



# Purification of radioactive waste water using a ceramic membrane

Master of Science thesis in Nuclear Engineering

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Cover: A tubular membrane

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# Abstract

Energy production by nuclear power produce radioactive waste water. In order to meet the demands from the authorities and follow the law the water must be treated before returned to the environment. Common techniques for water treatment are evaporation and ion exchange. Membranes can also be used for treatment of radioactive liquid waste.

A membrane is a semi permeable barrier which can be used for separation of substances in a fluid. One benefit with water treatment systems based on membranes is low energy consumption in comparison with evaporation.

The coolant water in light water reactors are continuously purified primarily by ion exchange resin and conventional filtration. Over time the resin becomes saturated and must be replaced with fresh resin. Water is used for transport the saturated resin to storage and this process will contaminate the water since radioactive material on the ion exchange resin will fall off during the transport process

Membrane technology has been used to treat the process water. The performance of a new type of tubular membrane has been tested in this project. A total volume of 2,4 m<sup>3</sup> with an average gamma activity of  $4,4\cdot10^5$  Bq/L has been reduced to 140 L of concentrate with an average activity of  $5,4\cdot10^6$  Bq/L. The produced permeate held an average activity of  $1,9\cdot10^3$  Bq/L.

A problem with membrane technology is depositing of unwanted material on the membrane surface. If the deposits are radioactive will it cause an increase of dose rate around the filter. A device called Back pulse hammer has been tested together with the membrane for solving this problem. When operating the Back pulse hammer during cleaning of the water, the dose rate build up was not as quick as without the device.

Key words: Membrane technology, Radioactive waste, Dose rate, Back pulse hammer, Fouling.

# Preface

The report which you are about to read is the results of my master thesis performed at Ringhals nuclear facility, Väröbacka, Sweden. The project has been sponsored by Vattenfall AB and been the last part of my education at Chalmers University of Technology.

A master thesis require hard work, perseverance and a piece of invention, and I would not been able to done it without the help and support of many people around me. I would like to show my gratitude and give a special thanks to:

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# Abbreviations

ALARA	As low as reasonably achievable		
BWR	Boiling water reactor		
CVCS	Chemical and volume control system		
DF	Decontamination factor		
HF	Hollow-fibre		
HLW	High level waste		
HPGe	High purity Germanium		
IAEA	International atomic energy agency		
IX	Ion exchange		
IXR	Ion exchange resin		
LLW	Low level waste		
MLW	Medium level waste		
MWCO	Molecular weight cut off		
NF	Nano filtration		
NPP	Nuclear power plant		
PWR	Pressurised water reactor		
RC	Reactor coolant		
RO	Reverse osmosis		
RWCU	Reactor water clean-up		
SiC	Silicon carbide		
SSM	Swedish Radiation Safety Authority		
TOC	Total organic carbon		
UF	Ultra filtration		

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# **1. Introduction**

Water consumption is fundamental for our existence and the supply of fresh water is a prerequisites for civilization. Today, all industry sectors accounts for approximately one fourth of the global water consumption. Some industries only use water for cooling and produce no waste water. However, many industries do create contaminated water. [1]

The motivation for waste water treatment are several. Reuse of waste water contribute to stable supply of fresh water and a sustainable society. The motivation can itself origin form law and regulations which can harm an industry economical if not followed. [1]

Nuclear power creates waste water which varies in chemical composition, radioactivity and volume. The waste water is treated with separation techniques found in other industries and water treatment facilities such as evaporation, adsorption and ion exchange, IX. Separation systems based on membrane technology has in the recent decades been developed and used in the nuclear industry with the benefits of low energy consumption (in comparison to evaporation) and low production of solid secondary waste. [2]

# 1.1. Background

The Swedish nuclear power plants (NPP) produce approximately 40 % of Sweden's electricity production. Today, 10 reactors are operating at three sites; Ringhals, Forsmark and Oskarshamn. Ringhals, located on West coast, is the largest and consists of 4 reactors, one boiling water reactor (BWR) and three pressurized water reactors (PWR). Approximately 20 % of Sweden's electricity is produced by the reactors at Ringhals. Appendix A provides fundamental information regarding radioactivity, dose and nuclear power. [3, 4]

Every nuclear installation (i.e. NPP, research facilities, hospitals etc.) in Sweden is controlled by the Swedish Radiation Safety Authority (SSM) which is the Managing Authority regarding radiation protection and nuclear safety under the Ministry of the Environment. If a licensee neglect the rules, for example release too much activity to the environment, SSM has the authority to sanction or suspend the operating license. [5]

#### 1.1.1 Radioactive waste categories and waste management in Sweden

Radioactive waste is not only produced while operating NPPs, but also from processes in hospitals and research facilities. How the waste should be managed, stored and transported is strictly regulated by laws elucidated by SSM via prescriptive regulations and direct rulings. [5]

Radioactive waste is categorised according to standards conducted by the International Atomic Energy Agency (IAEA) and is based on the activity content and the half-life of the present nuclides. There are three main categories of radioactive waste; low level waste (LLW), medium level waste (MLW) and high level waste (HLW). LLW and MLW are typically operation and demolition waste, and HLW is normally spent nuclear fuel. [6]

Operational waste refers to material and equipment used in the maintenance of the NPP which have been in contact with radioactive substances and cannot be decontaminated. It can for example be protective clothing or equipment used in the process. Approximately 85 % of all Swedish radioactive waste are operational waste and are classified as LLW. This type of waste is compressed into bales which is later combusted or stored in surface repositories for approximately 50 years. Operational waste can also be classified as MLW and can for example be filters and IXR used for water treatment. [6,7]

