## Simulations of Evaporation Plants in Kraft Pulp Mills

Including Lignin Extraction and Use of Excess Heat

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Heat and Power Technology
Department of Energy and Environment
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#### Cover illustration:

An industrial evaporation plant (Billerud Gruvön, photo by Metso Power) on the left, a screenshot from OptiVap on the right, and the algorithm for solving the equation system in OptiVap below. The screenshot and the algorithm are a bit distorted, as they may appear after too many hours in front of the computer...

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#### Abstract

In this thesis, evaporation plants at kraft pulp mills are simulated with the purpose of making them more energy efficient. The work is important since energy saving is assumed to be one of the major solutions for handling the world's energy demand in the future. Pulp and paper mills can do much in this respect; they represent almost 50% of the energy consumption in Swedish industry. In kraft pulp mills, the greatest energy demand is usually in the evaporation plant. To simulate the plants, an existing tool was developed and used; important parameters for the simulations are the amount of water evaporated, the number of evaporation effects, and the solids content profile of the evaporation train. The evaporation plants are assumed to be situated in a model mill resembling typical Scandinavian market pulp mills. To assure realistic assumptions, the project was conducted in cooperation with industrial representatives in a national research programme.

Previous research shows that great energy savings can be obtained in evaporation plants by reusing excess heat, provided that excess heat can be made available in the mill. Evaporation plants that reuse excess heat are called process-integrated (PI) plants. The energy surplus resulting from the savings could be exported from the mill to replace fossil fuels. For example, the surplus could be extracted as the energy-rich component lignin.

As an excerpt of the results, 26% of the live steam could be saved in the evaporation plant by employing a 7-effect PI plant (1.0 GJ/ADt of excess heat) instead of a modern 7-effect conventional plant. The additional profit for PI plants was  $0.3-1.5 \in /ADt$  in comparison with conventional plants (for the conditions in Paper 6). With predictably higher energy prices in the future, the profits from energy-saving measures could increase further.

As an example of the results for lignin extraction, an evaporation plant with 190 kg/ADt lignin extraction (LE) requires 12% more live steam than a plant without LE. Should the viscosity of lignin-lean black liquor be as low as recent experiments indicate, the investment cost for a plant with LE may be only 5% higher than that for a plant without LE. An overall conclusion from the cooperative work is that LE may be economically interesting for pulp mills, at least in connection with increased pulp production. However, the results depend greatly on the electricity and lignin prices.

**Keywords:** Energy-efficient evaporation, simulation, heat integration, process integration, excess heat, lignin extraction, energy saving, modelling, kraft pulp mill.

#### List of papers

The thesis is based on the following papers which are appended in Part II.

#### Bibliographic data of the papers

- Paper 1. Erik Axelsson, Marcus R. Olsson, and Thore Berntsson. Heat integration opportunities in average Scandinavian kraft pulp mills: Pinch analyses of model mills. *Nord. Pulp Paper Res.* J., 21(4):466–475, 2006.
- Paper 2. Marcus R. Olsson, Erik Axelsson, and Thore Berntsson. Exporting lignin or power from heat-integrated kraft pulp mills: A technoeconomic comparison using model mills. *Nord. Pulp Paper Res.* J., 21(4):476–484, 2006.
- Paper 3. Erik Axelsson, Marcus R. Olsson, and Thore Berntsson. Increased capacity in kraft pulp mills: Lignin separation and reduced steam demand compared with recovery boiler upgrade. *Nord. Pulp Paper Res. J.*, 21(4):485–492, 2006.
- Paper 4. Marcus R. Olsson and Thore Berntsson. A tool for simulating energy-efficient evaporation using excess heat in kraft pulp mills. In 2007 Engineering, Pulping & Environmental Conference, Jacksonville, FL, USA, 2007. TAPPI.
- Paper 5. Marcus R. Olsson and Thore Berntsson. Extracting lignin from black liquor: Consequences for the evaporation plant. *Submitted to TAPPI Journal*, 2009.
- Paper 6. Marcus R. Olsson and Thore Berntsson. Comparing conventional evaporation plants with plants using excess heat: A simulation study. Submitted to BioResources Online Journal, 2009.

The following publication, co-authored by the author of this thesis, is relevant to this work but not included in the thesis.

Erik Axelsson, Marcus R. Olsson, and Thore Berntsson. Opportunities for process-integrated evaporation in a hardwood pulp mill and comparison with a softwood model mill study. *Applied Thermal Engineering*, 28(16):2100–2107, 2008.

#### Statement of the author's contribution

Papers 1–3 are written by Olsson and Axelsson, the work being equally divided. Olsson conducted the simulations of the evaporation plants, whereas Axelsson was responsible for the pinch analyses of the energy system. The simulations required iterative work to set up the condensate treatment and the amount of lignin to be extracted. Furthermore, the simulation tool was thoroughly developed by Olsson during the work. Olsson is the main author of Papers 4–6. Professor Thore Berntsson supervised the work in all papers.

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# Review of the work

## Introduction

If history repeats itself, and the unexpected always happens, how incapable must Man be of learning from experience. /George Bernard Shaw (1856 - 1950)

uch has been done to reduce the energy consumption and environmental emissions in the pulp and paper industry since the 1970s and 1980s. However, while reading papers within this research field from those decades, the resemblance to the papers of today is striking. Typical phrases such as "continuously rising energy costs" and "increasing need for energy savings" are used still today. Most people believe that energy prices will continue rising and that a sustainable world must reach lower emission levels. Additionally, many back-casting models for the energy system of the world in the year 2100 presume that energy efficiency measures are vital to cover the energy demand in 2100. A presumption in some of these models is that only 50% of the energy demand can be covered by the supply in 2100, which means that energy efficiency measures must "cover" the remaining 50% of the demand. These problems we are facing are reasons why this thesis work is important. The objective of the present work is to present means to reduce the energy consumption of pulp and paper mills; the main contribution is examples of how to enhance the performance of evaporation plants.

#### 1.1 Background

Pulp and paper mills represent almost 50% of the energy consumption in Swedish industry (The Swedish Energy Agency, 2007). Since high energy consumption also means high costs, the mills can often achieve economic savings by implementing energy-saving measures. To cope with the competition from other mills, the energy consumption per tonne of pulp produced has to be reduced over time. In articles written around the year 1980, it

is clear that this is not a unique situation for today (von Matérn, 1981; Olauson, 1979).

The pulp and paper industry is, as are all industries, trying to find new methods to remain competitive. The "hot topic" of today is to redefine the way we look upon a pulp mill, i.e. wood enters the mill and the mill converts it into pulp. Instead, the pulp mill should be seen as a biorefinery: Wood enters the mill and the mill converts it to valuable products, such as pulp, ethanol, electricity, district heating, tall oil or lignin. In the present thesis, the biorefinery concept of extracting lignin from black liquor is compared with the conventional approach of increasing the electricity production.

To obtain an overview of the energy system in a mill, pinch analysis (Linnhoff, 1993) can be used. From pinch analysis, the theoretical heating and cooling demand of the mill is determined<sup>1</sup>. Parts of the cooling demand can be found at temperatures exceeding 80°C; this excess heat can be used in the evaporation plant, for example. Evaporation plants using excess heat are heat integrated, or process integrated, with the rest of the mill. In the present work, they are called process-integrated (PI) evaporation plants.

The evaporation plant is usually the process unit with the greatest steam demand in a kraft pulp mill. In many cases, the evaporation represents 30–35% of the entire mill's demand for process steam. By process-integrating an evaporation plant with other parts of the mill, energy savings of 50% or more can be obtained (in the evaporation plant) in comparison with well-performing evaporation plants in operation today (Algehed, 2002). Should process-integrated evaporation be implemented in all Swedish pulp mills and lead to the proposed savings above, a rough estimate is that 6 TWh<sup>2</sup> of steam could be saved annually. This is roughly equivalent to the heating of 300,000 small houses in Sweden.

#### 1.2 Objectives

The main objective of the work was to simulate process-integrated evaporation plants to compare their steam demand and profitability with those of conventional plants. To simulate the evaporation plants, an existing tool was used. During this Ph.D. project, the tool was developed further to make it more user-friendly and more versatile. Additionally, new modules were added to the tool, e.g. lignin extraction and condensate treatment. The improved simulation tool was used to evaluate the consequences for the evaporation plant of extracting lignin and of lowering the temperature of the surface condenser.

<sup>&</sup>lt;sup>1</sup> Pinch analysis is not included in the present thesis. In Papers 1, 2 and 3, the pinch analyses were conducted by one of the co-authors, Axelsson.

 $<sup>^2</sup>$  Average steam demand for evaporation: 5.1 GJ/ADt (FRAM, 2005). Swedish production of kraft pulp: 7.8 million ADt/year (Wiberg, 2007). Total steam demand:  $5.1 \cdot 7.8 \cdot 10^6$  GJ  $\approx 40$  PJ  $\approx 11$  TWh.

#### 1.3 Thesis outline

The thesis is divided into two parts.

Part I: Review of the work. This part is an extended survey of the work in the papers appended in Part II. Since the content of the papers is often limited in number of pages or words, the authors are forced to write concisely. In this review, the papers can be more thoroughly elaborated and previously cut sections are added for clarification.

Part II: Papers appended. The papers already published are included in the versions published, whereas the papers to be published are included in the version sent to the journal in question.

In Part I, Chapters 2 and 3 give an overview of the research field: The main concepts and problem statements are described in Chapter 2 and previously done research is referred to in Chapter 3. The objectives of the Ph.D. project are then specified in Chapter 4, including a description of the collaboration with industry and other researchers. In Chapter 5, the technical and economic conditions for the simulations are given. Some of these conditions are part of the simulation tool that is described in Section 5.2. The equation system of the simulation tool is described in more detail in the appendix. In Chapter 6, the results in the papers are summarised and discussed. In the final chapters, 7 and 8, the conclusions drawn from the work and suggestions for further work are given.

The scope of the papers in **Part II** spans from the large scale of the pulp mill to the relatively small scale of the evaporation plant, see Figure 1.1. Papers 1–3 are connected closely, since they concern the same model pulp mill developed within the FRAM programme. In these studies, the economic consequences for the complete mill are evaluated. In Paper 1, the pinch analyses conducted show the opportunities for heat integration in the mill. The amount of steam surplus and the investment costs necessary conclude the analyses. In Paper 2, the concluding results from Paper 1 are reused to compare the profitability of (a) utilising the surplus steam for additional electricity production in condensing turbines, and (b) extracting lignin from the black liquor. In Paper 3, the recovery boiler is assumed to be a bottleneck for increased pulp production. In this paper, the results from Paper 1 are scaled for a 25% increase of pulp production and two debottlenecking methods for the recovery boiler are compared: The unconventional method of extracting lignin from the black liquor and the conventional method of rebuilding the boiler.

In Paper 4, the simulation tool used for the evaporation simulations is described and illustrated with an example. In the example, the three objective functions of the tool are compared.

In Paper 5, the consequences for the evaporation plant of extracting lignin are investigated in more detail. The consequences regarding the necessary heat exchanger surface and the steam demand are included.

Paper 6 investigates further the mill consequences of process-integrated (PI) evaporation plants. The PI plants are compared with conventional plants in terms of steam demand and profitability. Additionally, the consequences of (a) lowering the surface condenser, and (b) extracting lignin are compared for PI plants and conventional plants.

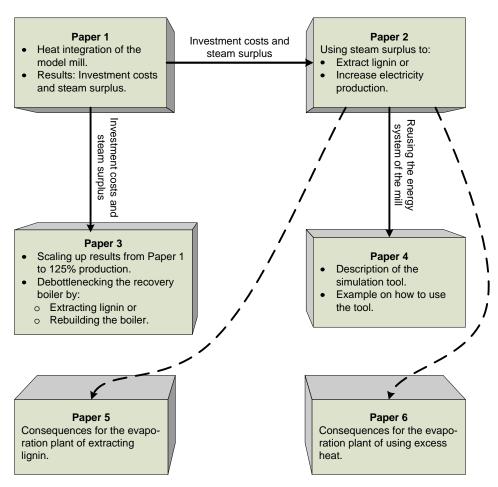


FIGURE 1.1: Overview of the papers and their connections to each other. The dashed lines indicate weaker connections than the continuous lines.

## Main concepts

his chapter gives an overview of the subject of the thesis by explaining the main concepts. First, the pulping process and the purpose of an evaporation plant are described schematically. Next, the more specific concepts of the thesis are introduced: process-integrated evaporation, lower surface condenser temperature and lignin extraction.

#### 2.1 Overview of the pulping process

In the pulping process, wood is converted to pulp that can be used to produce paper. Dry wood consists primarily of cellulose (41–46%), hemicellulose (25–32%) and lignin (26–31%).<sup>1</sup> The cellulose fibres are converted to pulp, whereas most of the hemicellulose and lignin is dissolved and separated. To separate the fibres from the other constituents, white liquor<sup>2</sup> and steam are added with the wood chips to the digester, see Figure 2.1. The pulp is then washed and screened before it is bleached. After drying, it is ready for transport to a paper mill. Alternatively, the paper mill can be integrated with the pulp mill, which makes drying before the paper production unnecessary.

For economic and environmental reasons, the chemicals in the white liquor are recovered in the recovery cycle which generally consists of an evaporation plant, a recovery boiler and a white liquor preparation plant, see Figure 2.1. The slurry from the digester is called black liquor and consists of chemicals, lignin, hemicellulose and water. Before the evaporation plant, the solids content of the black liquor is low, typically 15–18%; this black liquor is called weak liquor. The heating value of the black liquor must be increased to combust the liquor and thereby recover the chemicals. To raise the heating value of the black liquor, water is evaporated in the evaporation plant, resulting in a heavy liquor with typically 70–82% solids. To recover the chemicals in the black liquor, it is combusted in the recovery boiler. In

<sup>&</sup>lt;sup>1</sup> The percentages are valid for softwoods (Gullichsen and Fogelholm, 1999).

 $<sup>^2</sup>$  White liquor is an aqueous solution with chemicals, the most important components being NaOH and Na<sub>2</sub>S.

the recovery boiler, steam is produced to be used in other processes in the mill and also to produce electricity. The chemicals are found in the smelt in the bottom of the recovery boiler. The smelt is dissolved in weak white liquor to produce green liquor, which is then reconverted to white liquor in the white liquor preparation plant.

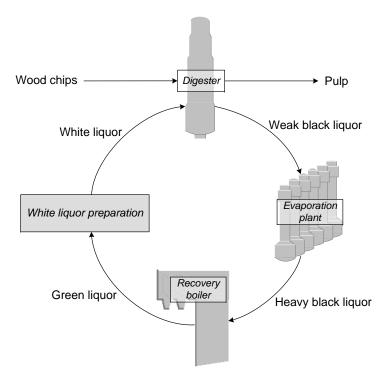


FIGURE 2.1: Overview of the pulping process and the chemical recovery cycle of a pulp mill. The pulping process in the figure is simplified, for example, the digester includes washing of the pulp.

