence on the overall efficiency. Lithium-ion and sodium-sulphur are emerging battery technologies with favourable characteristics in this respect.

The *environmental impact* of battery systems can be reduced by matching operating conditions and battery characteristics in a life cycle perspective. To decrease the environmental impact of battery systems, the development of battery technologies should aim at the *recycling of materials, increased service lives* and *higher energy densities*. To decrease the environmental impact arising from the use of *metals* in battery systems, metals with relatively *high natural occurrence* should be used, and regulations implemented to

decrease the need for virgin metals. To increase the overall *energy efficiencies* of battery systems, the development of battery technologies should aim at *higher charge-discharge efficiencies* and more *efficient production* and *transport* of batteries.

Different methods of assessing environmental aspects of battery systems have been applied. *Life cycle assessment* (LCA) is a comprehensive method of describing and identifying environmental aspects of battery systems. Life cycle inventory data for material groups can be used as estimates in life cycle assessments, when specific data on materials are not available. In this way, the environmental significance of a material can be identified at an early stage in the assessment. Following the identification of environmental hot spots, targeted methods may be applied. The eco-toxicity of metals is important when evaluating battery systems, and this is difficult to interpret in LCA. To thoroughly address eco-toxicity aspects, LCA can be combined with other methods, for example, environmental risk assessment (ERA), in order to model the exposure and response of organisms to different metals. ERA has extensive data requirements and the focus is to assess the risk of actual effects, that determine if measures have to be taken.

Substance flow analysis can be used for a simplified assessment of potential environmental impact when few data on the battery system are available. The method can be used to assess whether or not a technical option could solve a problem in principle on a macro level. By relating metal flows arising from the use of batteries to natural metal flows, the potential environmental consequences of current and future battery markets can be assessed. The method used is in line with the precautionary principle since it enables assessment early in the cause-effect chain, when few data on toxic effects are available. It can also be used to indicate whether environmental problems are simply shifted from one to another.

Industrial batteries have a higher turnover of energy than batteries in portable applications. Consequently, analysis of energy flows is important when assessing industrial batteries. *Energy analysis* can be used to assess the net energy output of renewable energy systems requiring energy storage in batteries. The energy return factor and the overall battery efficiency can be useful indicators of the battery system requirements of fossil fuels and electricity from a closed renewable system, respectively. The choice of conversion efficiencies for different energy qualities and system boundaries are crucial for a reliable assessment.

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Appendix

A. Critical assessment of LCA of battery recycling by ERM

In the LCA of battery recycling by ERM (2000), it is assumed that recycled materials can be modelled as virgin or recycled steel. Appendix A presents a sensitivity analysis of this assumption based on the available input data from ERM (2000). The conclusions and implications of the analysis are discussed in Section 4.1.7.

A1. Energy requirements for material production

To evaluate the consequences of using virgin and recycled steel in the battery waste management model by ERM (2000), primary energy is used as data category for environmental flows. Energy is descriptive for environmental flows since a significant fraction of the total air emissions and resource use is related to combustion of fossil fuels. A simple static linear model of end-of-life treatment of batteries is used to evaluate the sensitivity of different energy requirements for material production and collection rates.

The relative influence of a material depends on the produced quantity and the energy intensity for its production. The metals content of the projected waste arising in 2003 is made up of 22 643 ton consumer batteries with a composition of 40% unspecified materials, 23% iron and steel, 16% manganese and 14% zinc (ERM 2000, Table 2.16). Industrial and automotive are estimated at 158 kton with a composition of 61% lead, 23% unspecified materials, 8% iron and steel and 8% plastics (ERM 2000, Table 3.9). The large fraction of unspecified materials includes energy intense metals e.g. nickel, cobalt, zinc and copper. Nickel-containing iron scrap from NiCd batteries can be profitably recycled in the steel industry (Paper I).

The energy requirements for virgin production of steel from cradle to gate are 26-35 MJ/kg (IISI 2000), while it is 8-11 times higher for nickel (NIDI 2000), 1.2-1.6 times higher for lead (Boustead and Dove 1998), 1.5-2 times higher for zinc (Boustead and Dove 1998) and 2-4 times higher for different kinds of plastic (APME 1998).

The benefit of recycling is that recovered materials displace the need for producing the same material in another way. Recycled materials production has lower energy requirements than virgin materials production since energy for extraction from mines and reduction of metals are allocated to the first material life cycle. For recovery of materials to primary grade the energy requirements are 5-50% of virgin production (Boustead and Hancock 1979, Sunér 1996, Paper I and IV). Transportation for collection may contribute 1-40% of the total energy requirements for recovered materials (Boustead and Hancock 1979, Paper I).

Fig A2 and Fig A3 shows that energy requirements for production and recycling of materials are the most important activities for the total energy use. Fig. 7 shows the relative contribution of different activities when also battery manufacturing is included. The assumption by ERM (2003) that steel production and recycling can be used to model all

different kinds of materials is therefore not appropriate since this parameter explains 70-99.8% of the total energy use, as shown in Fig. A2 and Fig. A3.

Fig A3 shows that 60% materials recovery of separately collected batteries is more energy efficient than landfilling at collection rates higher than 11% for a worst case assumption when only lead is recovered.

Based on the estimated waste flow of industrial and automotive batteries in the UK 2003, 158 kton/year batteries are disposed of in landfills when batteries are collected with the municipal solid waste (MSW). In the case of 50% separate battery collection and 60% materials recovery, 79 kton/year is disposed of in landfills and 32 kton/year non-recoverable battery materials can be disposed of in controlled landfills. A benefit of separately landfilled metals is that they are potentially recoverable in the prospects of waste mining for resources in the future.

A2. Material flows of batteries

The battery waste flow in Fig. A1 is indicated by $m_{Battery waste}$, the fraction of the total battery waste stream that is separately collected is given by $C_{Battery}$, and the fraction of recovered battery materials is given by R.

Since the waste flow arises when the battery is being disposed of, there is a time lag (1-25 years) between material requirements for battery production and recovered materials. If the battery market is at steady state, closed loop recycling can be assumed. If the material requirements are lower for battery material production than the amount of recovered materials, open loop recycling can be assumed and recovered materials displace the need for virgin metals in other applications.



Fig. A1. Activities included for modelling end-of-life treatment of waste batteries at steady state. The mass of annual battery waste is indicated by $m_{Battery waste}$, the fraction of batteries collected separately is C, and the recovery rate of battery materials is R. For modelling of a long-term perspective, the system boundary may be extended to include the dashed box with leachate water treatment and soil remediation.

A3. Sensitivity analysis of energy requirements for production of battery materials

To evaluate the consequences on the total energy use depending on the energy requirements for virgin and recycled production of materials, waste management of automotive and industrial batteries is modelled for a case representing the UK 2003 (ERM 2000). Conclusions of the results are discussed in Section 4.1.7.

Using the model, direct landfilling of batteries is compared with 50% separate collection and 60% recovery of battery materials. The balanced materials flows are shown in Fig. A4 and the input parameters are shown in Table A1. Fig A2 shows the significance of the energy requirements for production of virgin materials, which is assumed to correspond to virgin steel production as in the study by ERM (2000). Virgin materials production contributes 71.1-99.8% of the total energy use when the energy intensity is 30 MJ/kg virgin steel and 10 MJ/kg for recycled steel (Table A1).

In the case of collection and recycling, transportation and recycling of battery materials are of the same magnitude. This is due to that transportation is assumed to be very long distance with a light truck, representing a worst case scenario where transportation and collection is 84 times more energy intensive than direct landfilling (Table A1). The transportation distance is assumed to be 20 times longer for separate collection and recycling than for direct landfilling (2 000 km vs. 100 km). The energy intensity for transportation is 4.2 times higher in the case of separate battery collection and recycling than for direct landfilling.

In the case of battery collection with the municipal solid waste (MSW), 158 kton batteries are disposed of in landfills (a fraction may be incinerated beforehand, but it has been excluded). If 50% of the waste batteries are collected separately, 79 kton is disposed of in landfills and 32 kton of non-recoverable battery materials can be disposed of in controlled landfills.

The total energy requirements are 2% lower for 50% collection than for 100% landfilling with the selected input parameters. When the collection rate exceeds 36%, the total energy requirements are lower than for direct landfilling. This means that even for very energy inefficient transportation for long distances, recycling is more energy efficient than landfilling.



Fig. A2. Energy use for end-of-life treatment of industrial waste batteries. The mass of landfilled metals is indicated by m, the fraction of batteries collected separately is C, and the recovery rate of battery materials is R. Energy requirements for virgin and recycled materials production are valid for steel. Input data is presented in Table A1.

To evaluate the consequences on the total energy use depending on the energy requirements for virgin and recycled production of materials, a more realistic assumption is made than in the study by ERM (2000). Instead of assuming that lead can be modelled as steel (see footnote ¹⁸), energy requirements for lead is used since it is estimated to contribute 61% to the total waste flow of automotive and industrial batteries (ERM 2000, Table 3.9). From the collected batteries, 60% of the material from is recycled and replace the need for virgin metal production. This is a low recovery rate since it assumes that 16.3% of the total separately collected batteries to be landfilled, which includes 8% plastics, 8% steel, 0.14% nickel and 0.12% cadmium. All these materials have higher energy intensity for virgin production than lead, and can also be efficiently recovered. Fig A3 therefore represents a worst case assumption.

The total energy requirements are 12% lower for 50% collection than for 100% landfilling with the selected input parameters. When the collection rate exceeds 11%, the total energy use is lower than for direct landfilling. Increasing collection results in decreased total energy use until a level when very long distances have to be driven in order reach very high collection rates. The reduction of battery materials to landfill is reduced in the same quantities as in Fig. A2.



Fig. A3. Energy use for end-of-life treatment of industrial waste batteries. The mass of landfilled metals is indicated by m, the fraction of batteries collected separately is C, and the recovery rate of battery materials is R. Energy requirements for virgin and recycled materials production are valid for lead. Input data is presented in Table A1.

A4. Summary of results and input data

Fig. A4 shows that the transportation distance is assumed to be 100 km and the energy intensity for transportation corresponds to a heavy truck with trailer in the case of the direct landfilling of batteries, (NTM 2003). The transportation distance is assumed to be 2000 km for battery collection (C=50%) and recycled materials (R=60%), respectively, and the energy intensity correspond to an inefficient light truck (NTM 2003). The energy intensity for transportation is assumed to be 84 times higher than for the case of direct landfilling. The inefficient truck transportation is assumed to include the extra energy requirements for production of collection bins and other components required for separate collection. No energy recovery is assumed for the case with direct landfilling.

No transportation distances were reported in the study by ERM (2000).



Fig. A4. Parameter settings for modelling of energy flows of waste industrial and automotive batteries. See Table A1 for details. Gray=0% collection, White= 50% collection, 60% material recovery

Table A1 shows the results from two parallel cases when the energy requirements for virgin and recycled material production are assumed to be steel and lead, respectively (bold numbers). For the case with direct landfilling, the total energy requirements are 2-13% higher than in the case with recycling.

	C= 0%	C=50%,	C=0%	C=50%,
		R=60%		R=60%
Mass battery waste (kton) ^a	158	158	158	158
Collection separate (%)	0	50	0	50
Recovery of battery materials (%)	0	60	0	60
Q virgin materials production (MJ/kg)	^b 30	^b 30	^c 41	^c 41
Q recycled material production (MJ/kg)	^d 10	^d 10	e 7	^e 7
E production virgin materials (TJ)	4 740	3 318	6 478	4 535
E production recycled materials (TJ)	0	474	0	332
E transport MSW collection (TJ)	11	114	11	114
E transport separate collection (TJ)	0	474	0	474
E transport recycled materials (TJ)	0	284	0	284
E total (GJ)	4 751	4 664	6 489	5 739
m landfilled batteries total (ton)	158	111	158	111
Fraction separate battery waste of total landfilled (%)	0	29	0	29

Table A1. Input parameters and results of waste management of automotive and industrial batteries in the UK 2003.

^a Estimated waste from automotive and industrial batteries in 2003 (ERM 2000, Table 3.9)

^b Virgin steel production 26-35 MJ/kg (IISI 2000). 32 MJ/kg (Ullman 1995).

^c Virgin lead production (Boustead and Dove 1998). 25 MJ/kg (Ullman 1995).

^d 10 MJ/kg (Boustead and Hancock 1979). Recycled steel has 36% of the energy requirements of virgin steel production (Sunér 1996). 15 MJ/kg (Ullman 1995). ^e 7 MJ/kg (Boustead and Hancock 1979). 8 MJ/kg (Ullman 1995).

B. Errata in the appended papers

Table A2. Discovered errors in the published papers.

Paper and page	Reads now	Should read
Paper I		
Y legend in Fig. 1.	No. of cell	No. of cells
p. 294, Row 6.	(Edwards and Schelling, 1999)	(Wiaux, 1999)
p. 298, Row 6.	The use of renewable energy remains constant at increasing recycling rates but non-renwable energy use de- creases by 5% when	The use of energy decreases by 13% when
p. 299, Fig. 3	(Black field in graph)	See: Page 26, Fig. 7
p. 307, References	J. Transp Res 2001;Port D(6):297-324	J. Transp Res 2001;Part D(6):297-324
Paper II p. 169, Column 2, Row 30	(Azar et al., 1996)	(Delete the reference)
p. 176, Column 2, Row 22	Fig. 4 shows that metals contributing >1% to the global metals flows are	Fig. 4 shows that metals contributing >1% to the global mine production are
p. 177	On the demand side, Cd is becoming a one-use metal, and approximately 70% is used in the manufacture of NiCd batteries (Plachy 2000b).	On the demand side, Cd is becoming a one-use metal, with approximately 70% used in the manufacture of NiCd batteries (Plachy 2000b).
p. 178, Text below Table 5	The change relative LEI99 is also given. Source: calculated from data in Tables 2-4.	<i>(Delete the sentence)</i> Source: calculated from data in Tables 2-4.
p. 178, Column 2, Row 6	The reduction is due to the low natu- ral occurrence of cadmium and the high metal intensity for storing elec- trical energy.	(Delete the sentence)
p. 178, Table 5, Columns: Low – high (%) Row: Cd	-11-51	-5111
p. 180, Below Table 6	The change relative LEI99 is also given.	(Delete the sentence)
p. 180, Table 6, Column: Low – high, Row: Cd	-8.0-38	-388.0
(The same figures in three columns)		
p. 182	The assessment of global metal flows arising from the use of portable re- chargeable batteries shows that Ni and Cd are of concern due	The assessment of global metal flows arising from the use of portable recharge- able batteries shows that Cd and Ni are of concern due

Continued: Table A2. Discovered errors in the published papers.

Paper and page	Reads now	Should read
Paper III		
Table 4, Row 7,	96	64
Column 4		
Fig. 1	(Arrows are missing)	(Vertical arrows in both directions should connect the boxes) Goal \leftrightarrow Inventory and Inventory () Impact associated
		Inventory \leftrightarrow Impact assessment

Paper I

Rydh, C. J. and Karlström, M. (2002) Life Cycle Inventory of Recycling Portable Nickel-Cadmium Batteries, *Resources, Conservation and Recycling* Vol. 34, No. 4, p. 289-309

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Resources, Conservation and Recycling 34 (2002) 289-309



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Life cycle inventory of recycling portable nickel-cadmium batteries

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Abstract

In this study, the environmental impact of recycling portable nickel-cadmium (NiCd) batteries in Sweden is evaluated. A life cycle assessment approach was used to identify life cycle activities with significant impact, the influence of different recycling rates and different time boundaries for emissions of landfilled metals. Excluding the user phase of the battery, 65% of the primary energy is used in the manufacture of batteries while 32% is used in the production of raw materials. Metal emissions from batteries to water originate (96-98%) from landfilling and incineration. The transportation distance for the collection of batteries has no significant influence on energy use and emissions. Batteries manufactured with recycled nickel and cadmium instead of virgin metals have 16% lower primary energy use. Recycled cadmium and nickel metal require 46 and 75% less primary energy, respectively, compared with extraction and refining of virgin metal. Considering an infinite time perspective, the potential metal emissions are 300-400 times greater than during the initial 100 years. From an environmental perspective, the optimum recycling rate for NiCd batteries tends to be close to 100%. It may be difficult to introduce effective incitements to increase the battery collection rate. Cadmium should be used in products that are likely to be collected at the end of their life, otherwise collection and subsequent safe storage in concentrated form seems to offer the best solution to avoid dissipative losses. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Nickel-cadmium batteries; Recycling; Life cycle assessment; Materials management

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1. Introduction

Portable nickel-cadmium (NiCd) batteries have been used in electronic products for many decades. In 1990, cadmium from portable NiCd batteries contributed 60% (84 ton Cd) of the inflow of cadmium in products to the Swedish society (Bergbäck, 1992). The use of portable NiCd batteries has decreased in Sweden during recent years but cadmium in portable and industrial NiCd batteries still contributes 90% (41 ton) of the inflow of cadmium-containing products in Sweden in 1998 (Olsson, 2000). The market for portable electronic products is expanding and the global production of portable rechargeable batteries grew at an annual rate of 14% during the past 10 years (Fig. 1). The total number of portable cells produced in 1999 was 2.9×10^9 cells (excluding $\approx 0.3 \times 10^9$ small sealed lead-acid cells). The introduction of new types of electrochemical systems (nickel-metal hydride, lithium-ion and lithium-polymer batteries) has decreased the global market share of NiCd batteries from almost 100% in 1992 to 49% in 1999. NiCd battery production in Japan, accounting for 42% of NiCd batteries manufactured globally 1998, increased by 29% during 1990–1994, but later decreased by 33% during 1994–1998 (Fujimoto, 1999).

In September 1997, the European Commission called for a ban on portable NiCd batteries to be introduced in 2008 because of poor recovery rates. Stakeholders (EPBA, 1999; Sempels, 1999; CollectNiCad, 2001) claim that the economic effects will be unacceptable and increased collection and reprocessing of portable NiCd batteries may reduce the environmental effects of NiCd batteries significantly.

In 1998, the Swedish battery ordinance came into effect, which states that all kinds of household batteries must be collected to avoid the spread of cadmium, mercury and lead. In 1998, approximately 1400 ton of mixed household batteries were collected in Sweden (Olsson, 2000). The only types of batteries sent for



Fig. 1. Number of rechargeable portable battery cells produced globally from 1989 to 1999 (portable lead-acid batteries not included) (Fujimoto, 1999; Noréus, 2000). (NiCd, nickel cadmium, NiMH, nickel-metal hydride, Li-ion, lithium ion).

materials recovery are NiCd (8 wt.% of the amount collected), small lead-acid batteries (3 wt.%) and nickel-metal hydride (1 wt.%), while the rest is landfilled (Renova, 1999).

Improved material management can lead to better utilization of refined materials, decreased use of primary materials and energy resources and a reduced need for landfill areas. The benefits of recycling materials from an economic, environmental and technical point of view depend on many parameters, such as transport distance, recycling processes and type of material. Louis et al. (1998) reviewed 314 articles on studies of the economics and environmental costs and benefits of recycling post-consumer waste. They concluded that there were major gaps in the literature regarding the assessment of the environmental costs of recycling, and no analyses supported any particular target level of recycling.

Recycling of NiCd batteries is interesting, since cadmium has been a political issue for many years, and the substance is toxic and geologically scarce. One of the proposed criteria for the EU environmental labelling of batteries (Baumann and Scholl, 1997) requires the importer of batteries to join a battery-recycling programme. However, only a few studies have quantified the benefits of battery collection and the recovery of metals (Hofstetter and Häne, 1990; Törnblom, 1996; Lankey, 1998; Karlsson, 1999). Conclusions from an evaluation of mature NiCd battery technology may be used to assess emerging new electrochemical systems.

2. Aim and scope

The aim of this study was to assess the environmental effects of recycling portable NiCd batteries in Sweden and to identify life cycle activities with significant potential environmental impact. The sensitivity of the NiCd battery system has been evaluated by varying recycling rates and emission factors for landfilled metals. Materials management of cadmium is discussed based on the results obtained.

The assessment was made using a life cycle inventory (LCI), which includes compiling an inventory of environmentally relevant inputs and outputs related to the functionality of a product (ISO, 1997). When using LCI methodology increased recycling normally will decrease energy use and metal emissions for most kinds of product systems. The contribution of this study is to quantify these benefits for a NiCd battery system with static modelling.

LCI was chosen as environmental analytical tool, because it includes if decreased emissions are shifted to other environmental problems and compared to other methods (e.g. material flow analysis, cumulative energy requirements analysis) it includes potential environmental impact connected to the material and energy flows.

Environmental impact related to metal emissions and energy use are regarded as the most significant activities for the batteries (Rydh, 2001). Environmental impact assessment was not performed since the environmental impact is strongly connected to these activities.

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The functional unit of the study was defined as 'a battery with an energy storage capacity of 1.0 W h electrical energy'. This corresponds to a cylindrical NiCd battery with a mass of 25 g (40 W h/kg).

Emissions and resource use in the user phase of the battery were excluded from the study since these do not influence the materials management of metals for the functional unit chosen. Various kinds of end-of-life treatment (recycling, landfilling and incineration) were considered. When possible, allocation procedures proposed by ISO, (1998) where applied to distribute environmental impact from activities with multiple outputs. Closed-loop recycling was assumed for cadmium and nickel.

It was assumed that the NiCd batteries were manufactured in Germany and used in Sweden. Data on raw materials extraction and refining (excluding capital and personnel) from cradle to gate are based on average data from manufacturers. Average transportation distances are estimated for materials production, collection and recycling of batteries in Sweden. Emissions from electricity generation (extraction, refining and combustion of fuels) were calculated for base case based on a country specific mix for electricity generation. The sensitivity of the model was evaluated by comparing the base case with hydro and coal power electricity generation, respectively.

Metal emissions from landfills to water are modelled for a surveyable time corresponding to 100 years, and in a long-term perspective, representing a worst case scenario or hypothetical infinite time when all metals have been completely released to the environment (Finnveden et al., 1995).

Inventory data were compiled from LCA databases, literature, interviews and reports on NiCd batteries (Kertes, 1996; Jensen and Petersen, 1999). Information on collection and sorting of batteries by municipalities was acquired through questionnaires.

3. Inventory

3.1. Manufacturing

Inventory data used for the calculations are presented in Rydh and Karlström (2001). The most common sizes of rechargeable portable battery cells are AA and Sub C cells (weight ≈ 22 and 50 g, respectively). Table 1 shows the material requirements corresponding to the functional unit of 1 W h, almost equivalent to a cell of AA size (average voltage = 1.2 V, capacity = 0.6–0.8 A h).

The average primary energy use for extraction and refining of cadmium (from zinc mining) and nickel has been estimated to be 70 MJ/kg Cd (Boustedt and Dove, 1998) and 159 MJ/kg Ni (Kirmayer, 1995), respectively. The primary energy requirements for manufacturing processes of batteries produced in Hagen, Germany were calculated to be 140 MJ/kg battery (Kirmayer, 1995). Metal emissions were approximated from the manufacture of stationary NiCd batteries in Sweden.

Table 1 Material requirements	for a 1 W	⁄h porta	ble nicke	el–cadmium ba	ttery (g)							
Components	Materia	ls										
	Steel	ïŻ	Cd	Hydroxides	H ₂ O (demin.)	KOH (30%)	ΡA	PVC	Co	Rubber ^b	Sum	Wt.%
Negative electrode	0.77	1.16	3.88	0.29							6.10	24.4
Positive electrode	1.65	2.93	0.21	1.74					0.12^{a}		6.65	26.6
Electrolyte					2.00	0.86					2.87	11.5
Separator							0.53				0.53	2.1
Can, header, cover	7.38										7.38	29.5
Vent										0.06	0.06	0.3
Current collector	1.02										1.02	4.1
Label								0.26			0.26	1.1
Gasket							0.13				0.13	0.5
Sum	9.80	5.11	4.09	2.03	2.00	0.86	0.65	0.26	0.12	0.06	25.00	100.0
Wt.%	39.2	20.5	16.4	8.1	8.0	3.5	2.6	1.1	0.5	0.3	100.0	
Source: processed data	a Kirmayei	r (1995).										
b Assumed to by po	dypropylen	e.										

3.2. Collection and recycling

The routes for final disposal are not well known for portable NiCd batteries since the in- and out-flows from the technosphere are difficult to quantify. It is uncertain how many NiCd cells enter the market since they may be assembled in packs containing variable numbers of cells. Given that NiCd batteries are used in power packs or are integrated in equipment, it is probable that large amounts of batteries are still being stockpiled and that steady state between in- and out-put has not yet been reached (Edwards and Schelling, 1999). It is thus uncertain where accumulated batteries will end up in the future. The registered annual supply of NiCd batteries to Sweden during the period 1990-1995 was 450-635 ton (mean 547 ton) (SCB, 2000). An average of 142 ton of NiCd batteries were collected annually from 1996–1999. Assuming a lifetime for NiCd batteries of 6–9 years gives a collection rate of 22-31% (mean 26%). Recent collection results indicate, however, that the amount of batteries collected is increasing. Another way to determine the recycling rate is to compare the amount of batteries collected with the amount found in municipal solid waste (MSW). However, sorting studies of mixed MSW have been carried out on a few samples only, which makes them less reliable. In the base case, it is assumed that end-of-life pathways for NiCd batteries are distributed between recycling (25%) and MSW. Of the MSW fraction, 60% is incinerated and 40% is landfilled. The fraction of batteries lost directly to the environment is not taken into account as the amount is so small (Rydh, 1999).

Modelling of consumer car transportation to recycling sites and local truck transports of batteries were adapted from a study of glass collection (Edwards and Schelling, 1999). The model considers that the recovery rate increases with increasing recycling site densities. Shorter distances per kg recovered battery have to be driven by private cars at higher site densities, thus leading to a minimum at recovery rates greater than 90%. The transportation distances involved in collecting mixed household batteries from battery collection boxes and taking them to a central point within a municipality vary in the range 30–250 km (average 100 km) for the different municipalities in Sweden. Batteries are often transported together with other waste. The fuel consumption per kg recovered battery for local truck transports is considered to be constant for recovery rates 10–85% (Edwards and Schelling, 1999). At higher recovery rates, the fuel consumption increases rapidly due to longer distances to cover all sites and the decreasing amount of material available per site. Energy use for production of battery collection boxes was excluded since it was found to be insignificant.

The municipalities are responsible for sorting the batteries into different fractions. Twenty-five percent of the portable batteries collected annually in Sweden is sent to Gothenburg for sorting in a semi-automatic sorting machine (Renova, 1999). Since NiCd and small lead-acid batteries are the reason for the collection of household batteries, the environmental impact of transportation of all household batteries is allocated to these types of batteries. The fraction of NiCd batteries (8 wt.%) of all household batteries collected is transported an average distance of 600 km to AB SAFT in Oskarshamn, Sweden for cadmium recovery. In 1998, 143 ton portable NiCd batteries, corresponding to 19 ton of cadmium, were recycled at AB SAFT, Oskarshamn, Sweden (SAFT, 1983–1998). Recoverable materials (76 wt.% of NiCd battery) are cadmium and nickel-iron scrap. The cadmium recovered is used in the production of new industrial NiCd batteries at SAFT, and nickel-iron scrap is sent to smelters for use as alloying metal in the steel industry. However, in this study, it is assumed that the cadmium recovered is used in the production of new portable batteries to avoid the use of different allocation procedures, which must be applied when recycling materials in cascade.

Heat energy is recovered from plastics (4 wt.% of NiCd batteries). The input of heat energy to the recycling process is recovered in the production of positive active material (drying of nickel hydroxide) in another process in the factory. Allocation of energy between the two product systems was made by system expansion (ISO, 1997), where the energy recovered from battery recycling is assumed to replace liquid petroleum gas which would otherwise have to be used at the plant if the recycling process had not been equipped with a heat exchanger.

3.3. Waste incineration and landfilling

In Sweden, approximately 60% of MSW (excluding coarse waste) is incinerated in waste incineration plants. The proportion of MSW incinerated is expected to increase in the future due to higher fees for the deposition of combustible waste. MSW incineration plants in Sweden are equipped with extensive air and water cleaning equipment to capture pollutants. However, small amounts of metals bound to aerosols may escape through filters. Metals are stored in bottom and flue ash. Organic sulphides are added to the ash for complexation of metals, and lime is added to maintain an alkaline environment. The stabilised sludge is landfilled at municipal sanitary landfills. Forty percent of the MSW is landfilled directly, which means that almost all metals entering the waste stream end up in landfills, if the metals are not recovered from the ash. Depending on the time perspective chosen. different amounts of metals will leached out into the environment giving rise to potential environmental impact. The rate of degradation and mobilisation of metals from batteries is very complex and depends on battery-specific parameters (type of casing and state of charge) as well as site-specific parameters (e.g. pH, redox potential, amount of oxidising agents, cation-exchange capacity, organic content, solid-to-liquid ratio and soil texture) (Jones et al., 1978). In degradation tests of NiCd batteries in landfills, Oda, 1989 concluded that cadmium from whole batteries would not be released for two to four decades in a landfill subjected to normal rainfall.

A crucial methodological issue in life cycle assessment is determining the system boundary for the cradle and the grave of a product system. Finnveden (1996) suggested that both a short- and long-term perspective should be considered. The short-term scenario is a surveyable time, which is the later part of the methane stage until a pseudo-steady-state has been reached. This period is of the magnitude of one century in northern Europe, but may be shorter in countries with higher average temperatures. Emission factors are calculated by considering the concentrations of metals in the leachate from landfills, the amount of leachate, and the metals in waste. In a short-term perspective the emission of metals from batteries may be very low.

Large amounts of heavy metals stored in landfills pose a potential threat in a long-term perspective. In this study, the long-term scenario is a hypothetical, infinite period, which is the period from landfilling until the landfilled material has been completely released to the environment and has become part of the biosphere (Finnveden, 1996). It is assumed that landfilled metals are corroded but immobilised as solid compounds, or in some cases as ions, adsorbed in the waste matrix or in the biomass, which prevents them from being biologically available. However, due to the laws of equilibrium, these products will be continuously leached out by percolation. Landfill erosion, fire or flooding may change the environmental conditions and cause a major release of metals.

The emission factors for a surveyable time are very small compared with the long-term perspective (landfilled metal to water: 5×10^{-3} kg Ni/kg Ni and 5×10^{-4} kg Cd/kg Cd) (Finnveden, 1996). Although metal emission factors found in the literature vary by several orders of magnitude, the values indicate that it will take thousands of years to mobilise and disperse accumulated metals.

4. Results

4.1. Activities with potential environmental impact

The following results represent the assessment of the current situation regarding battery handling in Sweden where 25% of NiCd batteries are recycled, 45% incinerated and 30% landfilled. Table 2 shows that 3.1% of the total primary energy is based on renewable energy, 65% of the primary energy is used in battery manufacture and 32% is used in the production of raw materials. The processes of electrolysis of the negative electrode and impregnation of the positive electrode contribute significantly to the energy use in the manufacturing phase. Of the total energy, 0.9% is used in battery distribution, collection and sorting.