Demolition waste consists of scrap metal, replaced reactor parts and equipment. This type of waste varies from highly radioactive (for example material from the reactor vessel) to low radioactive level. MLW is immobilized, often by mixing it in concrete and moulded into reinforced blocks. The moulds are stored for at least 500 years under ground. [7]

Discharged fuel elements accounts only for 5 % of all the waste from a NPP but holds around 95 % of the total activity of all the radioactive waste. They are classified as HLW and stored in storage pools where they are cooled down. The plan is to encapsulate the fuel and store it 500 m below ground in solid bedrock. [7]

#### 1.1.2 Radioactive waste water

All current nuclear reactors in Sweden are light water reactors. In the ideal world, a nuclear reactor would have no leakage of neutrons, fission products and actinides. However, this is not realistic and all reactors have some leakage which will contribute to radioactive particles in the cooling water. [8]

There are four ways of how radioactive material can leave the reactor vessel, carried by the coolant:

- Induced radioactivity of the cooling medium, for example:  ${}^{16}O + n \rightarrow {}^{16}N + p$
- Induced radioactivity of corrosion products from construction material, for example:  ${}^{54}Fe + n \rightarrow {}^{54}Mn + p$

 ${}^{58}Ni + n \rightarrow {}^{58}Co + p$ 

 $^{59}Co + n \rightarrow {}^{60}Co$ 

- Leakage of actinides and fission products due to damage in fuel elements, for example:  ${}^{238}U$ ,  ${}^{131}I$ ,  ${}^{137}Cs$
- Fission products from Uranium which has been leached from damaged fuel pins and deposit on the surface in the core (i.e. Tramp Uranium). [8]

Induced radioactivity is the process where radiation affects a material and makes it radioactive. Leakage of neutrons may lead to activation, i.e. neutron capture, in the cooling water or corrosion products. Alpha-emitting actinides and Beta-emitting fission products can also induce radioactivity. [8]

All activity outside the reactor vessel is unwanted and much effort is spent on containing the radioactive substances. It is not possible to contain everything and small releases of radioactivity to the environment will always occur. NPPs are allowed to release small amounts of radioactivity. At Ringhals, the target for water releases is on average <100 Bq/L<sup>1</sup>. [8]

The liquid radioactive waste (LRW) from a NPP varies in volume, radioactivity and chemical composition. There are several methods for treatment of this waste and they traditionally include evaporation, filtration, chemical cleaning, IX and adsorption. After the treatment, the remaining waste is often classified as MLW and solidified in concrete moulds or drums. [8]

#### 1.1.3 Reactor systems for purification of coolant

In PWRs, the Chemical and Volume Control System (CVCS) is used as a support system for the primary coolant circuit. CVCS is partially used for purification of the coolant (i.e. minimize the level of radioactive material or material which can be activated) and coolant water is continuously led through IX and filters. BWR has a similar system for cleaning the coolant; Reactor Water Clean-Up system (RWCU). RWCU is also based on IX and filters and cleans condensate water. The coolant needs to be cleaned from radioactive material (and material which can be activated) in order to decrease the dispersion of radioactive substances, avoid activation of corrosion products and decrease the risk of damage to fuel. [9]

An IX consists of resins which will exchange either anions or cations with OH/H<sup>+</sup>. The performance of the resins will decrease over time due to saturation of the resin. In order to keep the water cleaning process efficient, the saturated resins must be replaced with new ones. This removal process is called

<sup>&</sup>lt;sup>1</sup> According to Bernt Bengtsson, Senior Chemistry specialist at Ringhals AB. 2014-11-24

backflush; water is transferred into the tank in which the IX resin (IXR) resides and transport the resins to a storage tank. Due to the turbulent condition, some of the materials attached to the resins surfaces will fall off. When the resins settle, the water is decanted and the decant water must be purified before release. [7, 9]

# 1.2 Aim and goals

The first aim of this master thesis was to purify the decanted water from activation- and fission products. A new type of membrane was tested in order to evaluate its performance. The membrane was a ceramic-type with a Silicic carbide (SiC) composition and assembled in a pilot plant. It was also investigated how Boron, salts, Silicic acid and organic compounds were distributed between the permeate and concentrate streams.

The decanted water was assumed to contain radioactive particles and ions which may stick to the membrane surface. This would lead to an increase of dose rate from the membrane. Because of the ALARA-principle (`As low as reasonably achievable' concerning radiation exposure) it is important to solve this issue. The activation build up problem can be solved by reversed flow flushing the membrane. This method consumes water and could introduce new organic compounds due to issues with the quality of the water available for the process.

The second aim was to test an alternative method for solving the activation build up problem without the addition of water. A new type of device was connected to the membrane called 'Back pulse hammer' (BPH). The hammer worked as an oscillator which pulses the permeate back and forward over the membrane in order to remove radioactive substances stuck on the membrane.

This master thesis was built on and compared with two previous master thesis (Hjelmberg, née Kallerfelt, 2010 and El Tayara, 2013). [10, 11]

## **1.3 Delimitations**

The back pulse hammer was only connected to the SiC membrane and this membrane was the only one investigated in this thesis. When running the pilot plant, the flow of the feed was kept constant and the concentrate stream was re-circulated.