#### 2.2 The evaporation plant

As is described above, the evaporation plant is a part of the chemical recovery cycle of a pulp mill. The purpose of the evaporation is to separate water from the weak black liquor to raise its heating value before it is combusted in the recovery boiler. The water is normally evaporated by indirect heating with steam. Low-pressure (LP) steam is usually used up to 75% solids content, whereas medium pressure (MP) steam is used to concentrate the liquor above 75% solids (Olausson, 2008).

To lower the steam demand of the evaporation plant, the secondary steam is re-used in the subsequent evaporation effects, see Figure 2.2. The effects in the plant are numbered according to the steam flow. The black liquor flow can be counter-current or co-current to the steam flow, or a mix of these two. Each evaporation effect can consist of several physically separated evaporation bodies. These bodies are usually in parallel on the steam side, i.e. driven by the same steam temperature, but in series on the liquor side.

In Figure 2.2, it can also be seen that the weak liquor is mixed with intermediate liquor in a mixing tank. This is done to avoid foaming in the evaporation effects and is needed especially in pulp mills based on softwood; hardwood black liquors do not contain as much soap (Gullichsen and Fogelholm, 1999; Olausson, 2008). The target for the mixed liquor is normally 20% solids.

In the surface condenser, the secondary steam from the last effect is condensed on one side, whereas cold water is heated to produce warm water on the other side. The heated warm water is used elsewhere in the mill, which means that the water is a part of the mill's energy balance; if the temperature of the water is lowered, the heat must be covered in another way. In modern pulp mills, the steam is normally set to condense at 55°C, but there are mills in which the steam condenses at 45°C. In this Ph.D. project, the consequences of lowering the surface condenser temperature to 40°C are investigated.

The physical appearance of black liquor at solids contents below 40% (at room temperature) is similar to dirty, black water. When the solids content is raised above 55%, the liquor thickens rapidly. At 80% solids and room temperature, the black liquor may be compared to black rubber. Nevertheless, for temperatures above 125°C, it is possible to pump the black liquor even at these high solids contents.

There are different kinds of evaporators. In pulp mills, there are often old rising-film evaporators mixed with newer evaporators of the falling-film type. The predominant evaporator in newly built plants is of the falling-film type, since they have higher heat transfer coefficients (on the evaporating side) and are capable of higher solids contents (Gullichsen and Fogelholm, 1999).

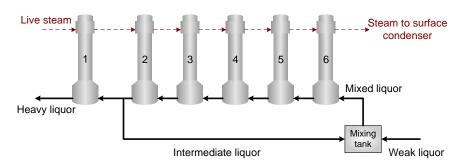


FIGURE 2.2: A counter-current evaporation plant with six evaporation effects. The weak liquor is mixed with intermediate liquor to form a mixed liquor with raised solids content.

The evaporation bodies with the highest solids content, the concentrators, can also be of the forced circulation type. The heat transfer surfaces in the evaporators can be tubes or plates (lamellas).

#### 2.3 The concept of process-integrated evaporation

In this work, process-integrated (PI) evaporation means that the evaporation plant uses excess heat from the rest of the mill to reduce the need for live steam (Algehed, 2002; Cripps et al., 1996; Smith, 1995); see Figures 2.3 and 2.4.

The excess heat can be supplied as steam to the evaporation effect where the driving steam is of the same or of a lower temperature. On one hand, it is the most energy-efficient to adjust the temperature profile in the evaporation train so that the excess heat is re-used in as many effects as possible. To achieve this, the  $\Delta$ Ts of the effects below the temperature of the excess heat should be as low as is technically possible. Consequently, the other evaporation effects will have  $\Delta$ Ts as high as is technically possible. On the other hand, to promote a low requirement for the total heat transfer area, the  $\Delta$ T should be nearly the same for the evaporation effects. Depending on the economic value of the live steam and the investment costs necessary, there is a trade-off for the optimal  $\Delta$ T of the evaporation effects. Although this trade-off is important in both conventional and process-integrated evaporation plants, it is especially relevant in process-integrated plants. The reason for this is that the demand for live steam changes stepwise when changing the temperature profile in process-integrated plants.<sup>3</sup>

An alternative way of using the excess heat is to preheat the black liquor between the evaporation effects. An advantage of this approach can be that the excess steam does not need to be reformed when foul; the excess heat does not even have to be in the form of steam. Another advantage is that the excess heat can be supplied at a lower temperature and still have the same steam saving potential. A drawback to this approach may be the difficulty in managing the fouling of black liquor in a heat exchanger (Redeborn, 2008). This alternative way of using the excess heat is not, however, included in the current version of the simulation tool, since it is not investigated in this study. This opportunity should be studied in further work, see Chapter 8.

The amount of excess heat available at the mill can be assessed by using pinch analysis. Pinch analysis is not included in the current thesis, but was done by Axelsson (2008) in Papers 1–3. Pinch analysis is a method to investigate the minimum heating and cooling load of the mill (Linnhoff, 1993) in order to take energy-efficiency measures. The pinch analysis divides

<sup>&</sup>lt;sup>3</sup> The temperature profile determines where the excess heat can enter the plant. If the temperature profile is changed so that excess heat is used in one more or one less evaporation effect, the steam demand changes stepwise.

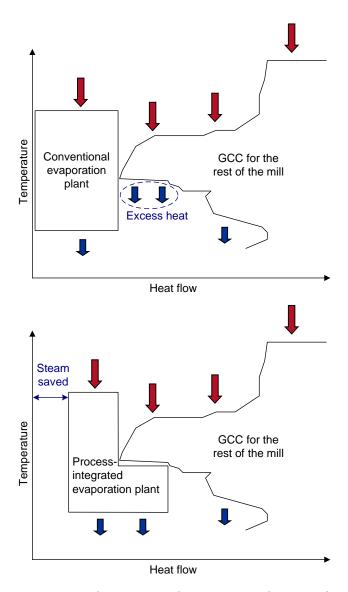


FIGURE 2.3: Top: A conventional evaporation plant together with the Grand Composite Curve (GCC) for the rest of the mill. Bottom: A process-integrated (PI) evaporation plant together with the same GCC; the resulting steam savings by using PI evaporation are shown to the left.

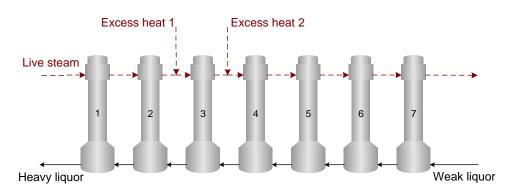


FIGURE 2.4: A process-integrated evaporation plant with seven evaporation effects and two levels of excess heat.

the energy system into two parts: one part (above the pinch temperature) with a heat deficit and one part (below the pinch) with a heat surplus. The analysis shows whether pinch violations exist in the mill, i.e. if the pinch rules below are violated:

- 1. No cold utility should be used above the pinch temperature,
- 2. No hot utility should be used below the pinch temperature, and
- 3. Heat should not be transferred from a stream above the pinch temperature to a stream below the pinch temperature.

Should pinch violations exist in the mill, solving them may affect the amount of excess heat available in the rest of the mill. If so, there is a trade-off between solving pinch violations and extracting excess heat. In this situation, the profitability of the PI evaporation plants depends on which pinch violations are solved; this means that the most profitable solution must be evaluated from case to case.

A common source of excess heat is black liquor flash steam in the digester plant, which arises when the liquor is cooled in flash tanks. The simplest arrangement is to flash the liquor in stages until it reaches its feed temperature in the evaporation plant. The flash steam is then used elsewhere in the mill. Another way of using the heat in the black liquor is to reuse the flash steam in the subsequent flashing stages, forming a pre-evaporation train. However, this generally means that the flash steam is reused in fewer stages than if reused in the main evaporation train as is done in this work.

#### 2.4 Lower temperature of the surface condenser

A lower temperature of the surface condenser raises the total available temperature difference in an evaporation plant. Greater temperature differences

in the evaporation effects mean stronger driving forces, hence less need to invest in heat transfer surface.

Unfortunately, there are also drawbacks to lowering the temperature of the surface condenser. First, a lower surface condenser temperature requires higher investment costs for piping, since the volume flows of the steam are higher in the cold end of the plant. Also, the surface condenser itself has to be larger due to the higher volumes. Second, the temperature of the warm water produced in the surface condenser is lower. This has to be taken into account when designing a new hot and warm water system for a mill. In our studies (Papers 1–3; Axelsson 2008, Paper VI), the amount of warm water was still sufficient, even after lowering the surface condenser temperature.

#### 2.5 Lignin extraction

Modern Scandinavian pulp mills usually have options to generate an energy surplus by making energy savings. Some of them already have an energy surplus. This surplus enables lignin to be extracted from the black liquor, since the energy content of the lignin is not needed in the mill. If the recovery boiler is a bottleneck in the process, lignin extraction can be used to debottleneck the boiler in order to enable increased pulp production. Another purpose of lignin extraction (however not included in this thesis) could be to enable lower investment in a new recovery boiler when replacing an old one. The lignin extracted can be sold as a biofuel or a chemical feedstock, or be used internally as a replacement for oil in the lime kiln.

When lignin is extracted in the evaporation plant, the evaporation capacity of the plant has to be raised, since filtrates from the lignin separation plant must be evaporated when recirculated to the evaporation plant; see Figure 2.5. This is described in more detail in Chapter 5 and Paper 5.

Moreover, the physical properties of the black liquor in the evaporators alter when lignin-lean filtrates are recirculated and mixed with ordinary black liquor. Previous research (Moosavifar et al., 2006; Wennberg, 1990) shows that the boiling point elevation (BPE) is affected only marginally, and that the viscosity of lignin-lean black liquors (LLBL) is lower than that of ordinary black liquors with the same temperature and solids content. The consequences of a lower viscosity are assessed in this thesis project.

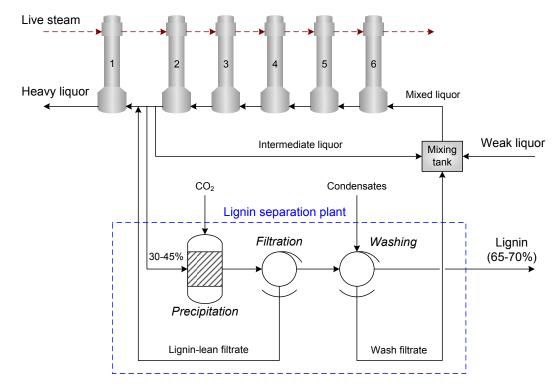


FIGURE 2.5: An overview of the lignin separation plant and its integration with the evaporation plant. The two filtrates from the lignin separation plant should probably be separated as in the figure, but other recirculation alternatives are also investigated in the papers.

## Previous work

Previous work by other researchers in this field is covered in this chapter. The thesis work concerns one of the most energy-intensive parts of a pulp mill, the evaporation plant, on which much work is being done at pulp mills around the world. Unfortunately, this work is seldom published openly in scientific journals for competitive reasons. This means that most information is found in trade journals as general descriptions or at conferences. The references in the following sections are mostly from scientific journals or conferences, since these sources can be considered relatively unbiased.

#### 3.1 Process-integrated evaporation plants

The concept of process-integrating an evaporation plant has been described generally by Kemp (1986, 2007) and Smith (1995). These studies concentrate on fully integrated evaporation plants, above and below the pinch temperature. Partially integrated plants (studied in the present work) are not considered by them, since their discussion is mainly focused on the most energy-saving integration. An overview of what is meant by fully and partially integrated evaporation plants is shown in Figure 3.1.

A fully integrated evaporation plant is seldom possible in the pulp and paper industry. Furthermore, in studies by Algehed (2002), Bengtsson (2004) and Wising (2003) it was shown that maximally integrated evaporation plants (a full integration was not possible) were less profitable than partially integrated plants.

Practical examples of PI evaporation in the pulp and paper industry are not common in the literature. In one of the few papers found, Cripps et al. (1996) describe how process integration was used in a real mill to find excess heat and to use it in a pre-evaporation train. This pre-evaporation train consists of 5 evaporation effects and the solids content is raised from 18% to 28%. The steam in Effect 1 condenses at 140°C, and the secondary vapour from Effect 5 condenses at 80°C. Cripps et al. also describe another mill in which excess heat was already being used in a similar fashion in 1978. That

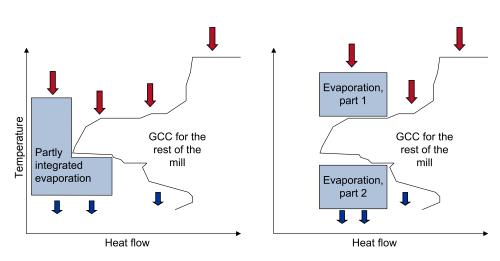


FIGURE 3.1: A partially integrated evaporation plant compared with a fully integrated plant.

particular evaporation plant was reported to have worked without problems since the start-up. Usage of excess heat in pre-evaporation trains is also described in other papers: Hadwaco Ltd. Oy (2000), Kayser et al. (1998), Olauson (1979) and von Matérn (1981). In these papers, the main source of excess heat is either digester flash steam in chemical pulp mills or steam from grinding in mechanical pulp mills. This kind of process-integrated pre-evaporation offers lower investment costs than process-integrating a new complete plant, but does not usually achieve as much steam savings<sup>1</sup>. In a recent case study at a real mill (Axelsson et al., 2008), the steam condensing in the surface condenser of a pre-evaporation train could be used as excess heat in a PI evaporation train. One of the conclusions in this paper is that producing hot water in the surface condenser of a pre-evaporation train can be viewed as a destruction of valuable excess heat (provided that the hot water can be produced with other heat sources).

Several studies have been made by our research group in which PI evaporation is compared with conventional evaporation. For example, Algehed et al. (2002) showed energy savings of 55% with a 7-effect PI evaporation plant<sup>2</sup> in comparison with a model plant of the best available technology (6 effects).

<sup>&</sup>lt;sup>1</sup> The steam savings depend on how many evaporation effects the steam is reused in. Pre-evaporation plants usually have fewer evaporation effects in series than main evaporation trains.

<sup>&</sup>lt;sup>2</sup> Including a low temperature of the surface condenser.

#### 3.2 Simulations of evaporation plants

Simulations of evaporation plants in pulp mills are probably primarily done by pulp mill personnel or by consultants. Usually, the results are not published in public journals since the mills want to avoid revealing too much to competitors. Furthermore, the gains of publishing the results are often limited, since these models are made specifically for the process studied in a particular mill. However, several papers have been published concerning simulations of evaporation plants in other industries, such as the desalination industry. These papers are not relevant to the present thesis work, since the conditions for these plants are different from the plants in the pulp and paper industry.