Fifty-five percent of the CO_2 emission originates from battery manufacturing, 44% from raw materials production and 0.8% from battery distribution, collection and sorting. Transportation in the materials production phase contributes 1% to the total CO_2 emission.

Fifty-six percent of the NO_x emission originates from materials production. Collection and sorting contribute 10% to this emission since short-distance transportation gives rise to high NO_x emission and the transported mass of batteries is high since all the household batteries collected are included. Assuming doubled transportation distances for collection and recycling increases the total NO_x emission by 4% while energy use and other emission change insignificantly.

Consumption of (metal) resources is ascribed to unit processes, which disperse resources (concentrated materials) and make them difficult to recover. Consequently, the greatest resource use is found in the incineration and landfilling activities.

Cd) (%)extraction/refining (%)manufact. (%)sorting (%)(%)landfill (%)Renewable energy0.1633.38.649.108.50.5(MJ/W h)8.649.108.50.5(MJ/W h)8.60.91.30.1Non-renew. energy4.9226.84.155.20.80.1(MJ/W h)0.3739.84.155.20.8-0.20.3 $(MJ/W h)$ 0.3733.622.030.810.2-0.20.3 $(MJ/W h)$ 0.5033.622.030.810.2-0.10.0 $(MJ/W h)$ 0.5033.62.74.6000 $(SW h)$ 3.10000000.0 $(g/W h)$ 3.800000100.0 $(g/W h)$ 3.800000100.0 $(g/W h)$ 3.800000100.0	Selected inventory da	Total	Raw mat'l (excl.	Cd	Battery	Collection and	Recycling process	Incineration and
$ \begin{array}{llllllllllllllllllllllllllllllllllll$			Cd) (%)	extraction/refining (%)	manufact. (%)	sorting (%)	(%)	landfill (%)
	Renewable energy (MJ/W h)	0.16	33.3	8.6	49.1	0	8.5	0.5
$ \begin{array}{cccccc} \dot{CO}_{a}^{a}\left(kg/Wh\right) & 0.37 & 39.8 & 4.1 & 55.2 & 0.8 & -0.2 & 0.3 \\ NO_{x}\left(g/Wh\right) & 0.50 & 33.6 & 22.0 & 30.8 & 10.2 & 3.1 & 0.0 \\ SO_{x}\left(g/Wh\right) & 4.17 & 92.6 & 2.7 & 4.6 & 0 & -0.1 & 0.0 \\ d\left(resource\right) & 3.1 & 0 & 0 & 0 & 0 & 0 & 100.0 \\ g/Wh & & & & & & & & & & \\ Ni \left(resource\right) & 3.8 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ Ni \left(resource\right) & 3.8 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ g/Wh & & & & & & & & & & & & & & & & & & &$	Non-renew. energy (MJ/W h)	4.92	26.8	4.8	66.0	0.9	1.3	0.1
$ \begin{array}{ccccccccc} NO_x \left(g/W h \right) & 0.50 & 33.6 & 22.0 & 30.8 & 10.2 & 3.1 & 0.0 \\ SO_x \left(g/W h \right) & 4.17 & 92.6 & 2.7 & 4.6 & 0 & -0.1 & 0 \\ Cd \left(resource \right) & 3.1 & 0 & 0 & 0 & 0 & 0 & 100.0 \\ \left(g/W h \right) & & & \\ Ni \left(resource \right) & 3.8 & 0 & 0 & 0 & 0 & 0 & 100.0 \\ Ni \left(resource \right) & 3.8 & 0 & 0 & 0 & 0 & 0 & 100.0 \\ SV h \end{array} $	CO_2^{a} (kg/W h)	0.37	39.8	4.1	55.2	0.8	-0.2	0.3
$ \begin{array}{ccccccccc} \mathrm{SO}_{x} \left(\mathrm{g}/\mathrm{W} \mathrm{h} \right) & 4.17 & 92.6 & 2.7 & 4.6 & 0 & -0.1 & 0 \\ \mathrm{Cd} \left(\mathrm{resource} \right) & 3.1 & 0 & 0 & 0 & 0 & 100.0 \\ \mathrm{g}/\mathrm{W} \mathrm{h} & & & \\ \mathrm{Ni} \left(\mathrm{resource} \right) & 3.8 & 0 & 0 & 0 & 0 & 0 \\ \mathrm{Ni} \left(\mathrm{resource} \right) & 3.8 & 0 & 0 & 0 & 0 & 0 & 100.0 \\ \mathrm{g}/\mathrm{W} \mathrm{h} & & & & \\ \mathrm{g}/\mathrm{W} \mathrm{h} & & & & \\ \end{array} $	NO_x (g/W h)	0.50	33.6	22.0	30.8	10.2	3.1	0.0
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	SO_x (g/W h)	4.17	92.6	2.7	4.6	0	-0.1	0
Ni (resource) 3.8 0 0 0 0 0 100.0 (g/W h)	Cd (resource) (g/W h)	3.1	0	0	0	0	0	100.0
	Ni (resource) (g/W h)	3.8	0	0	0	0	0	100.0

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4.2. Evaluation of different recycling rates

Table 3 shows a minimum at 90% recycling rate for energy use and NO_x emissions. The minimum is due to the fact that recycled materials and longer transportation distances have less impact than extraction and refining of virgin materials. At recycling rates greater than 90%, local transport for emptying collection boxes and delivery of batteries to sorting plants increases rapidly.

The use of renewable energy remains constant at increasing recycling rates but non-renewable energy use decreases by 5% when increasing the recycling rate from 25 to 90%. The difference between 25 and 90% recycling is a decrease of approximately 87% for cadmium and nickel resource use and emissions. CO_2 and SO_x emissions decrease by 30 and 80%, respectively. Extraction and refining of virgin nickel give rise to high SO_x emission, which decrease significantly at higher recycling rates.

The total NO_x emission decreases by 39% when comparing no recycling with 90% recycling (Fig. 2.). The contribution of transportation and sorting increases from 7.5 to 53%. The minimum total NO_x emission is found at a 90% recycling rate since it is modelled that increased local truck transportation for collection is needed to achieve very high collection rates. When no batteries are recycled, battery materials contribute most to the total NO_x emission (35%) followed by battery manufacturing Cd and mining/refining. At recycling rates greater than 80%, the recycling process leads to a net reduction in NO_x emission due to higher efficiency than the alternative use of liquid petroleum gas. At different recycling rates, the recycling activity contributes to 4 to -1% of the total NO_x emissions. No significant

Table 3

Selected inventory data for the NiCd battery life cycle (excl. user phase) for different end-of-life treatment methods

	Landfills 100%	Incineration 60%/landfills 40%	Recycling 90%/incineration 6%/landfills 4%	Recycling 100%
Renewable energy (MJ/W h)	0.16	0.16	0.14	0.14
Non-renew. energy (MJ/W h)	5.18	5.15	4.29	4.32
CO_2^{a} (kg/W h)	0.41	0.41	0.26	0.26
NO_x (g/W h)	0.56	0.56	0.34	0.47
SO_x (g/W h)	5.45	5.45	0.83	0.32
Cd (resource) (g/W h)	4.1	4.1	0.41	0
Ni (resource) (g/W h)	5.1	5.1	0.51	0

^a Carbon from fossil sources.


Fig. 2. NO_x emission from the NiCd battery life cycle (excl. user phase) at different recycling rates.

differences were found among the analysed data categories if other energy sources (natural gas, hydropower and coal power) were used in the recycling process instead of liquid petroleum gas.

Fig. 3 shows that an increase in recycling rate from 0 to 90% decreases the total primary energy use by 17%. As a percentage of the total energy use, collection and sorting energy increases from 0.6 to 5%, while energy use in raw materials production decreases from 36 to 15%. By using recycled metals, the energy for the



Fig. 3. Primary energy use of the NiCd battery life cycle (excl. user phase) at different recycling rates.

processing of battery raw materials is reduced by 65% compared with virgin materials only. Energy use in the battery manufacturing activity remains constant irrespective of the recycling rate.

The way of electricity generation may alter the absolute values of primary energy use. If all electricity is generated by hydropower, the total primary energy is 2.1–2.8 MJ/W h and NO_x emission 0.17–0.31 g/W h. Corresponding values for coal power is 8.4–10 MJ/W h primary energy and 91–107 g NO_x/W h. Compared to the country specific electricity mix, primary energy use is reduced by half or doubled depending on the energy conversion efficiencies of the different power sources.

To quantify the energy savings made by using secondary materials, the total energy required for recycling must be allocated between the nickel and cadmium recovered. Allocation on a mass basis is preferable to economic allocation since physical parameters are constant (ISO, 1998). Economic values of recovered metals fluctuate over time and do not follow open market prices since the Swedish battery fund pays for the recycling of NiCd batteries. Considering primary energy for collection and recycling of 1 kg of cadmium (mass allocation Cd/Ni = 44/56), 19 MJ is needed for transportation and 19 MJ for recycling. Corresponding values for 1 kg nickel are 20 MJ for collection and 20 MJ for recycling. Compared with extraction and refining of virgin metal, 54 and 75% less primary energy is needed to recycle cadmium and nickel, respectively. The corresponding values using economic allocation (metal prices US\$98/kg, Cd/Ni = 7.6/92 (USGS, 1999)) are decreases of 90 and 59%, respectively.

4.3. Metal emissions in short- and long-term perspectives

Total metal emission from different activities is shown in Table 4. At a 25% recycling rate, 0.23-0.26% of the potential metal emission occurs in a 100-year perspective. Most of the metal emission (96–98%) to water occurs in the incineration and landfilling activities, irrespective of whether short- or long-term emissions are considered. Two percent of the cadmium and nickel emission to water occur during battery manufacturing. The emission of cadmium to air takes place mainly (99%) in the incineration and landfilling activities. For nickel, 82% of the emission occurs during incineration and landfilling, while 15% originates from raw materials extraction.

A comparison of metal emissions over the NiCd battery life cycle, considering different methods of end-of-life treatment, shows that the greatest nickel emission to water occurs for 100% landfilling (Fig. 4). The nickel emission to water decreases by 52%, when comparing landfilling with incineration/landfilling. The reduction in emissions for incinerated batteries is due to the fact that metals are more stable in the ash after the addition of sulphides.

The emission of cadmium to air is greatest for incineration/landfilling. Comparing incineration/landfilling with recycling and landfilling, shows that the cadmium emission to air decreases by 75 and 98%, respectively. The nickel emission to air shows the same trends as for cadmium air emission.

	Total	Raw material (excl. Cd) (%)	Cd extraction/refining (%)	Battery manufacture (%)	Collection and sorting (%)	Recycling process (%)	Incineration and landfills (%)
Cd(act) (mg/W h)	1.4	0.0	1.9	2.0	0.0	0.1	96.0
Ni(ad) (mg/W h)	9.2	0.0	0.0	1.8	0.0	0.1	98.1
Cd_{rair} (mg/W h)	9.4	0.1	0.3	0.0	0.0	0.1	99.4
Ni _(air) (mg/W h)	2.8	15.1	0.4	2.0	0.0	0.1	82.4



Fig. 4. Total emissions of nickel and cadmium throughout the NiCd battery life cycle (excl. user phase) for different methods of end-of-life treatment (short-term perspective).

Incineration plants with extensive systems for flue gas cleaning lead to low emissions of metals to air and water. Uncontrolled incineration may, however, volatilise cadmium thereby increasing its mobility. Fires in landfills may cause considerable emissions of metals, but this scenario has not been considered in this study. The addition of organic sulphides to ash stabilises metals, but oxidising conditions can make sulphides soluble in water. The mobility of metals may also increase in acidic environments.

Of the total nickel and cadmium content in batteries, 0.08-0.52% of the metals is released to air and water in a 100-year perspective. The potential cadmium and nickel emissions in a long-term perspective are 4.1 g Cd/W h and 5.1 g Ni/W h, i.e. approximately 300-400 times greater than in the short-term perspective. It is, however, not possible to determine the distribution of different metal species and their potential bioavailability.

5. Environmental impact during the NiCd battery life cycle

Primary energy use and the emission of CO_2 are most significant during battery manufacturing, and significant SO_x and NO_x emissions arise in the extraction and refining of raw materials. At the estimated 25% recycling rate in Sweden, emissions and resource consumption of metals are significant in the end-of-life treatment activities.

Depending on the type of battery application, energy in the user phase of the battery and charger losses may contribute to the most significant energy use (and environmental impact) of the NiCd battery life cycle. Different user patterns for a 1 W h NiCd battery (e.g. cycle life 10–1000 cycles and charger efficiency 0.2-0.8) correspond to 0.045-18 MJ electrical energy (0.11-45 MJ primary energy at a conversion efficiency of 0.4). The total primary energy use in the production of NiCd batteries (4.4-5.3 MJ/W h) and battery charging is in the range 4.5-50 MJ/W h NiCd battery. Primary energy in the user phase may account for 2.0-91% of the energy during the NiCd battery life cycle.

Jensen and Petersen, (1999) concluded that the weighted potential environmental impacts were significant regarding energy use in stand-by mode of the charger. The weighted environmental impact was greater for the production of the battery charger than for batteries. When comparing the environmental impact of other rechargeable battery technologies (lead–acid, nickel–metal hydride and lithium– cobalt) with NiCd batteries, no significant differences were found apart from the higher toxicity of NiCd batteries (Kertes, 1996; Törnblom, 1996; Jensen and Petersen, 1999). The primary energy use for the manufacture of different battery types has been calculated to be 5.9 MJ/W h (NiCd), 3.3 MJ/W h (nickel–metal hydride) and 0.40 MJ/W h (non-rechargeable alkaline manganese) (Kirmayer, 1995). The differences depend mainly on the varying energy densities of the battery systems. Refining of raw materials and manufacturing of one alkaline–manganese cell requires 3–4 times less energy than equivalent NiCd or NiMH cells (Kirmayer, 1995).

Quantification of primary energy requirements for recycled metals relies on estimates and the values may vary depending on the system boundaries chosen. Lankey (1998) estimated the energy required for manufacture of batteries with recycled materials to be approximately half the energy needed to manufacture batteries using only primary materials. In this study, the energy reduction was calculated to be 16%. Lankey (1998) claims that 190 MJ/kg is needed for virgin cadmium production and 22 MJ/kg for recycled cadmium. However, these data are difficult to assess since the allocation principles and the use of different energy carriers were not explained.

Longer transportation distances and the use of recycled materials are compensated for by lower emissions in the raw material extraction and refining activities. Although the transport sector contributes significantly to the environmental impact in society, the transportation distance involved in the collection and recycling of materials is generally of very low environmental significance compared with other kinds of transportation (Hunhammar, 1995). The public awareness of transport is higher than for other sectors since transport is associated with high monetary costs and causes emission in urban areas. The LCI model shows that collection and recycling is beneficial from an environmental perspective, even at very low recycling rates. At high recycling rates, increased NO_x emission in urban areas must be compared with reductions in other emissions and resource use. Since the recycling process is not affected by contaminants, the optimum recycling rate for battery metals tends to be close to a hypothetical 100% recycling rate. At closed loop

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recycling, the cadmium recovery efficiency is 0.9997 mainly due a fraction of cadmium lost to nickel-iron scrap. Low overall metal losses are crucial for maintaining a stock of metals in batteries and metal resource availability (Andersson and Råde, 2000).

Uncertainties in the results depend on the choice of methodology and data source. Choices in methodology that could affect the results are, modelling of cadmium and nickel as closed-loop recycling, recycling of steel, choice of model for electricity production and the use of system expansion to allocate the useful waste heat from battery recycling. Uncertain data values include assumptions about metal emissions during battery manufacturing, load factor of trucks and transport distances. Sensitivity analyses have, however, shown that these parameters are of minor importance in the final result. The absolute values may be distorted by methodological choices and data values but the identified trends will remain the same.

By relating the use of portable NiCd batteries in Sweden (1998, 25% recycling) to societal material and energy flows the significance of batteries may be identified. Regarding the amount of MSW in Sweden in 1998, all NiCd batteries sold in 1998 would contribute 40–80 ppm to the total mass of MSW (SCB, 2000). In the case of primary energy, the total energy use during the life cycle (excl. user phase) of portable NiCd batteries in Sweden in 1998 contributed 15–23 ppb to the total primary energy turnover that same year. In a study of the material flows of metals in the municipality of Stockholm in 1995, it was estimated that portable NiCd batteries contributed 91% to the annual metal inflow (total 8.8 ton Cd) and more than 25% of the accumulated cadmium in goods and constructions (120 ton) during the period 1900–1995 (Lohm et al., 1997).

The total emission of cadmium from different sources in Sweden in 1995 were estimated to be 1.8 ton cadmium to water $(Cd_{(aq)})$ (39% from mining residue deposits, 31% from pulp and paper) and 0.78 ton cadmium to air $(Cd_{(air)})$ (42% incineration excl. waste, 22% metal smelters) (SCB, 2000). In this study, (25% recycling 1998), the cadmium emission from NiCd batteries was estimated to be 9.2 kg $Cd_{(aq)}$ and 60 kg $Cd_{(air)}$ during 100 years (cumulative metal emission of batteries before 1998 were not considered). Although the emission of cadmium from NiCd batteries is very low in the short-term perspective, continuous accumulation implies an increase of future emissions. The relative contribution of NiCd batteries to the total dissipative losses will probably increase in the future since cadmium use in other products is declining. Guinée et al. (1999) studied metal flows in the Netherlands and estimated that the cadmium emission from accumulated cadmium in the technosphere would increase by 30% from 1990 until a hypothetical steady state is reached. Accumulation in soil could lead to the exceeding of critical levels for human, terrestrial and aquatic ecotoxicity.

The comparison above shows that there is major concern related to the high toxicity of cadmium and the increasing contribution of NiCd batteries to future potential cadmium emission. Incineration plants and landfills in Sweden emit low levels of metals in the short-term perspective and the main problem is whether spent batteries end up in less controlled waste treatment systems or in the environment. Although not all metals are released at the same time from landfills, continued dissipative losses give rise to elevated background concentrations. To be able to make a risk assessment, the mobility of cadmium and other metals and human exposure to them in different environments would have to be evaluated.

6. Materials management of cadmium

The primary extraction of most metals is still increasing, and a higher degree of closed-loop recycling of metals could lead to a higher accumulation of metals in the technosphere. The emission from metals accumulated in the technosphere could increase in the future for example through corrosion and inadequately controlled incineration.

Voet et al. (1994) studied material flows of cadmium within the EU and evaluated the effects of different policies for controlling cadmium. They concluded that the proposed EU policy based on (1) end-of-pipe measures (2) phasing out of certain applications and (3) recycling of batteries and certain other products, does not appear to offer a sustainable solution to the dispersion of cadmium. This is primarily due to the inelastic nature of the cadmium supply since cadmium is extracted as a by-product of zinc production. On the inflow side, measures should not focus on cadmium but on the main products zinc and phosphate. Otherwise, recycling may only lead to increased accumulation of cadmium and thus to larger future losses to the environment. On the outflow side, the collection of waste and products containing cadmium and subsequent safe storage in a concentrated form seems to offer the best solution.

Although the results of this study show that increased recycling of NiCd batteries decreases the environmental impact, the end use of recycled cadmium must be considered. If recycled cadmium is used in new portable NiCd batteries, it is uncertain whether the battery will be collected again. Some important reasons why it is difficult to achieve high collection rates are that portable NiCd batteries are dispersed among many battery owners, the usage of batteries in diverse applications, the small size of battery units and low economic value of products (Rydh, 2001).

In order to create incentives for battery recycling a demand for spent materials must be created. A combination of historically low prices, limited growth in the use of cadmium metal and pending environmental legislation has made the use of cadmium uncertain. Zinc miners who produce cadmium as a by-product are now regarding the metal as a cost rather than an asset (Mining Journal, 1998). The market is already oversupplied, and new zinc mines under development in Australia and North America will exacerbate the situation. On the global level, the production of refined cadmium was 19.6 kton in 1998 (Plachy, 2000). On the demand side, cadmium is becoming a one-use metal, and approximately 70% is used in the manufacture of NiCd batteries (Mining Journal, 1998). The major share (75%, 10 kton Cd) of cadmium is used in the production of portable NiCd batteries, while the rest (3.4 kton Cd) is used in industrial NiCd batteries (Plachy, 2000). The price

of portable batteries is 2-10 times greater than for the same capacity of industrial NiCd batteries, which makes the market for portable batteries more profitable.

Recycled cadmium is more expensive than primary cadmium delivered from zinc smelters (Mattsson, 2000). The annual average price of cadmium on the open market has decreased from US\$4.05/kg in 1995 to US\$0.62/kg in 1998 (USGS, 1999). In 1999, the average nickel price was US\$6.16/kg (USGS, 1999). The nickel content of NiCd batteries accounts for the largest contribution to the value of scrapped NiCd batteries.

Material and energy resources generally have lower economic value than labour costs. Since recycling is more labour intensive than landfilling or waste incineration, it is a more expensive way of treating end products. Shapek (1996) studied the economic aspects of battery collection in Florida and concluded that the cost increased with higher collection rates. Greene (1995) evaluated actual and hypothetical scenarios for household battery collection and concluded that neither was cost effective. However, it was not possible to determine the economic benefits of reduced metal contamination.

To make recycled materials competitive with virgin materials measures can aim at increasing consumer demand for recycled material. Examples of such measures are introduction of minimum recycled content specifications, taxes on virgin material and subsidies for products containing recycled material. Another way is to increase the supply of available material for recycling. This can be done by modifying consumer behaviour by introducing e.g. compulsory take back, deposits, fixed target recycling and landfill bans. MacDonagh-Dumler (2000) argues for that supply-increasing policies are the most effective for portable NiCd batteries since the last user does not have sufficient incentive to return batteries to the collection system. The motivation to control toxic material fate is driven by public and environmental health concern, not resource efficiency and economic asset value maximization.

Several actions have been introduced or proposed to increase collection rate e.g. public information campaigns, import fees, refunds on batteries, labelling, disposal charges and improved statistics on battery stocks and flows (Ayres and Ayres, 1996; Environment directorate, 1999). A number of stakeholders (EPBA, 1999; Sempels, 1999) are opposed to the regulation of the battery market, and the implementation of stricter measures to encourage improved collection may thus be delayed. However, agreements implemented on the international market (e.g. extended producer responsibility) may diminish trade distortions.

The above-mentioned aspects make it less probable that high collection rates will be achieved for portable NiCd batteries. Toxic metals may instead be used in products with higher metal content and thus of higher economic value per kg recovered battery (e.g. stationary NiCd batteries), which are more likely to be recycled than portable NiCd batteries. Portable NiCd batteries have their niche in power tools and in emergency lightning due to their high power density and good operational characteristics at high temperatures. New battery technologies (NiMH, Li-ion) containing less toxic materials can replace portable NiCd batteries in most applications (Noréus, 2000). NiMH batteries have not entered the NiCd market niches since the main development goal has been to achieve high energy density by using foamed positive electrodes. However, higher power density in NiMH batteries can be achieved by using sintered positive electrodes. Another reason may be that some NiMH producers also manufacture NiCd batteries and therefore want to conserve the NiCd market niche (Noréus, 2000).

Uncertainties about future emissions and the mobility of metals for different products mean that it is important to perform further studies on emission factors to different media. An environmental assessment of the recycling of portable zinc-carbon and alkaline-manganese batteries would also be of interest to assess the possibility of decreasing the primary extraction of zinc.

7. Conclusions

Primary energy use and emission of CO_2 are most significant in battery manufacturing. Emissions and resource consumption of metals are significant in the end-of-life treatment, regardless of whether short- or long-term emissions are considered. Transportation for the collection of spent NiCd batteries has no significant environmental impact and thus NiCd batteries can be transported long distances for recycling and it would still be beneficial from an environmental perspective. From an environmental perspective the optimum recycling rate for NiCd batteries tends to be close to 100%.

Batteries manufactured with recycled cadmium and nickel have 16% lower primary energy requirements than if only virgin metals are used. Using recycled cadmium and nickel requires 46 and 75% less primary energy respectively, compared with extraction and refining of virgin metal.

There are considerable uncertainties associated with emissions of metals that may occur in the future. The potential cadmium and nickel emissions were 300–400 times greater than in a 100-year perspective. To avoid dissipative losses, cadmium should be used in products that will probably be collected at the end of their life.

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Paper II

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Impact on global metal flows arising from the use of portable rechargeable batteries

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Abstract

The use of portable rechargeable battery cells and their effects on global metal flows were assessed for the following three cases: (1) the base case, which reflects the situation in 1999 of the global production of batteries; (2) the global production of portable nickel-cadmium batteries in 1999, assumed to be replaced by other battery types; and (3) assessment of the projected battery market in 2009. The study included the following battery technologies: nickel-cadmium (NiCd); nickel-metal hydride (NiMH) (AB5, AB2); and lithium-based batteries (Liion: Co, Ni, Mn; Li-polymer: V). Based on the lithospheric extraction indicator (LEI), which is the ratio of anthropogenic to natural metal flows, and the significance of battery production related to global metal mining, the potential environmental impact of metals used in different battery types was evaluated. The LEIs and average metal demand for the battery market in 1999, expressed as a percentage of global mining output in 1999, were estimated to be as follows: Ni 5.6 (2.0%); Cd 4.4 (37%); Li 0.65 (3.8%); V 0.33 (6.5%); Co 0.18 (15%); Nd 0.18 (8.4%); La 0.10 (9.5%); Ce 0.083 (4.4%); and Pr 0.073 (9.4%). The use of Ni and Cd is of the greatest environmental interest, due to their high LEIs. In the case of complete replacement of portable NiCd batteries by NiMH or Li-based batteries, the LEI for Ni (5.6) would change by -0.1-0.5% and the LEI for Cd would decrease from 4.4 to 3.0 (-31%). Meanwhile, the mobilization of metals considered less hazardous than Cd (LEI<0.65) would increase less than 7%. Based on this assessment, the replacement of NiCd batteries would result in decreased environmental impact. To decrease the impact on global metal flows arising from the use of portable batteries the following points should be considered: (1) development of battery technologies should aim at high energy density and long service life; (2) metals with high natural occurrence should be used; and (3) metals from disused batteries should be recovered and regulations implemented to decrease the need for mining of virgin metals. The method used enables an assessment early in the cause-effect chain, when few data about toxic effects are available. It can also be used to assess whether environmental problems are shifted from one to another. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Rechargeable; Nickel-cadmium batteries; Nickel-metal hydride batteries; Lithium batteries; Environmental impact; Metal flows; Mining; Lithospheric extraction; Recycling

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1. Introduction

The emergence of new types of rechargeable (secondary) batteries has accelerated the replacement of portable nickel-cadmium (NiCd) batteries in many applications, mainly in northern Europe (ENS, 2001). Replacement has occurred because new battery technologies [nickel-metal hydride (NiMH) and lithium-based batteries (Li-ion and Li-polymer)] have better performance (e.g. higher energy density and no memory effect) in many applications (Moshtev and Johnson, 2000). Another important reason for the replacement of NiCd batteries is due to the toxic properties of Cd. The new battery technologies are based on metals that are believed to have lower environmental impact. This replacement has been seen as an example of action in accordance with the precautionary principle.

The new batteries contain metals (e.g. La, Nd, Co) for which only few toxicological or ecotoxicological data are available. Consequently, the introduction of new metals is associated with uncertainties regarding environmental impact, and a shift in the use of battery technology may induce a change from one problematic metal to another. The introduction of new battery technologies must be evaluated to determine whether the replacement of NiCd batteries will lead to other environmentally problematic metal flows.

The environmental aspects of metal flows resulting from the use of various battery technologies have been assessed in several studies (Hofstetter and Häne, 1990; Tillborg, 1999; Rydh, 2001; Rydh and Karlström, 2002). Resource availability regarding metals for established and emerging electric vehicle batteries has also been studied (Andersson and Råde, 2001). However, quantified risk assessments of metal use in emerging battery technologies are lacking. Many methods for environmental system analysis require comprehensive data from real production, e.g. environmental risk assessment and life-cycle assessment. This makes it difficult to compare different types of batteries that are not yet in full-scale production.

Assessment must be made early in the causeeffect chain before serious effects arise. The purpose of this study was to assess the potential effects on global metal flows if NiCd batteries were to be replaced by different types of batteries.

2. Scope and method

The use of sealed portable rechargeable battery cells and their effects on global metal flows were assessed within an interval for the following three cases: (1) the base case, which reflects the situation in 1999 of the global production of batteries; (2) the global production of portable NiCd batteries in 1999 is assumed to be replaced by other battery types (equivalent number of cells and equivalent energy capacity); and (3) assessment of the projected battery market in 2009.

Combinations of different commercial and experimental battery technologies that may replace portable NiCd-batteries were considered: nickelmetal hydride batteries (NiMH) (alloys of type AB_2 and AB_3); and lithium-based batteries. Lithium batteries (Li-ion: Co, Ni and Mn; Li-polymer: V) were collected into one group to cover the use of different metals. Electronic circuits for charge control were not included.

Portable batteries are defined as single cells with a maximum weight of 100 g/cell. Applications for portable batteries include e.g. emergency lighting, power tools, cameras, mobile phones and portable computers. In some battery applications, small sealed lead-acid (SSLA) batteries and nonrechargeable (primary) batteries may be used instead of nickel and lithium-based secondary batteries. Due to limited use of these battery types in fast growing technologies such as portable consumer electronics, they were excluded from this study.

The potential environmental impact resulting from the use of metals in batteries was assessed from its influence on lithospheric extraction indicators (LEIs). LEIs are calculated for each metal as the ratio between anthropogenic metal flows and natural metal flows (Benjamin and Honeyman, 1992). LEI and mining output for the year 1999 were used as the reference and all changes due to battery use were related to these values. Calculation of LEI for the year 1999 is shown in Eq. (1) where: F_{Mining} is the annual metal flow from mining (ktonnes/year); $F_{\text{Fossil fuels}}$ is mobilization

of metals due to combustion of fossil fuels (ktonnes/year); and F_{Natural} is the natural turnover of metals from weathering and volcanic activity (ktonnes/year).

$$\text{LEI}_{99} = \frac{F_{\text{Mining}} + F_{\text{Fossil fuels}}}{F_{\text{Natural}}} \tag{1}$$

The maximum potential impact of metals is based on the following general assumptions: (1) all mined metals will be released to the environment in the future due to geological processes; (2) anthropogenic emissions of metals are distributed in the same way as naturally occurring elements and have the same degree of bio-availability; (3) organisms and ecosystems have adapted to natural background concentrations of metals. The calculated LEIs do not represent absolute values, but provide a simple system for environmental ranking of different metals.