Since the laboratory work was performed at Ringhals, the measurements and analysis procedures were adjusted to their regulations and routines. The main aspiration of the laboratory work of this master thesis was not to disrupt the daily routines at the chemistry department and this was something that permeated trough the daily tasks during the thesis project work.

The focus of the laboratory measurements were to identify and quantify (if possible) the differences between feed, permeate and concentrate, rather than to precisely determine the water chemistry. The simplification that 1 g of the used water is equal to 1 mL was applied for the whole project.

# 2. Theory

This chapter gives an introduction to the theory behind the relevant concepts and processes. The first sub-chapter deals with membrane filtration which is the most central part in this thesis. Following section is a brief orientation about the use of membranes in the nuclear industry. A short description about the methods of analysis is also included.

# 2.1. Membrane filtration

Membrane filtration is a separation process which involves a membrane and a fluid mixture containing two or more species. The mixture could contain solids (particles) or dissolved substances such as ions. A membrane can be seen as a selective barrier between two phases which allows some substances in the fluid to pass the barrier but prevents others. [1]

The filtration process is either in cross flow (also known as tangential flow filtration) or dead-end geometry. Figure 1 shows a schematic representation of cross flow filtration. The flow into the membrane is often referred as the feed and the main part of the feed travels tangentially across the membrane. Some of the feed will cross the membrane and this stream is called the permeate. The part of the feed which do not cross the membrane is called the retentate. Depending on the situation, the desired product can be either the permeate or the retentate. Both the retentate and permeate are sometimes called concentrate in applications where some species is concentrated in the permeate or retentate. [1]



Figure 1. Cross flow filtration. Here, the feed flow enters from left and is divided into permeate and retentate.

In dead-end filtration (also known as normal once through filtration) the entire feed is pushed through the membrane in normal direction, see figure 2. The species which are rejected by the membrane will deposit on the membrane surface and a "filter cake" is created. No filter cake is created in cross flow filtration since the substances which cannot penetrate the membrane is transported away by the retentate flow. The filter cake will decrease the penetration ability for the substances in the mixture. [1]



Figure 2. Dead end filtration.

Membrane filtration processes exists in a wide variety and differ from application to application. This makes the process inflexible to changes of fluid properties and every membrane separation device must adjusted to the specific application. Another drawback with membrane filtration is fouling which is explained in section 2.1.4. In comparison with other separation processes, evaporation, distillation, ion exchangers etc., membrane filtration generally has the following benefits [1]

- Less energy consumption since the separation does not require any change of phase
- The process can be carried out continuously under steady state condition
- Chemical additives like coagulants and flocculants are seldom needed

Table 1 shows some examples of membrane filter types and under which driving force they are operated by. Driving force, Structure and separation mechanism are explained in the following subsections.

Table 1. Examples of different membrane types. [2]

Driving force	Membrane type	Structure	Separation mechanism	
Pressure	Reversed osmosis	Dense	Difference in diffusion rate between solvent and solutes	
	Nanofiltration	Dense	Combination of difference in diffusion rate and sieving through micropores (<2nm)	
	Ultrafiltration	Porous	Sieving through mesopores (2-50 nm)	
	Microfiltration	Porous	Sieving trough macropores (>50 nm)	
Concentration	Dialysis	Porous	Diffusion	
<i>Electrical potential</i> Electrodialysis		Charged, Porous	Difference in ionic size and strength	
Temperature	Membrane distillation	Porous	Difference in partial vapour pressure	

#### 2.1.1 Transport process and driving force

Transportation through a membrane is a resultant of the driving force acting on a fluid. Chemical and/or physical interaction between the membrane surface and the substances in the fluid (i.e. the solvent, particles, ions etc.) will occur. These interactions will differ between the substances and some substance will cross the membrane faster than others. [1, 12]

The separation driving force used for cross-flow filtration are a pressure gradient, concentration gradient, electrical potential gradient or a temperature gradient over the membrane. It is theoretically possible to use more than one driving force, these membranes are however under development and not commercially used. [2]

Pressure driven separations are mainly used in water treatment systems where the product is cleaned permeate water. For Ultrafiltration (UF) and Microfiltration (MF), the transport through the membrane is based on size exclusion i.e. sieving. The membrane is porous (contain small channels) and to large particles will not cross the membrane. Transport through dense membranes, such as Reversed osmosis (RO) and Nanofiltration (NF), is on the other hand based on diffusion; the components in the fluid are absorbed by the solid membrane, diffuse through it and desorbs on the other side. Diffusion is the spontaneous movement of substances from an area with high concentration to low concentration. The separation is achieved if the substances in the mixture diffuse of different speed through the membrane. The driving force speed up the diffusion and diffusion require larger driving force than for the sieving. Illustrations of sieving and diffusion trough a membrane is given in figure 3. [12]



Figure 3. Transport trough a membrane is either based on size exclusion (left) or diffusion (right).