Nevertheless, some simulations of evaporation plants at pulp mills have been reported in the literature. Stefanov and Hoo (2004) simulate a 5-effect evaporation plant with plates as heat transfer surfaces. The liquor flow is counter-current to the steam flow with the special arrangement that the weak liquor is fed to both Effects 4 and 5. Moreover, dynamic changes can be investigated with this model. Another dynamic model is presented by Costa and Lima (2003), who combine heat and mass balances with an empirical (neural networks) approach. They simulate a particular 6-effect plant with both falling-film, rising-film and forced circulation evaporators.

Ray and Sharma (2004), Bhargava et al. (2008), Gidner and Jernqvist (1997) and Bremford and Müller-Steinhagen (1996) present steady-state models of evaporation plants. Ray and Sharma simulate a 6-effect evaporation plant with mixed liquor flow according to the sequence Effect  $5 \rightarrow 6 \rightarrow 4 \rightarrow 3 \rightarrow 2 \rightarrow 1$ . The consequences of flashing only condensates and flashing both condensates and product liquor are investigated for rising-film evaporators. Bhargava et al. model a 7-effect evaporation plant with counter-current liquor flow. The raw material is straw and the solids content of the heavy liquor is relatively low: 53%. Gidner and Jernqvist present a flow sheet program developed for both design and evaluation of any type of evaporation plants. The program handles falling-film, rising-film and forced circulation evaporators. Bremford and Müller-Steinhagen simulate rising-film evaporators at a particular pulp mill. Two plants are simulated: a 4-effect plant and a 5-effect plant, both with counter-current liquor flow.

#### 3.3 Lignin extraction from black liquor

Lignin can be extracted from black liquor by ultrafiltration (Wallberg, 2005), acid precipitation (Öhman, 2006), a combination of ultrafiltration and precipitation, or a combination of electrolysis and precipitation (Davy et al., 1998; Loutfi et al., 1991; Uloth and Wearing, 1988). Uloth and Wearing found in laboratory tests that acid precipitation yielded lower costs and

better separation abilities than ultrafiltration. Davy et al. and Loutfi et al. investigated lignin separation in combination with increased pulp production. The study of ultrafiltration by Wallberg (2005), was concentrated on the separation of lignin in the digester, whereas the work regarding acid precipitation by Öhman (2006) investigates separation of lignin in the evaporation plant. In the present work, lignin is assumed to be separated in the evaporation plant using the method proposed by Öhman, who also took part in the FRAM research programme.

Wising et al. (2006) have conducted a study of lignin separation in a greenfield mill. Some of their conclusions were that the profitability depends greatly on energy prices and the CO<sub>2</sub> cost for the precipitation. One difference between the study of Wising et al. and the present work is that they used a greenfield model mill with state-of-the-art technology while the present work uses models of typical Scandinavian mills which are retrofitted. Furthermore, they investigated only a low electricity price (US\$25/MWh) and did not include the effect of policy instruments. Additionally, Wising et al. used preliminary technical data for the lignin separation, while the present work uses data based on a pilot plant in a real mill that has been in operation during the FRAM programme (FRAM, 2005).

#### 3.4 The gap in knowledge

In the above cited publications, the following gaps are covered in the present thesis:

- Simulations of evaporation plants for black liquor including the use of excess heat in the main evaporation train;
- Simulations of evaporation plants for black liquor including separation of lignin;
- Consequences for the evaporation plant of separately using excess heat or lowering the surface condenser temperature; and
- Consequences for the evaporation plant of lignin separation, e.g. the evaporation plant has to be adapted to the new conditions that an integrated lignin separation plant entails; analyses of these new conditions are presented here.

#### Objectives of the current work

he objectives of the thesis are treated in this chapter. The main features of the simulations are described, and the collaboration with industry and other researchers is stated. In applied research such as this, collaboration with industry is crucial to facilitate discussion of practical limitations in the processes studied and to ensure realistic assumptions. This kind of collaborative work is of mutual benefit, as the industry is interested in current advances in research. Collaboration with other researchers is also valuable in order to be updated in overlapping research.

### 4.1 Investigating more energy-efficient evaporation plants

The objectives of the first three papers are to evaluate the consequences, for an average Scandinavian kraft pulp mill, of extracting lignin or increasing the electricity production. In these studies, the economic consequences for a complete mill are evaluated. In Papers 5 and 6, more details regarding the specific consequences for the evaporation plant are investigated. The objectives of these papers are to examine the consequences for the evaporation plant of separately (1) extracting lignin, (2) using excess heat or (3) lowering the surface condenser temperature. The first three papers assess the combined effects of (1), (2) and (3). In Paper 4, the aims are to present the simulation tool and to show that the tool can apply three objective functions to design evaporation plants.

To conduct the simulations for the new evaporation conditions described in the previous paragraph, the tool had to be developed to suit these new conditions. The tool was made more user-friendly and new options were added, such as:

- Lignin extraction,
- Condensate treatment, and
- Optimisation with a selection of three possible objective functions.

During this Ph.D. project, the simulation tool was used to simulate not only evaporation plants in theoretical model mills but also in a real mill. However, since the evaporation aspects are not a core issue in the paper on the real mill (Axelsson et al., 2008), the paper is not included in this thesis.

### 4.2 Collaborating with industry and other researchers

Since the present work is a part of the FRAM programme, most of the collaborative work was done within the FRAM network, but other specialists were also involved when necessary.

The first three papers were done together with Axelsson (2008), who was responsible for the pinch analysis of the mills' energy systems. Without this collaborative work, the mill consequences of using excess heat, lowering the surface condenser and extracting lignin could not have been assessed (only the consequences for the evaporation plant). The iterative work with Axelsson was especially important in Paper 3, since the outputs from the evaporation simulations were inputs for the energy system calculations and vice versa.

In Papers 5 and 6, the viscosity model for lignin-lean black liquor was adapted to experimental research by Moosavifar (2008). In this way, it could be ensured that the estimations of the heat transfer surfaces are realistic when lignin is extracted from the black liquor. Since the work by Moosavifar was in progress in parallel with the present thesis work, much discussion took place between these Ph.D. projects.

The design of the lignin separation plant was discussed with Öhman (2008) and Delin (2008). Important parameters in this thesis are the solids content and temperature of liquor going to precipitation.

To ensure realistic assumptions regarding the placement of heat transfer area and investment costs, discussions were held with an evaporation expert (Redeborn, 2008) for Papers 5 and 6. In these papers, the final investment costs for the evaporation plants were estimated by Redeborn.

During the development of the simulation tool, practical assumptions were made after discussions with engineers at Metso Power (previously Kvaerner Power) and ÅF-Process.

### Conditions for the simulations

he technical conditions for the simulations concern how the evaporation train is modelled and how the physical properties of steam and black liquor are modelled. Important physical properties are the heat transfer coefficients, the viscosity of black liquor and the boiling point elevation. Since the simulation tool is a central part of this thesis, it is placed in its own section. The economic conditions comprise the estimations of investment costs for evaporation plants and the selling price of lignin and electricity.

#### 5.1 Technical conditions

#### 5.1.1 Key data for the evaporation plants simulated

The evaporation trains are simulated with counter-current liquor flow and 6–8 evaporation effects. Nine effects were simulated in a screening study indicating that too high investment costs were required. The evaporators are of the falling-film type and the heat transfer surfaces are tube bundles.

All of the simulated evaporation plants are assumed to be installed in a typical, Scandinavian, market pulp mill producing bleached kraft pulp from softwood. This particular mill has been developed as a computer model during the FRAM research programme and is called the FRAM Typical Mill; the mill is described more thoroughly in Paper 1 and FRAM (2005). The pulp production capacity of the mill is 327,000 ADt/year (1000 ADt/day).

Since the development of the tool is a continuous process, the simulations in the papers were conducted with the latest version of the tool at the time they were written (see Section 5.2.7 for more details regarding the development process). This means that it is difficult to compare specific details for the evaporation plants between the papers. The conclusions in the papers, however, should still be valid, since all of the evaporation plant simulations in a particular paper were simulated with the same version of the tool.

In Table 5.1, the key conditions for the evaporation simulations are summarised. The evaporation capacity (tonnes of water evaporated) is shown for the plants with and without lignin extraction. The higher values for the plants with lignin extraction depend on the recirculated filtrates that also have to be evaporated when extracting lignin; see Section 5.1.2. In Paper 6, two amounts of excess heat are considered and the temperature level of the excess heat is raised from 100°C to 105°C to facilitate the simulation work. The amount of lignin extracted depends on the amount of steam savings that can be achieved with the evaporation plants. With more evaporation effects, the steam savings are higher, thus enabling more lignin to be extracted. In Papers 5 and 6, the amount of lignin extracted is kept constant to make it easier to compare the plants.

The solids content of the heavy liquor is set to 80% in the first four papers, since this is a common choice for modern evaporation plants in Scandinavia today. For solids contents above 75%, MP steam has to be used in the concentrator<sup>1</sup> according to Scandinavian practice (Olausson, 2008). When more MP steam is used, the electricity production is decreased, which has to be taken into account in the profitability analysis for the entire Therefore, a prerequisite for conducting a profitability analysis, in the first four papers, was to include the entire energy system of the mill. The same reasoning in Papers 5 and 6 led to the simplification that only LP steam should be used, and hence that the solids content of the heavy liquor could not be raised above 75\%, since only the evaporation plants are included in these studies. In Paper 5, the consequences of extracting lignin are investigated in a moderately retrofitted evaporation plant, which led to the assumption that the solids content is kept at 73%. In contrast, in Paper 6, the entire plant is assumed to be replaced and it was decided to raise the solids content as much as possible without using MP steam.

The reasoning for the temperature of the surface condenser (SC) was not the same in all six papers. In Papers 1–3, the conventional plants with six evaporation effects were simulated with the SC temperature at 60°C. For more than six effects, the SC temperature was lowered to increase the total available  $\Delta T$  and thereby "make room for" the additional effects. In the simulations with process-integrated plants, the SC temperature was set to 40°C. This was done to investigate the potential of both lowering the SC temperature and using excess heat. In Paper 4, only process-integrated plants were simulated, hence, 40°C was used. In Papers 5 and 6, the reasoning was changed to facilitate comparison between the plants. In these simulations, the temperature of the warm water was set to the same temperature for all of the evaporation plants compared. In Paper 5, the original surface condenser at the mill was reused, which caused the varying SC temperature (see

<sup>&</sup>lt;sup>1</sup> The evaporation effect in which the liquor with the highest solids content is evaporated is often called the concentrator or the super concentrator.

TABLE 5.1: A summary of the conditions for the simulations. For Papers 1–3, only the values for the high water usage mill are shown in order to display comparable numbers. (For the low water usage mill, more excess heat could be found which meant that more lignin could be extracted.)

	Papers 1 & 2	Paper 3	Paper 4	Paper 5	Paper 6
Evap. capacity (tonnes/h)*	363/373–383	454/468-479	$363/\!-$	442/474	357/477
Excess heat $(GJ/ADt)$	1.0 @ 100°C	1.0 @ 100°C	1.0 @ 100°C	_	1.0 @ 105°C, 2.0 @ 105°C
Extracted lignin $(kg/ADt)^{**}$	110-210	110–210	_	190	190
Heavy liquor	80%	80%	80%	73%	75%
Surface cond. <sup>†</sup>	40 – 60°C	40 – 60°C	$40^{\circ}\mathrm{C}$	$60–63^{\circ}\mathrm{C}$	$40–55^{\circ}\mathrm{C}$
Live steam	MP&LP	MP&LP	MP&LP	$\operatorname{LP}$	$\operatorname{LP}$
Pulp prod.	100%	125%	100%	125%	100%/125%
Objective func.	Min steam	Min steam	$\mathrm{All}^{\ddagger}$	Min area	Min area

<sup>\*</sup> The amount of water evaporated, without/with lignin extraction. The range of the capacity for the plants with lignin extraction depends on the different amounts of lignin extracted.

Paper 5 for more details). In Paper 6, the consequences of lowering the SC temperature were compared for conventional and process-integrated plants.

Since the results from the first four papers show that lignin extraction is the most interesting in connection with a production increase, all simulations with lignin extraction in Papers 5 and 6 are considered with an increased pulp production of 25%. Regarding the objective functions, the first three papers aim to investigate the potential of the most energy-efficient evaporation plants, which means that the lowest steam demand is the goal. In Paper 4, all of the objective functions are compared for the process-integrated evaporation plants in Paper 2. In the last two papers, the evaporation plants are designed to be as close to real plants as possible, which in this case means that the  $\Delta$ Ts of the effects should be nearly the same. This is achieved by minimising the required heat transfer area.

#### 5.1.2 Method used to separate lignin from black liquor

All data concerning the lignin separation plant originate from experience obtained within the FRAM programme (Delin, 2008; Wallmo, 2008; Öhman, 2008); more information is available in Paper 5. In the simulations, lignin is assumed to be precipitated from the black liquor in the evaporation plant.

 $<sup>^{**}</sup>$  Dry lignin, 100% solids. The amounts of lignin extracted vary with the steam savings achieved.

<sup>†</sup> Temperature of the condensing steam.

<sup>&</sup>lt;sup>‡</sup> All objective functions: Min steam, Min area and Max profit.

A portion of the liquor is diverted from the evaporation plant at a position where the solids content is 30–45%, see Figure 2.5 (page 14). In Papers 1–3, the liquor is diverted at about 30% and in Papers 5–6 at about 45%. The reason for the difference in solids content is that the optimal solids content of the liquor going to precipitation is not easily determined. In general, the higher the solids content of the black liquor, the less the amount of  $CO_2$  needed and the more  $H_2SO_4$ .

When the black liquor enters the separation plant, the pH of the liquor is lowered by injecting CO<sub>2</sub>. This causes the lignin molecules to agglomerate, thus forming a precipitate. The precipitated lignin is separated and then washed with acidified condensate from the evaporation plant. The final lignin cake has a solids content of 65–70%; the filtrates from the filtration and washing stages are recirculated to the evaporation plant. The two filtrates should probably be returned separately to the evaporation plant as in Figure 2.5, but other recirculation alternatives are also investigated in the papers.

As is mentioned in Section 2.5, lignin extraction can be used to debottleneck the recovery boiler in order to enable increased pulp production. The minimum amount of lignin that must be extracted, for a given increase in pulp production, can be estimated by setting up material and heat balances over the recovery boiler. The balances include the solids content and the heat content of the firing liquor entering the boiler. The key details here are that the temperature in the recovery boiler should be high enough and that the amount of flue gases should not be too high. The maximum amount of lignin that can be exported from the mill is determined by the steam balance of the mill. If steam savings can be made, more lignin can be exported.