The advantage of the method is that data are obtainable early in the cause–effect chain of metals, and the ratio gives an early indication as to whether an activity may cause environmental impact. By assuming that the use of metals in batteries influences the amount of metals mined annually, while other metal use is constant, the LEI at a certain time (T1) and for a certain metal can be calculated from Eq. (2).

$$\text{LEI}_{\text{T1}} = \frac{F_{\text{A T0}} + F_{\text{Battery T1}} - F_{\text{Battery T0}}}{F_{\text{N T0}}}$$
(2)

 $F_{\rm A \ T0}$ is the total annual metal flow from mining (incl. metals in batteries) and the combustion of (ktonnes/year), $F_{\text{Battery T1}}$ and fossil fuels $F_{\text{Battery T0}}$ are the virgin metal flow due to battery production (ktonnes/year) in a specific year (T1) and a reference year (T0=1999), respectively. $F_{\rm N \ T0}$ is the natural turnover of metals from weathering and volcanic activity (ktonnes/year), and is assumed to be constant. The metals flow may end up in products or be emitted to air, soil, water and sediment. In order to calculate the net metal flow (net mobilisation) due to the manufacturing of batteries, the proportion of recycled metal used in batteries, F_{Recycled} (ktonnes/year) is subtracted from the gross metal flow in the manufacturing of batteries, $F_{\text{Battery gross}}$ (ktonnes/year) (Eq. (3)).

$$F_{\text{Battery}} = F_{\text{Battery gross}} - F_{\text{Recycled}}$$
(3)

The metal demand for manufacturing of a specific battery ($F_{\text{Battery gross}}$), was calculated from Eq. (4), where n_{Cells} is the number of annually manufactured batteries (cells/year), m_{Cell} is the mass of a battery cell (g/cell), c_{Metal} is the metal content of a battery (wt.%), $k_1 = 10^{-9}$ (ktonnes/g) and $k_2 = 10^{-2}$ (1/wt%).

$$F_{\text{Battery gross}} = n_{\text{Cells}} \times m_{\text{Cell}} \times c_{\text{Metal}} \times k_1 \times k_2 \qquad (4)$$

To calculate the metal demand for an equivalent energy capacity (Case 2), different energy densities and cycle life for the various battery technologies were considered in order to make the battery technologies comparable.

With the equations above, the potential change in anthropogenic mobilisation of metals for assumed cases concerning different battery types was calculated. The results were presented as LEIs for three different cases. The mining demand for each battery technology was related to the mining output 1999. The values for LEI_{T1} (Eq. (2)) were related to the reference year LEI₉₉ and the potential change compared with the reference year was given as (LEI_{T1}-LEI₉₉)/LEI₉₉×100%.

Metals of special concern were identified according to (1), the mining demand for battery metals as a fraction of total annual mining >1% and/or (2), LEI>1. The value of LEI was chosen as an arbitrary limit to distinguish between low and high impact on natural flows (Azar et al., 1996). LEI <1 shows that the anthropogenic emission of a metal is lower than the natural flow and that the use of this quantity of the metal will probably have a low environmental impact. LEI> 1 indicates a potential for increasing metal concentrations in the environment and thus increased environmental impact.

The aim of this method is to provide a simple assessment of technologies for which few data are available. The method does not take into account the chemical properties of different metals or their distribution between air, soil, water and sediment. It does not assess actual toxicological effects or temporal aspects. It can, therefore, not be assumed that a high LEI value for a certain metal will result in high environmental impact, since some metals



Fig. 1. Global production of portable rechargeable batteries 1989–1999 (Fujimoto, 1999; Noréus, 2000). The projected market development from 2000 to 2009 is described in Section 3.1.

may have low toxicity or are present as a chemical species that is not bio-available.

The metal flows resulting from the production and use of portable rechargeable batteries are assessed in Section 3 and the impact of rechargeable batteries on global metal flows is calculated in Section 4. In Section 5, the results are discussed.

3. Use of metals in rechargeable batteries

3.1. Market development for rechargeable batteries

The assessment of material flows in the different cases is based on the quantities of different battery

 Table 1

 Specifications for the different battery types

cells manufactured globally (Fig. 1). The total mass of batteries was calculated from the total number of cells and the average mass per battery cell (Eq. (4)).

Table 1 presents the technical specifications needed to calculate metal requirements for the different types of batteries.

When comparing different battery types, the functionality of the batteries must be the same. In some applications high energy capacity and many charge-discharge cycles are required. In applications where batteries are used for backup power, little energy is withdrawn and the energy storage capacity is less important than high reliability. To cover different modes of battery use and market

	NiCd	NiMH(AB ₅)	$NiMH(AB_2)$	Li-based ^c
1. Average mass per cell ^a (g/cell)	25-45	26-46	26-46	26-46
2. Gravimetric energy density (Wh/kg)	48-60	64-80	72-90	104-130
3. Volumetric energy density (Wh/dm ³)	40-90	230-250		250-300
4. Cycle life ^b (No. of cycles)	500-1000	300-800	300-500	100-600
5. Life cycle energy (kWh/kg) $(2.\times 4.)$	24-60	19-64	22-45	10 - 78
6. Adjusted NiCd life cycle energy	1.00	1.25-0.94	1.33-1.11	2.31 - 0.77
7. Global market share 1999 (%)	49	36–37	0-1	14

Sources: Vincent and Scrosati, 1997; Moshtev and Johnson, 2000; Hong, 2001; Råde and Andersson, 2001. Note: The battery types have different characteristics, e.g. power density, charge–discharge behavior, rate of self-discharge, robustness and temperature range. Therefore, the different types of batteries are not directly interchangeable in different applications.

^a Battery sizes: low estimate = AA cell, high estimate = Sub C cell.

^b 100% depth of discharge (DOD) and 80% of initial capacity. Cycle life increases with lower DOD.

^c Li-batteries: Li-ion (Co, Mn, Ni); and Li-polymer (V). Electronics required for charge control.

Table	2													
Mass	of each	cell	type :	and	number	of	cells	produced	globally	for	the	three	cases	studied

	NiCd	NiMH(AB ₅)	NiMH(AB ₂)	Li-based ^b
Energy capacity produced 1999 (TWh)	1688-3799	1693-3860	0-130	1101-2434
Case 1. No. of cells in 1999 ($\times 10^9$)	1.41	1.02 - 1.05	0-0.03	0.41
Case 2. No. of cells (NiCd replacement) ($\times 10^9$)	0	2.03-2.46	0.90 - 1.44	1.03 - 1.81
Case 3. No. of cells in 2009^{a} ($\times 10^{9}$)	0.38	3.08-3.18	0-0.10	2.49
Case 1. Mass of cells in 1999 (ktonnes)	35-63	26-48	0-1.0	11-19
Case 2. Mass of cells (NiCd replacement) (ktonnes)	0	53-113	23-66	27-83
Case 3. Mass of cells in 2009 ^a (ktonnes)	10-17	80-146	0-4	65-114

^a Projected development, see Section 3.1 and Fig. 1.

^b Li-batteries: Li-ion (Co, Mn, Ni); and Li-polymer (V).

development, three cases were evaluated. Quantitative data for the cases are presented in Table 2. The broad ranges are due to uncertainties in data for the average weights of cells and varying energy densities for each type of application.

3.1.1. Case 1: Metal demand for batteries in 1999

In Case 1, the impact on global mining requirements for rechargeable batteries for a mix of different battery technologies in the year 1999 was estimated. AB_5 alloys are used in NiMH batteries worldwide, while NiMH(AB_2) batteries are mainly used in the USA (Ye and Zhang, 2001). It is assumed that NiMH(AB_5) batteries make up 97– 100% of the NiMH market and that NiMH(AB_2) account for the remainder. Li-ion batteries based on positive electrodes made of Co dominate the market over Li (Ni), Li (Mn) and Li-polymer batteries (Vincent and Scrosati, 1997).

3.1.2. Case 2: Different battery types replace NiCd batteries

Case 2 is based on the assumption that different battery technologies NiMH(AB₅), NiMH(AB₂), or Li-based batteries, replace the market share of NiCd batteries. Depending on type of battery application, replacement can be based on energy capacity, life cycle energy, or total number of cells.

In the low estimate, it was assumed that new battery technologies were produced at an energy capacity equivalent to that of NiCd cells produced globally in 1999. Different electrochemical systems have different energy densities. The metal required by different batteries to supply equivalent energy storage capacities were based on average technical performance data (Table 1).

The gravimetric energy density of a battery defines its metal intensity. A battery with high energy density requires less metal to supply a given energy capacity than a battery with lower energy density. Table 1 shows that the gravimetric energy densities, expressed as share of NiCd, are 75% for NiMH(AB₅), 67% for NiMH(AB₂) and 46% for Li-based batteries. These percentages are reflected in the energy-related demand for metals. The low estimate in Case 2 (Table 2) shows that the mass of cells decreases proportionally to the energy density.

The high estimate is based on the assumption that the same numbers of cells are produced as the number of NiCd cells produced in 1999. Potential effects on metal flows due to the replacement of NiCd batteries by other battery technologies were calculated by assuming a replacement in the world market of 1.4×10^9 portable NiCd cells in 1999. This is motivated since many portable electronic products where rechargeable batteries are used have a short service life. Batteries in such applications do not reach their technical end of life.

In some applications, batteries are charged and discharged often and have to be replaced when the maximum cycle life of the battery has been reached. This means that the battery has to be replaced more often which leads to a higher turnover of batteries and metals. Row 5 in Table 1 shows the theoretical energy that can be withdrawn from different batteries considering the

maximum cycle life. Row 6 shows the adjusted NiCd life cycle energy factor for batteries compared with the NiCd battery. The cycle life of batteries can influence the metal flows of batteries by 0.77–2.31 (Li-based). The evaluation of metal flows for applications when maximum battery cycle life is reached is covered within the low–high estimate in Case 2.

3.1.3. Case 3: Projected battery market in 2009

In Case 3, a projected scenario was made for the battery market in the year 2009. The case for 2009 in Fig. 1 is based on annual scaling factors from the year 1999, with the following average values: NiCd=0.88; NiMH=1.14; and Li-ion= 1.22. Related to the year 1999, the market for NiCd batteries is projected to decrease by 73%, NiMH batteries to triple and Li-ion batteries to increase by 6.1 times. The total number of cells will increase 2.1 times. The use of NiMH batteries may increase due to good technical performance (Noréus, 2000), a growing market of electric hybrid vehicles (Anderman et al., 2000) and legislative bans or fees on NiCd batteries. The market for lithium batteries is likely to increase, since this technology provides several technical advantages compared with the other technologies. The operating voltage (three times higher than NiCd and NiMH), the relatively low self-discharge rate, the absence of memory effect, the smooth slope of the discharge curve and the high energy density have led to increasing consumer interest and consequently by a significant increase in manufacturing output (Vincent and Scrosati, 1997).

3.2. Content of rechargeable batteries

The composition of the different battery types was estimated within an interval (Table 3). The data include metallic constituents such as active materials, casing, performance enhancing chemicals and trace elements.

3.2.1. Nickel-cadmium and nickel-metal hydride batteries

NiCd batteries have a positive electrode made of nickel hydroxide and a negative electrode made of Cd. Sintered electrodes are used for high dis-

Table 3 Metal content of different portable batteries (% by weight)

Element	NiCd	$NiMH(AB_5)$	$NiMH(AB_2)$	Li-based
Al	0.019	0.50-2.0	0.50-1.0	4.6-24
Cd	15 - 20			
Ce		0.43-5.5		
Co	0.60	2.5 - 4.3	1.0-3.0	12-20 ^a
Cr	0.017	0.020-0.080	0-1.6	
Cu				5.0 - 10
Fe	29-40	20-25	23-25	4.7-25
La		1.4-6.6		
Li				1.5 ^b -5.5 ^c
Mn	0.083	0.81-3.0		10-15 ^d
Nd		0.96-4.1		
Ni	15 - 20	25-46	34-39	12-15 ^e
Pr		0.32-1.3		
Ti			2.2 - 3.9	
V			2.2 - 4.7	15-20°
Zn	0.060	0.092-1.6		
Zr			3.9-8.7	

Note: the data include all battery components i.e. active materials, current collectors and case. *Sources*: Lyman and Palmer, 1994; Ruetschi et al., 1995; Scholl and Muth, 1996; Murano, 1997; Zhang et al., 1999; Eriksson, 2001; Lain, 2001; Lee and Rhee, 2002.

^a Li-ion (Co).

^b Li-ion (Co, Ni, Mn).

^c Li-polymer (V).

^d Li-ion (Mn).

e Li-ion (Ni).

charge rates and foamed electrodes for high-capacity applications at lower discharge currents. Performance chemicals, e.g. $Co(OH)_2$ and $Ba(OH)_2$, can be added to the positive electrode to improve cell capacity and cycle life (Kordesh, 1999). The electrolyte of NiCd and NiMH batteries is typically of potassium hydroxide (KOH).

NiMH batteries are constructed with a negative electrode consisting of a hydrogen-storing alloy and a positive electrode of Ni. The negative electrodes are made of either AB_5 or AB_2 alloys. AB_5 alloy is the most common type of negative electrode in NiMH batteries (Ruetschi et al., 1995). AB_5 alloy contains mischmetal, which is a mixture of lanthanides (rare earths¹). NiMH(AB₅)

¹ Rare earths include the following 17 elements: scandium, yttrium (the lanthanides) lanthanum, cerium, praseodymium, neodymium, promethium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium and lutetium.

batteries contain 8-10% mischmetal and its composition corresponds to that of natural ores. It contains 50-55% Ce, 18-28% La, 12-18% Nd, 4-6% Pr, and other rare earth elements in small quantities, as well as metallic impurities. Zhang et al. (1999) have reported rare earths composition of 52% La, 33% Nd, 10% Pr and 3.4% Ce used in commercial cells.

In AB₂ alloys, A stands for a hydride forming element (e.g. zirconium or titanium) and B other metals, such as Ni, Co, V, Mn, Al, Cr, or Fe. The total content of Zr, Ti, Nd and Pr is 13-14%(Ruetschi et al., 1995). A review of different compositions is given in Råde and Andersson (2001). The low and high composition estimates are represented by alloys given by Shaju et al. (1999) and Venkatesan (1994), respectively.

3.2.2. Lithium polymer and lithium-ion batteries

Rechargeable lithium batteries can be divided into Li-polymer (Li-metal) and Li-ion batteries. Li-polymer batteries have a negative electrode made of a Li metal foil and the electrolyte consists of a polymer material (e.g. polyethylene oxide-PEO-LiClO₄). An experimental design of the positive electrode is based on vanadium oxide (e.g. V_6O_{13} or VO_x), in which Li ions have been chemically inserted (Vincent and Scrosati, 1997). Li-polymer batteries have lower energy density and cycle life than Li-ion batteries. The advantage is that the cells can be manufactured in different kinds of geometrical forms (Scrosati et al., 2001).

The negative electrode (anode) of a Li-ion cell is made of carbon. Different forms of carbon containing materials, such as graphite, coke and amorphous carbons, have been used. Li-ion batteries have Li ions intercalated at the negative electrode. In Li-ion batteries, the positive Li electrode (cathode) contains metal oxides of the composition LiMeO₂, where Me denotes Co, Ni or Mn (Vincent and Scrosati, 1997). LiCoO2 is used in most of the Li batteries in global production since it gives the highest capacity and cycle numbers (Vincent and Scrosati, 1997; Scrosati, 2000). Lithiated nickel oxide has higher specific capacity but lower stability and is used by the battery manufacturer SAFT for small and large batteries. The cycle life is shorter than LiCoO2 but the elements have higher availability and lower cost. A third type of positive electrode material is lithiated manganeseoxide. They have lower capacity and shorter cycle life, but they are low-toxic, stable and of low cost (Vincent and Scrosati, 1997). The electrolyte consists of a liquid organic electrolyte (e.g. polypropylene carbonate, PC) for carbon electrodes, solid polymer (solid electrolyte), or both (gel electrolyte). Several different Li salts have been used in commercial batteries. Most common are LiClO₄, LiPF₆ and LiBF₄ (Wakihara, 2001). Other elements used in Li-ion batteries include Fe, Cu and Al. Li-ion batteries usually have a casing made of aluminium or nickel-plated iron. Copper and Al are used as current collectors in the electrodes.

3.3. Recycling of metals from batteries

The relatively small quantities of metals available for recycling from portable batteries and the low metal value make it difficult to achieve commercial recycling. Many countries lack effective collection schemes for disused batteries. Hoarding of batteries (Wiaux, 2001a), the large number of battery users and the small size of portable batteries make it difficult to achieve high collection rates for portable batteries (Rydh, 2001). Regarding disused batteries available for collection, average collection efficiencies of 55% have been estimated for European countries (Wiaux, 2001b). Similar efficiencies have been estimated in Japan (Fujimoto, 1999). These estimates are based on the supply from a stock that has been built up by consumers for several years. Due to the uncertainties in calculating recycling rates, a broad interval is covered to reflect these uncertainties. NiCd and NiMH batteries are often collected together and they can be handled in the same recycling plant. Recycling rates at the global level are estimated to be 5-50% for NiCd and NiMH, and 2-10% for Li-ion batteries.

Recovered Cd is upgraded to high purity in metallic form and is used in new batteries and other applications such as pigments or coatings. In the production of industrial batteries by SAFT (Oskarshamn, Sweden) up to 20% of the Cd used has been recycled (Rydh, 1999). Nickel and Fe from NiCd and NiMH batteries are used in the steel industry (Lyman and Palmer, 1994; Wiaux, 2000). Recovery of nickel from batteries is profitable, which has led to increased interest in developing efficient recovery technologies (Zhang et al., 1999; Tenório and Espinosa, 2002). Mischmetal from NiMH(AB₅) batteries is used as an alloy in stainless steel.

Materials recovered from Li-ion batteries are Co, Cu, Al, steel and LiOH (Murano, 1997; Wiaux, 2000; Lain, 2001). Cobalt recycling is important since it is a rare and expensive metal. Cobalt is recovered as cobalt chloride, which is a basic material in Co compounds and alloys (Murano, 1997). Lee and Rhee (2002) have demonstrated a procedure for recovery of Li and Co followed by preparation of $LiCoO_2$ for use in Li-ion batteries.

The major proportion of metals used in the manufacturing of portable batteries is of virgin origin. Recovered metals from batteries are mainly used in the metals industry. Recovered metals can therefore be assumed to decrease the demand for mining of primary metal. This assumption does not apply for metals, which are mined as byproducts. Mining of Zn recovers Cd as a byproduct and it contributes 80% to the global production of refined Cd while the rest originates from recycled Cd (Plachy, 2000a). Although recycling of Cd does not lead to decreased demand of primary metal, it is assumed to do so, to allow investigation of the consequences of different recycling rates.

4. Rechargeable batteries impact on global metal flows

4.1. Natural and anthropogenic metal flows

Global metal flows have been quantified in several studies (Galloway et al., 1982; Nriagu and Pacyna, 1988; Nriagu, 1989, 1990; Azar et al., 1996). Some of these studies are based on inventories of emissions to various environmental media and the magnitude of activities. To reduce the complexity in estimating the metal flows, the mobilisation of metals in this study was calculated from data on global average values. Table 4 shows the estimated values of global mobilisation of metals, arranged in alphabetical order. Data for some other metals (As, Hg, In, Mo, Pb) are presented for comparison, since they are or can potentially be used in other types of batteries.

The natural turnover of metals is based on weathering and volcanic activity. Weathering has been estimated from average metal concentrations in the earth's crust and a geological turnover of 1.5×10^{16} g/year (Nriagu, 1990). The value chosen for geological turnover is 17% lower than the turnover of 1.8×10^{16} g/year calculated by Odum (1959). This assumption leads to a low estimate of natural turnover of metals. The natural metal occurrence in soils can be lower or higher than the natural occurrence in the earth's crust. Comparing all the metals studied (Table 4), the average natural occurrence is 31% lower in soil than in the earth's crust. The concentrations of metals in soils may be influenced by the deposition of metals from the air, chemical weathering and processes in the soil. These processes may cause greater leaching of some metals than others. The natural occurrence in soils was chosen as a basis for the calculations. This assumption leads to a low estimate of natural metal mobilisation.

Data on metal mobilisation are presented from several studies for comparison. Although these studies have not been performed using the same methodology, metals of concern can be ranked in each study. In the study by Galloway et al. (1982), Pb, Hg, Mo, Sb and As have the highest mobilisation factors. According to the studies by Nriagu (1990) and Azar et al. (1996) the metals with the highest LEIs are, in decreasing order, Cu, Pb, Hg, Mo and Zn.

Metals can also be ranked by comparison of their natural occurrence. The abundance principle indicates that there is an inverse relationship between the toxicity of elements and their natural occurrence in the environment (Håkanson, 1980). Studies by Wolterbeek and Verbrug (2001) imply that the natural abundance of metals or metal ions in the earth's crust may be regarded as a general comparative measure of metal toxicities.

A set of toxicity data, exemplified for *Daphnia magna* sp., shows that metals with very low soil concentrations have high aquatic toxicity (Fig. 2). Data from terrestrial and freshwater environments

								Î
Element	Conc. in soil ^a (mg/kg)	A. Weathering and volcanic activities ^b (ktonnes/year)	B. Mining ^c (ktonnes/year)	C. Fossil fuels ^d (ktonnes/year)	LEI ₉₉ ^e (B+C)/A	LEI ₉₀ ° (Azar et al., 1996)	LEI ₈₆ ° (Nriagu, 1989)	MF ₈₀ ^f (Galloway et al., 1982)
AI	72 000	1 100 000	23 000	34 000	0.050	0.048		
As	7.2	110	26	18	0.40	0.33	1.7	28
Cd	0.35	5.3	20	3.4	4.4	4.4	9.6	19
Ce ^g	40	1200	30	70	0.080			
Co	29	540	28	70	0.18		0.30	0.6
Cr	54	830	13 000	34	16	4.6	9.6	2
Cu	25	380	13 000	55	34	24	24	14
Fe	$26\ 000$	390 000	992 000	34 000	2.6	1.4		
Hg	0.03	1.4	2.3	10	8.8	6.5	20	275
In	0.24	4.5	0.2	0.05	0.060			
La^{g}	40	540	17	35	0.10			
Li	31	360	15	220	0.65	0.64		
Mn	550	8300	6700	170	0.83	1.1	3.7	0.5
Mo	0.97	15	129	17	9.7	8.5	13	46
s pN	40	750	12	120	0.18			
Ni	19	300	1100	570	5.6	4.8	4.5	4
Pb	19	290	3000	85	11	12	20	340
Pr^{g}	6	170	3.4	6	0.070			
Ti	2900	44 000	2400	1700	060.0			
^	80	1200	40	350	0.33	0.32	0.13	σ
Zn	60	910	7600	260	8.6	8.3	14	23
Zr	230	3500	820	140	0.27	0.3		
Sources	:: Galloway et al.	., 1982; Nriagu and Pa	icyna, 1988; Nriagi	u, 1989, 1990; Aza	r et al., 1996;	Magaw et al., 1999;	USGS, 2000; EIA	2001.
^a Conce	entration in soil (Azar et al., 1996; Sne	ller et al., 2000).				:	
^b Weath	nering mobilisatic	on is calculated using	average metal con	centrations in soil a	and a suspend	led sediment flux of 1	1.5×10^{16} grams po	er year in rivers (Nriagu,
1990). TŁ	he dissolved trace	metal flux is generall	y much lower than	the particulate flu	х.			
^c Data 1	for 1999 (USGS,	2000). Only a fraction	n of each metal mi	ned each year is re	leased into th	e environment in the	same year.	
				(

Table 4 Natural and anthropogenic global mobilisation of metals

^d (Azar et al., 1996). Production in 1999: coal 4.66 Gtonnes; oil 3.45 Gtonnes (EIA, 2001). Elemental composition in crude oil for rare earth metals (Magaw et al., 1999).

⁶ Lithospheric extraction indicator (LEI) = anthropogenic metal flows divided by natural metal flows (Eq. (1)). ⁷ Mobilisation factor (MF) = emission rate human sources/emission rate natural sources (Galloway et al., 1982). Anthropogenic emission = fossil fuels and industrial particulate fluxes. Natural emission = soil dust and volcanic-emanation. ⁸ The flow of each metal of the rare earth metal group is based on worldwide rare earth mine production (Hedrick, 2001) (1999: 82 ktonnes) and average concentrations in the source minerals bastnasite and monazite (Hedrick, 1999) (Ce 47.6%; La 24.4%; Ne 17.1%; and Pr 4.88%).



Fig. 2. Relation between lethal concentrations (LC_{50}) for *Daphnia magna* sp. and natural concentration in soil (Laveskog et al., 1976; KEMI, 1989; Dameron and Howe, 1998; Sneller et al., 2000; Table 3).

were compared due to data availability. Wolterbeek and Verbrug (2001) assessed terrestrial or freshwater data to have a low impact on the relationbetween toxicity and ships background concentrations. There is statistical evidence (χ^2 test: P > 99.9%) that the metals studied with concentrations in soil <1 g/tonne have $LC_{50} < 0.1$ mg/dm^3 (LC₅₀=Lethal concentration at which 50% of the test organisms die). For metals with higher concentrations in soil (>1 g/tonne), the relation between concentration in soil and toxicity is not significant.

Organisms and ecosystems have adapted to the natural concentrations of substances in the biosphere, i.e. the natural background levels. If a metal is present at low concentrations there may have been no need to develop adaptive physiological processes. Organisms and ecosystems are not equipped to deal with a significant increase in the concentration of metals and subsequent exposure of the organisms. Adaptation to the higher concentration level may require considerable time for biological evolution. High or increasing metal concentrations and exposure in ecosystems seem to be the major problem associated with metal use. For naturally occurring substances, the ultimate environmental target is that concentrations in the environment should be close to background values. LEIs were calculated (Eq. (1)) to compare the magnitude of anthropogenic metal flows. A high ratio between anthropogenic and natural metal flows indicates potential environmental impact of the specific metal. For several metals, e.g. Cu, Cr, Pb, Mo, Hg, Zn, Ni and Cd, the anthropogenic metal flows are 4.4–34 times higher than the natural metal flows (Fig. 3). Bearing in mind the relationship between toxicity and natural abundance, greatest environmental concern is for metals with low natural concentration in the earth's crust and high LEI, e.g. Hg, Cd, Mo and Cu.

4.2. Potential impact on global metal flows due to the use of rechargeable batteries

4.2.1. Case 1: Metal demand for batteries in 1999 Based on the total mass of batteries (Table 2) and their composition (Table 3), the total amounts of metals used in batteries were calculated (Eq. (4)). By relating the mass of battery metals to annual mining (Table 4), the metal demand for batteries can be obtained. Fig. 4 shows that metals contributing >1% to the global metal flows are (average contribution within parentheses): Cd (37%); Co (15%); La (9.5%); Pr (9.4%); Nd (8.4%); V (6.5%); Ce (4.4%); Li (3.8%); and Ni (2.0%).



Fig. 3. Lithospheric extraction indicator (LEI) related to natural concentration in soil.

Fig. 4 shows that Cd in portable NiCd batteries constitutes 13–60% of the total Cd mining, corresponding to 2.6–12 ktonnes Cd. The broad ranges are due to variations in average cell weights, energy densities and assumed recycling

rates. On the demand side, Cd is becoming a oneuse metal, and with approximately 70% is used in the manufacture of NiCd batteries (Plachy, 2000b). Of the Cd used in batteries, 75–80% is used in the production of portable NiCd batteries, while



Fig. 4. Metal demand of portable batteries as a percentage of global mining output in 1999. The bars indicate the demand of each battery technology. The lithospheric extraction indicator and the 1999 mining output for each metal is also given.

Table 5

Lithospheric extraction indicators for Case 2 assuming replacement of NiCd batteries with $NiMH(AB_5)$, $NiMH(AB_2)$, or Li-based batteries. The change relative LEI_{99} is also given

Element	NiMH(AI	B ₅)		NiMH(AI	B ₂)		Li-based		
	LEI _{Case 2}	Change LEI ₉₉ (%)	Low-high (%)	LEI _{Case 2}	Change LEI ₉₉ (%)	Low-high (%)	LEI _{Case 2}	Change LEI ₉₉ (%)	Low-high (%)
Cd	3.0	-31	-11-51	3.0	-31	-11-51	3.0	-31	-11-51
Ce	0.1	1.7	0.1-3.4	0.1	0	-	0.1	0	-
Co	0.2	1.4	0.5 - 2.4	0.2	0.6	-0.3 - 1.5	0.2	7.1	1.6-12.6
La	0.1	4.1	0.4 - 7.8	0.1	0	-	0.1	0	-
Li	0.7	0	-	0.7	0	-	0.7	0.8	0.1-1.5
Nd	0.2	1.0	0.1-1.9	0.2	0	-	0.2	0	-
Ni	5.6	0.5	0 - 1.0	5.6	0.2	-0.3 - 0.7	5.6	-0.1	-0.1 - 0.2
Pr	0.1	3.4	0.3-6.4	0.1	0	-	0.1	0	-
V	0.3	0	-	0.3	0.3	0-0.7	0.3	1.9	0.6-3.3
Zr	0.3	0	-	0.3	0.1	-0.1 - 0.3	0.3	0	-

The change relative LEI₉₉ is also given. Source: calculated from data in Tables 2-4.

the rest is used in industrial NiCd batteries (Plachy, 2000b; Nilsson, 2001).

The major end use of Ni is in the steel industry. Portable batteries account for 0.6-3.3% of the end use. Of this, NiMH batteries make up 53% and NiCd batteries 35%.

Cobalt use in batteries constitutes 7.2-22% of global Co mining. Fifty-eight percent of this cobalt is used in Li-ion(Co) and 33% in NiMH(AB₅) batteries.

Vanadium use in batteries constitutes up to 6.5% of global mining. The variation is due to the uncertainties in V content in Li-polymer (V) batteries, which can be up to 20%. Ninety-nine percent is used in Li-polymer (V) batteries, and the remaining 1% in NiMH(AB₂) batteries.

Ce, Pr, La and Nd are used in $NiMH(AB_5)$ batteries and constitute 0.38-18% of the global mining of these metals.

4.2.2. Case 2: Different battery types replace NiCd batteries

To assess the influence of portable rechargeable batteries on global metal flows, changes in LEI were calculated (Eqs. (1) and (2)). The changes were calculated for cases where NiMH(AB₅), NiMH(AB₂) or Li-based batteries were assumed to replace NiCd batteries. Table 5 presents $LEI_{Case 2}$ and the average change compared with LEI_{99} . It is further assumed that changes in demand

for metals on the battery market are reflected in corresponding changes in global mining production.

Replacement of NiCd batteries by the other battery technologies would result in a decrease in LEI for Cd of 11–51% compared with 1999. The reduction is due to the low natural occurrence of Cd and the high metal intensity for storing electrical energy. The natural occurrence of mischmetal in the earth's crust is approximately 100 times higher than that of Cd. Only one-third of the metal is required for storing electrical energy with mischmetal than with Cd. The metal intensity is 1.2 kg/kWh for mischmetal, 0.14–0.52 kg/kWh for Li and 3.4 kg/kWh for Cd (Råde and Andersson, 2001).