In electro dialysis, electric potential will move ions through ion-selective membranes whereas in dialysis small molecules will be separated from bigger by diffusion through a porous membrane. In membrane distillation, the membrane lies between a hot and a cold side. Volatile substances evaporates at the hot side, crosses the membrane and condense at the cold side. [1, 12, 13]

#### 2.1.2 Membrane structure and material

A membrane material can be dense or porous, homogeneous or non-homogeneous and organic or inorganic as well as having an electrical charge. The choice of structure and construction material is based on the application (for example fluid properties, demand on the separation, driving force) and dependent on the economy of the process. Membranes can also be surface modified in order to increase the material stability or the performance for example by making the material more hydrophilic. [1, 14]

The separation technique using porous membranes is based on size exclusion, i.e. sieving, which is a mechanical process. Dense membranes are on the other hand based on physical and/or chemical interactions between the fluid components and the membrane surface. [1]

Porous membranes are classified either by characterize pore size (measured in Angstrom, nm or  $\mu$ m) or by Molecular Weight cut-off (MWCO). The unit of MWCO is in Daltons (Da, where 1 Da is equal to 1 gram per mole) and it is rated on basis of the smallest molecule weight which will be at least rejected to 90% by the membrane. MWCO cannot be converted into pore size since they describe two different things. However, in practical situations, the pore size of 1  $\mu$ m is often approximated to be in the range of 500.000-2.000.000 Da<sup>2</sup>. Porous membranes are divided into three categories based on their pore size; micropores, mesopores and macropores. Another way to classify porous membranes is with the unit Micron (micrometer) which refers to the size of the particles that in some extent will be rejected by the membrane. [1, 12, 15]

Homogeneous (symmetric) membranes are made of one material. Theses membranes are made thick in order to ensure the desired mechanical strength. However, the thickness makes the flow resistance higher which decrease permeability and hence the performance of the membrane. The structure of asymmetric membranes change with depth. In general, asymmetric membranes consist of a "skin" which is a thin, selective layer, and a porous layer to support the skin. They are used for smaller molecules were homogeneous membranes often lack the ability to keep the permeability high. [2, 12]

A commercially used membrane is made of either by an organic or an inorganic material. Organic membranes are made of polymers which are generally cheaper to produce than inorganic membranes. The membrane must be chemically inert to the fluid otherwise it will be degraded over time. It must also be physically strong and cope with the operating temperature and pressure. Polymer membranes are limited to operation temperature below 200 °C and they are often sensitive to organic mixtures. The mechanical strength and chemical resistances of the membrane is often higher for the inorganic

<sup>&</sup>lt;sup>2</sup> Conclusion drawn by comparing information in [1], [12] and discussion with C-H Hansson at NORDCAP AB.

membranes which are metallic, ceramic or made of glass or carbon. There are also polymeric membranes coated with a thin, dense layer of metal. [1, 12]

#### 2.1.3 Configuration and membrane modules

The configuration of a membrane involves how the membrane area is oriented relative to the flow i.e. the geometry of the membrane surface. To stabilize and support the membrane, the membrane is placed inside a "house", i.e. a module. [1]

Membrane configurations are either based on a planar or cylindrical shape. Choosing the most optimal configuration involves the following considerations: [12]

- Membrane surface area in relation to the total module volume
- Turbulent flow in order to maximize the mass transport
- Design which enables cleaning
- Production cost

Planar configurations are plate and frame (also known as flat sheet) and Spiral-Wound module. The plate and frame module is based on flat membrane plates which are attached to support material, see figure 4. Between the membrane plates are spacers for mechanical support and separation between the plates. This configuration is mainly used for small-scale application. [1, 12, 14]



Figure 4. Schematic illustration of a Plate and Frame membrane module.



Figure 5. Schematic illustration of a Spiral-Wound module.

Spiral-Wound module appears to be cylindrical, but it is based on flat membranes plates wrapped around a central permeate tube which shows in figure 5. Sheets used for membrane support and for transporting the permeate to the permeate tube lies between the membrane plates. Spiral-Wound configuration is common in pressure-driven processes and has the advantage to be simple and robust in its design. [1, 12, 14]





Figure 6a. The principle of cylindrical membranes.

*Figure 6b. A Hollow-fibre module with a bundle of membrane fibres.* 

Tubular modules and Hollow-fibre (HF) module are cylindrical configurations. A schematic picture of cylindrical membranes is shown in figure 6a. Tubular modules have a similar appearance as shell and tube heat exchangers. The membrane covers the inside of one or several tubes. Tubular membranes have the advantage of high turbulence promotion. One disadvantage is the limited surface area in relation to module volume. [1, 12, 14]

Hollow-Fibre modules consist of capillary fibres arranged in a bundle, see figure 6b. These membranes are self-supporting and need only support in the bundle end. The cost is low due to its simple construction. [1]

#### 2.1.4 Fouling

In membrane technology, fouling is seen as a process resulting in decreased membrane performance as a result of unwanted deposits onto membrane external surface, in the pore-openings or within the pores. The deposit material can be suspended solids, particles or dissolved substances. Fouling on a membrane result in decline of flow through the membrane which in turn will lead to a higher energy demand for keeping the performance high. The overall cost for the separation will increase and the membrane life-time will decrease. [14]

Methods of minimize the problem of fouling are based on pre-treatment and cleaning of the membrane. A module design which enable cleaning must also be choose for applications where the feed is suspected to contain fouling components i.e. foulants. [14]

Pre-treatment of the feed can involve both chemical modification (for example pH-adjustment, aggregation of particles etc.) and pre-filtration. Membrane cleaning methods are based on chemically or physically breaking the bonds forces between the foulants and the membrane. The chemical reactions include hydrolysis, solubilisation and chelation. Membranes are often cleaned when the normal flux is reduced by 10-15 %. [2, 14]

Examples of physical cleaning methods are sonication (sound waves), vibration, scouring (with e.g. foam balls for tubular modules) and reversed flush. The process of reversed flush (also known as backward flush) changes the flow direction of the permeate in order to remove the foulants. Reversed flush may also be performed with another solution than the permeate, eg. distilled water. [14]