A production increase of 25% in the FRAM Typical Mill requires the extraction of at least 190 kg dry lignin/ADt when the solids content of the heavy liquor is 73% (Delin, 2008).<sup>2</sup> This amount of lignin corresponds to 35% of the lignin in the weak liquor, or 77,600 tonnes lignin/year (65% solids). In Papers 5 and 6, it was assumed that this amount of lignin could be extracted from the black liquor. In the first three papers, the solids content of the heavy liquor was instead assumed to be raised to 80%, thus requiring less lignin to be extracted.<sup>3</sup> In these papers, 150 kg dry lignin/ADt must be extracted to allow a production increase of 25%.

<sup>&</sup>lt;sup>2</sup> Given that the solids content of the heavy liquor is kept at 73% and that the amount of flue gases is the limiting factor of the recovery boiler.

<sup>&</sup>lt;sup>3</sup> A higher solids content of the liquor fired in the boiler means that less water is brought to the boiler. This, in turn, means that less water ends up in the flue gas. Hence, a higher solids content gives less flue gas and, thereby, less minimum lignin extraction.

#### 5.2 The simulation tool

#### 5.2.1 General description

The simulation tool (OptiVap), originally made in our research group by Algehed (2002), was developed further by the author of this thesis. The current version of the tool is described in more detail in Paper 4.

OptiVap uses spreadsheets in Excel for steady-state energy and material balances, and functions in Visual Basic for the physical properties of steam and black liquor. Important physical properties include BPE, viscosity and heat transfer coefficients. All of the properties are modelled with equations from the literature. Some practical assumptions were made after discussions with engineers at Metso Power<sup>4</sup> (previously Kvaerner Power) and at ÅF-Process<sup>5</sup> during the development of OptiVap.

The variables available for a simplified version of the tool are shown in Figure 5.1, in which the green circles indicate input data from the user and the blue circles indicate free variables. The equation system of the simulation tool is described in more detail in the appendix (page 67).

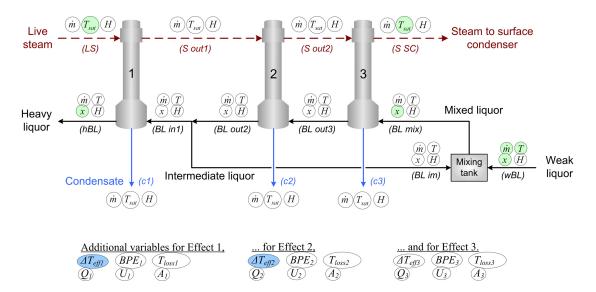


FIGURE 5.1: The variables for a simplified version of the reference sheet in OptiVap. The encircled variables with a green background are inputs from the user of the tool. The blue variables ( $\Delta T_{\rm eff1}$  and  $\Delta T_{\rm eff2}$ ) are the free variables in the equation system.

 $<sup>^4</sup>$  Metso Power is a supplier of chemical recovery systems for the pulp and paper industry.

 $<sup>^5\,\</sup>text{ÅF-Process}$  is a technical consultancy firm.

#### 5.2.2 Objective functions

The user can choose from three objective functions in OptiVap: Minimise the steam demand, Minimise the heat transfer area or Maximise the net annual profit. Maximising the net profit is of course the natural goal for a mill. However, it is not an easy optimisation problem, since the evaporation plants have differing optimal designs for different values of steam, and it is difficult to choose a "correct" value for the steam (see Section 5.3.3). Consequently, the entire energy system of the mill has to be modelled and included in a Max profit simulation. The steam that is saved in the evaporation plant could be used, for example, to increase the electricity production. The increased electricity production would then set a value on the steam saved, which would affect the overall profitability of the evaporation plant; thus it would change the evaporation plant design accordingly. Since minimising the heat transfer area has been shown to result in realistic evaporation plants, this is the objective function that should usually be used.

#### 5.2.3 Condensate treatment

The condensates from the evaporation plant can be reused in other parts of the mill. In OptiVap, an automatic distribution of the condensates is made, based on the cleanliness of the streams. Generally, the cleanest condensates (type A) are diverted to the bleaching plant, and the average foul (type B) to the white liquor preparation. The user decides how much condensate should be sent to the stripper column. In many mills, at least the foulest condensates (type C) are cleaned in the stripper.

The cleanliness of the condensates is based on Lindén (2001); the figures are valid for six evaporation effects. The percentages from Lindén were marginally altered after discussions with Olausson (2008):<sup>6</sup>

- Effects 2–4: 100% A,
- Effects 5–6: 90% B and 10% C, and
- Surface condenser: 80% B and 20% C.

The cleanliness of the condensates above is based on counter-current evaporation; thus it can be seen that the evaporated vapour from the beginning of the evaporation is the foulest steam. After the automatic condensate distribution, the user decides how to obtain the condensate temperatures that the other unit operations of the mill require. This step is done iteratively by mixing and flashing the condensates.

The condensate stripper can be of the stand-alone type or it can be integrated between Effects 1 and 2, 2 and 3, or 3 and 4. The heating media

<sup>&</sup>lt;sup>6</sup> All of these foul condensate percentages can be changed by the user.

of stand-alone strippers is LP steam. Integrated strippers use secondary steam from the previous effect for heating; the exiting steam at the top of the stripper is partly condensed in the subsequent effect. This primary condensation is assumed to be in a separate part of the effect. The heat from the secondary condensation is used for warm water production. This is a normal configuration for integrated strippers in the industry (Redeborn, 2008).

## 5.2.4 Boiling point elevation

A water solution that contains dissolved solids has a higher boiling point than pure water at the same pressure. The difference between the boiling point for the solution and that for pure water is called the boiling point elevation (BPE). In black liquor, the BPE is significant, and it increases with higher solids content. In the evaporation plant, the BPE generally increases from 1°C to 25°C as the black liquor solids content increases from 20 to 80%.

A consequence of BPE is that the total available temperature difference in the evaporation plant decreases by the sum of the BPEs in the evaporation bodies. This means that for a solution with high BPE, the available temperature difference is low, which leads to that the heat transfer areas have to be larger than for a solution with low BPE. In the papers, the BPE was modelled in the same way for all black liquors, see Eq. 1 (Gullichsen and Fogelholm, 1999).

$$BPE_P = (6.173S - 7.48S^{1.5} + 32.747S^2) \cdot (1 + 0.006 [T_{sat,P} - 373.16]) (1)$$

# 5.2.5 Viscosity of black liquor

The viscosity of black liquor is important for the heat transfer coefficients in evaporation effects: A low viscosity causes high heat transfer coefficients, which means a low requirement for heat transfer area. The viscosity tells how viscous a particular fluid is. In general, the viscosity increases with increasing solids content and decreases with increasing temperature. The viscosity of black liquor, which varies significantly from mill to mill, depends on wood species, cooking conditions and thermal treatment.

In the simulations, the viscosity of black liquor was modelled in the same way for all "ordinary" black liquors, see Eq.  $2^7$  (Wennberg, 1989).

<sup>&</sup>lt;sup>7</sup> The equation is valid for solids content up to 85% and temperatures of 25–175°C.

$$\mu = AT^{0.5}e^{\frac{B}{T-C}} \tag{2}$$

where

$$A = 1.024 \cdot 10^{-6}$$

$$B = 624.28 (1 - S) + 6.01D \cdot S$$

$$C = ([138.98 (1 - S)]^{n} + (D \cdot S)^{n})^{\frac{1}{n}}$$

$$D = 306.3$$

$$n = 2.178$$

The viscosity of the lignin-lean black liquor (LLBL) was modelled differently in the papers. In Papers 1–3, the viscosity model above (Eq. 2) was used for all black liquors, including the lignin-lean liquors. In Papers 5 and 6, the viscosity for the lignin-lean liquors was assumed to be lower than that for ordinary black liquor (BL), see Figure 5.2. The figure is based on early results by Moosavifar (2008), which indicated that the viscosity of LLBL was half of that of ordinary black liquor at 70-80% solids. This sets the viscosity ratio to 50% at 70–80% solids (point A in Figure 5.2). Moreover, the viscosities in Moosavifar's experiments for black liquor with lower solids content approached the value for ordinary black liquor, with the ratio 90% for 15–40% solids (point B). Since the share of LLDS is not the same in Moosavifar's experiments as in our simulations<sup>8</sup>, our viscosity ratios were estimated by linear interpolation between his values and the values for ordinary black liquor.

As an additional explanation to the figure, three alternative filtrate recirculations are compared in Paper 5:

- I. Sending both filtrates to the weak liquor mixing tank,
- II. Sending both filtrates to Effect 1, and
- III. Sending the filtrates separately to the mixing tank and Effect 1.

An important comment on Figure 5.2 is that current findings by Moosavifar show that the viscosity of lignin-lean black liquors is probably even lower than estimated in this figure. Moreover, the viscosity for the highest solids contents seems to be more curved than indicated in the figure.

#### 5.2.6 Heat transfer coefficients

The overall heat transfer in the evaporator tubes can be expressed as a function of five heat transfer resistances:

 $<sup>^8</sup>$  Moosavifar's experiments were performed on black liquor with about 60% lignin-lean dry solids (LLDS), whereas our simulations are around 30% and 50% LLDS, depending on where the filtrates are recirculated.

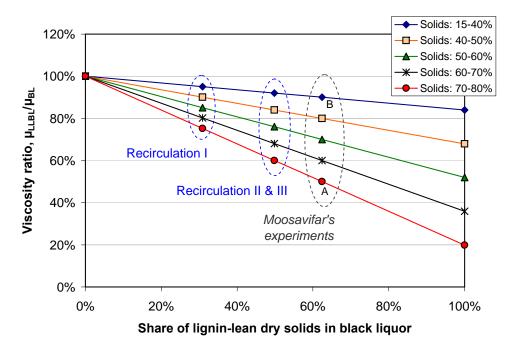


FIGURE 5.2: The viscosity ratio versus the share of lignin-lean dry solids (LLDS) in the black liquor. A viscosity ratio of 10% means that the viscosity of LLBL is only 10% of that of ordinary BL. The left hand side of the figure (0% LLDS) represents ordinary black liquor and the right hand side (100% LLDS) represents extrapolated values for pure filtrate.

- 1. The condensing steam film,
- 2. Fouling on the condensing steam side,
- 3. The tube material,
- 4. Fouling on the black liquor side, and
- 5. The black liquor film.

In this work, fouling has not been taken into account explicitly either on the black liquor side or on the condensing steam side. Instead, the final heat transfer areas have been increased to allow a safety margin for fouling. The heat transfer coefficient of the black liquor film has been modelled with Eq. 3 (Schnabel, 1984).

$$\alpha_{\rm BL} = {\rm Nu} \cdot \lambda_{\rm BL} \left(\frac{\nu^2}{g}\right)^{\frac{1}{3}}$$
 (3)

where

$$\begin{aligned} \mathrm{Nu} &= \sqrt{\mathrm{Nu_{lam}^2 + Nu_{turb}^2}} \\ \mathrm{Nu_{lam}} &= 0.9 \cdot \mathrm{Re^{\frac{1}{3}}} \\ \mathrm{Nu_{turb}} &= 0.00622 \cdot \mathrm{Re^{0.4} Pr^{0.65}} \\ \mathrm{Re} &= \frac{\dot{m}}{\pi \cdot d_o \cdot \mu} \\ \mathrm{Pr} &= \frac{\mu \cdot c_p}{\lambda_{\mathrm{BL}}} \end{aligned}$$

This equation is only valid for Prandtl numbers below 50, but no better model was found in the literature. For the conditions studied here, Pr > 50 means a solids content above 55%, which occurs only in Effect 1. In future versions of the simulation tool, a model by Johansson (2007) could be implemented.

# 5.2.7 The development of the simulation tool

#### Mapping the input variables

It became clear quite early in the project that the existing algorithm for the equation system was not as stable as one could wish, and that input variables were spread out in the calculation sheets and in the Visual Basic code. Therefore, all inputs were mapped and most of them were put in an extended version of the existing input sheet. Here, the benefits of elucidating the input data to the user had to be weighed against the disadvantage of showing too many details. On one hand, the tool is more user-friendly if all inputs are visible. On the other hand, the tool is difficult to use if too many detailed inputs have to be set by the user. Since the tool should be possible to use for making quick estimations, the user should not have to worry about too many details, for example the geometry of the heat transfer surfaces.

#### The condensate treatment

One of the first tasks was to survey the overall energy balance of the evaporation plant. During this work, ideas evolved concerning how to develop the condensate treatment in the plant. The condensates from the plant are sent to other parts of the mill, the main ones being the bleach plant, white liquor preparation, and waste water treatment. These condensate users require the condensates to be of a certain temperature and cleanliness. It was decided

that the user of the tool should set the cleanliness of the condensates from the various evaporation effects as inputs; an algorithm was then developed to automatically divert the appropriate amount to the condensate users. A manual step, however, still remains: To flash the condensates in order to adjust the temperatures to the desired levels. This process is difficult to automate, since flashing the condensates involves digital steps: To flash it or not to flash it. These digital steps cause problems for the equation solver.

#### Reducing the number of spreadsheets

The first version of the simulation tool (Algehed, 2002) consisted of nine calculation sheets: one sheet for each of the effects (1–9). The drawback to this structure is that when changing anything in the calculations, the changes must be made in all of the simulation sheets. This is why one of the major changes to the simulation tool was made: To use one sheet as the basis for generating all of the other calculation sheets. The change meant that six effects were used in a reference sheet, and that an automated procedure enabled the formation of calculation sheets with 6–9 evaporation effects.

The next step was to separate the calculations for the stripper column in their own part of the calculation sheet. This was done to make this part optional in the calculations.

### Adding the option to extract lignin

Since one of the objectives of the project was to simulate the consequences of separating lignin from the black liquor, a new optional module was added. Important questions regarding lignin extraction within the FRAM project were:

- What solids content should the liquor to precipitation have?
- What are the viscosity and BPE of the new mix of lignin-lean black liquor?
- Where should the filtrates from the lignin separation plant be recirculated in the evaporation plant?

These matters are discussed further in Section 5.1.2, where the lignin separation method is described, and in Sections 5.2.4 and 5.2.5 where the BPE and viscosity are discussed, respectively.

#### Adding objective functions

At this stage of the project, the process of conducting a simulation was found to be more and more strenuous and time consuming, because the newly added modules complicated the use of the tool. At that time, the

final evaporation design was determined by solving the equation system and then manually changing the required initial guesses. New simulations were conducted until all reasonable initial guesses had been covered. The solution to this problem was to automate the assignment of the most important initial guesses, and to add the option to optimise the evaporation plant for the conditions set by the user. The first objective function was set to minimise the steam demand of the evaporation plant at a specified evaporation capacity. Later, the options to minimise the heat transfer surface and to maximise the net profit were added to the simulation tool.

The evolution of the simulation tool described in the previous paragraphs is visualised in Figure 5.3. The large, indistinct, grey area shows all evaporation designs possible when considering the constraints given by the user; the most important constraints include:

- The number of evaporation effects in total,
- The solids content of the heavy liquor and the weak liquor,
- The temperature of the live steam and the surface condenser, and
- The low and high limits for the temperature differences in the evaporation effects.