The replacement of NiCd batteries by NiMH(AB₅) batteries would lead to the greatest increases in LEIs for La (4.1%), Pr (3.4%), Ce (1.7%) and Co (1.4%). Metal flows due to the introduction of NiMH(AB₂) batteries would result in increases in LEI for Co (0.6%), V (0.3%) and Ni (0.2%).

The use of Ni and Cd is of the greatest environmental interest, due to their high LEIs. The LEI for Ni is 5.6 and the introduction of other battery technologies would result in a small change only. Li-based batteries would reduce LEI for Ni by 0.1% while NiMH batteries would increase the



Fig. 5. Lithospheric extraction indicator for Case 2 and relative change compared with LEI₉₉. [Data for NiMH(AB₅), LEI_{Case} 2].

LEI by 0.5%. The LEI for Cd would decrease from 4.4 to 3.0.

For the other metals studied, which show an increase of 0.1–7.1%, the LEIs do not exceed 0.7, which indicates low potential environmental impact of these metals. Large-scale introduction of batteries containing mischmetal (Ce, La, Nd and Pr) would result in an LEI of 0.11, which indicates that the increase of environmental concentrations are low compared with background concentrations of these metals. Of the total annual mobilisation of mischmetal, 7.9% originates from the combustion of fossil fuels and 2.8% from mining (Table 4).

Replacing NiCd batteries by Li-based batteries would result in an increase in LEI for Co (7.1%) and V (1.9%). The LEI for Ni would decrease by 0.1%. This small decrease is due to the fact that Ni in portable NiCd batteries constitutes only 0.2–1% of annual Ni mining 1999.

To assess whether a change would lead to a potential decrease or increase in environmental impact, Fig. 5 can be used to assess material flows. If the LEI and the relative change are high and positive for a particular metal, the environmental concern would be high. For several metals, e.g. Pr, La, etc. the LEI is <1 and the relative change is <1%. The environmental concern regarding these metals is thus low.

In the case of Cd, the LEI is >1 (3.0) and the relative change is negative (-31%). This indicates a potential decrease in environmental impact and a positive response. The LEI for Ni is 5.6 and the relative increase is small. Measures should be aimed at decreasing the LEI for Ni by employing recycling technologies that decrease primary mining of Ni.

4.2.3. Case 3: Projected battery market in 2009

Metal flows were calculated for the projected battery market in 2009 in which the total number of cells has increased 2.2 times (Fig. 1). The assumed market shares are 6% for NiCd, 53% for NiMH and 41% for Li-based batteries. The assumed market share of Li-based batteries (+6.1 times compared with 1999) may increase the demand for mining of several metals (Fig. 6). The metal demand for the battery market 2009, expressed as a percentage of annual mining output in 1999, is in the range 32–103% for Co, 22– 57% for V and 6–41% for Li. For NiMH(AB₅) batteries, the demand of Ce, Pr, La, Nd is 1–54%.



Fig. 6. Projected metal demand of portable batteries in 2009 as a percentage of global mining output in 1999. The bars indicate the demand of each battery technology. The lithospheric extraction indicator and the 1999 mining output for each metal is also given.

The amount of Cd in NiCd batteries accounts for 4-16% of global mining, compared with 13-60% for 1999.

Batteries require 2–8% of mined Ni in the projected future market case. Although a reduction in the market share of NiCd batteries would decrease the use of Ni in batteries, the introduction of NiMH batteries would cause a net increase in Ni use compared with 1999.

Nickel and Cd are of greatest environmental concern since their LEIs are greater than one: 5.7 and 3.4, respectively. The LEIs for other metals are below 0.66, which means that their material flows may have a low impact on the global level.

The LEI_{09} and changes in LEI for different metals are presented in Table 6. For the projected market in 2009, the LEI for Cd would decrease by 23% compared with LEI_{99} . The LEI for Co

Table 6

Lithospheric extraction indicators for the projected battery market in 2009 (LEI_{09}) and contribution from different battery technologies. The change relative LEI_{99} is also given

Element	LEI09	Change in L	EI ₉₉ (%)	Mean cont	tribution (%)		
		Mean	Low-high	NiCd	NiMH(AB ₅)	NiMH(AB ₂)	Li-based
Cd	3.4	-23	-8.0-38	100	0	0	0
Ce	0.086	2.7	0.20-5.1	0	100	0	0
Co	0.21	15	7.0-23	0.42	22	0.30	77
La	0.10	6.3	0.70-12	0	100	0	0
Li	0.66	1.3	0.31-2.2	0	0	0	100
Nd	0.18	1.5	0.20-1.1	0	100	0	0
Ni	5.7	1.8	0.6-2.9	3.8	72	2.0	23
Pr	0.077	5.2	0.70-9.7	0	100	0	0
V	0.34	3.4	1.9-4.8	0	0	0.60	99
Zr	0.27	0.006	0-0.012	0	0	100	0

The change relative LEI₉₉ is also given.

would increase by 15% from 0.18 to 0.21. Lipolymer (V) batteries contribute 78% to the battery-related Co flow.

For the other metals studied, the relative changes in LEI are below 6.3%. The use of Ni in NiMH batteries may be of environmental concern since its LEI is high (5.7), and the relative increase is 1.8% compared with 1999.

5. Discussion

The results show that the replacement of portable NiCd batteries by NiMH or Li-based battery technologies may lead to a decrease in the LEI for Cd of 11-51%. Meanwhile, the mobilisation of metals considered less hazardous (LEI < 0.66) than Cd would increase less than 7%. Based on this assessment, the replacement of NiCd batteries would result in decreased environmental impact.

Cadmium and Ni used in batteries are of concern due to their high LEI (4.4–5.6), and battery requirements constitute 37 and 2% of total mining 1999, respectively. An increase in the use of NiMH batteries may lead to a higher demand for Ni, resulting in an increase in the LEI of 0.6-3%. Mischmetal in NiMH(AB₅) batteries is deemed to be of low environmental concern due to its low LEI.

A growing battery market (Case 3) shows that portable batteries may be an important use for Co, Nd, La, Ce, Pr and Li. Increasing demand for these metals may result in higher metal prices, which may limit the growth of Li-ion(Co) and NiMH(AB₅) technologies.

The calculation of LEIs allows a simple environmental assessment of different metals, which can be used for early decision making. For example, molybdenum oxide has been be used as material in the positive electrodes of experimental Li-polymer batteries (Niawa, 2001). The LEI for Mo is 9.7, which indicates that its toxicological effects should be further evaluated before largescale introduction of technologies based on Mo.

Experimental Li-polymer batteries have been tested with negative electrodes based on Cu_0Sn_5 (Vincent, 2000). The high LEI (34) for Cu makes it important to consider its use in products. Even though Li-batteries demanded less than 0.014% of

the global Cu mine production in 1999, it is important to establish collection and recovery of metals from disused batteries. Iron, Zn and Cr have LEIs of 2.6–16 and their uses in products should be considered to assure their proper materials management.

The major uncertainty in metal use is the actual mobility of the metal and the exposure of organisms, especially when considering a long-term perspective. Depending on the type of application, economic value and physical properties of the metal, it may be more or less dispersed. Anthropogenic mobilisation of metals may result in the release of metals in a more or less toxic form than if naturally released. If anthropogenically emitted metals are more bio-available than naturally occurring metals, the environmental impact would be higher than that indicated by the magnitude of the LEI.

Weaknesses of the method are that regional and local effects are not considered. Even if the use of a metal is small compared with natural emissions, there may be high concentrations locally due to, e.g. mining, factory emissions, or leakage from landfills. Potential effects are considered that may lead to overestimates of environmental effects. The method used provides no full environmental risk assessment since dose–response relationships are not considered (Upham, 2000). The method used is one element of environmental risk assessment and it should be complemented with other methods of assessment.

In an ongoing risk assessment (De Win, 2000), it has been concluded that a market restriction on portable NiCd-batteries would address 2% of the total Cd emission to the environment within the European Union. The ratio of Predicted No Effect Concentrations (PNEC) to Predicted Environmental Concentrations (PEC) for different environments was calculated to be 38-225 for Cd. Assuming that NiCd batteries contribute 10% to the total Cd emission, the PNEC/PEC ratio for NiCd batteries would be 380-2250. One uncertainty affecting these results is how Cd emission will change in the future when consumers discard hoarded batteries. Most industrialized countries have implemented extensive control measures to immobilize metals treated in municipal solid waste,

which means that predicted emissions are valid for these countries only. PNEC values have been estimated for certain organisms only and synergetic effects with other substances have not been considered.

To be able to provide a reliable environmental risk assessment, a large number of parameters concerning metals must be determined including bio-availability, metal species, toxicity, exposure of organisms, synergistic effects and response. The oxidation state of the metal is one of the key problems to be overcome in future metal-ion toxicity studies (Wolterbeek and Verbrug, 2001). Limited knowledge and uncertainties concerned with these parameters motivate the use of a simplified method. The strength of the method is that it provides a fast means of analysis with small data needs. The focus is early in the cause-effect chain and gives an early warning of the potential impact of a new technology. Quantitative data are used which makes it possible to assess temporal development. The method is flexible for further development and studies at different geographical levels.

Alternative approaches to evaluating trace metal concentrations in natural systems and comparing trace metal release and transport rates from natural and anthropogenic sources have been suggested (Benjamin and Honeyman, 1992). One approach is to compare the composition of atmospheric particles with that representing the average composition of the earth's crust to indicate whether certain elements are enriched in atmospheric particulates. Another approach is to calculate the metal loading on the environment from specific human activities (e.g. the discharge of wastewater) and compare the results with natural release rates. These approaches have not been deemed appropriate for the assessment of the impact of batteries due to difficulties in obtaining the data required.

The calculated low-high values for metal flows presented in this study show the potential for improvement of metals management in batteries. Development of battery technologies should aim at high energy density and long service life. Metals from disused batteries should be recovered and regulations implemented to decrease the need for mining of virgin metals. In order to control the mobilisation of certain metals, interactions with other metal use must be addressed. For example, the demand for Zn also leads to mining of Cd, since it is a byproduct of Zn. A way of avoiding the mobilisation of metals is to deposit lowdemand metals at the extraction mine to avoid dissipative losses. Problems associated with the collection of disused batteries and the small quantities of metals available for recycling make it difficult to realize commercial recycling of batteries. Replacing NiCd batteries would lead to a decrease in the demand for Cd metal and consequently lower prices and costs for safe storage. Regulations would have to be implemented to ensure that Cd in products and from mining were collected and stored safely. The use of Cd in industrial NiCd batteries, a product application where high collection efficiencies are achievable, may give market incentives for continued collection of Cd containing products.

In order to decrease the uncertainties in the results, further studies could focus on the monitoring of metal flows and estimating the fluctuations. Existing indicators can be further developed and refined in order to allow assessment on different temporal and geographical scales. Metal flows arising from small sealed lead-acid batteries and different types of non-rechargeable (primary) batteries should be assessed. The bio-availability of metals and the exposure of organisms must also be further explored.

6. Conclusions

The assessment of global metal flows arising from the use of portable rechargeable batteries shows that Ni and Cd are of concern due to their high LEIs (4.4-5.6) and that battery metal demand constitute 37 and 2% of global mining 1999, respectively.

In the case of complete replacement of portable NiCd batteries by NiMH or Li-based batteries, the LEI for Ni (5.6) would change by -0.1-0.5% and the LEI for Cd would decrease from 4.4 to 3.0 (-31%). Meanwhile, the mobilisation of metals considered less hazardous than Cd (LEI < 0.65) would increase less than 7%. An increase in the use of NiMH batteries may lead to a higher

demand for Ni, resulting in an increase in the LEI of 0.6–3%. Based on this assessment, the replacement of NiCd batteries would result in decreased environmental impact.

A growing battery market shows that portable batteries may be an important use for Co, Nd, La, Ce, Pr and Li. Increasing demand for these metals may result in higher metal prices, which may limit the growth of Li-ion(Co) and NiMH(AB₅) technologies. Higher prices for metals used in batteries may create incentives for battery collection and recycling.

To decrease the impact on global metal flows arising from the use of portable batteries the following points should be considered: (1) development of battery technologies should aim at high energy density and long service life; (2) metals with high natural occurrence should be used; (3) metals from disused batteries should be recovered and regulations implemented to decrease the need for mining of virgin metals.

The method used enables an assessment early in the cause–effect chain, when few data about toxic effects are available. It can also be used to assess whether environmental problems are shifted from one to another.

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Paper III

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Environmental assessment of vanadium redox and lead-acid batteries for stationary energy storage

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Abstract

The environmental impact of both the vanadium redox battery (vanadium battery) and the lead-acid battery for use in stationary applications has been evaluated using a life cycle assessment approach. In this study, the calculated environmental impact was lower for the vanadium battery than for the lead-acid one. The net energy storage efficiency of the vanadium battery was greater due to lower primary energy needs during the life cycle. Favourable characteristics such as long cycle-life, good availability of resources and recycling ability justify the development and commercialisation of the vanadium battery. © 1999 Elsevier Science S.A. All rights reserved.

Keywords: Life cycle assessment; Lead-acid secondary batteries; Vanadium redox batteries; Applications/stationary energy storage

1. Introduction

To overcome daily and seasonal varieties in the supply of, e.g., photovoltaic cells and wind energy, these technologies have to be combined with energy storage systems which improve their usefulness as electric power sources. In remote places, an autonomous electrical system may be less expensive, have greater efficiency and less impact on the environment than if the load has to be connected to a high capacity grid [1].

Secondary batteries used as storage systems provide several favourable characteristics in energy distribution [2,3]. Their fast response time makes them suitable for dynamic system operations. Batteries connected to the electricity grid can improve power quality and reliability. Lower environmental impact may also be achieved by load levelling and peak load reduction. Thus, it is possible to achieve a high utilisation rate for generating facilities which have high production efficiency.

To assess the environmental characteristics of energy storage in batteries, the efficiency and the environmental impact during the life cycle of the battery has to be considered. Several authors [4-6] have made life cycle assessments of lead-acid batteries as well as other batteries to be used in electric vehicles. The energy and environmental impacts of lead-acid batteries have also been studied [7].

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To increase the share of renewable energy, energy storage systems need to be inexpensive and large. Classical secondary batteries suffer from limited system life and high cost. Systems with a combination of water electrolysis, hydrogen storage and fuel cells have low overall energy efficiency. One interesting battery under development is the vanadium redox flow battery (vanadium battery). It offers high overall efficiency and the cost for additional storage capacity is limited to the active materials and storage tanks. In this paper, the environmental impacts of both the vanadium battery and the lead-acid battery have been compared for use in a back-up power system.

2. Vanadium and lead-acid batteries

Demonstration units of the vanadium battery have been built in Japan and Thailand [8] where the system has been tested for load levelling and as back-up power in photovoltaic plants. The battery consists of two electrolyte containers with the electrolytes of V^{2+}/V^{3+} and V^{4+}/V^{5+} in sulphuric acid solution, two electric pumps and a battery stack. The electrolytes are pumped into the stack where they are separated by an ionic membrane. The electron exchange takes place at carbon felt electrodes in the aqueous phase according to the following reaction [9] (discharge in the right direction):

Positive electrode: $VO_2^+ + 2H^+ + e^- \Leftrightarrow VO^{2+} + H_2O^{2+}$

Negative electrode: $V^{2+} \Leftrightarrow V^{3+} + e^{-}$

One advantage over other redox flow batteries (e.g., the $Fe^{2+/3+}-Cr^{2+/3+}$ system) is that cross contamination of the electrolytes does not damage the system since the metal ions are of the same element. Electrolyte service life is thus indefinite. At the current stage of development, the stack membranes must be replaced approximately every 5 years, but cycle-life for the whole system is expected to be rather high. Favourable features are that the capacity can be increased by enlarging the size of the storage tanks and the power output can be raised by increasing the flow rate or enlarging the stack assembly. The battery can be recharged electrically, or by exchanging the electrolytes by charged ones.

The lead-acid battery is the most commonly used battery in terms of kW h. It is well established since it is less expensive than other batteries, has high peak-power and is recyclable. In stand-by power applications, service life can be more than 10 years and batteries built for cycling can have a cycle-life of 1500-2000 cycles. Deep discharges as well as over-charging shorten cycle-life and the battery requires maintenance by water refilling due to gas evolution [10].

3. Methodology

In this paper, a life cycle assessment (LCA) approach was used to compare the batteries. LCA is a technique for

Table 1

Specifications of the model systems for storage of electrical energy



Fig. 1. Phases of a life cycle assessment [11].

assessing the environmental aspects and potential impacts associated with the life cycle of a product [11]. The phases within this work compile an inventory of relevant inputs and outputs of a product system (Fig. 1). The environmental impacts associated with the inputs and outputs are evaluated and interpreted to the objectives of the study.

The inventory of emissions and energy was assisted by the software LCA inventory tool [12]. The impact assessment was made by classifying and characterising the inventory results with regard to global warming potential, photo-oxidant formation, acidification, eutrophication and resources. The weighting (aggregation of all the inventory results to a single impact value) was made by means of the quantitative methods of Environmental Theme (ET) and Environmental Priority Strategies (EPS) in product design [13]. In the ET method, the different impacts are weighted against political emission goals for various impact cate-

	Lead-acid battery	Vanadium battery
References	[15,16]	[17,18]
Design and assembly	Traction battery with tubular plates, 4–9 wt.% Sb, $C_5 = 1125$ Ah cell ⁻¹	4 stacks \times 80 cells (serial)
	2 (parallel) \times 100 cells (serial)	
Positive electrode	$PbO_2(s)/PbSO_4(s)$	$VO_2^+(aq)/VO^{2+}(aq)$
Negative electrode	$Pb(s)/PbSO_4(s)$	$V^{2+}(aq)/V^{3+}(aq)$
Electrolyte	1.295 kg dm ⁻³ , 5 M H ₂ SO ₄	1.8 M V in 4.2 M H ₂ SO ₄ ^a
Ion membrane	Polyethylene separator	Selemion membrane
Discharge voltage 5-90% SOC (V)	180-207	310-500
Operating temperature (°C)	- 10 to 40	10 to 45
Energy efficiency (%)	70-80	72-88 ^b
Specific energy at 20°C (W h kg ⁻¹)	37	(28) ^a 20
Energy density at 20°C (W h dm ⁻³)	105	(47) ^a 30
Cycle-life (cycles)	1800°	≫ 2000 ^d
Total system volume (dm3)	4300	(9600) ^a 15 000
Mass of one system (kg)	12 100	(16 100) ^a 22 200

Nominal power 50 kW and energy storage capacity 450 kW h.

^aPossible to have 3 M vanadium in 5 M H₂SO₄: related improvements are shown in brackets

^bIncluding 3% energy loss to pumps. ^cAt 35% depth of discharge (1200–1500 cycles at 70–80% DOD).

^dNot determined. Shelf life is very high but at current state of development, maintenance includes replacement of membranes approximately every 5 years.
Table 2

		-					-	
cycling f	for 20 years (72	300 cycl	es) deliver	ing	150 kW l	h day ⁻¹		
Material	requirements ^a	for the	operation	of 1	ead-acid	batteries	on	daily

Material	Component	Mass (kg)	Wt.%
Lead	Active material,	29 400	61.2
	grids and poles		
Water	Electrolyte (dilution	6400	13.3
	to 1.295 s.g.)		
Sulphuric acid (pure)	Electrolyte	4600	9.6
Polypropylene	Cases and covers	3888	8.2
Sb, Sn, As	Grid alloys	1012	2.1
Polyethylene	Separators	960	2.0
Polyester	Tubular mats	144	0.3
Copper	Connectors	130	0.3
Others	Expander and oxygen	1440	3.0
	in PbO ₂		
Total		47974	100.0

^a4×200 cells. Recycled materials not taken into account.

gories (e.g., acidification, greenhouse effect). The EPS method, on the other hand, is based on the willingness to pay to restore five safeguarded objects (e.g., human health, natural resources etc.) to their normal status.

4. Goal definition and scoping

The scope of this paper is to assess and compare the environmental impacts of the vanadium and lead-acid batteries. The net energy storage capacity and the availability of vanadium and lead resources are compared.

For the lead-acid battery, the influence of 50 and 99% secondary lead-acid use and different maximum cycle-life is assessed. The functional unit (FU) is defined as an electricity storage system with a power rating of 50 kW, a storage capacity of 450 kW h and an average delivery of 150 kW h electrical energy per day for 20 years. These specifications exemplify the electricity requirements for several remote houses for 10-70 persons [2] and provide an autonomous system for 3 days. The analysis is restricted to the energy storage systems and space require-

Table 3

wirements^a for the operation of the vanadium battany for 20

Material requirements for the operation of	the valiadium battery for 20 years			
Material	Component	Mass (kg)	Wt.%	
Water	Electrolytes (1.8 M V, 4.2 M H ₂ SO ₄)	11 251 ^b	47.7	
Sulphuric acid (pure)	Electrolytes	6103 ^b	25.9	
Vanadium pentoxide	Electrolytes	2369 ^b	10.0	
Steel	Pumps, motors, racks, bolts	2516	10.6	
Polypropylene	Electrolyte containers	600	2.5	
Polypropylene, rubber, carbon black	Flow frames, bipolar plates	328	1.4	
Copper	Connectors, end electrodes	184	0.8	
Polysulphone and fluoride ^c	Ionic membranes	104	0.4	
Carbon felt, graphite	Electrodes	60	0.3	
Others		86	0.4	
Tutal		22 601	100.0	

^aRecycled materials are not taken into account.

^bIncludes 1–2% addition due to losses during use

^c Polystyrene manufacturing assumed.

ments, losses in converters, control units etc. are assumed to be equal. The environmental impact of electricity production to charge the batteries is not included since equal energy efficiencies is assumed. The energy storage system is furthermore assumed to be assembled and used in the Gothenburg region, Sweden. The vanadium battery is not yet in full-scale production, but is assumed to be assembled in Sweden and materials are manufactured by domestic suppliers, whenever possible. Important construction materials are analysed, by collecting available data, from the extraction of resources to the final deposit. Materials which are recycled and re-used in other products are allocated with the 50/50 method [14]. Post operation, the system is disassembled and the materials re-used or deposited.

The specifications of the systems studied are shown in Table 1. The lead-acid battery is of traction design with antimony grid-alloys for long cycle-life.

5. Results

5.1. Inventory

The energy and material requirements for the vanadium battery were based on a hypothetical manufacturing scenario and these data may differ for a future production. The vanadium electrolyte is assumed to have very long life and its only treatment is filtering before re-use. Vanadium can be extracted in different ways, e.g., by mining or recovery from petroleum residues. Due to the lack of data, a scenario for the vanadium recovery from boiler soot needed to be estimated. This resource was assumed to be a secondary material since it would otherwise have been deposited.

Data for the manufacturing of lead-acid batteries were collected from factories supplying Swedish battery manufacturers. The batteries were recycled at Boliden Bergsöe in Landskrona, Sweden, where lead and alloying metals are recovered. Less than 50% secondary lead is used in

23

24 Table 4

Energy requirements of the battery life cycles and to cover losses during use of the lead-acid and vanadium battery systems (20 years and delivering 1095 MW h)

	Lead-acid			Vanadium			
	50% seconda	ary Pb	99% seconda	ary Pb	99% secondary V ^a		
	Electricity (MW h)	Other primary energy (MW h)	Electricity (MW h)	Other primary energy (MW h)	Electricity (MW h)	Other primary energy (MW h)	
(1) Material	49	118	21	108	7	30	
(2) Transport	0	18	0	15	0	3	
(3) Battery production	64	25	96	25	29	10	
(4) Recycling	6	14	6	14	3	-4	
(5) The sum of (1) to (4)	120	175	92	162	39	39	
(6) Primary energy (5) × 0.4 ^b	70		65		16		
(7) Electricity losses during use ^c	365		365		365		
(8) The sum of electricity $(5) + (6) + (7)$	555		522		420		
(9) Net electricity delivered from the battery during 20 years	1095		1095		1 095		
(10) Gross energy requirement (8) + (9)	1650		1617		1 515		
(11) System efficiency (9)/(10)	0.66		0.68		0.72		

^a99% secondary vanadium assumed to make the systems comparable.

 5 7 9 10 d. Efficiency in the conversion of primary energy to electricity. c At 75% total efficiency (losses in batteries and converter) and 1095 MW h.

new batteries since the lead oxide production requires high purity.

The material requirements for the lead-acid battery are shown in Table 2. Over 20 years, 200 cells will be replaced four times and approximately 25000 l of water will be added intermittently. The data for the vanadium battery (Table 3) include electrolyte adjustment, pump maintenance and replacement of ionic membranes every 5 years.

dium battery since it uses more and heavier materials. For both batteries, heat is recovered from the recycling process when polypropylene containers are incinerated, which gives an energy gain for the vanadium battery. For the lead-acid battery, an increase in the use of secondary lead from 50-99%, the electricity and primary energy needed in the material production decrease by 43% and 8%, respectively.

To compare the overall energy requirements, primary energy (specific heat value) was recalculated as electrical energy. The energy efficiencies of the batteries were assumed to be equal since it varies with the rate of discharge and no definite values were found for the chosen systems. The net system efficiency values in this study vary with the chosen allocation method. The lead-acid battery re-

5.2. Impact assessment

Table 4 shows that the energy used for transportation is considerably greater for the lead-acid than for the vana-

Table 5	
Emissions of selected substances and contributions from the different	processes during the life cycle of the lead-acid and vanadium batteries

Emitted substances and contributing processes	Lead-acid 50% secondary lead	Lead-acid 99% secondary lead	Vanadium 99% secondary V ^a	
$\overline{\text{CO}_2 (\text{ton FU}^{-1})}$	29.3	25.4	8.5	
Mtrl/Trp/Prod/Recyc (%)	43/18/19/20	36/19/22/23	67/11/16/6	
SO_2 (kg FU^{-1})	215	147	28	
Mtrl/Trp/Prod/Recyc (%)	38/7/8/48	11/7/11/70	61/23/20/-4	
$CO(kg FU^{-1})$	57	42	5.2	
Mtrl/Trp/Prod/Recyc (%)	35/38/34/-7	21/42/46/-9	41/87/7/-35	
CH_4 (kg FU^{-1})	33	32	8	
Mtrl/Trp/Prod/Recyc (%)	16/6/20/57	14/5/21/60	102/6/1/-8	
NO_{x} (kg FU ⁻¹)	242	172	45	
Mtrl/Trp/Prod/Recyc (%)	48/39/6/6	38/45/9/9	47/45/6/2	
N_2O (kg FU ⁻¹)	0.72	0.52	0.15	
Mtrl/Trp/Prod/Recyc (%)	34/47/16/3	17/57/22/4	19/49/29/2	

Emissions from electricity production to charge the batteries are not included.

^a99% secondary vanadium assumed to make the systems comparable. FU = 150 kW h day⁻¹ for 20 years, $E_{\rm max}$ 450 kW h. Mtrl = materials, Trp = transport, Prod = battery production, Recyc = recycling.

Table 6

Results of the characterisation of five impact categories for lead-acid and vanadium batteries (electricity production to charge the batteries is not included) Impact categories Lead-acid 50% Lead-acid 99% Vanadium 99% Ratio Ratio

Impact categories	Lead-acid 50%	Lead-acid 99%	Vanadium 99%	Ratio	Ratio	
	secondary Pb	secondary Pb	secondary V ^a	Pb _{50%} /V	Pb _{99%} /V	
Global warming potential ^b	21617	17 366	8929	2.4	1.9	
Photo-oxidant formation ^c	116	96	29	4.0	3.3	
Eutrophication ^d	32	23	6	5.3	3.8	
Acidification ^e	229	180	59	3.9	3.1	
Resources ^f	648	258	70	9.3	3.7	

^a99% secondary vanadium assumed to make the systems comparable.

^bCarbon dioxide equivalents (CO₂, CO, N₂, CH₄).

^cEthene equivalents (C₂H₄, CO, CH₄, hydrocarbons)

^dChemical oxygen demand equivalents (BOD, COD, NH₃, NO_x, N_{tot}, P_{tot}).

^eSulphur dioxide equivalents (SO_x, NO_x, NH₃, HCl, H₂S). ^fUse to reserve ratio.

quires 2.9–3.5 times more energy than the vanadium battery in the production and recycling phase. The energy losses of the batteries make up 22-24% of the total energy requirements during the life-cycle. For the vanadium battery, the net system efficiency can be increased to 0.89 if the whole energy storage capacity (450 kW h) is utilised daily. A higher utilisation of the potential capacity is not possible for the lead-acid battery, since deep discharges shorten its cycle-life.

Table 5 shows that the carbon dioxide emissions for the lead-acid battery were mainly due to the production of materials (43 and 36%), where lead mining and polypropylene production were the largest contributors. Also for the vanadium battery, the production of materials (67%) was the largest emission of carbon dioxide, mainly from steel production.

In the recycling processes, polypropylene is incinerated and the energy is used for district heating, which is assessed as a decrease in emissions of SO_2 , CO and CH₄. For the lead-acid battery, the smelting process used to produce secondary lead needs considerable amounts of primary energy, which also cause large SO₂ emissions.

The emission values are multiplied with characterisation indices [14] to get a value of the impact on a certain category. The results show that the vanadium battery has a lower impact (1.9-9.3) compared to the lead-acid battery (Table 6). For the lead-acid battery, the use of resources shows the greatest difference between 50% and 99% sec-

ondary lead use (60%). For the other impact categories, a doubling of the secondary lead use decreases the environmental impact by 17-28%.

Weighting showed that the vanadium battery had a lower environmental impact value than the lead-acid battery (Table 7), mainly due to less use of oil resources and lower carbon dioxide emissions. The largest environmental impact for the vanadium battery originated from the production of polypropylene tanks and flow frames as well as steel stacks. For the lead-acid battery, lead mining and refining contributed most to the impact, followed by polypropylene production.

In contrast to ET and other weighting methods, the EPS method evaluates the outtake of primary resources. The degradation of metals resources is included in the second row in Table 7. The resource weighting is based on the geochemical occurrence in the Earth's crust which gives scarce metals a high index (240 ELU kg⁻¹ for Pb and 28.3 ELU kg⁻¹ for V, respectively). Therefore, the extensive use of lead is the dominant contributor to the environmental impact value of the lead-acid battery. The use of secondary material decreases the environmental impact for the product examined since the burden is shared with (allocated to) the products which used the material first. The ratios between the batteries range from 2.9 to 682. When 100% primary vanadium is considered, the EPS resource ratios are 3.7 and 85 (99% and 50% secondary Pb).