#### 2.1.5 Membrane performance

Flux and conversion are two parameters relevant when evaluating the performance of a membrane. A high flux result in a large value of the conversion. [1]

The flux over a membrane refers to the amount of material which passes through a unit area of the membrane per unit time. Flux is also called 'permeate velocity' and the SI unit is  $m^3m^{-2}s^{-1}$ . The driving force is (in theory) proportional to the flux and inversely proportional to the membrane resistance. Equation 1 describes the flux over a pressure driven porous membrane, where *J* is the flux,  $\Delta P$  the pressure difference (i.e. the driving force),  $\mu$  the viscosity of the fluid and  $R_m$  the membrane resistance. The membrane resistance is affected by fouling, the hydrodynamic condition at the membrane-fluid interface, the porosity and the physical shape of the pores. [1]

$$J = \frac{\Delta P}{\mu R_m} \tag{Eq. 1}$$

Conversion, also known as recovery, is the percentage of the feed which goes through the membrane. The parameter is mainly used for cross flow and is derivatives from the mass balance over the membrane which is written as equation 2. Q refers to volume flow of the feed, permeate and retentate. [1]

$$Q_F = Q_P + Q_R \tag{Eq. 2}$$

Conversion, *C*, is written as equation 3.

$$C = \frac{Q_P}{Q_F}$$
(Eq. 3)

The mass balance given in equation 2 can also be written with regard to a certain compound, *i*, using the volume flow rate  $\dot{V}$  of the feed, permeate and retentate and the concentration of the compounds in the feed, permeate and retentate:

$$C_{F,i}\dot{V}_F = C_{P,i}\dot{V}_P + C_{R,i}\dot{V}_R \tag{Eq. 4}$$

## 2.2 Membranes in the nuclear industry

Liquid waste from nuclear fuel cycle operations (power reactors in operation, fuel enrichment and conversion, mining, decommissioning) must be treated before disposal. The management of the waste must be achieved at a reasonable cost by the use of appropriate separation technologies. Traditionally, liquid waste from nuclear power reactors are treated with direct evaporation, ion exchangers, conventional filtration, solvent extraction or a combination of these separation techniques. The goal of the treatment is not only to purify the water which will be discharged to the environment, but also to concentrate the radioactive compounds by volume reduction which enables solidification and recovery of useful components which can be reused (for example Boron). It is possible to reach these goals by applications of membrane technology. [2, 16]

Pressure driven membrane processes (such as RO, NF, UF and MF) have become the most common one in the nuclear industry over other membrane processes (such as temperature and concentration driven processes). The pressure driven processes are well proven and mature technologies, with a large number of full-scale applications. Other membrane processes have been used mainly in pilot scale applications with a few exceptions. [2]

One drawback with membranes is their lack of flexibility since they are not versatile (i.e they are best suited for one type of fluid). Every active waste treatment system must be tested in pilot scale in order to understand the application and identify future problems. [2, 16]

#### 2.2.1 Process design considerations

Designing and running a cleaning facility based on membrane technology in the nuclear industry faces the same difficulties as in any industry. The initial steps in the process design require information about the feed water quality and the end point of the separation, i.e. the desired quality of the permeate and retentate, in order to select a membrane to begin with pilot-scale testing with. [2]

A detailed characterisation of the feed water is an important part of the design of the membrane process. The characterisation often involves analysis of radioactivity, pH, dissolved substances (for example minerals and organic matter), salinity and colloids/particles. A well performed characterisation may identify potential foulants. How the feed water quality varies, the range of the important parameters, should also be determined in the characterisation. [2]

The goal of the separation in the nuclear industry is most commonly to reduce the activity contents. Membranes used for radioactive waste water treatment are often evaluated by their decontamination factor, DF. The DF often relates to the total activity of the permeate and feed but it can also relate to specific nuclides. This parameter is defined as equation 5. [2]

$$DF = \frac{A_{Feed}}{A_{Permeate}}$$
 (Eq. 5)

Fouling is a severe problem for membranes used in the nuclear industry since the foulants could be radioactive. This will lead to a build-up of active species on the membrane which will increase the dose to the surrounding area. Pre-treatment of the feed water to reduce the foulants and cleaning methods are therefore especially important questions in the design of membrane processes in the nuclear industry. Feed water in RO and NF systems is often pre-treated with MF or UF in order to reduce colloids and particles. Chemical cleaning methods creates waste which must be treated, pre-treatment and physical cleaning are therefore favourable. [2]

When cleaning waste water from nuclear facilities, the membranes will be exposed to radiation. This can lead to a decrease of lifetime of the membrane and decrease of performance since the radiation may alter the membrane structure. The risk of structural changes due to radiation is considered to be bigger for polymer membranes than for ceramic and metallic. [2]

#### 2.2.2 Examples of membrane technology installations

RO is used in many nuclear applications around the world for treatment of liquid waste. One application was developed in 1970 in Chalk River Laboratory, Canada. It consisted of two RO tubular membrane working in parallel. The feed contained suspended solids which resulted in fouling. Sponge balls were used for membrane cleaning. Decontamination factors were high (>1000) for <sup>85</sup>Sr, <sup>134</sup>Cs and <sup>60</sup>Co. [16]

A well-known application for NF is recovery of Boron from liquid waste. NF enables separation of ions, mainly monovalent ions from multivalent once. In Bugey NPP, France, NF is used for separation of Boron and ionised silica from radioactive ions ( $\text{Sn}^{2+}$ ,  $\text{Ag}^+$ ,  $\text{Co}^{2+}$ ). The recovery of Boron is 16,5 % and 92% for Silicon. [17]