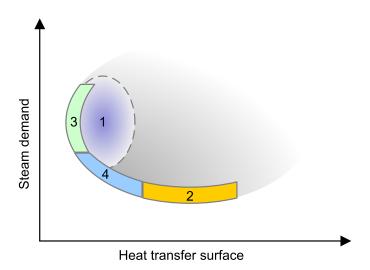


FIGURE 5.3: The gradual development of the simulation tool, in terms of possible evaporation designs resulting from a simulation. The grey area illustrates all possible solutions to the optimisation problem with the given constraints. The numbered coloured shapes show where the resulting evaporation designs are placed in the diagram when using the objective functions available.

The numbers in the coloured shapes in Figure 5.3 are explained below:

- 1. The first simulation procedure by Algebed could, theoretically, lead to any of the possible solutions covered by the grey area. However, after the user changes the initial guesses and tightens the constraints, the design would probably be somewhere within the oval marked 1.
- 2. Using the objective function *Minimise the steam demand*, the possible solutions were narrowed to the yellow segment marked 2.
- 3. If the lowest investment cost is the goal for the optimisation, the objective function *Minimise the heat transfer surface* should be used; this is symbolised by the green shape marked 3.
- 4. The blue shape marked 4 symbolises the objective function *Maximise* the annual profit for a given evaporation plant. To use this objective function, more information on the entire mill is needed than for the other two objective functions.

In this context, it should be noted that the grey area in the figure is generally considered small for an evaporation plant with the constraints stated above. It is commonly assumed within the industry that, if the amount of evaporated water  $(Q_{\text{tot}})$  and the number of effects (n) are given, the live steam demand is  $Q_{\text{live steam}} = Q_{\text{tot}}/n$ . This is almost true in practice, provided the evaporation is divided reasonably equally between the effects and that there are no heat losses. Nevertheless, the difference in steam demand between two plants with differently placed heat transfer surfaces is not negligible, as is shown in this work. The reason why differently placed heat transfer surfaces affect the steam demand is that the temperature profile is changed, which in turn determines how much vapour is flashed into the effects and how much preheating of black liquor is needed between the effects.

#### Changing the algorithm to solve the equation system

To make the convergence of the tool more stable and to reduce the number of equality constraints, the entire equation system was surveyed. This work required that a new algorithm to solve the equation system be produced, which was the most demanding part of this Ph.D. project. The effort required was not only to set up the equation system and to produce the new algorithm, but also to make it function in the Excel spreadsheets. Some of the results from this particular work are presented in the appendix on page 67.

The new algorithm was formulated for three evaporation effects. This algorithm is used in the reference sheet of the tool. Using this reference sheet, it is possible to generate calculation sheets with 3–9 evaporation effects. There is no theoretical upper limit for the number of effects; nine effects were chosen as it was shown to be a reasonable upper limit after

making profitability analyses in the first three papers. With a future higher economic value of steam, together with a lower temperature of the surface condenser, nine or more effects may become profitable alternatives. The completely new equation system and the new algorithm for solving it offer a more reliable optimisation and stable convergence.

#### 5.3 Economic conditions

## 5.3.1 Profitability analyses

Profitability analyses are conducted in Papers 2, 3, 4 and 6. The net annual profit from an investment is calculated with the annuity method, see Eq. 4:

annual profit = revenue 
$$-a \cdot \text{investment cost} (-\text{operating cost})$$
 (4)

where

$$a = \frac{i}{1 - (1+i)^{-n}} \tag{5}$$

The operating costs for the evaporation plant (electricity for pumps, for example) are assumed to be the same in the plants compared, and are therefore excluded in the economic comparisons. The annuity factor (also called the capital recovery factor), a, is set to either 0.1 or 0.2: The annuity factor 0.1 is used for strategic, long-term investments and 0.2 for more short-term investments. For example, 0.1 is used for a complete new plant, whereas 0.2 is used for retrofits of existing plants. In some papers, both 0.1 and 0.2 are shown to let the reader decide which one to choose.

The annuity factors applied have been agreed upon within FRAM after discussions with industrial representatives. With Eq. 5, a = 0.1 is equivalent to an economic lifetime n = 25 years and a discount rate i = 9% (excluding taxes), for example, whereas a = 0.2 is equivalent to 7 years and 9%.

The revenue is calculated from the value of the resulting steam savings. In Papers 2–4, the steam is valued indirectly, according to either the electricity price or the value of lignin. In Paper 6, the steam is valued directly. Since the value of steam can be calculated in several ways, the profit is plotted versus steam values ranging from 0 to  $25 \in /MWh$ . Estimation of steam values is discussed further in Section 5.3.3.

<sup>&</sup>lt;sup>9</sup> In fact, there is in practice an upper limit besides the limit defined by the economy: With a certain number of effects, all of the available temperature difference is "consumed" by BPE and pressure losses, which means that there is no "room for" the evaporation effects.

## 5.3.2 Investment costs

In the investment estimations in this work, the greatest effort was put into obtaining reasonable additional costs of using more evaporation effects or larger heat transfer areas. In other words, the absolute values of the investments are more uncertain than the difference between two investments. Furthermore, the investments are not calculated in the same way in all of the papers. Thus, the investment costs cannot be compared between the papers; the costs can only be compared for plants within the same paper.

In Papers 1–4, the investment model for the evaporation plants was developed by Algehed (2002) using investment estimations made by Kvaerner Pulping in 2002. Since then, the price of raw materials has gone up, especially that of steel. In Papers 5 and 6, the investment costs are estimated by an evaporation expert at ÅF-Process (Redeborn, 2008). The investment costs in Paper 6 are significantly higher than the others, since the evaporation plants in Paper 6 are assumed to be complete new plants which require more investments in auxiliary equipment and preparation.

A new investment model is developed in this work. This model is matched to the investments estimated by Redeborn in Paper 5. In Figure 5.4, the investment model developed in this work is shown together with the model by Algehed and the estimated costs by Redeborn in Paper 5. In the figure, the evaporation plants on the x-axis are denominated by labels used in Paper 5. All of these plants are results from simulations using the objective function  $Minimise\ the\ heat\ transfer\ area$ , which is indicated by the b at the end of the labels. In the figure, a reference plant without lignin extraction, Ref, is compared with three plants with lignin extraction: I, II and III. Moreover, the plants are retrofitted in either of two ways, represented by 1 or 2. For more details, see Paper 5.

The investment model by Algehed (2002) is presented in Eq. 6. Since Algehed's model is given in US\$, the exchange rate 1.37 US\$/ $\in$ <sup>10</sup> was used to convert the model to euro.

$$I_{\text{evap}} = C_1 \cdot n + C_2 \cdot A \tag{6}$$

where

$$C_1 = 0.62 \text{ M} \in$$
  
 $C_2 = 243 \cdot 10^{-3} \text{ M} \in /\text{m}^2$ 

The investment model developed in this work is shown in Eq. 7. The greatest difference between this new model and the one by Algebed is that the new model has set a higher investment for Effect 1, due to the higher

<sup>&</sup>lt;sup>10</sup> Average rate during 2007 (The Swedish Central Bank, 2008).

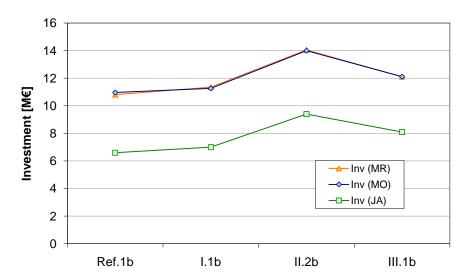


FIGURE 5.4: The investment costs for retrofitting the evaporation plants in Paper 5. Three models are shown: a model by Algehed (2002, JA) used in Papers 1–4, Redeborn's estimations (MR) used in Paper 5 and a model by the author of this thesis (MO) adjusted to Redeborn's estimated investments.

complexity of this effect. As for the validity of the model in Eq. 7, it is only valid in the near future and for retrofit situations in which the number of new effects and the amount of new heat transfer area are close to those used in the present work, i.e. 3-4 evaporation bodies and 2700–6400  $\rm m^2/new$  body. The model is quite rough; Redeborn stated that his investment estimations are valid at best within  $\pm 30\%$ . The model is not applicable to a completely new evaporation plant since additional costs including control systems, pumps and storage tanks are not included for the present retrofitted plants.

$$I_{\text{evap}} = C_1 \cdot (1.3n_1 + n_{2-6}) + C_2 \cdot (1.3A_1 + A_{2-6})^{0.6} \tag{7}$$

where

$$C_1 = 1.82 \text{ M} \in$$
  
 $C_2 = 17.3 \cdot 10^{-3} \text{ M} \in /\text{m}^2$ 

In Paper 6, the old evaporation plant was assumed to be completely replaced by a new plant. The investment costs were estimated by Redeborn (2008). Redeborn started by estimating the investment cost for the conventional plant with 7 effects. The costs for the other plants were estimated by assessing the deviations from this plant in economic terms.

It is important to keep in mind that it is difficult to make reliable investment models for evaporation plants in real mills. Naturally, it is even more difficult to make investment models that should be valid for arbitrary plants in fictitious mills. Every plant is unique in the sense that many costs are site specific, such as the distance for piping, the state of the existing equipment and the complexity of the control system. The uncertainty of an investment cost model becomes greater over time, especially with such volatile prices of raw materials as there are today. In a real mill, estimations by consultants should be within  $\pm 25\%$  of the final investment cost, which gives a picture of the level of uncertainty.

#### 5.3.3 The value of steam

The value of low pressure (LP) steam in a mill can be estimated in different ways; three possible ways are:

- The value of fuel that can be saved by not having to produce the steam, minus the value of the possible decrease in electricity production;
- The income from selling heat produced by the steam (e.g. district heating); and
- The value of electricity that can be produced by the steam in a condensing turbine.

As an example, Reese (2006) states that the value of LP steam<sup>11</sup> was 5–20  $\in$ /MWh<sup>12</sup> for American conditions in 2005. In the model mill studied here (FRAM, 2005), the value of LP steam is 8–13  $\in$ /MWh provided that:

- All of the surplus steam can be used to produce electricity,
- 1 MWh of LP steam can generate 0.19 MWh of electricity, and
- The electricity price is 40–70 €/MWh.

In real mills, the steam can be valued differently depending on the amount of steam surplus (Towers, 2005). For example, if the turbines are run close to their limit, only some of the LP steam surplus can be used to produce electricity in condensing turbines. Additional steam surplus cannot be used for electricity production; hence it has a different value.

<sup>&</sup>lt;sup>11</sup> 5.5 bar(a), originally stated as 65 psi(g).

 $<sup>^{12}</sup>$  Originally stated as \$5/klb with the given span of \$2–\$8/klb. This is equivalent to 3–13 €/tonnes or 5–20 €/MWh for our mill conditions.

# Results and discussion

he results of the thesis work concern principally the evaporation plant at kraft pulp mills, but also the simulation tool used and developed during the Ph.D. project. The consequences for the evaporation plant of process-integrating it, of extracting lignin from it and of lowering the surface condenser are discussed. The consequences for the entire mill have been studied more thoroughly in a related Ph.D. thesis: Axelsson (2008).

# 6.1 Process-integrated evaporation

For the conditions studied in Paper 6, and steam values between 8 and  $13 \in /MWh$ , the additional profit for PI plants is  $0.1\text{--}0.5 \text{ M} \in /\text{year}$  (0.3–1.5  $\in /ADt$ ) in comparison with conventional plants (an overview is presented in Figure 6.2 to aid comparisons). With higher energy prices in the future, the additional profit with PI plants could increase further. In the overall profitability analysis for a mill, the investments for extracting the excess heat must also be taken into account. This is done in all papers that include profitability analyses for PI plants. All of the investment costs necessary are given in Papers 1–3, and some of these costs are reused in Papers 4 and 6.

When simulating process-integrated (PI) evaporation plants, there are several factors that influence the final assessment of the profitability of the plants. Important factors include the number of evaporation effects and the amount of excess heat available for the plant. In the model mill studied, 7 evaporation effects are often more profitable than 6 or 8 effects. Moreover, 1.0 GJ/ADt was shown to be a commonly occurring amount of excess heat in the pinch analyses. Hence, the subsequent analyses of the consequences of PI evaporation are simplified by only including results for 7 evaporation effects and 1.0 GJ/ADt of excess heat.

Selected evaporation plants from Papers 1 and 6 are summarised in Table 6.1. Here, it can be seen that the steam demands are 26% lower for the PI plants than for the conventional ones. Moreover, it is also clear that the

steam demand is the same for the two PI plants and nearly the same for the two conventional plants, despite the fact that more water is evaporated in the Paper 1 evaporation plants. In this respect, it is important to keep in mind that the steam demands in the table are not fully comparable, since the conditions for the simulations are slightly different: the condensates do not leave the plants at the same temperatures, and the heavy liquors have different enthalpies because of their different solids contents. Apart from this, the objective function applied in the simulation tool differs: in Paper 1, the steam demand was minimised, whereas in Paper 6, the heat transfer area was minimised.

Another result from both Paper 1 and Paper 6 is that the steam demands for the PI plants with 6 and 7 evaporation effects are lower than for the conventional plants with 7 and 8 effects, respectively. However, this does not automatically mean that the overall steam demand for the mill is lower with the PI plants. Here, the key is the source of the excess heat. Should some of the excess heat originate from the stripper column, the mill's steam savings with a PI plant would not be as great (compared with a conventional plant) as is discussed above, since the conventional plants could integrate the stripper and thereby count in also this steam saving. In such cases, PI plants and conventional plants have nearly the same profitability (at current electricity prices; see Section 5.3.3 and Papers 1–3). In a real mill, the most profitable alternative may be a PI plant where the stripper column is integrated.

In a wider perspective, the potential for reduced CO<sub>2</sub> emissions, resulting from energy savings proposed in Paper 2, is of the same magnitude as the Swedish annual goal for the years 2008–2012. This is true provided the same energy savings can be made in all Swedish kraft pulp mills (Axelsson, 2008). This may be unattainable; nevertheless, it gives a rough estimation of the potential of reduced steam demands in Swedish pulp mills.

# 6.2 Sensitivity to the temperature of the excess heat

The temperature of the excess heat determines where the heat can enter the evaporation plant. The higher the temperature of the excess heat is, the more effects can make use of the heat, since the steam temperature decreases stepwise along the evaporation train. In the plants simulated, excess heat of 100–105°C generally enters Effect 3.

In supplementary simulations for Paper 6, the consequences of excess heat entering Effect 2 or 4 are investigated. For these examples, the temperature has to be above 121°C or 94°C, respectively, for PI plants with 7 effects. With excess heat at 121°C, the steam demand decreases by 2 MW (7%), whereas at 94°C it increases by 2 MW (7%). The investment costs

Table 6.1: Live steam demands with 7 evaporation effects for conventional and PI evaporation plants. Other key data are added to aid comparisons. The PI plants make use of  $1.0~\mathrm{GJ/ADt}$  of excess heat at  $100-105^{\circ}\mathrm{C}$ .