Table 7

Total environmental impact of lead-acid and vanadium batteries after weighting with the Environmental Priority Strategies (EPS) in product design and the Environmental Theme (ET long-term goals) method

	Lead-acid 50% secondary Pb	Lead-acid 99% secondary Pb	Vanadium 99% secondary V ^a	Ratio Pb _{50%} /V	Ratio Pb _{99%} /V
EPS excluding resource weighting (ELU)	10259	7068	2117	4.8	3.3
EPS including resource weighting (ELU)	1.8×10^{6}	78084	2639	682	30
ET long-term goals	3.3×10^{6}	2.7×10^{6}	9.4×10^{5}	3.5	2.9

^a99% secondary vanadium assumed to make the systems comparable



Fig. 2. Environmental impact during the life cycle of the lead-acid battery assuming different operational cycle-lives at 30% DOD and secondary lead use (weighted with the EPS method including resource out-take).

Fig. 2 shows how the environmental impact of the lead-acid battery, weighted with the EPS method, depends on cycle-life and use of secondary lead. The impact can be decreased by 94% if the rate of secondary lead use increased from 50 to 99%, at a cycle-life of 1800 cycles.

5.3. Use and resources of vanadium and lead

The capacity-determining elements in the batteries studied are the active materials vanadium and lead. Table 8 shows the theoretical energy storage capacity calculated by assuming that all reserves were used to build batteries with the system specifications and thereafter kept in functional batteries. This storage capacity is greater for the vanadium battery if the known vanadium reserves were used.

Vanadium has a wide industrial usage, e.g., in metallurgy, electronics and dyeing. Vanadium gives additional strength and resilience to alloyed steel and its use in metallurgical applications accounts for 97% of the total use in the USA [19]. The availability of vanadium and the economics of its production are intimately connected with the particular co-product involved. About 60% of the world's supply of vanadium originates from vanadiferous

Table 8

Annual production rates and reserves of lead and vanadium [23]

magnetite deposits from which vanadium is extracted as a by-product of iron.

Vanadium is a trace metal in petroleum products and a major anthropogenic source of vanadium is the combustion of fossil fuels. Reserves in oil sands worldwide are estimated to supply vanadium for an exceedingly long time into the future. Vanadium recovery plants, e.g., in Japan (Kashima-Kita) and Germany (Gfe), have been constructed to treat fly ash from boiler plants fired by the Venezuelan crude oil (Orimulsion). Orimulsion has an average vanadium content of 310 ppm and its increasing use and recovery may contribute to an annual production of 4200 tons of vanadium by the year 2000 [20].

More than 70% of lead production in the western world is used in batteries and the remaining 30% in chemicals, extrusions and alloys. Stationary batteries account for 15%, whereas 75% is used for starting, lighting and ignition (SLI) batteries in cars and the rest (10%) in traction batteries [21]. About 55% of the refined lead production of the western world (total 5000 ktons in 1997) consisted of secondary lead. When secondary lead is used in batteries, impurities can promote self-discharge and accelerate water losses due to electrochemical dissociation. For low-maintenance, high-quality batteries (e.g., submarine and stand-by power), a maximum of 20 to 60% secondary lead is used to fulfil the lead specifications. However, traction batteries which are regularly discharged and daily overcharged can be built with up to 100% secondary lead without affecting quality [22].

6. Discussion

The lead-acid battery is well-established and the inventory data were taken from real production which makes the results reliable. The vanadium battery is still under development and no large scale production data are available. There are considerable uncertainties about manufacturing and maintenance, and unidentified impacts can be expected. To obtain more reliable data on the vanadium battery, the development and possible future commercial introduction of this battery must be followed. The manu-

Annual production rates a	innual production rates and reserves or lead and vanadium [25]											
	Primary production worldwide 1997 (ktons pure element year ⁻¹)	Reserve ^a (ktons)	Static reserve life ^b (years)	Material requirement ^c (kg pure element kW ⁻¹ h ⁻¹)	Theoretical energy storage capacity ^d (TW h)							
Lead	2900	68 000	24	16	4.2							
Vanadium (mining)	35	10 000	286	3	3.3							
Vanadium (recovery)	3.4 ^e	6000	1974	3	2.0							

Vanadium can be either mined or recovered from petroleum residues and spent catalysts [23].

^aIdentified resources mined economically at prevailing prices

^bNumber of years the reserve will last at present mining rate.

^ckg material kW⁻¹ h⁻¹ installed capacity of the studied system

^dReserve divided by material requirement.

^ePartly secondary vanadium from spent catalysts.

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facturing and recycling of lead-acid batteries in Sweden as well as electricity production are considered to have low emissions compared to other production sites in the world. Thus, the environmental impact of the lead-acid battery has to be classified as a low impact scenario in comparison to a lead-acid battery life cycle in another country.

Depending on the type of electricity production (e.g., coal fired boiler or hydropower) to charge the batteries, the environmental impact to cover energy losses over a battery life cycle is often higher than the energy used in the manufacturing of a battery. If the production of electricity for charging the battery causes high emissions, the development of a more efficient battery requiring greater energy in the production phase could prove worthwhile.

The vanadium battery had a higher net energy storage capacity than the lead-acid battery. The difference would increase more if the greater specified energy efficiency of the vanadium battery had been considered and if the whole potential capacity would have been fully utilised.

The results of the impact assessment indicate that the vanadium battery provides energy storage with lower environmental impact than the lead-acid battery. System improvements with regard to the environmental impact of the lead-acid battery would be most effective with greater use of secondary lead and improved battery life. This may be achieved by the development of less expensive secondary lead refining techniques and by new lead specifications [24]. The cycle-life may be improved by optimised lead alloys and charging regimes as well as by introducing mechanical pressure on the electrodes.

In a stationary application, the weight of the battery system is of limited importance. However, process and transport emissions are almost proportional to weight and environmental benefits are achieved by reducing the material requirements and transportation needs. The mass of the vanadium battery system is mainly made up by water (48 wt.%). This water can be distilled and added to a concentrated electrolyte at the site of use. The development of electrolyte with higher concentration can reduce the voluume of the storage tanks and the space requirements for the installation.

For both batteries, the use of fossil fuels and release of carbon dioxide, mainly in the processes of metal extraction and plastic production, have a significant impact.

Improvements should be directed towards sustainable processes if the batteries are to be competitive in the long run. This implies material substitution to the least harmful alternative and higher efficiency in material and energy utilisation. Measures should also be taken to sustain or improve long-term productivity and biodiversity [25].

The toxic properties of lead have been known for thousands of years and this has led to strict environmental regulations. Mayer and Wilson [26] have reviewed the exposure ways and health effects of lead, concluding that lead-in-blood levels have decreased during recent decades, at least in the more industrialised countries. In contrast to lead, vanadium is considered to be essential for a number of organisms. The toxicity of vanadium compounds decreases with both decreasing oxidation state and solubility [27]. The toxic effects have been reported to be acute, never chronic.

Extraction of resources from the lithosphere increases the risk of future contamination. The EPS weighting method tries to include this impact by evaluating the outtake of resources. As indicator of global sustainability, the ratio between anthropogenic extraction from the lithosphere (i.e., mining, fossil fuels) and natural flows (i.e., weathering and volcanic processes) gives an index of the human impact on bio-geochemical cycles. If this ratio exceeds one, anthropogenic extraction can have a major influence on the natural flows and quantities in the ecosystem. The ratios were calculated to be 12 and 0.32 for lead and vanadium, respectively [28]. Materials accumulated in the technosphere tend to leak to the ecosphere and these values suggest that we can expect increasing background lead concentrations in the future.

The batteries studied consist of relatively pure active materials which makes these batteries more suitable for recycling than many others. The recycling rate of used SLI batteries in many European countries is 80-95% [29] and even higher for industrial batteries. Metals can be locked up by their use in batteries, keeping them concentrated in the technosphere and thus preventing them from being dispersed in uncontrolled flows (Hoover technology) [30]. However, this requires high recycling rates, low process losses and high levels of re-use. The technological opportunities to achieve a closed technospheric flow of lead were investigated by Karlsson [31]. He concluded that, with very high recovery of lead-acid batteries in Sweden, the lead losses to the environment from production and recycling processes were very small compared to natural lead flows and historical lead losses during industrialisation.

Provided that no other means for SLI batteries than lead-acid becomes commercially available, the increase in cars worldwide will compete with stationary batteries for lead reserves. However, by utilising a dual battery system in cars, the use of lead can be reduced substantially [32]. Lead in other products will probably decrease in accordance with political policy. The Swedish Parliament has adopted government bill 1990/91:90 [33] with the aim of phasing out the use of lead in the long run, mainly through voluntary measures.

A large-scale introduction of vanadium batteries would increase the demand for vanadium and its mining. Vanadium recovery from boiler soot has great potential and is growing in importance due to stricter environmental legislation. This treatment gives better resource utilisation and may substitute the need for vanadium (and e.g., nickel) mining and reduce the need for landfill areas and the risks of contamination. On the other hand, the combustion of oil does release, for example, huge amounts of carbon dioxide

and some of its environmental impact could be allocated to products using vanadium, in order to indicate that the resource is not free of pollution.

The result of this environmental assessment provides no definite answers since it changes with the analysed system. New processes developments and environmental findings can change the evaluation of certain emissions and materials. The LCA methodology can, however, be used to find processes where significant improvements can be made, as well as new questions generated. In this study, the vanadium battery was found to make less environmental impact and have higher energy efficiency than the lead-acid battery. Favourable characteristics such as long cycle-life, good availability of resources, and recycling ability justify the development and commercialisation of the vanadium battery.

7. Conclusions

In this study, the calculated environmental impact was lower for the vanadium battery than for the lead-acid one. The net energy storage efficiency of the vanadium battery was greater due to lower energy losses during the life cycle. Favourable characteristics such as long cycle-life, good availability of resources and recycling ability justify the development and commercialisation of the vanadium battery.

The energy requirements (recalculated as electricity) for the production and recycling phase were 2.9–3.5 times greater for the lead-acid battery than for the vanadium battery. The resulting net energy efficiency was 0.68 for the lead-acid and 0.72 for the vanadium battery.

With the EPS weighting method, the greatest environmental impact of the vanadium battery originated from the production of polypropylene and constructional steel. For the lead-acid battery, lead extraction contributed most to the environmental impact, followed by polypropylene production.

The recovery factor of lead-acid batteries and the rate of re-use of secondary lead in new batteries is of major importance for the environmental impact. To avoid accumulation of lead in the ecosphere and material constraints, secondary smelters should focus on producing primarygrade metal. Battery manufacturers should also endeavour to develop new lead alloy specifications and batteries of improved design with operational procedures for long battery life.

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Paper IV

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Life cycle inventory data for materials grouped according to environmental and material properties

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Abstract

Environmental data are presented for material groups to be used in simplified life cycle assessments at an early stage in product design. Life cycle inventory (LCI) data from cradle to gate was evaluated for 214 material cases used in mechanical design. Based on their environmental and physical properties materials they were structured into 17 different groups. The environmental characteristics for each material group were expressed in terms of LCI data as well as characterised and weighted inventory data. LCI data categories contributing significantly to environmental impact were identified. Multivariate analysis showed weak correlation between material properties and environmental impact. The environmental data presented provide averages of LCI data for each material group and can be used as estimates when LCI data for specific materials are missing.

Keywords: screening LCA, Life cycle inventory, Environmental impact, Material characteristics, Material classification, Material selection, Multivariate analysis, Eco-indicator 99

1. Introduction

To consider environmental aspects in product development there is a need for easily accessible environmental information. Early in product design only a limited amount of information is available, making it difficult to assess the potential environmental impact of products. By using basic characteristics of materials or products, classes with different properties can be identified. A classification of materials or products early in the conceptual design phase can be used to identify where significant environmental impact may occur. Hereby the further design process can focus on reducing the environmental impact of the most important aspects.

Numerous studies have been conducted in order to identify classes and relate their characteristics to environmental impact. The literature includes studies on chemicals [1], materials [2-7], products [8-15] and firms/ industrial branches [16, 17].

For products, which can be classified as passive (i.e. energy is not required for their function), the major environmental impact throughout the life cycle is due to the production

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of materials [14] or the disposal. The environmental impact of materials can be calculated by multiplying the mass of the material with an environmental index. Weighted life cycle inventory data for materials and processes can be used to perform simplified environmental assessments of different designs [18-20].

To select a material with minimal environmental impact for the function of a product, a wide range of parameters has to be considered. By relating different material properties to each other, material charts can be used to select materials which fulfil certain criteria, e.g. density, thermal conductivity, energy content [3, 21-23]. Holloway [4] developed material selection charts to present the relationship between material properties (elasticity and strength) and corresponding air and water pollution. A limitation of Holloway's chart [4] of air pollution is that it does not include CO₂ emissions. Since environmental impact of materials occurs in many different ways, it can not be assured that optimisation for minimum energy use, water- or air pollution are the most important environmental aspects. It is therefore relevant to present weighted [24] environmental impact of different groups of materials.

Early in the design process, little data is available on specific materials and amounts of materials, which are going to be used in a new design. Average environmental impact data for material classes may be used as a first estimate. In a study by the authors [7], Eco-indicator 99 (ECO'99) weighted LCI data for 400 material cases were grouped based on their similar environmental and physical properties. Seventeen material groups were proposed to provide optimum accuracy. However, detailed LCI data for these material groups has not been analysed or presented. Correlations between material and environmental properties may be used to predict environmental performance of materials. The literature review shows that there is a need to increase the understanding of environmental impact of different materials.

2. Goal and scope

The goal of this study is to present environmental data for material groups. The data is intended to be suitable for simplified life cycle assessments of products in mechanical design at early phases in product development and as estimates when detailed LCI data is missing. LCI data for a material may be estimated by assigning it to a group, based on its belonging to a material class or its physical properties.

The study further aims at identifying LCI data categories and environmental damage categories contributing significantly to weighted environmental impact of different material groups. The most important LCI data categories for each material group may be used when compiling life cycle inventories for specific materials.

A third goal is to analyse correlations between different environmental parameters and material properties. If relationships exist, material properties can be used to predict the environmental performance of a material.

The scope of the study is to analyse different material classes used in mechanical design (e.g. polymers, metals, porous ceramics, woods and composites). Due to the great variability of recycled materials and lack of data for engineered ceramics, their environmental properties were discussed only.

3. Method

Grouping of materials and identification of the number of material groups that can be used to present average environmental data with low standard deviation have been presented in a previous study [7] by the same authors. The materials were grouped to increase data availability for product designers and be representative for the environmental performance of materials. The criteria for grouping were based firstly on environmental properties and, secondly, on the mechanical and physical properties of the materials. Cluster analysis and scatter plots of weighted environmental impact were used to identify groups of different materials. Data consistency for the LCIs of material cases was ensured regarding the system boundaries and allocation principles used [7]. If a particular group of materials had a standard deviation greater than 30% of the group average, analysis of material properties (e.g. material composition or density) was conducted for further sub-division [7]. The grouping resulted in 17 groups the average values of which represent weighted environmental impact with an average standard deviation of 22% [7]. The standard deviation represents the variability of the average LCI data for production of different materials and similar materials at different sites.

Average data were presented for density, elasticity modulus and yield strength. LCI data from cradle to gate was analysed for 397 material cases, covering country specific and global average production during the 90's [25]. The databases included e.g. ETH-ESU 96, Buwal 250, Idemat 2001, Franklin US LCI and industry data [25]. The life cycle inventories included emissions for energy generation and transportation related to the production of materials. The environmental performance for each material was expressed in terms of LCIs of data categories as well as characterised and weighted inventory data. Default indices were used for the weighting method ECO indicator'99 (ECO'99, H, A) [26, 27] since the method is comprehensive and provides indices for many data categories. ECO'99 was based on normalisation values for Europe in a hierarchist (H) valuation perspective (balanced time perspective, consensus among scientists determines inclusion of effects) and average weighting (A) (human health 40%, ecosystem quality 40% and resources 20%). Weighting with EPS2000 [28, 29] was evaluated to assess the consequences on material grouping and identification of significant environmental impact.

LCI data categories contributing significantly to ECO'99 weighted environmental impact were identified for each of the 17 material groups. Average LCI data and standard deviation for each material group were presented for the environmentally significant data categories. Average ECO'99 weighted data and the pathways for environmental impact were presented for each material group. Activities in the production of materials causing significant environmental impact were not identified.

It can be assumed that e.g. rare elements require more energy for extraction and refining, resulting in increased environmental impact. The hypothesis that there exist correlation between environmental or material properties and environmental impact was therefore tested. Linear regression analysis and multivariate analysis were applied to identify correlation between different parameters e.g. density, elasticity modulus, tensile strength and primary energy, CO_2 , NO_x and weighted environmental impact.

In Section 4.1, mechanical and environmental descriptors are reviewed and selected. The material groups are described and their mechanical properties are presented in Section 4.2. LCI data (Section 4.3) and weighted LCI data (Section 4.4) for material groups are presented. In Section 4.5, relationships between parameters are analysed. Application and uncertainty of the data is discussed in Section 5.

4. Results

4.1. Review and selection of parameters for describing properties of materials

4.1.1. Physical parameters

Materials can be classified according to their similar physical properties. In order to compile relevant data, which can be useful for finding relationships between materials, parameters for describing materials were reviewed. Parameters for describing physical properties of materials can be divided into the following groups [3, 30]: economic, general physical, mechanical, thermal, electrical, magnetic, resistance, production and aesthetic. Data was compiled for density, elasticity modulus and tensile strength since they are of high importance in mechanical design [3, 31] and are easy to determine for different materials.

4.1.2. Environmental parameters

The initial compilation of environmental data included all available data categories from life cycle inventories of materials. Inventory data were selected and presented for data categories, which had highest contribution to ECO'99 weighted environmental impact. Primary energy use was calculated based on the lower heating value of the energy carriers used. Primary energy is energy embodied in natural resources (e.g. coal, crude oil, sunlight, uranium) that has not undergone any anthropogenic conversion or transformation.

Parameters such as processing, maintenance needs and material service life, reuse and recyclability may have high importance for the total environmental impact of materials. The consequences of using recycled materials on the environmental performance were evaluated. Processing of materials and maintenance were excluded since these parameters are specific for a product design and not within the scope of this study.

4.2. Mechanical properties and description of material groups

If no specific environmental data is available for a material, it can be estimated by classifying the material into one group. A material can be assigned to a group based on (1) its belonging to a material class, (2) its name or (3) its physical properties. In some cases the materials of different groups have similar technical properties but the name of the material can be useful to define its belonging to a particular group. If a specific material has not been selected, but the material specifications are known, its environmental properties can be predicted based on its mechanical properties.

Grouping of materials according to their ECO'99 weighted environmental impact and physical properties resulted in 17 groups. The difference to the grouping in [7] is that one group of woods and non-ferrous metal, respectively, have been excluded and the groups of composites, rubber and thermosets foam have been added to give higher resolution in inventory data. Table 1 shows average values for mechanical properties calculated from data of 214 material cases [23].

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Class ^a	Group ^b	Total	Density	CV	Elasticity	CV	Yield strength	CV
		(n)	(Mg/m^3)	(%)	modulus (GN/m^2)	(%)	(MN/m^2)	(%)
Metals	Non-ferrous	22	8.2	30	166	53	593	42
Metals	8 (Cu etc)	26	5.0	10	126	66	223	17
wietais	5 (Al etc)	20	5.0	47	120	00	223	7/
Metals	Ferrous Ni>5%	12	7.7	3.9	193	5.1	362	40
Metals	Ferrous Ni<5%	10	7.7	2.1	204	5.3	627	38
Metals	Ferrous Ni=0%	40	7.8	1.9	201	4.2	452	69
Composites	Composites ^c	2	1.6	17	103	45	-	-
Glasses	Glasses	2	3.1	23	94	-	3600	-
Porous	Porous	5	2.5	6.7	58	45	173	85
ceramics	ceramics							
Polymers	Thermosets	2	2.0	54	2.1	55	65	-
Polymers	epoxy Thermoplasti	50	1.1	22	2.2	59	33	47
-	cs							
Polymer	Thermosets PLIR foam	6	0.060	-	0.040	-	2	89
Elastomers	Rubbers	3	0.90	5.5	0.0052	28	-	-
Woods	Woods High	5	0.87	-	15		-	-
Woods	Woods Medium	4	0.81	20	11	24	-	-
Woods	Woods Low	5	0.67	20	10	9	-	-
Woods	Cardboards	10	0.60	-	0.80	-	-	-
Woods	Papers	7	0.60	-	0.80	-	-	-

Table 1. Mechanical properties for 17 material groups [23].

CV, Coefficient of variance= standard deviation/average x 100%

^a Classes from [3] Table 4.1, except for cardboard and paper

^b Groups from [7]

^cGlass fibre reinforced polymer or carbon fibre reinforced polymer

Metals, which do not contain iron, were assigned to one of three groups depending on density. The group of Non-ferrous metals 8, with an average density of 8.2 Mg/m³ (5.7-10.7), included Cu, Ni, V, Ti, Mo and alloys of Cu, Ni and Ti. Cobalt, tin and platinum group metals were not assigned to any groups since their weighted environmental impact was several times higher all other groups. The weighted impact was 2.4 and 6.8 times higher for Co and Sn, respectively, than the group of Non-ferrous 8. Specific LCI data therefore has to be used for these materials.

The group Non-ferrous metals 5 had an average density of 5.0 Mg/m³ (1.8-7.5) and included Al, Cd, Cr, Mg, Mn, Si, Zn and alloys of Al, Mg and Zn. Two exceptions were Pb and W (density 11 and 19 Mg/m³, respectively), which was included in this group according to their environmental properties.

For metals containing iron, three different groups were distinguished depending on their content of nickel (Eq. 1). The ECO'99 weighted environmental impact, $EI_{ECO'99}$ (Pts/kg), can be estimated from the nickel content of the metal (c_{Ni} , wt%). The regression coefficient is 0.79 and the linear relationship gives a fairly good estimate for environmental impact.

$$EI_{ECO'99} = 0.0314 c_{Ni} + 0.0855 \tag{1}$$

Ferrous metals with nickel concentrations >5wt% and <5wt% made up two different groups. The third group of Ferrous metals contained no nickel. Stainless steels could be found in all three groups due to the use of alloying metals other than nickel.

Composites included glass fibre reinforced polymer (GFRP) and carbon fibre reinforced polymer (CFRP). Data availability was low for this group but it was included to provide an estimate. The group Porous ceramics included ceramics, cement and concrete. The group of Glasses included sodium and SiO₂ glass.

Data for the group of Thermosets was limited, and the average value could be presented for epoxy-based thermosets only. Foams of thermosets were included in the group Thermosets polyurethane (PUR foam).

The group of thermoplastics included a broad range of polymers e.g. ABS, HDPE, PA, PC, PE, PMMA, PP, PS, PVC and PET, which are further explained in [32].

The class of paper and woods was divided into Paper, Cardboard and three groups of woods. The grouping of woods was based on environmental properties only, since no mechanical properties made it possible to distinguish between groups of woods [7]. The weighted environmental impact of woods was widely scattered depending on the production method's impact on biodiversity. The LCI data for woods were of poor quality, which made the results uncertain. Low impact (LI) woods were defined as Ash, Aspen, Beech, Birch, Cedar, Hickory, Larch, Oak, Pine, Silver fir, Spruce and Teak. Medium impact (MI) woods were Afzelia, Blue gum, Bubinga, Mahogani, Silver fir and Willow. Rare species of tropical wood were assigned to the group high impact (HI) woods, which included Avodire, Baboen, Guaiacum, Olon and Wenge.

4.3. Life cycle inventory data for material groups

Average life cycle inventory data for production of materials may be used as an estimate if LCI data for a specific material is not available and to compare the magnitude of data from different origins. LCIs of materials were mainly representative for the global and European situation during the 90's and average technology level. The averages of LCI data is presented as global since they are based on data originating from different countries. Market shares of global production have not been considered for averages of materials originating from different countries.

Production of certain materials includes addition of secondary materials. The recycled content for materials was estimated to be 55-99% for glass, 15-45% for aluminium and 20-60% for iron and steel. In spite of the content of recycled material, the data is defined as primary production since it represents the common practice in the production of these materials.

Average data is presented for LCI data categories contributing significantly to

environmental impact according to ECO'99 weighting (see 4.4). Selected data categories are provided for resources (Table 2), land use (Table 4) and emissions to air (Table 3) and water (Table 4).

Table 2 shows the use of primary energy, energywares and mineral resources. LCI data for energy resources is included since these data categories contribute significantly to weighted environmental impact. The total primary energy use also includes other energy resources (i.e. hydro and nuclear power) in addition to coal, oil and natural gas. It is therefore not possible to calculate the total primary energy use based on the data in Table 2 only.

The use of Ni (ore) is greatest for the group Non-ferrous 8 followed by the groups of Ferrous metals. Non-ferrous metals have the greatest use of Cu (ore).

Table 2. Energy and resource use for material groups. Average life cycle inventory data for primary production from cradle to gate in different countries during the 90's [25].

Group	E prim	CV	Coal	CV	Crude	CV	Natural	CV	Ni(ore)	CV	Cu(ore)	CV
-	(MJ/kg)	(%)	(kg/kg)	(%)	oil	(%)	gas	(%)	(g/kg)	(%)	(g/kg)	(%)
					(kg/kg)		(kg/kg)					
Non-ferrous 8	375	85	3.0	79	1.9	47	1.9	144	271	138	330	125
Non-ferrous 5	171	40	2.3	52	0.67	76	0.96	112	0.021	105	6.6	191
Ferrous Ni>5%	65	35	0.86	28	0.33	36	0.34	32	99	12	0.21	343
Ferrous Ni<5%	28	13	0.60	10	0.14	15	0.0073	3.7	17	48	0.0015	3.6
Ferrous Ni=0%	27	38	0.67	46	0.12	35	0.051	178	4.7	223	0.0077	301
Composites	200	1.0	1.6	59	0.84	66	2.3	141	-	-	-	-
Glasses	13	0.7	0.046	8.2	0.19	1.9	0.011	3.0) _	-	-	-
Porous ceramics	5.8	47	0.035	191	0.016	76	0.075	209	-	-	-	-
Thermosets epoxy	180	54	0.20	n/a	2.5	141	1.2	n/a	-	-	-	-
Thermoplastic s	80	40	0.26	156	0.59	110	0.72	290	-	-	-	-
Thermosets PUR	96	23	0.47	85	0.42	83	0.94	139	-	-	-	-
Rubbers	97	3.9	0.16	113	1.2	38	0.57	173	-	-	-	-
Woods HI	13	21	0.027	70	0.24	18	0.024	87	-	-	-	-
Woods MI	14	5.6	0.030	41	0.27	9.2	0.021	62	-	-	-	-
Woods LI	9.3	54	0.028	64	0.17	62	0.016	64	-	-	-	-
Cardboards	18	59	0.025	65	0.046	65	0.055	111	-	-	-	-
Papers	25	36	0.078	50	0.092	55	0.14	73	-	-	-	-

CV, Coefficient of variance= standard deviation/average x 100%. - = insignificant. n/a= not

applicable. The number of materials for which the average has been calculated is shown in Table 1, column 3.

Table 3 shows that the CO₂ emissions are 1.3-4.1 kg CO₂/kg for Ferrous metals, 3.3-4.5 kg CO₂/kg for Polymers and 10-20 kg CO₂/kg for Non-ferrous metals. The groups Thermosets epoxy, Thermosets PUR foam, Thermoplastics and Rubbers generally have higher CH₄ emissions than the other groups. SO_x emissions are significant for the group of Non-ferrous 8 and Ferrous Ni>5%. The average NO_x emission for all material groups is 4.4 g NO_x/kg CO₂ and the linear correlation factor is 0.21. The average value can be used for assessing the relative magnitude of NO_x emission of a material.

Group	CO_2	CV	NO _x	CV	SO _x	CV	CH_4	CV	Ni	CV	Dust	CV
-	(kg/kg)	(%)	(g/kg)	(%)	(g/kg)	(%)	(g/kg)	(%)	(mg/kg)	(%)	(g/kg)	(%)
Non-ferrous 8	20	69	10	90	658	76	1.4	187	0.26	201	2.9	158
Non-ferrous 5	10	57	20	61	65	37	9.7	77	2.8	195	5.9	243
Ferrous Ni>5%	4.1	33	3.0	22	152	16	0.22	32	0.038	33	1.2	30
Ferrous Ni<5%	1.5	15	4.4	16	26	46	0.19	3.7	0.034	3.0	0.94	11
Ferrous Ni=0%	1.3	36	4.4	20	9.5	152	1.1	259	0.79	323	0.56	145
Composites	12	3.5	36	20	23	19	12	0.59	0.02	15	1.5	12
Glasses	0.76	1.4	2.3	0.61	2.3	26	0.79	2.1	0.41	1.2	1.2	12
Porous ceramics	0.41	82	1.5	65	0.94	71	0.42	134	0.065	162	1.9	173
Thermosets epoxy	3.5	97	26	49	11	120	37	n/a	-	-	8.5	125
Thermoplastics	3.3	55	16	45	18	62	13	59	1.3	242	3.2	77
Thermosets PUR	4.5	17	20	32	51	57	7.1	99	3.3	238	3.6	115
Rubbers	1.7	61	10	22	15	39	5.0	141	8.7		1.4	4.3
Woods HI	0.88	20	7.7	28	7.5	31	0.25	89	0.036	89	0.16	7.0
Woods MI	1.0	5.6	9.6	12	9.5	11	0.28	63	0.041	63	0.16	7.6
Woods LI	0.65	56	5.2	85	5.1	83	0.28	64	0.042	64	0.12	55
Cardboards	0.56	46	2.1	43	3.7	107	0.89	52	0.50	41	1.1	68
Papers	0.64	131	3.9	26	5.1	60	2.0	18	1.2	122	1.9	37

Table 3. Air emissions for material groups. Average life cycle inventory data for primary production from cradle to gate in different countries during the 90's [25].

CV, Coefficient of variance= standard deviation/average x 100%. - = insignificant. n/a= not applicable. The number of materials for which the average has been calculated is shown in Table 1, column 3.

A comparison among the groups shows that BOD emissions are significant for the groups Cardboards and Papers followed by Thermosets and Thermoplastics (Table 4). Land conversion is significant for the groups of Woods HI and MI.

Table 4. Water emission and land use for material groups. Average life cycle inventory data for primary production from cradle to gate in different countries during the 90's [25].

Group	BOD (mg/kg)	CV (%)	Land conversion (m ² /kg)	CV (%)	
Non-ferrous 8	0.39	51	0.089	246	
Non-ferrous 5	1.3	190	0.19	313	
Ferrous Ni>5%	0.20	49	0.052	159	
Ferrous Ni<5%	0.090	25	0.059	149	
Ferrous Ni=0%	15	304	0.060	198	
Composites	20	0.35	0.11	n/a	
Glasses	0.5	0.74	-	-	
Porous ceramics	0.11	109	0.015	129	
Thermosets epoxy	616	134	0.033	n/a	
Thermoplastics	352	204	0.040	11	
Thermosets PUR	857	57	0.091	16	
Rubbers	30	6.3	0.027	12	
Woods HI	0.69	63	4.8	89	
Woods MI	0.97	3.9	2.0	63	
Woods LI	0.79	52	0.027	119	
Cardboards	2700	104	-	-	
Papers	3500	91	-	-	

CV, Coefficient of variance= standard deviation/average \cdot 100%. - = insignificant. n/a= not applicable. The number of materials for which the average has been calculated is shown in Table 1, column 3.