The relative large pores of UF membranes  $(0,001-0,1 \ \mu m)$  allow dissolved substances to pass through the membrane and hinders colloids and suspended matter. UF can used for application where the radioactive material is in colloid or suspended form. In RO-processes, UF can be used for pre-treatment. Another way to use UF is to bind radioactive ions with complex agents which can be rejected by the UF-membrane. [17]

MD has been used for cleaning of operation and production waste in pilot scale at the Institute of Nuclear Chemistry and Technology in Warsaw, Poland. Spiral-Wound modules were operated at feed-temperatures between 35-80 °C. High decontamination factors were reached during the experiments since there was almost 100 % rejection of the radioactive ions present in the feed. [18]

# 2.3 Analysis techniques

The theory behind the analysis techniques used in the execution of this project are described in this section. Section 3.1 gives a brief description of the methods of the analytical procedures.

#### 2.3.1 Gamma spectroscopy using a HPGe-detector

Emission of gamma rays, i.e. gamma decay, occurs as a result of transition between energy levels within a nucleus. When a radioactive nucleus decays, for example by alpha or beta decay, the daughter nuclide is often left in an excited state decays to its ground state by gamma decay, in one or several steps. The energy of the gamma rays takes specific values which can be utilized to identify the mother nuclide. [8, 19]

Gamma spectroscopy is the study of gamma rays over a wide energy spectra. The appearance of the spectra is affected by several factors which include the intensity of the sample and geometry of the detector. [19]

One type of detector used for gamma spectroscopy is High Purity Germanium, HPGe. It consists of a germanium (Ge). HPGe detectors have high resolution but low sensitivity (i.e. it takes fairly long time to generate a spectrum). [19]

The HPGe-detector is a semiconductor. When gamma rays are absorbed in the Ge-crystal, electron carries (which are pairs of electron-ions and electron holes) are created which give rise to electric current. This current is recorded and the voltage of the current is proportional to the energy of the incoming gamma rays. [19]

#### 2.3.2 Determination of Boron concentration

Boric acid,  $H_3BO_3$ , is used as a neutron poison for control of the reactivity in PWRs. Boric acid dissolves to Boron and B-10 absorb neutrons. The concentration of Boron acid is highest in beginning of cycle of PWRs (i.e. when the fuel is fresh) and decrease with time. Too high concentration of Boron in liquid radioactive waste is a problem since Boric acid inhibits the curing process of the concrete used for moulds<sup>1</sup>. [8]

A method of measuring the concentration of Boron is titration with addition of the sugar mannitol. Boric acid is a weak acid and together with mannitol it forms a complex which is a medium strength acid which has an equilibrium point around pH 8,5. [20]

#### 2.3.3 Determination of Silicic acid concentration

Silicon in aqueous solution normally takes the form as Silicic acid,  $SiO_2$ . NPPs which uses ocean water for cooling of coolant will have Silicic acid in the cooling system since Silicic acid is naturally abundant in the ocean. Silicic can also be released from construction materials in the cooling circuit<sup>1</sup>.

By adding ammonium molybdate to Silicic acid (in the pH-range 1-2) a complex is formed. If this complex is reduced, a new complex with a clear blue colour is formed. The colour intensity of the solutions which contains the blue complex is proportional to the concentration of Silicic acid and is measured with spectrophotometry. [21]

#### 2.3.4 Conductivity

Electric current in aqueous solutions is transported by ions. Conductivity is a measure of a materials ability to transport current and the conductivity of a solutions is therefore related to the ion concentration. The SI unit of conductivity is Siemens per meter (S/m). [22]

<sup>&</sup>lt;sup>1</sup> According to Bernt Bengtsson, Senior Chemistry specialist at Ringhals AB. 2014-11-24

#### 2.3.5 Determination of Total organic carbon

Total organic carbon (TOC) is a measure of the total content of organic carbon in aqueous solution. Results from TOC analysis gives the measured value in concentration of carbon, e.g. mg organic carbon per litre. TOC, in a similar way as Boron, affects the curing process of concrete<sup>1</sup>. [23]

One method of determining the concentration of TOC is based on oxidation of TOC to carbon dioxide,  $CO_2$ . The produced  $CO_2$  is transferred to a liquid indicator and a change in colour due to acidification will occur which is measured with spectrophotometry. [23]

#### 2.3.6 pH

pH is the negative logarithm of the chemical activity of hydrogen ions in a solution. It can be measured with a pH-meter which basically consist of a probe and electronic meter. The probe measures the electro-chemical potential between a known liquid (which is inside the probe) and the solvent on the outside. [24]

#### 2.3.7 Particle counting

The amount of dissolved particles in the reactor coolant is monitored at Ringhals using a method based on a laser technique. A continuous flow of coolant is lead through the monitor. When particles passes the laser, the intensity of the light changes which enables particle counting (number of particles per mL) in the range 2-900  $\mu$ m. The method is not validated and only used for trends of the size distribution of the particles. [25]

#### 2.3.8 Ion Exchange process

Reactions which involve ion exchange may be defined as the reversible exchange of ions between a solid phase and a solute. The solid phase is the ion exchanging (IX) material which is insoluble to the solution. If the solutions contains cations,  $A^+$ , and the IX of a fixed material,  $M^-$ , and another type of cation,  $B^+$ , as the exchanging ions, the reaction which takes place can be represented by the following equation:

$$M^{-}B^{+} + A^{+} \rightleftharpoons M^{-}A^{+} + B^{+}$$
(Eq. 6)