	Paper 1		Paper 6	
Type of evap.	Conv.	PI	Conv.	PI
Steam (MW)	35.8 26.4		$35.5 \stackrel{\frown}{\sim} 26.4$	
(GJ/ADt)	3.09	2.28	3.07	2.28
Heavy BL, solids	80%	80%	75%	75%
Evap. $H_2O$ (kg/s)	100.9	100.9	99.1	99.1
(t/h)	363	363	357	357
Surface cond.	$50^{\circ}\mathrm{C}$	$40^{\circ}\mathrm{C}$	$55^{\circ}\mathrm{C}$	$55^{\circ}\mathrm{C}$
Objective func.	Min steam	Min steam	Min area	Min area
Cond. stripper	Integrated	Stand-alone	Stand-alone	Stand-alone

necessary are roughly the same as for 105°C. With the higher or lower excess heat temperature, the yearly profit would be of the order of  $0.1-0.2 \text{ M} \in$  higher or lower for a PI plant with 7 effects.

# 6.3 Lignin extraction

In Paper 3, the conclusion is drawn that lignin extraction is economically interesting in connection with increased pulp production. Without increased pulp production, as in Papers 1–2, it seems better to utilise a steam surplus to increase the electricity production rather than to extract and sell lignin. These are general conclusions from the collaborative work with Axelsson (2008).

As far as the recirculation of filtrates from the lignin separation plant is concerned, separate recirculation is the most feasible. With this recirculation, the filtrates are mixed with black liquor having a similar solids content, and the lignin-lean solids are not sent through the lignin separation plant again. For the conditions studied in Paper 5, an evaporation plant with lignin extraction and separate recirculation of filtrates involves a 12% greater steam demand and a 12% higher investment cost than a plant without lignin extraction.

Recent experiments by Moosavifar (2008) show that the viscosity of LLBL may in fact be even lower than assumed in Papers 5 and 6 (the assumptions regarding the viscosity of LLBL in these papers originate from preliminary experiments by Moosavifar). In Paper 5, the viscosity used for lignin-lean black liquor (LLBL) in the best alternative with lignin extraction was set to 60% of that of ordinary black liquor. To investigate the consequences of a lower viscosity, this evaporation alternative was simulated with

a viscosity only 20% of that of ordinary black liquor. The results show that the total heat transfer area for the evaporation plant with the lower viscosity decreased by 17%, which translates into a reduction in the total investment of roughly  $0.7 \text{ M} \in (6\%)$ . This means that the plant with lignin extraction would require an investment only 5% greater than the best plant without lignin extraction (see the overview in Figure 6.2).

It should be noted that the results depend on the fact that the solids content of the mixed liquor to Effect 6 is the same (i.e. 20%) in all of the simulations. This is achieved by adding the right amount of intermediate liquor (see Figure 6.1) and is done to avoid foaming problems in the plant. If the weak liquor is sent directly to Effect 6 (i.e. without a mixing tank<sup>1</sup>), and filtrates from the lignin plant are added, the solids percentage of the liquor entering Effect 6 can be either raised or lowered. Thus, in the absence of a mixing tank, the lowest heat transfer area would probably be achieved by recirculating the filtrates to the weak liquor, since the filtrates are added at a stage where their solids content is higher than that of the ordinary liquor. The other two recirculation alternatives (II and III in Figure 6.1) would probably result in a generally higher solids content of the liquor (in Effects 2–6). This gives higher BPEs and lower heat transfer coefficients, which leads, in turn, to larger heat transfer areas.

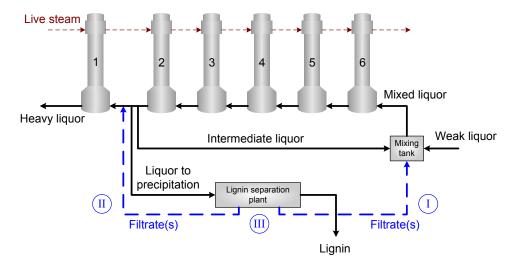


FIGURE 6.1: Three ways of recirculating the filtrates from the lignin separation plant: (I) both filtrates are sent to the mixing tank; (II) both filtrates are sent to Effect 1; (III) the filtrates are sent separately to the mixing tank and Effect 1.

In the simulations in Paper 5, it is interesting to note that even if the amount of water evaporated and the number of effects are the same in two

 $<sup>^{1}\,\</sup>mathrm{This}$  is not uncommon for evaporation plants where the black liquor is based on hardwood. (Olausson, 2008)

retrofitted plants, the differences in steam demand are not negligible. In these simulations, heat transfer areas placed differently can affect the steam demand by at least four percentage units. In general, it was evident that a low steam demand was achieved in the evaporation plants that had larger heat transfer areas at the cold end of the evaporation train.

An important difference between the simulations in Papers 5 and 6 is that the evaporation plant is only retrofitted in Paper 5, whereas it is completely replaced in Paper 6. An assumption in this regard is that the surface condenser is kept as it is in Paper 5, whereas it is replaced by a new one in Paper 6. The increased steam demand with lignin extraction means that more secondary vapour must be condensed in the surface condenser. When the surface condenser is kept, the increased cooling demand is handled by increasing the  $\Delta T$  over the surface condenser. The increased  $\Delta T$  over the surface condenser reduces the total temperature difference that is available to the evaporation effects. This means that the heat transfer areas in the effects must be larger with lignin extraction than without lignin extraction. If the surface condenser is replaced by a new one, this disadvantage for the plants with lignin extraction is removed. These assumptions about the surface condenser mean that the plants with lignin extraction have an intrinsic disadvantage in Paper 5 but not in Paper 6.

# 6.4 Process-integrated evaporation with lignin extraction

For the conditions studied in Paper 6, the additional profit for PI plants is  $0.3-1.5 \in /\mathrm{ADt}$  in comparison with conventional plants (see Section 6.1). When lignin is extracted, the additional profit for PI plants is higher than that:  $0.3-0.7 \,\mathrm{M} \in /\mathrm{year}$  ( $0.7-1.7 \in /\mathrm{ADt}$ ). The reason for the especially high profits with PI plants when extracting lignin is that the heat transfer areas for the PI plants do not need to be increased as much as without LE. All of the PI plants require more area than the equivalent conventional plants, but in the PI plants with LE, the solids content profile is lower than for the PI plants without LE, see Figure 6.3. This is because the excess heat is the same for both PI plants, but the liquor flow is higher with LE due to the recirculated filtrates. The higher liquor flow means that the water evaporated by the excess heat does not increase the solids content as much as without the recirculated filtrates.

 $<sup>^2</sup>$  This is done to maintain the temperature of the warm water produced in the surface condenser.

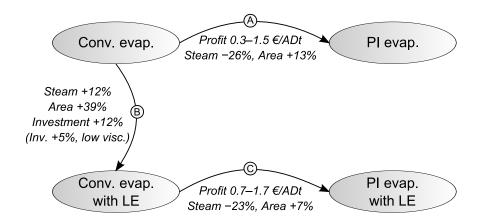


FIGURE 6.2: Overview of the results from the simulations. (A) and (C): The increased profit from using PI plants (1 GJ/ADt of excess heat), as well as the decrease in steam demand and increase in heat transfer area. (B): The increase in steam demand, heat transfer area and investment cost caused by adding a lignin separation plant to a conventional evaporation plant. Here, the investment will be lower if the viscosity for lignin-lean black liquor is as low as recent experiments indicate.

# 6.5 Lower surface condenser temperature

The consequences for an evaporation plant of a lower (than normal) temperature in the surface condenser (SC) are analysed in Paper 6, while the consequences for the mill are covered in Papers 1–3. In Paper 6, the evaporation plants are assumed to be new, complete plants. For the conventional plants with 7 and 8 evaporation effects, the additional profit from decreasing the SC temperature is 0.2–0.3 M $\in$ /year (0.6–0.9  $\in$ /ADt). For these plants, the profit is independent of the steam value, since the steam demands for normal and low SC temperatures are equal. In contrast, with 6 effects, there is no additional profit from lowering the SC temperature, since the steam demand for the low SC plant was marginally higher than for the normal SC plant. The difference in steam demand depends on marginal effects of a lower surface condenser temperature; these effects usually cancel each other:

- More heating is needed to reach the saturation temperature of the liquor in the effects, causing a higher steam demand;
- More vapour is flashed from the black liquor before entering the first evaporation effect, causing a lower steam demand; and
- The temperature of the surplus condensates to the purification plant becomes lower, causing a lower steam demand.

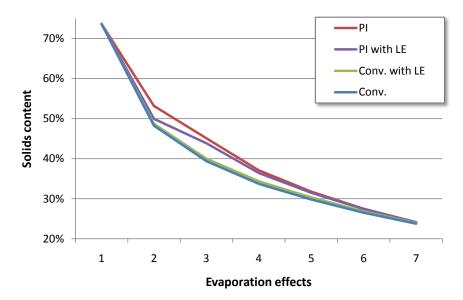


FIGURE 6.3: The solids contents of the black liquor exiting the 7 effects in the evaporation train. Conventional plants are compared with PI plants, both with and without lignin extraction (LE). The plants were simulated in Paper 6.

For the PI evaporation plants, the trend is the same for 6 and 7 effects as for the conventional plants. However, for 8 effects, it was possible to use the excess heat in one more effect in the *low SC temperature* plant than in the *normal SC* plant, resulting in a lower steam demand. This results in a  $0.5-0.6 \ \mathrm{M} \in \mathrm{higher}$  profit for steam values between 8 and  $13 \in \mathrm{/MWh}$ .

# 6.6 Objective functions

The natural goal for a mill is to maximise the profit from an investment. For various reasons it is difficult to find an evaporation plant that maximises the overall profit for a mill, see Section 5.2.2. An objective function that maximises the net annual profit is, nevertheless, available in the simulation tool. The other two objective functions in the tool minimise the steam demand and minimise the heat transfer area, respectively.

In the papers it is clear that the objective function which minimises the heat transfer area results in evaporation plants with reasonable  $\Delta T_{\rm eff}$  in the effects, thus making them realistic for use in the design of real plants. The objective function that minimises the steam demand can be used as a comparison, although it tends to suggest unreasonably high  $\Delta T_{\rm eff}$  in Effect 1 and unreasonably low  $\Delta T_{\rm eff}$  in the last effects (5–6). The user must therefore be more careful when setting reasonable limits for the minimum and maximum  $\Delta T_{\rm eff}$  in the effects.

The steam demand for evaporation plants with a given number of evaporation effects and a known amount of water to be evaporated varies, naturally, only slightly in differently designed plants. The heat transfer area, however, can vary significantly for differently designed plants. For this reason, the objective function that minimises the heat transfer area should be used in most simulations. Another reason for using this objective function is that the heat transfer area is automatically distributed evenly within the evaporation plant, which has been shown to be favourable in practice.

# 6.7 Visualisation of evaporation plants in Grand Composite Curves

The evaporation plant is, as a simplification, usually represented by a rectangle in a Grand Composite Curve (GCC), see Figure 2.3. However, the rectangular shape seldom gives a true picture of the amount of condensed steam in the intermediate effects and the surface condenser. To get the real picture of the evaporation plant in the GCC, simulations are needed.

In Figure 6.4, two 7-effect evaporation plants are shown in a T/Q diagram. For the conventional plant, the heat transferred from the condensing steam decreases between Effects 1 and 4. The reason is that the black liquor is not preheated before entering the effects, which means that some of the steam has to be used to heat the liquor to the boiling point. In the following stages (Effects 5–7 and the surface condenser), the transferred heat is increasing since the black liquor feed is flashed into the effects before entering the first evaporation effect (here Effect 7). For the PI plant, the trends are the same as for the conventional plant, except that excess heat enters Effect 3. Furthermore, it is clear that when using a PI plant, more water is evaporated in the cold end of the plant.

The results show that, without preheating the liquor between the effects in a counter-current plant, valuable heat is "wasted"; the reason is that the heat used for preheating cannot be used for producing secondary vapour that is transferred to the subsequent effect. This means that more heat than necessary must be transferred in the effects, thus requiring larger heat transfer surfaces. However, with preheating between the effects, other heat transfer surfaces are needed. The advantage of preheating in separate heat exchangers could be that it may be possible to use low-quality heat.

# 6.8 Complementary sensitivity analysis

#### 6.8.1 Expanding to 7 evaporation effects (Paper 5)

In Paper 5, the aim is to investigate the consequences of integrating a lignin separation plant with an existing evaporation plant. In that paper, it was

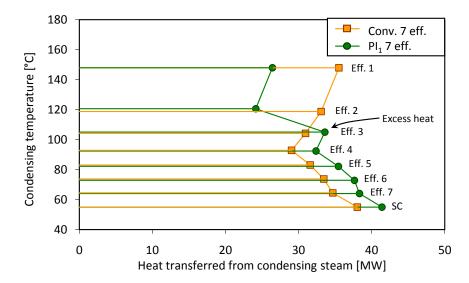


FIGURE 6.4: Representation of a conventional and a PI evaporation plant (1 GJ/ADt of excess heat entering Effect 3) in a T/Q diagram. The labels show in which effects the steam condenses; the steam at the bottom condenses in the surface condenser (SC).

decided to rebuild the evaporation plant as little as possible; thus, the evaporation plant was enlarged from five to six evaporation effects. To evaluate whether the conclusions are different for seven evaporation effects, supplementary simulations of the best conventional plants with and without lignin extraction were performed.

In summary, the supplementary simulations indicate that the conclusions are the same for six and seven evaporation effects. The investment costs for the plants with lignin extraction increased by virtually the same percentage for seven effects (+9%) as for six effects (+10%), when compared with the same plant without lignin extraction. The steam demand is also increased by almost the same percentage for seven effects (+11%) as for six effects (+12%) in comparison with the same plant without lignin extraction. As additional information, when expanding from six to seven effects, the investment necessary for the evaporation plant was increased by about 30\%, while the steam demand was decreased by about 15% (both percentages are valid whether lignin is extracted or not).

## 6.8.2 Consequences of using a simplified model for Effect 1

In a real plant, Effect 1 is generally divided into three or four sections that are in parallel on the steam side and in series on the liquor side. The decision was made to simplify the model of Effect 1 in OptiVap to avoid too many details. Effect 1 has thus been modelled as one section, which meant that the solids content and temperature of the liquor along the tubes were estimated

to be the same as the solids content and temperature of the heavy liquor. In reality, this is an overestimation, since the solids content and temperature along the tubes are lower than that of the heavy liquor in the greater part of Effect 1.

Redeborn (2008) estimated a reasonable expansion alternative for the reference plant in the present work so that the consequences of separate sections in Effect 1 could be assessed. This expansion alternative for the reference plant meant that the investment cost could be reduced by around  $1 \,\mathrm{M} \oplus$  for a conventional plant. This decrease in investment is probably valid for all of the simulated plants, since Effect 1 was modelled in the same way for them all.