LCI data for engineered ceramics and recycled materials has not been presented in separate groups due to limited data availability and to reduce the number of groups and complexity. General data is presented for these cases to show the variability and to allow estimates of environmental impact.

The group of engineered ceramics includes alumina, diamond, silicon carbide and zirconia [3]. Zirconia had 3-17 times higher LCI data than the group of Non-ferrous 8 [3]. Inventory data for silicon carbide was 70-97% lower than the values of the group Non-ferrous 8.

Production of recycled materials has lower environmental impact since the impact for extraction from mines and reduction of metals is allocated to the first material life cycle. By using 100% recycled glass, the LCI data was 20-40% lower than for virgin glass. For recycled steel and aluminium, LCI values were 41-61% and 80-95% lower, respectively, than for production from virgin materials.

4.4. Weighted LCI data of material groups

LCI data for the materials was weighted in order to provide a single environmental value for each material group. It also made it possible to identify the relative impact on damage categories and the relative importance of contributing data categories.

Fig. 1 shows that different groups can be distinguished depending on their weighted environmental impact and elasticity modulus. The graph can be used to identify materials with a desired elasticity modulus and lowest environmental impact. The error bars indicates how closely related different groups are to each other. Merging of groups would result in greater standard deviation. The weighted environmental impact for Woods High Impact, Woods Medium Impact and Non-ferrous 8 are greater than 1 ECO'99 Pts/kg.



Fig. 1. ECO-indicator 99 weighted environmental impact and elasticity modulus for material groups. The error bars indicate the standard deviation.

Table 5 shows average figures for weighted environmental impact for material groups which may be used in simplified environmental assessments. The average standard deviation among all groups is 25%. Since only a few cases were available on engineered ceramics and the difference between the material cases was high, it was not adequate to calculate an average for this group. The weighted environmental impact was 0.22 Pts/kg for silicon carbide and 15 Pts/kg for zirconia.

LCI data for recycled materials depends on the allocation principles used. It was not possible to determine the allocation principles used in each case of recycled materials. By using 100% recycled materials, e.g. steel, the weighted environmental impact can be reduced by 43-51% compared to virgin materials. Recycled aluminium has a 90-95% lower impact than one way use of virgin aluminium.

Table 5. ECO-indicator 99 weighted environmental impact, contribution of environmental damage categories and most important data categories for material groups. Average for primary production from cradle to gate in different countries during the 90's. Processed data from [25].

Group	No of	ECO'99	CV	Damage category contribution		Most important data	
	cases (n)	(Pts/kg)	(%)	to ECO'99 (%)		categories	
	(11)			Human health	Ecosystem R quality	Resources	5
Non-ferrous 8	44	2.5	30	47	9	44	SO_x , metal ore, oil, CO_2 ,
Non-ferrous 5	56	0.56	21	43	9	48	natural gas, oil, SO_x , CO_2 , NO_x
Ferrous Ni>5%	13	0.45	20	58	7	35	SO _x , Ni _(ore) , natural gas, oil, CO ₂ ,
Ferrous Ni<5%	11	0.15	28	52	12	36	SO_x , NO_x , oil, $Ni_{(ore)}$, CO_2
Ferrous Ni=0%	57	0.077	21	45	14	41	NO_x , oil, SO_x , CO_2 , natural gas
Composites	2	0.99	43	25	5	70	oil, natural gas, NO_x , CO_2 , SO_2
Glasses	6	0.057	10	32	13	55	oil, NO_x , Pb, SO_x , CO_2
Porous ceramics	7	0.028	28	47	13	40	NO_x , Natural gas, CO_2 , oil. SO_x .
Thermosets epoxy	2	0.76	22	22	4	74	oil, natural gas, NO_x , dust, CO_2
Thermoplastics	66	0.36	28	28	3	69	oil, natural gas, NO_x , SO_x , CO_2
Thermosets PUR	17	0.43	10	45	6	49	natural gas, oil, NO_x , SO_x , CO_2
Rubbers	4	0.31	12	25	4	70	oil, natural gas, NO_x , SO_x , CO_2
Woods HI	10	12	16	1	99	0	land use
Woods MI	44	6.3	34	1	98	1	land use
Woods LI	30	0.58	30	7	85	8	land use, oil, NO _x
Cardboards	23	0.035	26	44	7	49	natural gas, oil, NO_x , SO_x , CO_2
Papers	12	0.066	26	43	6	51	natural gas, oil, dust, NO _x , SO _x

CV, Coefficient of variance= standard deviation/average x 100

ECO'99 weighting scores are made up of the potential impacts on the damage categories resources, ecosystem quality and human health [26, 27]. Impact on damage categories occur through different pathways. The damage category resources are measured as use of extra energy required for future extraction of minerals and fossil fuels. Impact on human health through respiratory effects, cancerogenesis, global warming, stratospheric ozone depletion and ionising radiation is expressed in disability adjusted life years. Ecosystem quality is measured as the percentage of vascular plants that potentially can disappear from

an area due to ecotoxicity, acidification/eutrophication and changes of habitat size.

Table 5 shows that Porous ceramics and groups of metals have significant impact on human health. The dominating damage pathway is through respiratory effects caused by emissions of SO_x and NO_x . Woods have significant impact on ecosystem quality due to land use.

Resource use is the most significant damage category for Composites, Thermosets epoxy, Thermosets PUR foam, Thermoplastics, Rubbers, Cardboards and Papers. Impact on resources occurs mainly through the use of fossil fuels (natural gas and oil).

The groups Non-ferrous metals 5, Ferrous Ni=0% and Porous ceramics also have high impact on resources through the use of fossil fuels. Non-ferrous 8 and Ferrous Ni>5% metals have high impact on resources by significant use of minerals.

The relative contributions of data categories to the total weighted impact were presented in order to identify the most important ones for each material group. Table 5 shows that oil, natural gas, NO_x , SO_x and CO_2 are some of the top contributors. For Woods, conversion of land is the dominating data category. Based on ECO'99 weighting, 26-100% (average 81%) of the total environmental impact is caused by the top five data categories.

4.5. Relationships between parameters

4.5.1. Correlation between environmental parameters

By establishing relationships between environmental parameters, quantified data for one parameter can be used to predict values for others. Regression analysis of different parameters showed high correlation between primary energy and CO_2 emissions. The relationships exist since energy generation is mainly based on combustion of carbon containing fuels. The amounts of emission depend on the energy conversion efficiency and carbon content of different fuels. The linear equation (Fig. 2) can be used to estimate CO_2 emissions for a material if the primary energy use is known. Since the data points are highly scattered below 200 MJ/kg, estimates in this region are uncertain. The data represents emission factors for a global average mix of energy generation. The average carbon intensity for energy use of the material groups was 52 g CO_2/MJ . The group of Rubbers had the lowest carbon intensity (18 g CO_2/MJ) and Porous ceramics the highest (71 g CO_2/MJ).



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Fig. 2. Relationship between primary energy use and CO_2 emissions for different materials (n=214).

Weak linear relationships (correlation factors 0.38 and 0.87) existed between primary energy and emissions of SO_x and NO_x , respectively. The variability is great among the material cases since the treatment of combustion gases depends on the technological level of manufacturing processes. NO_x emissions from combustion processes have been reduced considerably the last decade by the use of more efficient combustion and NO_x reduction technologies (catalytic converters etc).

Relationships between BOD and COD showed high variability among the different materials except for the groups of Woods. Values for COD for Woods can be estimated by multiplying BOD by 3.

4.5.2. Multivariate data analysis of relationships between physical and environmental parameters

Multivariate data analysis [33] was applied to identify relationships between the compiled data for materials. The explanatory value and the relative importance of the parameters were also evaluated.

Multivariate data analysis of LCI data and weighted environmental impact was used to determine the most important data categories. Through analysis of data for the 17 material groups, data categories with greatest importance were found to be SO_x , natural gas, crude oil, E_{Prim} , CO_2 and NO_x , coal, methane, $Cu_{(air)}$ and $Ni_{(air)}$. These results are in accordance with the data shown in Table 5.

The results of the multivariate data analysis showed that data for density, elasticity modulus and tensile strength explain up to 20% of the variability in weighted environmental impact. The modelling was very sensitive to extreme data values, which made the results uncertain. Due to the complexity of environmental interactions, it may be difficult to identify a few parameters with high explanatory value.

For metals, it was concluded that there was little or no correlation between concentration of metals in the earth's crust and ECO'99 weighted environmental impact, primary energy or CO_2 emissions. For all different material groups, there was weak correlation between weighted environmental impact and NO_x , SO_x or primary energy use.

5. Discussion

5.1. Use and validity of LCI data for material groups

The standard deviations for material groups for energy are 1-114% (average 41%) and for ECO'99 weighted environmental impact 6.5-50% (average 25%). A comparison of LCAs conducted with specific material data and material group data [7], respectively, showed that the weighted environmental impact was 12-26% higher with data based on averages for material groups. These results indicate that the average data can be used as estimates in many cases of simplified environmental assessments and provide sufficient accuracy.

To assess the validity of the data presented, the results were compared with literature data on primary energy. Primary energy was chosen for the comparison due to high data availability and its high contribution to environmental impact. Energy data presented in other studies [2, 5] for the groups Ferrous Ni<5% and Ferrous Ni>5% is 1.2-9.5 times higher than the results in this study. The group Non-ferrous 8 is 1.4-1.9 times higher than the average data calculated in [2, 5]. The differences in the data are due to differences in technological and geographical system boundaries in the studies. The energy data presented by [1, 34] originate from the 70's and 80's and since then, both material production technologies and energy analysis methodology have improved. The comparisons indicate that the data presented in this study is similar to other studies and may be used as a first estimate in simplified life cycle assessments.

LCI data shows that woods have low impact compared to other material groups. The ECO'99 weighting method weights conversion of land and following impact on biodiversity highly. This makes some groups of woods into high impact materials. The results imply that forestry methods should be considered when selecting wood materials. The uncertainties of these results are high since LCI data on land conversion is often lacking or difficult to estimate for all kinds of materials. Assessment of environmental impact of woods is improving by different research initiatives [35, 36].

The weighted environmental impact of material groups is similar to the study by Rombouts and Hennessey [6]. However, they found that polymer foams had higher environmental impact, which may be due to CFC emissions that were used in the production of foams in the early 90's.

This study identified oil, natural gas, NO_x , SO_x and CO_2 as data categories highly descriptive for the total environmental impact of different material groups. A study of metal production [37] identified the following significant descriptors for different metals: energy, CO_2 , NO_x , SO_x , VOC, dust and noise. Eisenhard et al. [11] suggested an abbreviated LCI list including energy, solid material, CFC, halon, Pb, Cd, Cr, Ni, PAH, suspended particulate matter, CO_2 , SO_2 , NO_x , CH_4 , C_xH_y , COD, $Ntot_{(aq)}$. Berkhout et al. [16] identified metal emissions as sector specific parameters for the metals industry and C_xH_y and VOC for the plastic industry. When conducting life cycle inventories, significant environmental impact is likely to be expressed with these data categories.

5.2. Uncertainties and limitations of LCI data for material groups

With the data used in this study, multivariate analysis showed weak correlation between physical material properties and environmental parameters. Better understanding of manufacturing processes and weighting methods may result in groups which are useful for simplified environmental assessments.

Uncertainties of the grouping and average values are due to low data quality for some materials. The different origins of LCI data made it difficult to assure consistent data quality for all materials. Characterisation and weighting methods are complex and contain phases of subjective valuation. Toxicity of substances (e.g. metals) is difficult to include in weighting methods. However, weighting methods must be considered as best available practice to interpret and make tradeoffs between different environmental impacts.

To assess how the results would change if a weighting method other than ECO'99 were used, a comparison was made with LCI data weighted with EPS2000 [28, 29]. The comparison showed that the trends for the material groups were similar except for the groups of woods. It was concluded that EPS2000 is very sensitive for CO_2 and the use of mineral and energy resources. The high weighting of metal resources may lead to that this part dominates and that other data categories hardly influence the total weighted impact.

The environmental data presented is limited to cover the production phase of the materials only. When comparing average environmental data for material groups with each other, it must be stressed that it should be done in a life cycle perspective. To select the best material for a product, the usage and waste treatment of the product and material also has to be assessed. Materials selection can influence a range of areas including energy efficiency, product lifespan, maintenance needs, reuse and recyclability.

Composites and engineered ceramics are heterogeneous material classes and only a few samples of materials could be included in this study. Since these materials are becoming more commonly used in mechanical design future research should aim to provide high quality data for these materials. Further studies of woods should aim at analysis of inventory data, grouping criteria and weighting models.

6. Conclusions

Average data for mechanical properties and environmental impact was presented for 17 material groups. The data presented for material groups showed average standard deviations of 25% for ECO'99 weighted environmental impact and 41% for energy. The environmental data provided for material groups may be used as estimates in life cycle assessments when specific data on materials is not accessible. Hereby the environmental significance of a material can be identified.

This study identified oil, natural gas, NO_x , SO_x and CO_2 as data categories highly descriptive for the ECO'99 weighted environmental impact of different material groups. By using 100% recycled glass, steel and aluminium, the environmental impact can be reduced by 20-95%. Future studies should aim to provide more comprehensive environmental data for woods, composites and engineered ceramics.

The impact in various environmental damage categories and damage pathways for material groups were identified. Porous ceramics and all groups of metals have their main environmental impact on human health, which is due to respiratory effects (emissions of SO_x and NO_x). Woods have significant impact on ecosystem quality due to land use. Due to the use of fossil fuels, depletion of resources is the most significant category for Composites, polymers (Thermosets epoxy, Thermosets PUR foam, Thermoplastics), Rubbers, Cardboard and Paper.

Linear relationship was identified between primary energy and CO_2 emissions. With the data used in this study, multivariate analysis showed weak correlation between physical materials properties and environmental impact.

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Paper V

Rydh, C. J. and Sandén B. A. (2003) Energy Analysis of Batteries in Photovoltaic Systems, Submitted for publication in *Energy Conversion and Management*



Energy analysis of batteries in photovoltaic systems

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Abstract

Energy return factors and overall energy efficiencies are calculated for a stand-alone PVbattery system at different operating conditions. Eight battery technologies are evaluated: lithium-ion (Ni), sodium-sulphur, nickel-cadmium, nickel-metal hydride, lead-acid, vanadium-redox, zinc-bromine and polysulfide-bromide. With a battery energy storage capacity three times higher than the daily energy output, the energy return factor for the PV-battery system ranges from 0.64 to 12 for different batteries and assumptions. This means that 8.1%-156% of the energy output is required to produce the PV-battery system. For a PV-battery system with a service life of 30 years, the energy payback time is 1.6-3.0 years for the PV array and 0.55-43 years for the battery, showing the energy related significance of batteries and the large variation between different technologies. The overall efficiency, including energy requirements for production and transport of the charger, the battery and the inverter, is 0.23 - 0.82. For some batteries the overall battery efficiency is significantly lower than the direct energy efficiency of the charger, the battery and the inverter (0.50-0.85). The importance of operating conditions influencing the battery service life (depth of discharge, battery temperature and air conditioning), transportation mode, the use recycled battery materials and battery performance parameters (charge-discharge efficiency and gravimetric energy density) are assessed.

Keywords: energy analysis, life cycle assessment, efficiency, renewable energy, photovoltaic, battery production, lithium-ion, lead-acid, nickel-cadmium, nickel-metal hydride, polysulfide-bromide, Regenesys, sodium-sulphur, vanadium redox flow, zinc-bromine

1. Introduction

To warrant support, new technologies need to prove that they have a potential to solve the problems they are designed to solve. For example, if a renewable energy technology shall be able to contribute substantially to energy supply, it needs to be able to convert significantly more energy from the renewable energy source than what is used for the production of the conversion system itself. Over time, the inability to do so will become evident, but in the meantime a lot of effort and money could have been wasted on an

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inferior technology. The ability to generate positive net energy has recently been at the heart of a debate over the benefits of producing ethanol from corn [1, 2]. The energy balance of solar photovoltaics (PV) has also been used as an argument against PV dating back to a study in 1972 that claimed that it took 40 years for a PV module to generate the electricity that was required to produce it, i.e. the energy payback time was 40 years [3]. It should be noted that for some small energy loads, it is of less importance to supply net energy. In some cases it is enough that a device works as a battery and move energy from one place to another. This could for example be the case for PV in satellites or in small solar home systems in poor rural environments.¹

With current technology and production methods, the energy payback time for PV modules has been estimated at 1.1-5 years depending on technology and solar intensity [4-9]. Over a lifetime of 25 years, PV modules thus generate 5-23 times the energy required to produce them. Balance of systems (BOS) components can add significantly to energy payback times. Heavy support structures could increase energy payback times by over 6 years [10]. Depending on application, PV systems have to be equipped with auxiliary components such as inverters, charge regulator and energy storage systems. Contributions to energy requirements from such components are normally small for grid-connected systems. Inverters usually add only a few months [10].

In many types of stand-alone systems, batteries are required to even out irregularities in solar irradiation and concentrate solar energy to higher power. In a study of solar home systems, Alsema [11] concluded that batteries contribute significantly to the gross energy requirements. The lead-acid batteries' addition to the energy payback time of the solar home systems was 10-11 years, and even more, 15-19 years, without recycling of materials. Rydh [12] compared the energy requirements for lead-acid and vanadium redox flow batteries for stationary energy storage but other battery technologies have not been assessed in this context.

The literature review shows that batteries are important components of PV-battery systems but energy analysis and comparisons of emerging energy storage technologies are lacking. The purpose of this study is to provide an energy analysis to enable comparison of different battery technologies in renewable energy applications. By quantifying energy efficiencies and energy requirements for manufacturing of different systems, increased awareness may lead to improved energy management of energy storage systems. Identification of important parameters can be used to direct research and product improvements and a comparison of different battery technologies can be used to guide battery selection for specific user conditions.²

¹ Likewise, if corn based ethanol is produced to function mainly as a low percentage additive to gasoline to reduce regulated emissions and not as an alternative fuel, the net energy argument is less relevant.

² A detailed description of the data used in this paper is presented by Rydh [13].

2. Goal and scope

The goal of this study is to analyse the energy efficiencies of different battery technologies when used in stand-alone PV-battery system at different operating conditions. The contribution of different PV-battery components to the gross energy requirement and important parameters is identified for each battery technology. The performance of the PVbattery system is evaluated by the energy return factor and the overall battery energy efficiency. Energy is one aspect of a PV-battery system and financial costs are not included.

The following battery technologies are evaluated: lithium-ion Ni (Li-ion), sodium-sulphur (NaS), nickel-cadmium (NiCd), nickel-metal hydride AB_5 (NiMH) and lead-acid (PbA). Three types of redox flow batteries (regenerative fuel cells) are included namely polysulfide-bromide (PSB), vanadium-redox (VRB) and zinc-bromine (ZnBr). The battery parameters investigated are battery charge-discharge efficiency, service life, gravimetric energy density and energy requirements for production and transport of batteries (see Section 4).

The study includes energy requirements from the cradle to the grave for production of PV arrays (PV modules, module frames and roof integrated array supports), batteries, inverter, charge regulator and air conditioning (AC). Transport of PV-battery system components from manufacturing to the site of use and return at the end of life is included. The standalone system has three days of autonomy and the average solar irradiation is 1.7 MWh/m^2 year. To make energy storage technologies with different characteristics comparable, they are normalised to fulfil a functional unit. The functional unit is defined as "an electricity storage system with a power rating of 50 kW, a storage capacity of 450 kWh and an output of 150 kWh electricity per day".

The choice of functional unit defines that the depth of discharge (DOD) of the battery is 33% at daily cycling (150 kWh/ day/ 450 kWh). Since it is difficult to determine the service life at a specific DOD accurately as well as the life-limiting factor for batteries, the battery performance was evaluated for service life limited by cycle life and float life, respectively. In a reference case (Case 1), the battery service life is assumed to be limited by cycle life due to the daily cycling. It can be assumed that the DOD may be higher than 33% during longer periods resulting in cycle life limiting conditions.

To assess the uncertainties and improvement potential of different technologies, battery specifications are given for best demonstrated performance, presented as high, and average or normal performance, presented as low. When cells are stacked together into battery modules the performance values decrease due to addition of structural materials, effects of unmatched cells and increased resistance in wires etc.

The effect of self-discharge is not included since the batteries are assumed to be cycled. Cooling requirements corresponding to energy losses in charger, battery and inverter are included when air conditioning is turned on. The housing of batteries is assumed to be equal for different battery systems and is not included in the energy analysis. It is assumed that no energy for transport is required for maintenance of the PV-battery system.

The uncertainties of the energy return factors and the overall battery efficiencies are presented in Section 5.1 for a reference case when battery service life is limited by cycle life and the temperature is 25°C. The contribution of different components to the gross

energy requirement is presented. To evaluate the importance of the battery service life influenced by life determining factors, three alternative cases are analysed in Section 5.2. The effect of using AC is evaluated to assess for which batteries it can be motivated to install AC from an energy perspective. Consequences of using virgin instead of recycled materials and plane instead of truck for battery transportation are analysed in Section 5.4. In Section 5.5, the effects of improved battery performance are evaluated by sensitivity analysis regarding: service life (t_3), gravimetric energy density (d), charge-discharge efficiency (η_3) and energy requirements for production (q_{P3}). The influence of the system design and external parameters is evaluated in Section 5.6.

Since all calculations are done in a spreadsheet computer model, results can be easily updated. The model is described in the following section.

3. Measures of energy efficiency

Energy efficiency can be defined in many ways, but in all cases it is a measure of the amount of energy resources (inputs) that is needed to provide an energy service (output). In this paper we will use two complementary measures of energy efficiency, the *energy return factor* and the *overall efficiency*.

3.1. Direct and indirect energy requirements

The gross energy requirement E_G of an energy conversion device with the energy output E_O can be decomposed into two parts (Fig. 1): the direct input of energy during operation E_D and the indirect energy requirement E_I , i.e. the energy used to produce the device and transport it to the site of operation.

$$E_G = E_D + E_I \tag{1}$$



Fig. 1. The general energy balance of an energy conversion device.

From these energy flows three measures of energy efficiency can be calculated: the direct (or normal) energy efficiency of the device

$$\eta = \frac{E_o}{E_D}, \qquad 0 < \eta < 1 \tag{2}$$

the overall energy efficiency

$$\eta^* = \frac{E_O}{E_G}, \quad 0 < \eta^* < 1$$
(3)

and the energy return factor

$$f = \frac{E_O}{E_I} \qquad f > 0 \tag{4}$$

The fact that energy may take different forms poses a problem. The gross energy requirement and the indirect energy requirement are normally made up of many different kinds of energy inputs. To be able to define single measures for the overall efficiency or the energy return factor, different energy forms need to be converted to a common energy currency.

The *energy return factor* (sometimes given in the form of an energy payback time or as net energy output)³ is normally used as an indicator of how efficiently a device (such as a PVbattery system) or a conversion system (such as ethanol production) uses non-solar energy in comparison to an alternative method of producing the same service. In our case the alternative means of producing electricity locally would be a diesel generator. The data available for indirect energy requirement are given in terms of primary energy. Therefore, we use primary fossil energy (indicated by the index *pf*) as the energy currency when we calculate the energy return factor. This is obviously a somewhat coarse approximation since there are great differences between coal, oil and gas (for example with regard to CO_2 emissions or resource availability). However, in principle, the diesel oil saved could be used for the heat, electricity and motor fuel production required for the production and transport of the PV-battery system.

The *overall battery efficiency* will be used as a measure of efficiency of the battery system (charger, battery and inverter). Since the direct energy input and the output are electricity, electricity (indicated by the index *el*) will be used as the energy currency for the calculation of this measure.

3.2. Energy return factor

The most common way to describe the energy balance of a PV-system (or any other energy flow conversion technology) is by calculating energy payback times, t^* [4]. A diesel generator continuously converts fossil energy into electricity. To produce a PV array and a battery that could replace the diesel generator requires energy (we will here assume fossil energy) while only solar energy will be used during operation (Fig. 2). After a certain time,

³ The net energy (output) is defined in some studies as E_O-E_I , in relation to the indirect energy requirement $(E_O-E_I)/E_I = f-1$ or to the energy output $(E_O-E_I)/E_O = 1-1/f$. The indirect energy requirements can also be expressed as percentage of the energy output, $E_I/E_O = 1/f$.

the energy payback time, the energy that was used to produce the PV-battery system is paid back by not using the diesel generator.

Both systems have the same output E_{use} (MJ_{el}/year). The average annual gross primary fossil energy use of the diesel system E_{G0} (MJ_{pf}/year) is calculated from

$$E_{G0} = \frac{E_{use}}{\eta_0^*} \tag{5}$$

where, η^{*_0} , is the overall efficiency of the diesel generator. The energy payback time is then calculated from

$$t^* = \frac{Q_{pf}}{E_{G0}} \tag{6}$$

where $Q_{\rm pf}$ (MJ_{pf}) is the primary fossil energy that is required to build the PV-battery system.



Fig. 2. The energy flows of the PV-battery system and the reference system (the diesel generator).

The energy return factor, f, is then the ratio between the service life of the PV-battery system t (year), and the energy payback time, or to put it differently, the ratio between the replaced fossil energy (diesel) and the fossil energy required to produce the PV-battery system

$$f = \frac{t}{t^*}, \qquad f = \frac{E_{G0} \cdot t}{Q_{pf}} = \frac{E_{G0}}{E_{I,pf}}$$
 (7a, 7b)

where $E_{I,pf}$ (MJ_{pf}/year) is the average annual energy required to produce and replace the PV-battery system. Eq. 7b is a version of Eq 4 where the energy flows are translated into a common energy currency.
Since the service life of the components in the PV-battery system differ, the meaning of an energy payback time becomes ambiguous. The energy return factor is then a better measure and for the more detailed representation of the system in Fig. 3 the energy return factor can be calculated from 7b and from

$$E_{I,pf} = \sum_{i=1}^{4} E_{I,i,pf} + \delta_1 \cdot E_{I,5,pf} = \sum_{i=1}^{4} \frac{Q_{i,pf}}{t_i} + \frac{\delta_1 \cdot Q_{5,pf}}{t_5}$$
(8)

where $\delta_1 = 1$ in cases where air conditioning (AC) is used, otherwise $\delta_1 = 0$.



Fig. 3. The PV-battery system components, indirect energy requirements, *E* (annual) and *Q* (total), lifetimes *t*, efficiencies η , and conversion factors α .

3.3. Overall efficiency of the battery system

The overall efficiency of the battery system, η_B^* , is the ratio between the output from the battery system, E_{use} (MJ_{el}/year), and total inputs translated into an electricity equivalent, E_G (MJ_{el}/year) (Fig. 4).

$$\eta_B^* = \frac{E_{use}}{E_G} \tag{9}$$

When calculating the energy return factor the solar electricity was implicitly regarded as an abundant free resource. When calculating the overall efficiency the electricity input is seen as the scarce resource worth saving.⁴

This could be a relevant measure of efficiency in a closed solar energy system. For a designer of a PV-battery system, the direct efficiency of the battery system is of interest. A large direct efficiency would save resources (materials, energy, capital and labour) used to produce the PV-system and space used by the PV arrays. In a world that to a larger extent rely on solar energy, the production of the battery system must also be produced from solar energy (we can not borrow fossil fuels to build the system any more). Thus more PV-systems (or other solar energy technologies) would have to be produced. Total electricity inputs can thus be interpreted as the output from PV arrays at the site and from PV arrays producing electricity that is used to produce and transport batteries. The closed solar energy system is just an example. The overall efficiency measure is valid for electricity produced from any energy source.

The gross electricity input E_G can be written as the sum of electricity output E_{use} and the energy loss in the charger E_{W2} , in the battery E_{W3} , and in the inverter E_{W4} and the energy used for the production and transport of the charger $E_{I2,el}$, the battery $E_{I3,el}$, and the inverter $E_{I4,el}$ and the direct and indirect energy requirement of the air conditioner E_{D5} and $E_{I5,el}$ when in use.

$$E_{G} = E_{use} + E_{W2} + E_{W3} + E_{W4} + E_{I2,el} + E_{I3,el} + E_{I4,el} + \delta_{1}(E_{D5} + E_{I5,el})$$
(10)

where

$$E_{W2} = E_{use} \left(\frac{1 - \eta_2}{\eta_2 \cdot \eta_3 \cdot \eta_4} \right) \tag{11}$$

⁴ The overall efficiency of the PV-battery system could be calculated by taking into account the direct efficiencies and indirect energy requirements for the PV array. This would then become a measure of how efficiently the solar irradiation is used. However, here the focus is on battery performance.



Fig. 4. The gross energy requirement of the battery system E_{GB} is the sum of the energy output, direct losses in the battery system and the indirect energy requirement for production and transport of components.

$$E_{W3} = E_{use} \left(\frac{1 - \eta_3}{\eta_3 \cdot \eta_4} \right) \tag{12}$$

$$E_{W4} = (E_{use} + \delta_1 \cdot E_{D5}) \cdot \left(\frac{1 - \eta_4}{\eta_4}\right)$$
(13)

$$E_{I,i,el} = \frac{Q_{i,el}}{t_i}, \ i = 2, 3, 4, 5$$
(14)

$$E_{D5} = \frac{E_{W2} + E_{W3} + E_{W4}}{\alpha_5} \tag{15}$$

where α_5 is the air conditioning conversion efficiency of electricity to heat. The air conditioner uses electricity from the PV array that has passed the inverter but not the charger and the battery. The cooling requirement of the battery room depends on the heat losses from the charger, the battery and the inverter. Heat transmission from the ambient air is not considered since it is estimated to have rather low influence in a battery room located below ground level.

3.4. Battery service life

The battery service life is limited either by cycle life ($\delta_2 = 0$) or float life ($\delta_2 = 1$)

$$t_{3} = \begin{cases} (1 - \delta_{2}) \cdot \frac{N}{n} \cdot \sigma(T) \\ \delta_{2} \cdot t_{float} \cdot \sigma(T) \end{cases}$$
(16)

where t_{float} is the float life at 25°C and N is the maximum number of charge-discharge cycles at 25°C and at a specified DOD. The service life is dependent on the temperature T (°C) and $\sigma(T)$ is a temperature dependent correction factor (see Section 4.3.2).