Equation 6 describes the exchange of cations. This type of IX, which has a cations as the exchanging ion, is called cation IX. Commonly, cation IX exchange  $H^+$ . If the IX consist of a fixed material which is positively charge and can exchange anions, the IX is of anion exchange type and commonly exchange  $OH^-$ . [26]

Several types of IX exist but the most common type is synthetic organic polymer based resins functionalised with active groups. Their spherical shape and polymer-network structure makes the surface to volume ratio high which increase the capacity of the IX. [12]

<sup>&</sup>lt;sup>1</sup> According to Bernt Bengtsson, Senior Chemistry specialist at Ringhals AB. 2014-11-24

# 3. Experimental

The practical work was based on experiments with the membrane connected to the pilot plant and analysis in laboratory (i.e. Ringhals chemistry department RDC1). Decanted water were transferred to tanks (i.e. a feed tank) which were connected to the pilot plant, see the schematic representation in figure 7. A sample of the decanted water from each tank was taken and used for characterisation. During an experiment with the pilot plant samples of permeate and concentrate were taken. After the experiment, when the level of the feed water was too low, the procedure were repeated for a new experiment. The permeate and concentrate samples as well as the characterisation samples were analysed in the RDC1-laboartory with methods presented in section 3.1.



Figure 7. An overview of the execution of the project.

# 3.1 Method of analysis

All analysis has been performed at Ringhals chemistry department GR-RDC1.

#### 3.1.1 Gamma spectroscopy with HPGe-detector

Gamma spectroscopy has been conducted using HPGe-detectors of two types; GEM-25P4-76-SMP (7 pcs) and GEM-20190 P (1 pcs). Both types were manufactured by ORTEC. The GEM-20190 P detector was electrically cooled whereas the others were cooled with Nitrogen. Calibration control (energy and peak shape) was performed 2-3 times per week by the chemistry engineers working at Ringhals. A full calibration was performed only when larger events affects the detectors (for example loss of vacuum).

All detectors uses the software GammaVision v.6.08 which was programed to calculate the uncertainties for every peak. The program was based on standard methods and used daily at Ringhals. Background radiation was measured every week for 28 h and the results were added to GammaVision which automatically does background corrections.

All samples (which are described in section 3.3 to 3.5) have been analysed in 50 mL Cerbo containers which was a standard geometry used for HPGe measurements. The samples were shaken and 50,0 g ( $\pm$ 0,05 g) was transferred to the Cerbo containers by the assumption that 50,0 g corresponds to 50,0 mL.

Measuring time depended on the radioactivity level of the sample and was decided after discussion with the chemistry engineers which were working with Gamma spectrometry. A longer measuring time results in lower uncertainties but was not always possible. In general were the permeate samples measured for 3-8 h and the feed and concentrate samples measured for 2-3 h. The samples after the sequence of filtrations (which is described in section 3.1.7) were all measured for 3 h.

The results from the HPGe measurements were given as the activity in Bq/L together with relative uncertainty for each nuclide. When adding uncertainties (i.e. the uncertainty of the total activity of the sample) a Kragten spreadsheet was used. [27]

#### 3.1.2 Determination of Boron concentration

Analysis of Boron were conducted using Mettler Toledo T70 with the method BORpH1. All samples were analysed using this method. The chemistry engineers at Ringhals calibrate the equipment once a week. The uncertainty for the method was 2% at 20 °C.

#### 3.1.3 Determination of Silicic acid concentration

Concentration of Silicic acid was determined with spectrometry with the instrument DR5000 from Hach Lange and the method 645 Kisel ULR. The instrument was calibrated by Hach Lange. At a concentration of 500  $\mu$ g/L was the uncertainty 2,6 %.

#### 3.1.4 Conductivity

The instrument WTW inoLab Cond 730 with the electrode LR 325/01 were used to measure conductivity. The electrode had an uncertainty of 2 % at 25 °C.

#### 3.1.5 Determination of Total organic carbon

TOC concentration was measured with DR3900 with kit LCK385 (3-30 ppm) and LCK386 (30-300 ppm) manufactured by Hach Lange. The method has an uncertainty of 4,6 %.

#### 3.1.6 pH

Measurements of pH were conducted using WTW inoLab pH 730. The electrode was calibrated on a weekly basis using standards at pH 4, 7 and 10. Calibration control using the standards was performed every day the electrode was used. The accuracy of the electrode was  $\pm 0.01$  in pH range 0-14.

#### 3.1.7 Filtration with Millipore and Vivacell100

A sequence of filtrations was performed according to figure 8. Approximately 200 mL of the sample was filtered with a 10  $\mu$ m Millipore-filter. 50 mL of the permeate was removed for HPGe-analysis. The remaining 150 mL was then filtered with a 0,8  $\mu$ m Millipore-filter. Vivacell100-filters with a MWCO of 300.000 Da and 100.000 Da were used after the 0,8  $\mu$ m Millipore-filter. The Millipore-filters were disposable items whereas the Vivacell filters were recyclable. The used Vivacell filters was cleaned with SQ-water and leached over night between the filtrations.



Figure 8. Evaluation of size distribution of radioactive substances.