# Conclusions

n overall conclusion is that kraft pulp mills can save steam by employing a PI evaporation plant instead of a modern conventional plant, given that excess heat can be made available at the mill. As an example, in a model mill resembling typical Scandinavian market pulp mills, 26% of the live steam could be saved in the evaporation plant by employing a 7-effect PI plant (1.0 GJ/ADt of excess heat, 105°C) instead of a modern 7-effect conventional plant. In some of the plants studied, it is even possible to employ one less evaporation effect and still achieve a lower steam demand with a PI plant than with a conventional one. For steam values between 8 and  $13 \in /MWh$ , the additional profit for PI plants was  $0.1-0.5 \text{ M} \in /year (0.3-1.5 \in /ADt)$  in comparison with conventional plants. With higher energy prices in the future, the additional profit of PI plants could increase further.

Another conclusion is that evaporation plants with lignin extraction (LE) require more live steam and larger heat transfer surfaces than plants without LE. The simulations show, for example, that an evaporation plant with extraction of 190 kg/ADt lignin (dry) requires 12% more live steam and a 12% higher investment cost than a plant without LE. However, should the viscosity of lignin-lean black liquor be as low as recent experiments indicate, the investment cost for a plant with lignin extraction could be only 5\% higher than for a plant without lignin extraction. As far as the recirculation of filtrates from the lignin separation plant is concerned, separate recirculation is the most feasible. An overall conclusion, from the cooperative work during the Ph.D. project, is that LE is economically interesting for pulp mills, at least in connection with increased pulp production. Without increased pulp production, the results indicate that it is more advantageous to utilise a steam surplus to increase the electricity production than to extract and sell lignin. However, the results depend greatly on the electricity and lignin prices.

When extracting lignin, the gains are especially high with a PI evaporation plant instead of a conventional plant. A PI plant with lignin extraction was  $0.3-0.7 \text{ M} \in \text{/year} (0.7-1.7 \in \text{/ADt})$  more profitable than a conventional

plant with lignin extraction, under the conditions studied here.

To achieve a lower investment cost for an evaporation plant, the temperature of the surface condenser could be lowered. In the present simulations, a lowered surface condenser temperature meant that the profit could be raised by 0.2-0.3 M --/year (0.6-0.9 --/ADt) for both conventional and PI plants. This means that the extra costs for the higher volume flows are low compared with the savings from a greater total available temperature difference.

The steam demand for evaporation plants with a given number of evaporation effects and a known amount of water to be evaporated varies, naturally, only slightly in differently designed plants. The heat transfer area, however, can vary significantly for differently designed plants. For this reason, the objective function that minimises the heat transfer area should be used in most simulation cases. Another reason for using this objective function is that the heat transfer area is automatically distributed evenly within the evaporation plant, which has been shown to be favourable in practice.

# Further work

s in all doctoral thesis research, there is a time limit. This means that everything could not be investigated, no matter how interesting it might be. Nevertheless, new research ideas keep popping up during the work; fortunately, so do new Ph.D. students.

# 8.1 Practical mill consequences

The present thesis work involves theoretical modelling. To validate the results, practical testing should be carried out to answer some questions.

- Scaling: Does scaling increase in the evaporation plant when extracting lignin? This could be so, since the share of inorganic substances is higher when some of the organic substances (mostly lignin) are removed. Another question is whether using excess heat may affect the incrustation tendency due to the changed solids content profile. These matters are investigated in an ongoing Ph.D. project of our research group: Gourdon (2007).
- Lower surface condenser temperature: The consequences of lowering the surface condenser must be studied further. In further work, the practical consequences should be investigated in more detail, as well as the economic consequences regarding the operating cost due to the lower pressure. According to the evaporation experts who have been consulted in the present work (Olausson, 2008; Redeborn, 2008), a lower surface condenser temperature should not be a problem from a technical point of view. Possible issues that must be managed are that larger metal constructions must be transported and handled in the building process, and that the amount of warm water produced must cover the mill demand. The latter has not been a problem in the mills studied in the current work.

# 8.2 Process-integrated evaporation

The considerations for a mill when introducing process-integrated evaporation have to be mapped more thoroughly:

- If the integration between the evaporation plant and the source of excess heat is done with a low  $\Delta T$ , a backup plan might be necessary to handle sudden changes of the excess heat temperature. In any case, the need for proper control systems and management, together with good cooperation between different divisions of the mill, is even more important in mills with process-integrated evaporation.
- Another issue regarding the dynamics of the excess heat is the variations over the year. The availability of the excess heat source as well as the temperature of it must be investigated for a whole year, if seasonal variations occur.
- An alternative way of using the excess heat is to preheat the black liquor between the evaporation effects. An advantage of this approach can be that the excess steam does not need to be reformed when foul; the excess heat does not even have to be in the form of steam. Another advantage is that the excess heat can be supplied at a lower temperature and still have the same steam saving potential. A drawback to this approach may be the difficulty in managing fouling of black liquor in a heat exchanger (Redeborn, 2008).

# 8.3 The lignin separation method

The method of separating lignin from black liquor has been developed over a period of several years; knowledge of the process is well surveyed and documented. Nevertheless, the process is still being developed; some of the assumptions in this work concerning the separation of lignin may be questioned and improvements will probably be made. Suggestions for future studies include the following:

- The liquor that is diverted to precipitation can be heat-exchanged with the filtrate entering Effect 1 instead of with the mixed liquor, as is assumed here.
- It may be possible to increase the solids content of the heavy liquor, since its viscosity is lower than that of ordinary black liquor. A viscosity model formulated by Moosavifar (2008) showed that the viscosity

<sup>&</sup>lt;sup>1</sup> In practice, the viscosity determines the solids content possible for heavy liquor, (Redeborn, 2008).

of lignin-lean black liquor at about 75% solids was equivalent to the viscosity of ordinary liquor at 73%.<sup>2</sup>

## 8.4 The simulation tool

The simulation tool used in the present work could be developed in several ways.

- A new heat transfer model by Johansson (2008) could be implemented in the tool to better predict the heat transfer coefficients on the black liquor side of the tubes.
- The viscosity of lignin-lean black liquor could be modelled with equations from experimental work by Moosavifar (2008).
- The resulting evaporation plant from a simulation should be visualised to aid the understanding of the results. Unnecessary details could be hidden until the user wants to reveal them. In this regard, it is always difficult to weigh the positive aspects of making it clearer for the user against the risk of hiding possible warnings of error.

 $<sup>^2\,35\%</sup>$  of the lignin was assumed to be removed and the temperature assumed for heavy liquor was 140°C.

# Nomenclature

### Abbreviations and variables

```
Annuity factor (1/year), also called the capital recovery factor,
a
          Eq. 4
A
          Heat transfer area (m<sup>2</sup>)
          New heat transfer area (m<sup>2</sup>) in Effect 1, Eq. 7
A_1
A_{2-6}
          New heat transfer area (m<sup>2</sup>) in Effects 2–6, Eq. 7
ADt
          Air-dried tonnes of pulp (90% solids)
BL
          Black liquor
BPE
          Boiling point elevation (°C)
BPE_{P}
          BPE at pressure P, Eq. 1
C_1/C_2
          Parameters in Eqs. 6 and 7
          Specific heat capacity (J/kg K)
c_p
          Outside tube diameter (m)
d_{\rm o}
Conv.
          Conventional, as in conventional evaporation
eff.
          Effects, as in Evaporation effects
Evap.
          Evaporation
FRAM
          A Swedish national research programme called the "Future
          Resource-Adapted Pulp Mill"
FRAM Typical Mill
          A computer model of a typical, Scandinavian, market pulp mill
          producing 327,000 ADt/year bleached kraft pulp from softwood.
          Acceleration due to gravity (m/s<sup>2</sup>)
g
GCC
          Grand Composite Curve
H
          Enthalpy (J/kg)
HP
          High-pressure steam; in this study 61 bar(a)
          Discount rate of an investment, Eq. 5
          Investment cost for the evaporation plant, Eqs. 6 and 7
I_{\text{evap}}
LE
          Lignin extraction
LLBL
          Lignin-lean black liquor
```

LLDS Lignin-lean dry solids LPLow-pressure steam; in this study 4.5 bar(a)  $\dot{m}$ Black liquor mass flow per tube (kg/s) MP Medium-pressure steam; in this study 11 bar(a) Nu Nusselt number (-). The indexes lam and turb in Eq. 3 refer to laminar and turbulent flow, respectively. Economic lifetime of an investment (year), Eq. 5 nNumber of new evaporation bodies, Eq. 6 nNumber of new evaporation bodies in Effect 1, Eq. 7  $n_1$ Number of new evaporation bodies in Effects 2–6, Eq. 7 OptiVap The simulation tool described in this thesis PIProcess integrated, as in process-integrated evaporation  $PI_1/PI_2$ PI evaporation using 1 or 2 GJ/ADt of excess heat  $\Pr$ Prandtl number (–) QHeat transferred from the condensing steam to the evaporating liquor in an evaporation effect (W) Re Reynolds number (–) SSolids content of black liquor (kg solids/kg total), Eqs. 1 and 2 SCSurface condenser Tonne TTemperature (K)  $T_{\text{sat},P}$ Saturation temperature (K) of water at pressure P, Eq. 1 Overall heat transfer coefficient in an evaporation effect  $(W/m^2K)$ , see the appendix Solids content of black liquor (kg solids/kg total), see the apxpendix

### Greek letters

$\alpha$	Heat transfer coefficient (W/m <sup>2</sup> K)
$\Delta T_{eff}$	Effective temperature difference in an evaporation body; also
	called the driving force between the condensing steam and the
	evaporating black liquor.
$\Delta T_{tot}$	Total available temperature difference for an evaporation plant
$\lambda$	Thermal conductivity (W/m K)
$\mu$	Dynamic viscosity of black liquor (Pa s)
$\nu$	Kinematic viscosity of black liquor (m <sup>2</sup> /s)

# Subscripts in the appendix

All of the subscripts below concern the equation system in Appendix A. The variables are shown in Figure 5.1 (page 25) and the algorithm is visualised in Figures A.1 and A.2 (pages 68 and 69).

1, 2, 3 Evaporation Effect 1, 2 or 3 BL im Intermediate black liquor BL in Black liquor entering an effect BL mix Mixed black liquor (after the mixing tank for weak liquor) BL out Black liquor exiting an effect  $\mathbf{c}$ Condensate hBLHeavy black liquor loss Loss, as in temperature loss due to pressure drop LSLive steam S out Secondary steam leaving an effect SSCSecondary steam to the surface condenser

sat Saturation, as in *saturation temperature* wBL Weak black liquor

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# References

- **Algehed J.** (2002). Energy efficient evaporation in future kraft pulp mills. Ph.D. thesis, Dept. of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden.
- **Algehed J., Wising U. and Berntsson T.** (2002). Energy efficient evaporation in future pulp and paper mills. In: 7<sup>th</sup> Conference on New Available Technologies. SPCI, Stockholm, Sweden.
- **Axelsson E.** (2008). Energy export opportunities from kraft pulp and paper mills and resulting reductions in global  $CO_2$  emissions. Ph.D. thesis, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden.
- Axelsson E., Olsson M.R. and Berntsson T. (2006a). Heat integration opportunities in average Scandinavian kraft pulp mills: Pinch analyses of model mills. *Nord. Pulp Paper Res. J.*, 21(4): 466–475.
- **Axelsson E., Olsson M.R. and Berntsson T.** (2006b). Increased capacity in kraft pulp mills: Lignin separation and reduced steam demand compared with recovery boiler upgrade. *Nord. Pulp Paper Res. J.*, 21(4): 485–492.
- **Axelsson E., Olsson M.R. and Berntsson T.** (2008). Opportunities for process-integrated evaporation in a hardwood pulp mill and comparison with a softwood model mill study. *Applied Thermal Engineering*, 28(16): 2100–2107.
- Bengtsson C. (2004). Novel process integration opportunities in existing kraft pulp mills with low water consumption. Ph.D. thesis, Dept. of Chemical Engineering and Environmental Science, Chalmers University of Technology, Göteborg, Sweden.
- Bhargava R., Khanam S., Mohanty B. and Ray A.K. (2008). Simulation of flat falling film evaporator system for concentration of black liquor. *Computers and Chemical Engineering*, 32(12): 3213–3223.

- Bremford D.J. and Müller-Steinhagen H.M. (1996). Multiple effect evaporator performance for black liquor: Part II: Development of improved steady state models. *Appita Journal*, 49(5): 337–346.
- Costa A.O.S. and Lima E.L. (2003). Modelling and control of an industrial multiple-effect evaporator system. *Canadian Journal of Chemical Engineering*, 81(5): 1032–1040.
- Cripps H.R., Melton A.W., Capell R. and Retsina T. (1996). Pinch integration achieves minimum energy evaporation capacity. In: 1996 Engineering Conference, vol. 1, (399–402). Chicago, ILL, USA.
- Davy M.F., Uloth V.C. and Cloutier J.N. (1998). Economic evaluation of black liquor treatment processes for incremental kraft pulp production. *Pulp Paper Can.*, 99:2: 35–39.
- **Delin L.** (2008). Senior consultant, ÅF-Process AB, Stockholm, Sweden. Personal communication.
- **FRAM** (2005). Final Report Model Mills (Future Resource-Adapted Pulp Mill Research Programme). Tech. Rep., STFI-Packforsk, Stockholm, Sweden.
- Gidner A. and Jernqvist A. (1997). Simulation of integrated systems of evaporation plants and different energy conservation techniques. *International Journal of Modelling and Simulation*, 17(2): 131–136.
- Gourdon M. (2007). Black Liquor Scaling Propensity: Experimental Investigation in a Pilot Evaporator. Licentiate thesis, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden.
- Gullichsen J. and Fogelholm C.J., eds. (1999). Chemical Pulping (Book 6B). Papermaking Science and Technology. Fapet Oy, Helsinki, Finland.
- **Hadwaco Ltd. Oy** (2000). From effluent to fresh water by cost-effective surplus heat or electrically driven evaporation. Tech. Rep., Helsinki, Finland.
- **Johansson M.** (2007). An implementation-oriented heat transfer model for black liquor evaporation. In: *Proceedings of the International Chemical Recovery Conference 2007*, vol. 1, (155–158). Quebec City, Canada.
- **Johansson M.** (2008). Heat transfer and hydrodynamics in falling film evaporation of black liquor. Ph.D. thesis, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden.
- Kayser M., Kokotovich S. and Retsina T. (1998). Pinch analysis reveals potential at International Falls Mill. In: *Proceedings of the TAPPI*

- International Engineering Conference, vol. 3, (1159–1166). Miami, FL, USA.
- **Kemp I.** (1986). Analysis of separation systems by process integration. *J. Separ. Proc. Technol.*, (7): 9–23.
- **Kemp I.C.** (2007). Pinch Analysis and Process Integration: A User Guide on Process Integration for the Efficient use of Energy. Elsevier Ltd., Oxford, UK, 2<sup>nd</sup> edn.
- **Lindén E.** (2001). Energieffektiv kondensathantering i framtidens massabruk. M.Sc. Thesis, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden.
- **Linnhoff B.** (1993). Pinch analysis A state-of-the-art overview. *Chemical Engineering Research and Design*, 71(A5): 503–522.
- Loutfi H., Blackwell B. and Uloth V. (1991). Lignin recovery from kraft black liquor: Preliminary process design. *TAPPI J.*, 74(1): 203–210.
- von Matérn F. (1981). Swedish trends for evaporation techniques to save energy and abate pollution. In: *International Conference on Recovery of Pulping Chemicals*, (287–292). Vancouver, Canada.
- Moosavifar A. (2008). Lignin Extraction from Black Liquor: Properties of the Liquors and Sulphur Content in the Lignin. Ph.D. thesis, Forest Products and Chemical Engineering, Chalmers University of Technology, Göteborg, Sweden.
- Moosavifar A., Sedin P. and Theliander H. (2006). Viscosity and boiling point elevation of black liquor: Consequences when lignin is extracted from the black liquor. *Nord. Pulp Paper Res. J.*, 21(2): 180–188.
- **Olauson L.** (1979). Heat economy improves with Lockman preevaporation using digester flash steam. *Pulp and Paper Canada*, 80(3): 45–48.
- **Olausson L.** (2008). General Manager, Technology & Engineering, Metso Power. Personal communication.
- Olsson M.R., Axelsson E. and Berntsson T. (2006). Exporting lignin or power from heat-integrated kraft pulp mills: A techno-economic comparison using model mills. *Nord. Pulp Paper Res. J.*, 21(4): 476–484.
- Olsson M.R. and Berntsson T. (2007). A tool for simulating energy-efficient evaporation using excess heat in kraft pulp mills. In: 2007 Engineering, Pulping & Environmental Conference. TAPPI, Jacksonville, FL, USA.