3.5. Energy requirements for production and transport of the components

Data for indirect energy requirements are given in the energy currency primary (fossil) energy. The energy requirement for producing and transporting component *i* is calculated from the mass m_i and the gravimetric energy requirement for production $q_{P,i,pf}$ (MJ/kg), and transport to the site of operation $q_{T,pf}$ (MJ/kg):

$$Q_{I,i,pf} = \begin{cases} (q_{P,i,pf} + q_{T,pf}) \cdot m_i & i = 1,2,4,5 \\ ((1 - \delta_3 \cdot y) \cdot q_{P,i,pf} + (1 - x - \delta_3 \cdot y) \cdot q_{T,pf}) \cdot m_i & i = 3 \end{cases}$$
(17)

where x is the weight percentage distilled water in the batteries. It is assumed that the water is filled at the site of operation. The factor δ_3 is the rate of recycling. Only the cases $\delta_3 = 0$ and $\delta_3 = 1$ will be reported in this paper. The factor y is equal to 0 for all components except for the redox-flow batteries. The factor y is the weight fraction of the electrolyte and active materials, excluding distilled water, in the redox-flow batteries. It is assumed that the electrolyte in these batteries is not sent back for recycling at the end of life but is reused at the site.

The gravimetric energy for transportation of components is

$$q_{T,pf} = 2L \cdot ((1 - \delta_4) \cdot e_{truck} + \delta_4 \cdot e_{air})$$
⁽¹⁸⁾

where *L* (km) is the length of the one way journey to the site of operation, e_{truck} and e_{air} (MJ_{pf}/kgkm) is the energy intensity of truck and air planes, respectively. The factor δ_4 is the fraction of the journey travelled by air. Only the cases $\delta_4 = 0$ and $\delta_4 = 1$ will be reported in this paper. It is assumed that all components is returned after use (or sent an equal distance) no matter if they are recycled or not, hence the factor two in the formula.

The effect of recycling is not investigated for non-battery components. For batteries the gravimetric energy requirement for production is divided into energy requirement for material recycling q_{R3} , and battery manufacturing q_{M3} . Materials may be of virgin q_{RV3} or recycled q_{RR3} origin:

$$q_{P3,pf} = q_{R3} + q_{M3} = (1 - \delta_3) \cdot q_{RV3} + \delta_3 \cdot q_{RR3} + q_{M3}$$
(19)
10

In the recycling case the electrolyte in the redox-flow batteries is not sent back for recycling at the end life but is reused at the site of operation.

The required battery mass is given by the gravimetric energy density d (Wh/kg) and the required battery capacity C (Wh)

$$m_3 = \frac{C}{d} \tag{20}$$

where

$$C = \frac{k \cdot E_B}{n}, \ C = \frac{k \cdot E_{use}}{n \cdot \eta_4}$$
(21a, 21b)

where k (-) is a factor indicating the over capacity (or the number of days of storage capacity) and n (year⁻¹) is the number of charge-discharge cycles per year to the depth of discharge (DOD) 1/k. In this application n = 365 year⁻¹.

For the PV array (i = 1), the required mass is calculated from

$$m_1 = \frac{w}{H \cdot \eta_1} \cdot \left(\frac{E_{use}}{\eta_2 \cdot \eta_3 \cdot \eta_4} + \frac{\delta_1 \cdot E_{D5}}{\eta_4} \right)$$
(22)

where $w (kg/m^2)$ is the mass per square metre of array and H is the annual solar irradiation at the site of operation (MJ/m²year).⁵

The required mass of the charger (i = 2), inverter (i = 4) and air conditioning (i = 5) is related to the output power of each component. The output power of the charger is related to the maximum solar irradiance S_p (W/m²), the inverter is related to maximum power that is required at the point of use, P_{use} (W) and the air conditioning is related to the daily reference yield from the PV array

$$m_2 = v_2 \cdot \frac{S_P}{H} \cdot \left(\frac{E_{use}}{\eta_3 \cdot \eta_4}\right),\tag{23}$$

$$m_4 = v_4 \cdot P_{use} \tag{24}$$

⁵ It is assumed that the battery is fully charged from the PV array every day, corresponding to E_{use} . In the case of cloudy days, the battery will not be completely charged, and the stored energy in the battery will gradually decrease at continued cycling. To enable the battery to be charged to full capacity again, the power rating of the PV array has to be increased. The maximum PV array rating required is when the battery can be fully charge during one day.

$$m_5 = v_5 \cdot \frac{E_{D5}}{\eta_4} \cdot \frac{S_p}{H}$$
(25)

where v_i (kg/W) is the mass per installed unit of output power. The extra power requirement of the inverter when the AC is turned on is neglected since it has no influence on the final results.

3.6. Energy quality and conversion factors

To calculate the overall efficiency of the battery system we need to convert the energy requirements for production and transport given in the currency primary fossil in Section 3.3 into electricity equivalents. To do this, it is assumed that electricity and thermal energy are used for the production of the components, and transport fuel is used for transport.

$$q_{P,i,el} = q_{P,i,pf} \cdot \left(\beta_i \frac{\alpha_{pf,el}}{\alpha_{el,el}} + (1 - \beta_i) \frac{\alpha_{pf,th}}{\alpha_{el,th}}\right)$$
(26)

$$q_{T,el} = q_{T,pf} \cdot \frac{\alpha_{pf,tr}}{\alpha_{el,tr}}$$
⁽²⁷⁾

where the conversion factors α are given in Table 1. The factor β_i is the estimated proportion of primary fossil energy used to generate the electricity used in the production of component *i*.

Table 1 shows that the average conversion efficiency for electricity generation from fossil fuels is estimated to be 0.35 [52]. Losses in distribution and conversion of primary fossil fuel to thermal energy result in a conversion efficiency of 0.95. The efficiency for refining and distribution of primary fossil fuel to diesel for transportation is 0.88 [14].

When PV generated electricity is used to produce the PV-battery system, electricity has to be converted into thermal energy and fuels for transportation (diesel or hydrogen), since different forms of energy are required in the production processes. Two extreme cases are investigated for the conversion of solar electricity, the *Reference case* and the *All PV case*.

In the *Reference case*, the PV-battery system is open to other fuels and the electricity produced can be used to replace a certain amount of primary fossil energy, which in turn can be used to produce heat and transport fuel. When electricity replaces the need for the combustion of fossil fuels for electricity generation, the conversion factor for electricity to heat is 2.71 ($\alpha_{el,th}$ =0.95/0.35). For transportation, the primary fossil fuel includes 12% losses for refining and distribution of diesel fuel [14], resulting in an energy conversion factor of 2.51.

The *All PV case* corresponds to a closed renewable energy system where solar electricity cannot be traded and has to be used directly to produce heat and a transport fuel. Electricity is then used directly for the generation of high temperature thermal energy (heat pumps not

considered) with the conversion factor 1. Transportation in a renewable energy system may be based on vehicles powered by fuel cells and electrical motors where hydrogen is used as a motor fuel. The conversion efficiency of electricity to hydrogen by electrolysis of water is estimated to be 80% and the efficiency in the distribution of hydrogen fuel is 80% [14]. A fuel cell vehicle could thus be 33% more energy efficient per ton km transported goods than a conventional diesel truck [14], resulting in a conversion factor of 0.85.

Table 1. Energy conversion factors.

		Conversion from primary fossil energy		Conversion electr	on from icity
				Reference case	All PV case
Electricity	$\alpha_{pf, el}$	0.35	$\alpha_{el, el}$	1.0	1.0
Thermal energy	$\alpha_{pf, th}$	0.95	$\alpha_{el, th}$	2.71	1.0
Transport fuel	$\alpha_{pf, tr}$	0.88	$\alpha_{el, tr}$	2.51	0.85

4. Performance and energy requirements of the PV-battery system components

4.1. Description of the batteries

Of the battery technologies studied (Table 2), PbA and NiCd batteries are most widely demonstrated for use in PV-applications. The relatively high production cost of Li-ion and NiMH batteries have made them little employed in applications for storage of several hundreds of kWh. At poor maintenance conditions batteries with gelled electrolyte have longer service life than flooded cells [15]. At temperatures above 30°C, gelled electrolytes may dry out, while flooded cells may lose some heat by decomposition of water and thereby getting longer service life. The construction of NiCd and PbA batteries were therefore based on flooded electrolyte pbA batteries have showed good performance at extensive cycling and high temperatures [15], the high values for PbA battery service life are representative this battery type. The NiMH battery is valve regulated, which means that evolved hydrogen and oxygen is recombined to water during charging, and the electrolyte has to be added on the site of manufacture. Li-ion batteries are only available as sealed cells with immobilised electrolyte.

The NaS battery uses sodium ion-conducting solid electrolyte, operating at 310-350°C to maintain the electrodes in a molten state and to obtain adequate electrolyte conductivity [16]. The NaS battery is commercially produced and is used in applications for power quality and uninterrupted power supply.

The PSB, VRB and ZnBr batteries are redox flow batteries based on liquid electrolytes which are pumped into the battery stack. The size of the stack determines the power rating and the volume of the electrolyte determines the storage capacity. The PSB battery technology has not yet been demonstrated in commercial operation but plants are currently being constructed with a power rating of 12-15 MW and an energy capacity of 120 MWh [17, 18]. The VRB battery technology has been demonstrated by different manufacturers for load-levelling and PV-applications with power ratings up to 1.5 MW and an energy

capacity up to 5 MWh [19]. Demonstration units of the ZnBr battery have been built for PV applications with the ratings of 50 kW/100 kWh and 250 kW/500 kWh [20]. It is assumed that the different technologies of redox flow batteries can be constructed to meet the battery requirements of the PV-battery system in this study.

Technology	Abbre-	Model	Description	Positive	Electro-	Negative
	viation			electrode/ catholyte	lyte	electrode/ anolyte
Li-ion	Li-ion	SAFT Li- ion VL 50 E. Mixed oxide: LiNi0.8(Co +M) 0.2 O ₂ ^a	cylindrical, sealed maintenance free cells	Li _{1-x} MeO ₂ / LiMeO ₂ ^a	PC, LiPF ₆	Li _x C/C
Sodium- sulphur	NaS	NGK- TEPCO E50 module, 50 kW, 430 kWh/ module	384 T5 cells, (8S x 6P) x 8S, 128 V	xS/S_x^{2-} (x= 5 - 3)	β-Al ₂ O ₃	Na/ Na ⁺
Nickel- cadmium	NiCd	SAFT SBM 1150	pocket plate, vented, low rate thick electrodes, maintenance free	NiOOH/ Ni(OH) ₂	20% KOH (1.19 g/dm ³)	Cd/ Cd(OH) ₂
Nickel- metal hydride	NiMH	SAFT NH12.3, 12 V module	EV battery plates, valve regulated, foam electrodes	NiOOH/ Ni(OH) ₂	КОН	MmH/ Mm ^b
Lead-acid	PbA	Tudor Exide 16OGi 1260	vented, pasted flat plates, flooded electrolyte	PbO ₂ / PbSO ₄	1.3 kg dm ⁻³ H ₂ SO ₄	Pb/ PbSO ₄
Polysulfide bromide	PSB	Regenesys	redox flow	NaBr ₃ (aq)/ 3 NaBr (aq)	$\mathrm{H}_2\mathrm{SO}_4$	$2 \operatorname{Na}_2 S_2(aq)/$ $\operatorname{Na}_2 S_4(aq)$
Vanadium	VRB	Sumitomo Electric Industries	redox flow, 4 stacks x 80 cells (serial)	$VO_2^+(aq)/VO^{2+}(aq)$	1.8 M V in 4.2 M H ₂ SO ₄	$V^{2+}(aq)/V^{3+}(aq)$
Zinc bromine	ZnBr	ZBB research	redox flow	Br ₂ (aq)/ 2 Br ⁻ (aq)	2.25 M ZnBr ₂	Zn/Zn ²⁺ (aq)

Table 2. Description of the batteries used in the PV-battery system.

Sources: [16, 21-27]

^a Me= Mixed oxide lithiated cathode LiNi0.8(Co+M)0.2 O₂. M= Different combinations of Mn, Al and other metals are used.

^b Mm= Misch metal. AB₅ alloy of rare earth metals.

4.2. *Energy efficiencies of the components and direct energy use of air conditioning*

PV modules based on multi crystalline silicon (mc-Si) PV modules are assumed to be used in the PV-battery system since they are produced at the highest volumes today. Table 3 shows that the energy efficiencies were estimated to be 0.12-0.13 for the PV modules, 0.90-0.95 for the charge regulator and 0.92-0.94 for the inverter. Correction of efficiencies for power or temperature deviation, incomplete utilisation of irradiation etc. is not considered for these components.

The charge-discharge efficiency is highest for the Li-ion battery (Table 3). For batteries requiring pumps and auxiliary components, these losses are included. Depending on the efficiency of the inverter, the battery capacity has to be increased to cover the losses. The maximum electricity output per cycle defined by the functional unit (450 kWh) was divided by the energy efficiency of the inverter, which sets the battery energy capacity to 479 - 489kWh. The sizing has not been corrected for capacity changes with operating temperature. Likewise, the battery energy efficiency influences the power of the charge regulator.

Components	η_1	η_2	$\eta_3^{\ a}$	η_4
1. PV (mc-Si)	0.12 - 0.13			
2. Charge regulator		0.90 - 0.95		
3. Batteries				
Li-ion			0.85 - 0.95	
NaS ^b			0.75 - 0.83	
PbA			0.70 - 0.84	
NiCd			0.65 - 0.85	
NiMH			0.65 - 0.85	
VRB ^c			0.60 - 0.80	
ZnBr ^d			0.60 - 0.73	
PSB ^d			0.60 - 0.65	
4. Inverter				0.92 - 0.94

Table 3. Energy efficiencies of the PV-battery system components.

Sources: [16, 19, 22-34]

^a DC/DC efficiency. T = 20-25°C. Low value= 100% SOC charge and 100% DOD cycles, chargedischarge rates of C/1-5 h. High value= shallow cycling at C/10 h currents.

^b 3-4% lower absolute efficiency included due to loss in operation of heaters required at low cycling operation. The battery is cycled between 10-90% SOC. [°] The lower value includes losses in pumps etc. The higher value is based on suppressed pump losses

[19] ^d Including losses in pumps etc. η for ZnBr is projected to be 0.80-0.85 [32].

The value of the electricity output, E_{use} , in terms of primary fossil fuel can only be determined after comparison with a reference energy system, which electricity generation is displaced by the PV-battery system. Electricity supply by a diesel generator was chosen as a reference because it is generally applicable in stand-alone systems and back-up systems. The efficiency of the diesel generator, including its production, is assumed to be 0.20 (η^*_0).

The annual solar irradiation (*H*) was assumed to be 1.7 MWh/m² year, representing medium irradiation levels which can be found in Southern Europe and large parts of the USA [6]. The maximum solar irradiance (S_p) is assumed to be 900 W/m².

The temperature of the battery room can be held constant with passive or active systems. Passive systems do not actively change the temperature and are based on insulation, heat driven fans or water circulation systems. An active system was considered to enable evaluation of its energy related significance. The air conditioning systems provide active temperature regulation and require electricity for compressor, fan and electronic regulation. It is assumed that the daily cooling requirement is generated during daytime directly from the PV array and energy is stored as ice or cold water, which enables cooling at a constant level during day and night.

The average heat load and corresponding cooling requirement by air conditioning was calculated to 19 kW. The conversion efficiency of electricity to heat (α_5) is 3 for the AC unit, resulting in 6.3 kW electrical power (average). When the AC is turned on, it is in operation for 5.2 hours per day which equals the reference yield of the PV array.

4.3. Service life of the components

4.3.1. Float life and cycle life of batteries and service life of PV array, charger and inverter

The end of battery service life is when the battery capacity has reached 80% of initial capacity or when it fails to function. The effects of ambient temperature on the performance and service life of redox flow batteries and the NaS battery are limited since their operating temperatures are regulated by pumping of the electrolytes or by thermal management systems.

When Li-ion, NiCd, NiMH and PbA batteries are used in applications with shallow cycling, their service life normally will be limited by float life [35].⁶ In systems where the cycling is deep, but occurs only a few times a year, temperature dependent corrosion processes is the normal life-limiting factor, even for batteries with low cycle life [36]. In systems with deep daily cycling, the cycle life determines the service life of the battery [35]. Battery service life limited by cycle life is used in the reference case since the batteries are assumed to be daily cycled at DODs below 33%.

⁶ Float life testing is used to estimate the service life of batteries due to corrosion processes. Float charge is defined as a method of maintaining a battery in a charged condition by continuous, long-term constant voltage charging, at a level sufficient to balance selfdischarge [21]. Determination of float life includes capacity testing by full discharges at certain intervals (e.g. every 20 days). Float service life of redox flow batteries may be difficult to estimate since these technologies have been mainly demonstrated in cycling operation. For longer periods of storage of the battery, the stacks can be drained from electrolyte to limit corrosion processes.

Due to the uncertainties in specifying service life and to assess the variability of different modes of cycling and temperatures, calculations are made for both float life and cycle life limited batteries at different temperatures.⁷ Table 4 shows that NiCd and VRB batteries have the highest float service life while Li-ion, VRB, NaS and ZnBr have the highest cycle life at 33% DOD. For the redox flow batteries, the number of cycles does not influence the service life and in the. The cycle life of redox flow batteries corresponds to the number of cycles, which can be achieved with one cycle per day until battery float service life is reached.

Component	ti	N ₁₀₀ ^a	N ₈₀ ^b	N ₃₃ ^c	t ₃ , _{cycle} ^d	t ₃ , _{float} ^e
	(yea	(1 000 x	(1 000 x	(1 000 x	(years)	(years)
	rs)	cycles)	cycles)	cycles)		
1. PV array (mc-Si)	30					
2. Charge regulator	10					
3. Batteries						
NiCd		1.6 - 2.0	2.0 - 2.5	4.8 - 6.0	13 - 16	20 - 25
VRB ^f		2.8 - 3.0	3.0 - 4.0	7.0 - 8.0	^g 15 - 20	15 - 20
Li-ion		3.0 - 5.0	5.0 - 7.0	7.0 - 10	^h 19 - 27	14 - 16
NaS		2.3 - 2.5	4.5 - 5.0	6.8 - 7.5	^g 14 - 16	14 - 16
PSB ⁱ		9.0 - 10	9.0 - 10	9.0 - 10	^g 14 - 15	14 - 15
PbA ^j		0.32 - 0.80	0.40 - 1.0	0.90 - 2.0	2.5 - 5.5	8.0 - 12
ZnBr		1.5 - 2.5	2.5 - 3.0	4.0 - 5.0	^g 8.0 - 10	8.0 - 10
NiMH		0.60 - 1.0	0.80 - 1.2	2.8 - 3.0	7.7 - 8.2	8.0 - 10
4. Inverter	10					
5. Air conditioning	8					

Table 4. Service life of PV-battery system components.

Sources: [12, 15, 16, 19, 22, 24, 25, 27, 32-35, 37-39]

^a Cycle life at 100% DOD and 20-25°C.

^b Cycle life at 80% DOD and 20-25°C.

^c Cycle life at 33% DOD and 20-25°C.

 d t_{3, cycle}= N₃₃/ n (Equation 16), where n= 365 cycles/year at 33% DOD.

^e Battery service life at 20-25°C at no-cycling (float charge).

^f Ionic membranes have to be replaced every 10 years.

^g Limited by float service life when cycled one cycle per day. Cycle life= float life (2 900 - 7 300 cycles).

^h 5 years shown in practice for Li-ion (Co) [39]. If limited by float service life *f* decrease 9% and η^*_B decrease 1.4%.

ⁱ Electrolyte management assumed for this cycle life performance [17]. Float life may be up to 20 years.

^j The higher values represent the performance of advanced gelled electrolyte PbA [15].

⁷ Battery specifications stated for calendar life is assumed to be equivalent to float service life. Data for shelf life, which is the duration of storage at the end when a battery still retains the ability to give specified performance, was not considered.

4.3.2. Temperature corrected service life of batteries

Temperatures can vary considerably throughout the days and seasons where PV systems are situated. To generate large amounts of solar energy, PV systems need be located at the sunniest conditions possible, which also mean that the ambient temperatures can be very high. The temperature on the PV module can be 30-35°C above the ambient temperature.

The values in Table 5 present extreme temperatures under which the batteries preferably should work for only short periods. Elevated temperatures result in accelerated ageing but also higher available capacity. NiCd has a robust mechanical design and its service life is relatively little affected of ambient temperatures above 40°C. The NaS battery is operated at 310-350°C and insulation and heaters are used to maintain its temperature.

The charge regulator and the inverter require cooling of electronics. Cooling is achieved by fans, which energy use is assumed to be included in the efficiency of inverter and charger.

Table 5. Ambient temperatures for battery operation and factors for temperature corrected battery service life.

Technology	Temperature (°C)	σ _{30°C}	σ _{35°C}	σ _{40°C}	σ _{45°C}	σ _{50°C}
Li-ion	-20 - 50	0.72	0.55	0.40	0.30	0.23
NaS ^a	-40 - 50	1.0	1.0	1.0	1.0	1.0
NiCd	-50 - 50	0.90	0.80	0.73	0.65	0.57
NiMH	0 - 40	0.85	0.75	0.65	0.52	0.35
PbA	-30 - 40	0.69	0.51	0.37	0.25	0.14
PSB	^b -40 - 50	1.0	1.0	1.0	1.0	1.0
VRB	^c 15 – 40	1.0	1.0	1.0	N/A	N/A
ZnBr	^c 10 – 40	1.0	1.0	1.0	N/A	N/A

Sources: [22, 33, 39-41]

Note: σ = temperature correction factor. Change in service life relative 25°C for battery cycle life and float service life. N/A = not applicable

^a The battery operating temperature is 310-350°C for the NaS battery

^b Heat generated in the battery prevent the electrolytes from freezing.

^c Optimal operating temperature is 25-30°C. Heat exchanger has to be operated at T > 30°C.

As temperature increases, the electrochemical activity of the battery increases and as well as the speed of the natural ageing of the active material increases. Accelerated life tests at elevated temperature, correlated with corrosion studies, provide a basis for estimating service life. Service temperature is the key factor in determining corrosion. As a rule for PbA batteries, every 10°C increase in temperature, service life at 20°C and >10 years at 25°C [42]. The rate of ageing for NiCd batteries is about 20% reduction in life for 10°C increase in temperature [42]. NiCd is less affected than PbA since increased electrochemical activity has little affect the steel structural components of the NiCd electrode assembly.

To include the effects of operating temperature on the battery service life, temperature correction factors, σ , were estimated relative 25°C (Table 5). The temperature correction

factors are assumed to be equal for both cycle life and float service life. Operational data is available for PbA and NiCd while data is uncertain for NiMH and Li-ion. No empirical data was available for the relative change in service life from 20°C.

The PSB and VRB batteries are little affected by varying ambient temperatures since pumps are used to circulate the electrolyte to heat exchangers which maintain their operating temperatures. No degradation of the NaS battery has been documented but the temperature of the electronic control systems has to be controlled. Elevated temperature may degrade plastic materials of the ZnBr battery, but no degradation has been quantified.

4.4. Energy requirements for production and transport of the PV-battery system components

4.4.1. Mass requirements of the components

Material intensities are used to calculate the mass of components that is required to give a certain service. The gravimetric density for the PV array including module frame was estimated to 9 kg/m² for roof integrated system and 12 kg/m² for ground mounted system. Table 6 shows that the mass of the PV array is 2.6-4.9 tons depending on the energy efficiency of the battery technology used. The PSB and VRB are the heaviest ones (24-49 tons) followed by PbA (15-24 tons). The mass of these systems is 5-8 times higher than the Li-ion battery, which has the lowest weight. The weight fraction of the electrolyte active material, *y*, is 0.32-0.40 for the redox flow batteries (PSB, VRB and ZnBr) and 0 for the other batteries (Eq. 17).

Component	$w_1 (kg/m^2)$	V _i (kg/kW _{el})	d ₃ (Wh/kg)	mass, m _i ^a (tons)
1. PV array (mc-Si)	^b 9.0			2.6 - 4.9
2. Charge regulator		10		0.56 - 0.91
3. Batteries				
NaS			103 - 116	4.1 - 4.8
Li-ion			80 - 120	4.0 - 6.1
ZnBr			70 - 85	5.6 - 7.0
NiMH			35 - 55	8.7 - 14
NiCd			22 - 30	16 - 22
PbA			20 - 32	15 - 24
VRB			15 - 20	24 - 33
PSB			10 - 15	32 - 49
4. Inverter		10		0.50
5. Air conditioning		° 42		0.11 - 0.36

Table 6. Densities and mass of complete PV-battery system components.

Sources: [12, 16, 21, 22, 24, 25-27, 44]

^a C = 479-489 kWh, $P_{use} = 50$ kW, $P_5 = 1.8-8.6$ kW.

^b Including module, frame and roof mounted array supports

^c Electrical power of air conditioning unit. (45 kg/ $3.2 \text{ kW}_{\text{heat}}$) x 3 kW_{heat} / kW_{el} = 42 kg/W_{el} [45]

4.4.2. Energy requirements for production of the components

The energy requirements for mc-Si modules are estimated to 4 200 MJ_{pf}/m^2 or 32 MJ_{pf}/W_p [5]. The energy requirements for production of single crystalline silicon (sc-Si) and amorphous silicon (a-Si) modules are 16-20% higher and 0-17% lower, respectively, than for mc-Si.

Module frames are assumed to use 2.5 kg Al/m² module corresponding to 500 MJ_{pf}/m^2 [5]. Energy requirements for array supports in roof integrated and ground mounted systems have been estimated to be 700 MJ_{pf}/m^2 and 1 800 MJ_{pf}/m^2 , respectively [5]. The total energy requirements for production of PV arrays (module, frame and array support) were calculated to be 5 400 MJ_{pf}/m^2 (roof integrated) and 6 500 MJ_{pf}/m^2 (ground mounted), corresponding to 45-54 MJ_{pf}/W_p (Table 7).

Energy requirements for producing inverter, charger and AC were estimated to be 1-3 MJ_{pf}/W_{el} , resulting in 70-100 MJ_{pf}/kg (Table 8). The power required of the PV array to operate the AC (P_5 = 1.8-11 kW_{el}) via an inverter is 2.1-12 kW, which means that the PV array rating has to be increased by 5-19% if AC is to be used.

Energy requirements for production of batteries were assessed from cradle to gate, including materials production and battery manufacturing. To enable a thorough evaluation of different LCA studies, they need to report the following parameters: (1) battery mass, (2) battery capacity, (3) choice of system boundaries (geographical, temporal, technological) (4) electricity's share of gross primary energy requirements, assumed efficiency of electricity production and method for adding up different energy qualities, (5) recycling rate of used batteries, (6) energy requirements for battery manufacturing processes and production of virgin and recycled materials, respectively, (8) battery design, (9) battery material composition, and (10) the allocation principles for multi-output processes. Based on published life cycle assessments and estimates [13], energy requirements for battery production were scattered between studies and there are uncertainties on the material requirements and manufacturing processes, particularly for the Li-ion, NaS and all the redox flow batteries. The best estimates are presented in Table 7.

The energy requirements of batteries with active materials in solid phase are assumed to be independent of the required PV-battery system output power by changing the cell configuration and voltage of the battery strings. Energy requirements for redox flow batteries have to be divided into stack production (MJ_{pf}/W) and energy storage capacity electrolyte (MJ_{pf}/Wh) , respectively. The energy requirements for redox flow batteries have been aggregated corresponding to a stack of 53-54 kW (*C*/*P_{use}= 9.8*), which makes it necessary to revise the data for assessments of applications with other power-capacity ratios.

Component	Materials and manufacturing (MJ _{pf} /m ²)	Materials and manufacturing (MJ _{pf} /W _{el})	Recycled materials recovery (MJ _{pf} / Wh)	Virgin materials recovery (MJ _{pf} / Wh)	Manufacturing (MJ _{pf} / Wh)
1. PV array (mc-Si)	^a 5400		· · ·	· · ·	
2. Charge regulator		^b 1.0			
3. Batteries					
Li-ion			0.31	0.67	1.2
NaS			0.29	0.80	0.60
NiCd			1.0	2.0	2.1
NiMH			0.60	1.6	2.1
PbA			0.45	0.77	0.42
PSB ^c			1.1	1.7	0.59
VRB ^c			1.4	2.1	0.74
ZnBr ^c			0.30	1.2	0.60
4. Inverter ^b		^b 1.0			
5. Air conditioning		^d 3.0			

Table 7. Energy requirements for production of the PV-battery system components.

Sources: [12, 13, 46-50]

^a Incl. module, frame and roof mounted array supports. mc-Si multi crystalline silicon η = 12-13% (Alsema 2000)

^bBased on 3 kW module [5] and on 500 kW converter [51].

 c C= 479-489 kWh, P_{use}= 50 kW

^d Estimated based on inverter data

The energy requirements for production of batteries range from 0.87 MJ_{pf}/Wh (PbA based on recycled materials) to 4.1 MJ_{pf}/Wh (NiCd based on virgin materials) (Table 7). Differences are partly explained by the energy intensity of materials recovery. For the NiCd battery, steel and nickel contribute 60-70% to the energy requirements of materials recovery. Production of nickel requires 2-8 times more energy than lead used in PbA batteries. Energy requirements for manufacturing processes contribute 33-78% of the gross energy requirements for battery production, resulting in relatively small changes between virgin and recycled materials.

Energy requirements for battery manufacturing are assumed to be constant while energy requirements for materials vary depending on the recycling of materials. The recovery of recycled materials requires 32-75% less energy than virgin materials since energy for extraction from mines and reduction of metals are allocated to the first material life cycle (Table 7). For redox flow batteries, it is assumed that the energy requirements for production of stack and other battery components are the same as for electrolyte production.

At the end of the service life of redox-flow batteries, they can be renovated by renewing the stacks while the electrolyte with active material is assumed to be used indefinitely (y= 0.32-0.40). Batteries based on active materials in solid phase (Li-ion, NaS, NiCd, NiMH, PbA) need to be manufactured and replaced completely.

Table 8 shows the energy requirements expressed per kg component. The fraction β_i of primary fossil energy requirement that is used for producing the electricity required for production of batteries, charge regulator and AC was estimated from LCA reports.

Component	Materials and	Recycled	Virgin materials	Manufacturing,	β_i^{a}
	production, q _{P,i}	materials	recovery, q _{RV3}	q_{M3}	
	(MJ _{pf} /kg)	recovery, q _{RR3}	(MJ_{pf}/kg)	(MJ _{pf} ∕ kg)	
		(MJ_{pf}/kg)			
1. PV array (mc-Si) ^b	600				
2. Charge regulator	100				0.50
3. Batteries					
Li-ion		25 - 37	53 - 80	96 - 144	0.75
NaS		30 - 34	82 - 93	62 - 70	0.70
NiCd		22 - 30	44 - 60	46 - 63	0.68
NiMH		21 - 40	54 - 102	74 - 139	0.68
PbA		9.0 - 14	15 - 25	8.4 - 13	0.65
PSB		11 - 17	17 - 26	5.9 - 8.9	0.50
VRB		21 - 29	32 - 42	11 - 15	0.41
ZnBr		21 - 26	84 - 102	42 - 51	0.50
4. Inverter	100				
5. Air conditioning	70				0.50

Table 8. Energy requirements for production of the PV-battery system components expressed per kg component.

Sources: Table 6 and Table 7

^a Share of primary energy used to generate electricity

^b Incl. module, frame and roof mounted array supports. mc-Si= multi crystalline silicon

4.4.3. Energy requirements for transport of the components

The transportation distance, L, was set to 3 000 km and transportation was done by heavy truck ($e_{truck}=0.72 \text{ MJ}_{pf}$ /tonkm) or by plane ($e_{air}=20 \text{ MJ}_{pf}$ / tonkm) [52].

Distilled water was assumed to be available on the site of use and did therefore not have to be transported or returned at the end of battery life. For redox flow and flooded batteries the mass made up by water was subtracted from the total battery mass requiring transportation. The mass of the transported battery was reduced with a factor (x) for the following batteries: NiCd (26-27%), PbA (12-13%), PSB (45-48%), VRB (45-48%) and ZnBr (40-45%).

5. Results

5.1. Uncertainties and contributing components

To present the uncertainties and the contribution of different components to the gross energy requirement, the following section presents the results for the reference case (Case 1) when the battery service life is limited by cycle life and the temperature is 25° C. It is

assumed that the batteries are produced from 100% recycled materials and that the different components are transported 3 000 km by heavy truck.

Fig. 5 shows that the energy return factor for the PV-battery system ranges from 2.3 for NiMH batteries to 12 for Li-ion batteries. This means that 8.1-44% of the energy output is required to produce the PV-battery system. The Li-ion battery has the highest average energy return factor (9.8), which means that the PV-Li-ion battery system will replace 9.8 times more energy throughout its life time than the energy required for its production.

For a PV-battery system with a service life of 30 years and taking into account the different service lives of the components, the energy payback time is 2.4-13 years, depending on the battery technology used. The PV array excluding batteries has an energy return factor of 10-19. With a service life of 30 years, the energy payback time is 1.6-3.0 years for the PV array. The energy payback time is 0.55-10 years for the different battery technologies, showing the energy related significance of batteries in PV-battery systems.

The uncertainty in the average value of the energy return factor is +/- 14-53%. The greatest uncertainty in f is found for the PbA battery due to its high variability in cycle life performance. Fig. 5 shows the importance of using specific data when comparing different battery technologies.



Fig. 5. Energy return factors for the PV-battery systems. Case 1: $T=25^{\circ}C$, 100% recycled battery materials, service life limited by cycle life, and transportation by heavy truck. The variation in the average value is +/-14 to 53%.

Production and transportation of batteries contributes 25-70% to the total production energy of the PV-battery system, also underlining the energy related significance of batteries in PV systems (Fig. 6). The relative contribution due to the production of batteries is lowest for the ZnBr battery and highest for the NiMH battery.

The contribution of production and transport of the PV array is 26-70% (NiMH-ZnBr). The highest absolute energy requirement for PV array production is 80-87 GJ/year for the redox

flow batteries due to their relatively low efficiency, resulting in the need for a larger PV array and charge regulator. Production and transport of the charge regulator contribute 1-4% (NiMH-ZnBr) to the gross energy requirement. The corresponding figures for the inverter are 2-5%.

The contribution of transport of all the components to the gross energy requirement is low (1.0-9.2%) for 3 000 km transport by heavy truck. The lowest energy requirement for transport is for the ZnBr battery due to its high energy density and the possibility of recycling the electrolyte. The transport of PbA batteries contributes 9.2% to the gross energy requirement since these batteries have a relatively low energy density and cycle life, and therefore a larger mass of batteries has to be transported.



Fig. 6. Energy requirements for production and transport of various PV-battery systems. Case 1: T=25°C, 100% recycled battery materials, service life limited by cycle life, and transportation by heavy truck. The uncertainty is +/- 14 to 53%.

Fig. 7 shows that the overall battery efficiency for the PV-battery system ranges from 0.41 for the NiMH battery to 0.82 for the Li-ion battery. The uncertainty is $\pm - 9$ to 24%. The greatest uncertainty is for the NiMH battery due to the great variability in energy density.

The direct efficiency of the charger, battery and inverter is 0.50-0.85. When considering the charger-battery-inverter system in a life cycle perspective, the corresponding figure for the overall battery efficiency is 0.41-0.82, which is an average decrease by 8.7%. The average efficiency of the NiMH battery decreases by 18%, from 0.65 to 0.53, which shows the effect of high energy requirements for production on the overall efficiency.



Fig. 7. Overall battery efficiencies including production and transport of the charger, battery and inverter. Case 1: service life limited by cycle life, $T=25^{\circ}C$, 100% recycled battery materials and transportation by heavy truck. The variation around the average value is +/- 9 to 24%.

Fig. 8 shows the gross electricity requirements for the battery system where the energy requirements for production and transport of the charger, the battery and the inverter have been converted to electricity.

The output energy from the batteries is the same for all batteries, corresponding to the functional unit. The relative contribution of output energy is 50-72% for the different battery technologies. The relative contribution of output energy decreases with the increasing number of days of storage capacity, i.e. the size of the battery.

Energy losses in the batteries contribute 9-33% of the gross energy requirement. Li-ion has the highest energy efficiency (0.85-0.95) where energy losses are 9% of the total. VRB, ZnBr and PSB have efficiencies of 0.60-0.80 and battery losses contribute to 27-33% of the gross energy requirement.

The ranking of batteries according to direct or overall efficiency depends mainly on battery production and transport, which contribute 3-19% of the gross energy requirement. The highest relative contribution for battery production and transport are for PbA and NiMH, causing a large difference between their direct energy efficiency and their overall battery efficiency.

Losses in the charger and inverter are 6-8% and 4-5% of the gross energy requirement, respectively. Production and transport of the charger and inverter contribute less than 1% of the gross energy requirement.





Fig. 8. Gross electricity requirements for the charger, battery and inverter including their production and transport. Case 1: service life limited by cycle life, T=25°C, 100% recycled battery materials and transportation by heavy truck. The uncertainty is +/- 9 to 24%.

5.2. Summary of results for the different cases

Table 9 and 10 presents a summary of the energy return factors and overall battery efficiencies for seven different cases. To evaluate the influence importance of depth of discharge and battery temperature on *battery service life*, four different cases were analysed. The effects of using AC on the overall battery efficiency were evaluated to assess for which batteries the installation of AC can be motivated.

Battery service life is limited by cycle life, except in Case 2, where it was limited by float service life. The battery temperature in the various cases was set to $(1) 25^{\circ}$ C, $(2) 25^{\circ}$ C, float (3) 25°C using active cooling with AC and, (4) 40°C. The other parameter settings are the same as in Section 5.1, i.e. it is assumed that the batteries are produced from 100% recycled materials and the different components are transported 3 000 km by heavy truck.

The consequences of *material recycling* and *transport* by plane instead of truck from the battery manufacturer to the site of operation, is analysed in Case 5 to 7. The temperature assumed was 25°C and the transportation distance for the PV-system components 3 000 km.

Technology	1. Cycle, 25°C	2. Float, 25°C	3. Cycle, 25°C,	4. Cycle, 40°C,	5.0% recycling,	6. 100% recycling, plane	7.0% recycling,
			AC on	AC off	truck		plane
Li-ion	9.8	8.4	9.3	6.8	9.2	7.9	7.5
NaS	9.2	9.2	N/A	9.2	8.0	6.9	6.2
NiCd	5.5	6.7	5.2	4.6	4.7	3.4	3.1
NiMH	4.0	4.4	3.8	3.0	3.3	2.4	2.1
PbA	5.1	7.8	4.9	2.7	4.4	1.6	1.5
PSB	7.3	7.3	N/A	7.3	5.6	5.0	2.9
VRB	7.8	7.8	7.1	N/A	6.0	6.1	3.8
ZnBr	8.1	8.1	7.2	N/A	5.3	7.2	4.2

Table 9. Energy return factor for the PV-battery systems for the different cases. The uncertainty is +/- 14 to 61%.

N/A = Not applicable

Table 10. Overall efficiencies of the PV-battery systems for the different cases. The uncertainty is +/- 9 to 33%.

Technology	1. Cycle, 25°C	2. Float, 25°C	3. Cycle, 25°C,	4. Cycle, 40°C,	5.0% recycling,	6. 100% recycling, plane	7.0% recycling,
			AC on	AC off	truck		plane
Li-ion	0.73	0.72	0.68	0.69	0.72	0.71	0.70
NaS	0.65	0.65	N/A	0.65	0.64	0.63	0.61
NiCd	0.57	0.60	0.52	0.55	0.56	0.52	0.50
NiMH	0.53	0.54	0.48	0.49	0.50	0.46	0.44
PbA	0.56	0.62	0.51	0.46	0.54	0.38	0.37
PSB	0.51	0.51	N/A	0.51	0.49	0.49	0.43
VRB	0.57	0.57	0.51	N/A	0.55	0.55	0.50
ZnBr	0.55	0.55	0.48	N/A	0.52	0.54	0.50

N/A = Not applicable

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5.3. Influence of service life

Considering on cycle per day, float service life is longer than cycle life for NiCd, NiMH and PbA batteries. The energy return factors are in this way increased by 10% for NiMH, 23% for NiCd and 52% for PbA. For Li-ion batteries, the energy return factor decreases by 14% for float service life compared with cycle life. Float service life will be the life-limiting factor for NaS, PSB, VRB and ZnBr batteries.

Fig. 9 shows that the energy return factor for the PV-battery system ranges from 2.7 (PbA at 40°C) to 9.8 (Li-ion at 20°C). The uncertainty is +/- 14 to 61%. The greatest uncertainty is for the PbA battery due to the great variability in cycle life performance.

Increasing the temperature to 40° C from 25°C, causes the energy return factor to decrease 0-47% for the different battery technologies. For batteries whose service life is temperature dependent, cooling of the batteries by operation with AC improves the service life compared with operation at 40°C. The effect of increased battery service life must be related to the energy losses due to operation with AC, to ascertain whether if active cooling results in improved overall battery efficiency.

Operation with AC increases the energy return factor for NiCd from 4.6 to 5.2 (+14%), for NiMH from 3.0 to 3.8 (+27), for Li-ion from 6.8 to 9.3 (+37%), and for PbA from 2.7 to 4.9 (+79%). AC is therefore beneficial for these technologies. NaS and PSB batteries are excluded from Case 3 with external AC, since these technologies have built-in thermal management systems. VRB and ZnBr batteries are excluded from Case 4 since these technologies require AC at temperatures above 30°C, and it is therefore not possible to evaluate the use of AC.



Fig. 9. Energy return factors for the PV-battery systems at different operating temperatures. The uncertainty is $\pm/-14$ to 61%.

The difference in overall battery efficiency between service life limited by cycle life and float life is -2 to 10%. Float service life is longer than cycle life for NiCd, NiMH and PbA batteries, resulting in 2-10% higher overall energy efficiency for these batteries. Fig. 10 shows that the overall efficiency of the PV-battery system ranges from 0.46 to 0.73. The uncertainty is +/-9 to 33%.

When considering the energy return factor, operation with AC is motivated for NiCd, NiMH, Li-ion and PbA batteries but this situation changes when evaluating the overall battery efficiency. A comparison with AC turned on with 40°C shows that AC results in higher overall efficiency for PbA batteries, while it decreases the overall efficiency for Li-ion, NiMH and NiCd batteries (Fig. 10). A high energy return factor will be important in the expansion phase of PV-battery systems, while the overall battery efficiency is important when considering the long-term perspective when renewable energy has to be used efficiently. A high energy return factor is important in an expansion phase of PV-battery systems while the overall battery efficient use of solar energy in a long-term perspective.

The direct energy requirement for operating AC is $21-60 \text{ GJ}_{el}$ /year, corresponding to 98% of the direct and indirect energy requirements of the AC unit. To improve the overall efficiency, it is most effective to improve the usage phase of the AC system.



Fig. 10. Overall battery efficiencies at different operating temperatures. The uncertainty is +/-9 to 33%.

5.4. Influence of material recycling and transport

Table 9 shows that the energy return factor for the PV-battery system ranges from 3.8 to 9.8. The greatest uncertainty (+/- 57%) of the energy payback time is for the PbA battery due to the great variability in cycle life performance. Considering the uncertainty interval, the lower values of the energy return factor is below one for the PbA battery when transported by plane.

When using virgin material the energy return factor decreases 7-35% compared with recycled material recovery. The greatest change of the energy return factor is for the ZnBr battery, which decreases from 8.1 to 5.3.

Fig. 11 shows that batteries with low energy density and short cycle life are most influenced by air transport. Air transport decreases the energy return factor for PbA batteries by 70%. Although redox flow batteries have a low energy density, the energy requirements for their transport are reduced since distilled water can be supplied at the site

of operation and the active materials in the electrolyte can be recycled on site. With no recycling, the energy return factor of the ZnBr battery is reduced by 35%.



Fig. 11. Relative changes in the energy return factor for different recycling rates and modes of transportation compared with Case 1.

Fig. 12 shows that the overall battery efficiency decreases by 1-6% when using virgin material compared with recycled material. The greatest change in the overall battery efficiency is seen for the ZnBr battery, for which the value decreases from 0.55 to 0.52 (Table 10). Air transport decreases the overall battery efficiency by 3-33%. The overall battery efficiency of the PbA battery decreases from 0.56 to 0.38 (Table 10).



Fig. 12. Relative changes in the overall battery efficiency for different recycling rates and modes of transportation compared with Case 1.

5.5. Sensitivity analysis of battery performance

To improve the energy efficiency of batteries, it is important to identify those parameters, which have the greatest influence. The relative importance of different battery parameters

was therefore identified with sensitivity analysis. The following battery parameters were analysed: service life (t_3), gravimetric energy density (d), charge-discharge efficiency (η_3) and energy requirements for production (q_{P3}). Case 1 was used as reference.

Improvement in the battery efficiency has the greatest influence on the energy return factor for Li-ion, NaS, PSB, VRB and ZnBr batteries (Table 11). The battery efficiency is the second most important parameter for NiMH, NiCd and PbA batteries. An improvement in the battery efficiency by one percent results in an increase in the energy return factor of 0.40-0.73%. Since losses in the battery must be compensated by higher energy input, low battery efficiency results in a larger PV array and charger, which means higher indirect energy requirements for their production.

For NiMH, NiCd and PbA batteries, the energy density and the service life have the greatest influence (0.51-0.63 % f / $\%\Delta\eta_3$). The substantial influence of these parameters is explained by the energy requirements for battery production, which is 56-70% of the gross energy requirement for NiMH, NiCd and PbA compared with 25-35% for the other battery technologies (Fig. 6). Low energy density results in higher material intensity and energy requirements for the production of materials as well as a higher battery mass to be transported. Short battery service life means that batteries have to be replaced more often, resulting in higher energy requirements for battery production and transport.

The energy requirement for battery manufacturing is an important parameter for NiCd and NiMH batteries. Energy requirements for battery production, q_{P3} , have 4-16% smaller influence than *d* and $t_{cycle,3}$, since this parameter does not influence the energy requirement for battery transport.

Technology	η_3	d, t _{cycle,3}	q_{P3}
Li-ion	0.42	0.28	0.28
NaS	0.65	0.26	0.25
NiCd	0.40	0.52	0.50
NiMH	0.30	0.63	0.61
PbA	0.41	0.51	0.43
PSB	0.66	0.26	0.25
VRB	0.62	0.29	0.28
ZnBr	0.69	0.23	0.23

Table 11. Percent change in the energy return factor when changing various battery parameters (% f / % $\Delta\eta_3$).

Note: Case 1: T=25°C, 100% recycled battery materials production, service life limited by cycle life, and transportation by heavy truck. $t_{cycle,3}$ = cycle life, d= gravimetric energy density, η_3 = charge-discharge efficiency, q_{P3} = energy requirements for battery production. **Bold** face indicates the highest values.

Table 12 shows that the charge-discharge efficiency of the battery has the greatest influence (0.69-0.96 %) on the overall battery efficiency. This due to the high energy turnover through the battery (50-72% of the total turnover). Changes in the efficiency of the Li-ion

battery have a relatively small influence compared with the other battery technologies. This is because the losses in the Li-ion battery correspond to 9% of the total gross energy requirement, compared with 19-33% for the other technologies. This results in a smaller improvement than for the other battery technologies and a relatively small change compared with the already high efficiency of the Li-ion battery.

Improvement of energy requirements for production, service life and gravimetric energy density by one percent change the overall battery efficiency by 0.03-0.15%. All these parameters influence the indirect energy requirements for production and transport. Since the electricity output from the PV array is partly converted to other energy carriers (thermal energy and transport fuel), it gives 2.5-2.7 times higher energy yield which decreases the relative importance of production and transport of the batteries and charger.

Table 12. Percent change of the overall battery efficiency when changing different battery parameters ($\% \eta *_B / \% \Delta$).

Technology	η_3	d, t _{cycle, 3}	q_{P3}
Li-ion	0.69	0.04	0.04
NaS	0.96	0.03	0.03
NiCd	0.88	0.10	0.09
NiMH	0.81	0.15	0.15
PbA	0.85	0.13	0.11
PSB	0.95	0.03	0.03
VRB	0.95	0.04	0.04
ZnBr	0.96	0.03	0.03

Note: Case 1: service life limited by cycle life, T=25°C, 100% recycled battery materials production and transportation by heavy truck. $t_{cycle,3}$ = cycle life, d= gravimetric energy density, η_3 = chargedischarge efficiency, q_{P3} = energy requirements for battery production. **Bold** numbers indicate the highest values.

5.6. Sensitivity analysis of external parameters and PV-battery system design

5.6.1. Influence of solar irradiation on the energy return factor

Depending on the distance to the equator, different levels of irradiation can be distinguished⁸. At low irradiation levels, the power rating of the PV arrays has to be increased to produce the same amount of energy. This results in higher indirect energy requirements for production of PV arrays. The level of solar irradiation influences the energy return factor for the PV array proportionally. The importance of the level of solar

⁸ High irradiation (2.2 MWh/m² year) can be found in south western USA and Sahara, medium (1.7 MWh/m² year) levels can be found in large parts of USA and Southern Europe, and low levels (1.1 MWh/m² year) can found in the middle of Germany [6]. A PV module installed in the middle of Germany therefore has 55% lower energy return factor than a module in Southern Europe.

irradiation on the total energy return factor depends on the relative contribution of the PV array to the gross energy requirements of the PV-battery system. For Case 1, an increase of the solar irradiation by 30% to 2.2 MWh/m² year, the energy return factor increases 8-19% for the different battery technologies. The solar irradiation has low influence on the overall battery efficiency since it only affects the power rating of the charger.

For the conditions in Case 1, the energy return factor equals one at irradiation levels below 100-250 kWh/m² year. This low irradiation is not relevant for practical applications and may only be found indoors. The results show the importance of locating PV plants at highest possible irradiation levels to increase the efficiency of PV-battery system. The solar irradiation may be increased by using reflectors thereby decreasing the size of PV arrays.

5.6.2. Influence of the electricity conversion efficiency

The value of electricity depends on the conversion efficiency form primary fuel to electricity. The valuation of electricity increases with decreasing conversion efficiency. Depending on which electricity generation technology the PV system replaces, different efficiencies can be assumed.

A conversion efficiency of 0.20 was selected as a default value to represent a diesel generator since it is likely that a PV-battery system can replace its use in an off grid application. If the efficiency of the diesel generator is 0.25, it results in a reduction of the energy return factor by 20% (0.20/ 0.25)). Assuming that the PV-battery system replaces a grid-connected system where the electricity conversion efficiency is 0.35, the energy return factor decreases by 43%.

The absolute values of the energy return factor has no effect on the relative comparisons of batteries. Considering higher conversion efficiency of the replaced electricity generation, some battery technologies will have energy return factors below one at certain conditions.

5.6.3. Influence of energy conversion efficiencies in a closed renewable energy system

The results presented above are based on the assumption that electricity generated by the PV-battery system replaces fossil fuels which otherwise would have been used to generate the required form of energy (the *Reference case*). Assuming that electricity generated by the PV array is used for production of PV-battery systems in a closed renewable energy system (the *All PV case* in Table 1), the overall battery efficiencies decrease by 3-15% to 0.35-0.80. This reduction in overall battery efficiency means that energy efficient production of the PV-battery system becomes more important when considering a closed energy system.

5.6.4. System design- best and worst cases

To evaluate the improvement potential and extreme values a case with transportation is done by electrical train (other parameters the same as in Case 1) and a worst case, which include 40°C battery temperature, virgin materials production and 6000 km transportation by plane.

For the best case, the energy return factor improves by 1-7% (1.5-11.5) and the overall battery efficiency is improved by less than one percent compared with Case 1. This is due to that transport by electrical train is 75% more efficient than truck.

For the worst case, the energy return factor decreases by 9-73% (0.1-4.9) compared with Case 1. The overall battery efficiency decreases from 0.39-0.87 to 0.082-0.73.

5.6.5. System design- relationship between depth of discharge and cycle life

Depending on the characteristics of the battery, deep discharges may reduce the cycle life of the battery. This is particularly an issue for PbA batteries since deep discharges lead to morphological changes of the active material and creation of lead-sulphate crystals with low conductivity may make it difficult to recharge the battery to full capacity. A PbA battery is therefore dimensioned not to exceed 80% DOD, resulting in a larger battery and thus higher energy requirements for its production. Case 1 is based on 33% DOD, resulting in a battery three times larger than a design, where the battery is discharged to 100% DOD. This means that the battery has to be replaced more often and therefore more energy is required for battery production. It may be possible to optimise the over capacity a PbA battery to achieve maximum overall battery efficiency.

Table 13 shows that 80 and 100% DOD has 4-8% higher energy return factor than at 33% DOD. Higher DOD also increases the overall battery efficiency. It is therefore more energy efficient to install a smaller PbA battery, which is discharged to 80% DOD and thereby having shorter cycle life. The relative change depends on the DOD versus cycle life characteristics of the battery.

Capacity	DOD at 150	Cycle life, N (cycles)	Energy return	Overall battery
battery	kWh/day (%)	(from Table 4)	factor, f	efficiency, η_{B^*}
(kWh)				
450	33	$900 - 2\ 000$	2.4 - 7.8	0.43 - 0.69
188	80	$400 - 1\ 000$	2.5 - 8.4	0.44 - 0.70
150	100	320 - 800	2.5 - 8.4	0.44 - 0.70

Table 13. Energy return factor and overall battery efficiency for the lead-acid battery at different depth of discharge.

6. Discussion

6.1. Uncertainties

The low and high values indicate the uncertainties in the results as well as the improvement potential of different technologies. Input data with high influence and large uncertainty interval is the battery charge-discharge efficiency and the battery service life. The uncertainty in output results for different battery technologies vary between 8% and 61%. The difference between low and high values of input data is 1.1-2.2 times, where the highest variability is for NiMH and PbA. Since all battery technologies, except for PbA and NiCd, are immature for PV applications there are uncertainties about their performance. The absolute values of the energy return factors are use dependent due to the high influence

of the conversion efficiency to electricity. With higher efficiency of the replaced reference system for electricity generation ($\eta_0 = 0.35$ instead of $\eta_0 = 0.20$) the absolute values of the energy return factor decrease by 43%.

The performance ratio, which is the ratio between the final yield and the reference yield of a PV system, is a common indicator for expressing the efficiency from PV array to useful energy. Typical values on performance ratios are 0.20-0.60 in off-grid PV-systems and 0.60-0.85 in on-grid PV-systems [53, 54]. The direct efficiency is 0.50-0.85 in this study, which is partly explained by that the rated PV efficiency is used in the calculation. In practice the PV array works below maximum rated performance due to the effects of heat and irregular irradiation. The high energy efficiency of the Li-ion battery and that optimised charging is assumed also contribute to the difference. In practice batteries may be float charged occasionally resulting in lower performance ratios.

The energy return factor in this study is given for a battery storage capacity three times higher than the daily energy output. Since the energy payback time for the battery and battery transport increases linearly with the battery storage capacity of the PV system, it can be recalculated to other storage capacities. Alsema [11] calculated the energy payback time for PbA batteries in a solar home system (SHS) to be 10 - 19 years. The number of days of autonomy is 6.8-20 days (battery voltage 12 V, battery capacity 70-100 Ah and array output 60-124 Wh/day). Recalculated to 3 days of autonomy, the energy payback time is 1.5-8.4 years, which can be compared with 1.8-9.6 years in this study. The service life for the starting-lighting-ignition (SLI) battery is assumed to be 3 years by Alsema [11] and 2.5-5.5 years in this study when cycle life limited at one cycle per day). In SHS without charge regulator and poor battery maintenance, the service life may be less than 3 years batteries when standard SLI- PbA batteries are used. Improvement of the charge/discharge strategy can extend the service life PbA batteries in PV applications [55].

Only a small number of demonstration units have been built of VRB, PSB, ZnBr and NaS batteries and mass production is likely to improve the production efficiency of batteries. Energy requirements for production of batteries may vary considerably depending on material requirements, where and how they are manufactured. Uncertainties are due to restricted availability of information since companies manufacturing batteries protect their technology from competitors. Material requirements for immature technologies can change fast in the course of development. However, this analysis indicates the technical potential of different technologies. Since the performance of a technology depends on the system design, data have to be compiled corresponding to the specific conditions of a particular application.

6.2. Implications of the overall energy efficiency of PV-battery systems

The overall efficiency can be used as a complementary indicator to the energy return factor, which provides a measure of the total energy turnover of the system. For batteries with a relative high decrease in the overall battery efficiency compared with the direct energy efficiency, measures need to be directed on reducing the energy requirements for production of the PV-battery system.

If the energy return factor is less than one, the energy used to produce and replace the device is larger than the output. In PV applications located at latitudes with low solar

irradiation, the energy output may be too low to pay back the energy required to produce the system. The function of providing low maintenance and reliable performance, e.g. PVsystems in lighthouses, means that energy is transferred from one place to another. Where PV-battery systems are used for providing reliability in power supplies, the energy output is minimal and the system has to be seen as a part of a larger energy supply system, since it does not supply any energy directly. In applications were the energy turnover is supposed to make a significant contribution to total energy supply the energy return factor needs to be higher than one.

Assuming that the world's PV systems have to produce their own energy for manufacture, Lysen and Daey Ouwens [56] estimated that the first world-wide net kWh on an annual basis was produced in 2002 and on a cumulative energy basis this will occur in 2007. To realise that PV and PV-battery systems will contribute to a renewable energy system, it is important to focus on improving the energy efficiency of all components of the energy system. Calculation of the energy return factor and overall battery efficiency can be used to monitor the development of energy technologies and identify areas for improvement.

The energy payback time for wind turbines has been determined to be 2-3 months for an average modern turbine at an average site [57]. With a service life of 25 years, the energy return factor is 150. The energy payback time is shorter for wind turbines than for PV since the conversion efficiency of wind is higher than for solar irradiation and the energy requirements for production are lower. However, comparisons between different generating technologies can be difficult since they may have different energy generation potential. Since the solar energy flow is greater than wind energy, there is a potential to construct more PV systems, which reduce the importance of short energy payback time. Another aspect that makes comparisons difficult is that technologies with the same energy return factor can have different environmental impact depending on if energy for their production originates from fossil or renewable sources. Regardless of the origin of energy, the energy return factor provides a measure of efficiency, which can be used for benchmarking in the development of different PV and battery technologies.

6.3. Future research

Data on energy requirements for production of batteries originate from different sources, which make comparisons unreliable because the system boundaries may be inconsistent. Further work is therefore needed to improve the data quality on material and energy requirements of batteries. For the redox flow batteries, energy requirements need to be expressed for power and storage capacity, respectively, in order to enable evaluation of the potential benefits of independent sizing of power and capacity of redox flow batteries. The influence of temperature on battery service life has to be further analysed. The effects of different battery charge-discharge efficiencies on the battery temperature and cooling requirements have to be further investigated. The energy model of this study can be further developed with functions for assessing the influence of the rate of battery self-discharge as well as different DOD due to different sizing of the battery capacity. The model can be extended to include assessment of resource use, emissions and potential environmental impact of different technologies. Flywheels, fuel cells with hydrogen storage in PV systems may be evaluated and compared with batteries.

7. Conclusions

The energy return factor and the overall battery efficiency were estimated for eight different battery technologies used in a stand-alone PV-battery system. With a battery energy storage capacity three times higher than the daily energy output, the energy return factor for the PV-battery system ranges from 0.64 to 12 for the different cases. This means that 8.1%-156% of the energy output is required to produce the PV-battery system. If the value of the energy return factor is less than one, the indirect energy used to produce and replace the device is larger than the direct energy output. In this case the device works as a non-rechargeable battery moving energy from one place to another.

In the reference case, production and transport of batteries contribute 25-70% to indirect gross energy requirement. The contribution of production and transport of the PV array is 25-66% depending on the battery technology used. For a PV–battery system with a service life of 30 years, the energy payback time is 2.4-13 years depending on the battery technology. The energy payback time is 1.6-3.0 years for the PV array and 0.55-10 years for the battery, showing the energy related significance of batteries and the large variation between different technologies.

The contribution of all transport to the indirect gross energy requirements is low (1.0-9.2%) with 3 000 km transportation by heavy truck. When transportation is done by plane, transport may contribute up to 74% of the gross energy requirements for batteries with low energy density (<30 Wh/kg) and short cycle life (<3 000 cycles at 33% DOD).

The direct energy efficiency of the charger, the battery and the inverter is 0.50-0.85. When considering the overall battery efficiency, including the production and transport of charger, battery and inverter, the values are 0.23 and 0.82 for different battery technologies and operating conditions. The overall battery efficiency can be useful for identification of important parameters for improvement of the efficiency of the PV-battery systems from a life cycle perspective.

The magnitudes of the influence of different operating conditions on the energy return factor were found to be as follows: (1) active cooling with air conditioning to 25° C compared with 40°C (+14 to +79%), (2) float life instead of cycle life (-14 to +52%), (3) 40°C instead of 25°C (-0 to -47%) (4) virgin instead of recycled materials (-7 to -35%), and (5) air instead of road transport (-12 to -70%). When comparing battery technologies, specific data must be used corresponding to the particular application since the charge-discharge efficiency and service life of batteries depend on operating conditions.

The battery charge-discharge efficiency has high influence on the energy return factor for batteries with relatively low energy requirements for production and transportation (Li-ion, NaS, VRB, ZnBr, PSB). Service life, gravimetric energy density and battery production are equally important for NiCd, NiMH and PbA batteries. The overall battery efficiency is significantly influenced by the charge-discharge efficiency.

PV-battery systems can be made more energy efficient by matching operating conditions and battery characteristics in a life cycle perspective. The energy efficiency of the PVbattery system can be increased by: (1) optimised charging algorithms, (2) passive temperature regulation to $\sim 20^{\circ}$ C, (3) increased utilisation of the active battery material, (4)

lower material requirements for battery production and, (5) efficient production and transport of PV-battery system components.

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