- Olsson M.R. and Berntsson T. (2009a). Comparing conventional evaporation plants with plants using excess heat: A simulation study. *Submitted to BioResources Online Journal*.
- Olsson M.R. and Berntsson T. (2009b). Extracting lignin from black liquor: Consequences for the evaporation plant. Submitted to TAPPI Journal.
- Ray A.K. and Sharma N.K. (2004). Simulation of multi-effect evaporator for paper mill Effect of flash and product utilization for mixed feeds sequences. *IPPTA: Quarterly Journal of Indian Pulp and Paper Technical Association*, 16(4): 55–65.
- **Redeborn M.** (2008). Segment Manager, Chemical Pulping, ÅF-Process AB, Stockholm, Sweden. Personal communication.
- Reese D. (2006). Energy: Feeling the pinch. Paper 360°, Sept. 14–18.
- Schnabel G. (1984). VDI-Wärmeatlas, section Md: Wärmeübertragung an senkrechten berieselten Flächen. VDI-Verlag GmbH, Düsseldorf, Germany, 4 edn.
- Smith R. (1995). Chemical Process Design. McGraw-Hill, New York, USA.
- **Stefanov Z. and Hoo K.** (2004). Distributed parameter model of black liquor falling-film evaporators. 2. Modeling of a multiple-effect evaporator plant. *Ind. Eng. Chem. Res.*, 43: 8117–8132.
- The Swedish Central Bank (2008). Interest and exchange rates, www.riksbank.se (Feb 20th, 2008).
- The Swedish Energy Agency (2007). Energiläget 2007 (in Swedish). Tech. Rep.
- **Towers M.** (2005). Energy reduction at a kraft mill: Examining the effects of process integration, benchmarking, and water reduction. *TAPPI Journal*, 4(3): 15–21.
- Uloth V.C. and Wearing J.T. (1988). Kraft Lignin Recovery: Acid Precipitation vs. Ultrafiltration. In: 74th Annual Meeting, Technical Section, Canadian Pulp and Paper Association, vol. A, (13–22). CPPA, Technical Section, Montreal, Canada.
- Wallberg O. (2005). Extraction of Lignin from Kraft Cooking Liquor by Ultrafiltration. Ph.D. thesis, Dept. of Chemical Engineering, Lund University, Lund, Sweden.

- Wallmo H. (2008). Lignin Extraction from Black Liquor: Precipitation, Filtration and Washing. Ph.D. thesis, Forest Products and Chemical Engineering, Chalmers University of Technology, Göteborg, Sweden.
- Wennberg O. (1989). Reological properties of black liquor. In: *International Chemical Recovery Conference*. Ottawa, Canada.
- Wennberg O. (1990). Sulfat- och sulfitlutars fysikalisk-kemiska egenskaper. Slutrapport etapp II, STU projektnr 88-03869 (in Swedish). Tech. Rep.
- Wiberg R. (2007). Energiförbrukning i massa- och pappersindustrin 2007 (in Swedish). Tech. Rep., Skogsindustriernas Miljö- och Energikommitté.
- Wising U. (2003). Process integration in model kraft pulp mills: Technical, economic and environmental implications. Ph.D. thesis, Dept. of Chemical Engineering and Environmental Science, Chalmers University of Technology, Göteborg, Sweden.
- Wising U., Algehed J., Berntsson T. and Delin L. (2006). Consequences of lignin precipitation in the pulp and paper industry. *TAPPI Journal*, 5(1): 3–8.
- Öhman F. (2006). Precipitation and separation of lignin from kraft black liquor. Ph.D. thesis, Division of Forest Products and Chemical Engineering, Chalmers University of Technology, Göteborg, Sweden.
- Öhman F. (2008). Senior Research Associate, STFI-Packforsk, Stockholm, Sweden. Personal communication.

#### **Appendix**

s is described in Section 5.2, OptiVap has a hidden calculation sheet that is used as a basis when generating new calculation sheets. The algorithm for the reference sheet can thus be applied for all calculation sheets, in spite of the fact that the reference sheet has three evaporation effects and the calculation sheets can have up to nine effects<sup>1</sup>. In the following sections, the algorithm is described for a simplified version of the equation system.

#### A Equation system for three evaporation effects

#### A.1 The algorithm for solving the equation system

For three effects, the simplified equation system has 60 unknown variables and 58 independent equality constraints, see Figure 5.1 (page 25), which means that there are two degrees of freedom.<sup>2</sup> As free variables, the effective temperature differences in the first two effects were chosen:  $\Delta T_{\rm eff1}$  and  $\Delta T_{\rm eff2}$ . Furthermore, some variables are calculated in an iterative manner, for example,  $\dot{m}_{\rm BL~out2}$  and  $\dot{m}_{\rm BL~out3}$ ; see Figures A.1 and A.2. The other iterations necessary are not shown explicitly in the figures.

The algorithm for solving the simplified equation system can be divided into 5 levels (the variables on each level are encircled in Figures A.1 and A.2):

- 1. The 7 input variables give directly 7 variables.
- 2. An initial guess of  $\dot{m}_{\rm BL~out2}$  means that 8 variables can be calculated.
- 3. An initial guess of  $\Delta T_{\rm eff1}$  means that 10 variables can be calculated.
- 4. An initial guess of  $\Delta T_{\rm eff2}$  means that 20 variables can be calculated.
- 5. An initial guess of  $\dot{m}_{\rm BL~out3}$  means that 13 variables can be calculated (including  $\dot{m}_{\rm BL~out2}$  and  $\dot{m}_{\rm BL~out3}$  after iterations).

<sup>&</sup>lt;sup>1</sup> In fact, there is no upper limit for the total number of effects to be used in a simulation. Nine effects have been the economic upper limit in the current work.

 $<sup>^{2}</sup>$  A simulation with n effects has n-1 degrees of freedom with the current algorithm.

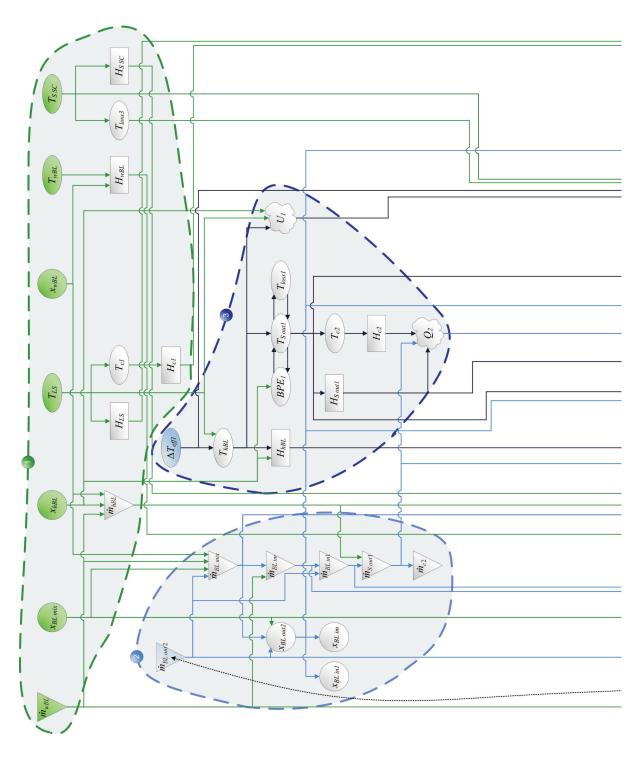


FIGURE A.1: The algorithm for the reference sheet with three evaporation effects in OptiVap.

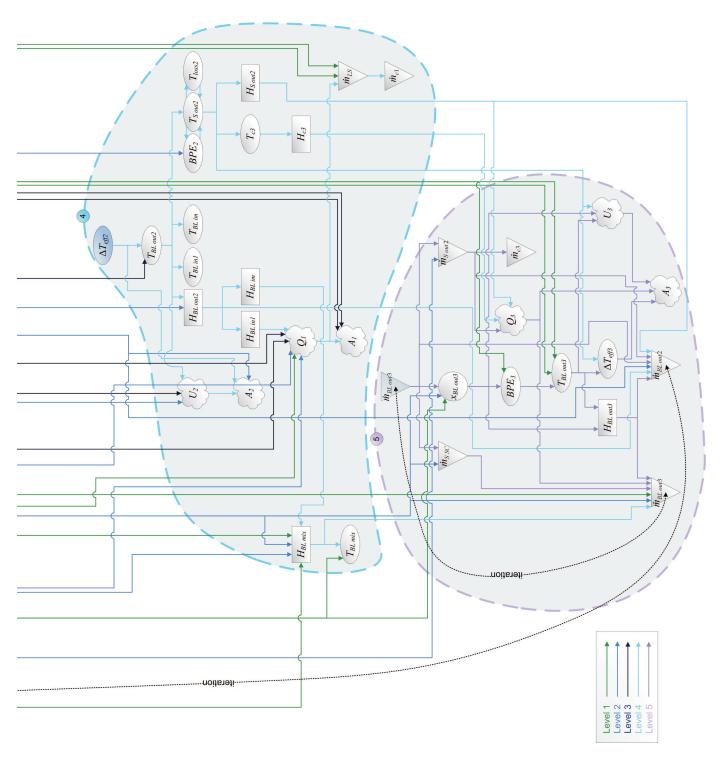


FIGURE A.2: The algorithm for the reference sheet with three evaporation effects in OptiVap.

In Figures A.1 and A.2, the algorithm is shown with these five levels encircled in five colours. Each dependence (the arrows) is coloured accordingly. The variables are explained further in Figure 5.1 and in the Nomenclature (page 55).

The equation system in Figures A.1 and A.2 is solved from the left to the right, or from the top to the bottom, if the book is turned 90° clockwise. The green variables at the top are input data. The dark blue variables ( $\Delta T_{\rm eff1}$  and  $\Delta T_{\rm eff2}$ ) are the free variables and the light blue variables ( $\dot{m}_{\rm BL~out2}$  and  $\dot{m}_{\rm BL~out3}$ ) are guessed values as described above.

The variables are divided into groups, represented by the geometric shapes surrounding them:

**Triangles.** Mass flow rates (kg/s),  $\dot{m}$ ;

Circles. Solids content (mass fraction) of black liquor, x;

**Ellipses.** Temperatures ( ${}^{\circ}$ C), T or Boiling Point Elevations, BPE;

**Rectangles.** Enthalpies (J/kg), H; and

"Clouds". Heat transferred from the condensing steam to the evaporating liquor in an evaporation effect, Q(W); overall heat transfer coefficients in the effects,  $U(W/m^2 K)$ ; or heat transfer surfaces,  $A(m^2)$ .

As a comment, the initial guess of the mass flow in Level 5 is not necessary, since a material and energy balance over the last two effects would give  $\dot{m}_{\rm BL~out3}$ . As this algorithm was difficult to incorporate in the Excel sheet, the more convenient algorithm with five levels was kept.

#### A.2 Simplifications to the equation system

To simplify the description of the algorithm, some parts of the evaporation plant have been omitted:

- The foul condensate stripper,
- The use of excess heat,
- The lignin separation plant,
- The surface condenser, and
- The flash tanks for the mixed liquor and the secondary condensates.

# Papers appended

Heat integration opportunities in average Scandinavian kraft pulp mills: Pinch analyses of model mills

Erik Axelsson, Marcus R. Olsson and Thore Berntsson

Published in Nordic Pulp and Paper Research Journal Volume 21, no. 4, pp. 466–475, 2006

Exporting lignin or power from heat-integrated kraft pulp mills: A techno-economic comparison using model mills

Marcus R. Olsson, Erik Axelsson and Thore Berntsson

Published in Nordic Pulp and Paper Research Journal Volume 21, no. 4, pp. 476–484, 2006

Increased capacity in kraft pulp mills: Lignin separation and reduced steam demand compared with recovery boiler upgrade

Erik Axelsson, Marcus R. Olsson and Thore Berntsson

Published in Nordic Pulp and Paper Research Journal Volume 21, no. 4, pp. 485–492, 2006

## A tool for simulating energy-efficient evaporation using excess heat in kraft pulp mills

Marcus R. Olsson and Thore Berntsson

Submitted to Pulp and Paper Canada.

In Proceedings of
2007 International Chemical Recovery Conference,
Quebec City, QC, Canada, pages 141-146
and
2007 Engineering, Pulping & Environmental Conference
Jacksonville, FL, USA.

**Comment:** The paper has been marginally updated compared with the conference proceedings.

#### Extracting lignin from black liquor: Consequences for the evaporation plant

Marcus R. Olsson and Thore Berntsson

 $Submitted\ to\ TAPPI\ Journal.$ 

## Comparing conventional evaporation plants with plants using excess heat: A simulation study

Marcus R. Olsson and Thore Berntsson

Submitted to BioResources Online Journal.