#### 3.1.8 Particle counting

The size distribution of particles was evaluated with the particle counter Chemtrac PC24001 with the software Trac Ware 2001. This method gives the amount of particles per volume in the range 2-4  $\mu$ m, 4-7  $\mu$ m, 7-14  $\mu$ m, 14-20  $\mu$ m, 20-25  $\mu$ m, 25-100  $\mu$ m and 100-900  $\mu$ m. The Chemtrac device was not calibrated and was therefore not used to record the actual amount of particles but only to monitor the size distribution and comparison between samples.

# 3.2 The pilot plant

The pilot plant was a test bed used for projects involving evaluation of membrane filtration. It has been used in many projects, such M. Hjelmberg, née Kallerfelt, 2010 and K. El Tayara, 2013, and can be adjusted to fit a certain application and membranes. An instruction for operating the pilot plant is given in Appendix B where the original process scheme is included. Pictures of the pilot plant and the membrane are given in Appendix C. Product data sheets of the membrane, its housing and the BPH are also included in Appendix C.

A simplified process-scheme of the pilot plant is given in figure 9. The feed (green line) enters the membrane from beneath and divides as retentate and permeate. A tank collects the permeate and the retentate was recirculated. Because of the recirculation, the rejected species were concentrated in the feed/retentate.



Figure 9. A simplified process-schematic of the pilot plant.

#### 3.2.1 The membrane

The membrane which was connected to the pilot plant was a tubular cross flow membrane. It had a surface area of  $0,3 \text{ m}^2$  in total and had a pore size of 70.000 Da. Its SiC surface maked it hydrophilic. The membrane resided in a stainless steel housing with openings for feed, permeate, concentrate and BPH. Both the membrane and the housing as well as the BPH were manufactured by LiqTech International.

#### 3.2.2 Back pulse hammer

The back pulse hammer, BPH, was connected to the membranes permeate side in the middle of the cartridge, see figure 9. It was driven by air at 3 bar.

During operation, the BPH was filled with permeate and pressurised air. When a pulse was about to start, BPH control system throttle back the permeate flow (the blue line in figure 9) and shortly after send a pulse of permeate and air which had resided in the BPH over the membrane. During the pulse no permeate was produced.

#### 3.2.3 Dose monitoring

Probes were attached to the top and bottom of the membrane housing in which the membrane resides. The probes were measuring the dose rate on the membrane which enabled monitoring of activation build up. Dose rate from background radiation was also measured with a probe which was placed in the same room as the pilot plant at a distance around 5 m from the membrane. The probes were connected to a measuring station, Probe-multiplexer 861,1, which in turn was connected to a computer which uses the software PROMUX-S to record the dose rate. The probes was of type Gamma Probe 6150 AD 18.

Dose rate was also measured with a GM device of type Automess 6150 AD 6R. The GM was used to measure dose on the feed tank, permeate tank and the membrane.

#### 3.2.4 Operation parameters

The operating parameters for the pilot plant and BPH were:

- Operation pressure
- Driving force
- Permeate flow
- Difference pressure
- Time interval

Operation pressure refers to the pump and how "hard" it is working. The pump was frequency controlled at 17 Hz and with an operation pressure of 1,0 bar resulting in a flow of approximately 1600 L/h.

The driving force was the difference in pressure on the permeate and feed/retentate side of the membrane. A large permeate flow require a large driving force. Difference pressure was the pressure difference over the length (from inlet to outlet of the feed/retentate) of the membrane. The BPH "shoots" a pulse of permeate, the time between the shoots was referred to the time interval for the BPH.

# 3.3 Experiments using the pilot plant

In total were three tanks of decanted water connected to the pilot plant and used for experiments. The first tank was used for two experiments whereas the other two tanks were used for one experiment each, see figure 10.



Figure 10. A schematic overview of the experiments using the pilot plant.

#### 3.3.1 Tank 1

Approximately 800 L of decant water was transferred to a tank i.e. the feed tank. The water was filtered with a 120  $\mu$ m-filter and the tank was connected to the pilot plant. The water in the tank was characterised and used for test 1A and test 1B.

A sample of approximately 1 L was taken from the top of the tank by hand and used for characterisation. The stirring device had been on for around 10 minutes before the sample was taken.

50 mL of the sample was measured with HPGe for 3 hours. Analysis of Boron, Silicic acid, pH, conductivity and TOC were carried out according to the method instructions. The sample was also analysed with the sequence of filtrations described in chapter 3.1.7 and the particle counter.

#### 3.3.2 Test 1A - Flux test

In test 1A were different permeate flows tested. The pilot plant was operated with constant values of operation pressure (1,00 bar), driving force (0,50 bar) and difference pressure (0,5 bar). There was no concentration of the feed since the produced permeate was returned to the feed tank. The concentrate had a constant flow of 260 L/h. Samples were taken during the test according to table D1 in Appendix D.

During test 1A, only the top probe was used since the bottom probe had not yet been installed on the membrane housing. A reversed flush of  $3 \times 20$  seconds was preformed after the test

#### 3.3.3 Test 1B - Concentration of the feed

Test 1B was carried out over four days with a total operation time of 20 h. The permeate was sent to an empty tank; the permeate tank as seen in figure 9. The operation parameters were kept constant during the test; The operation pressure at 1,00 bar, driving force at 0,50 bar, difference pressure at 0,5 bar and permeate flow on 30 L/h. Table D2 in Appendix D lists the samples taken during test1B.

The dose rate was measured with GM at each start up, at every sample time and before and after each reverse flush operation. Reverse flush of  $2 \times 10$  was preformed after day 1-3 and  $3 \times 20$  after the last day. The probes were also used to monitor the change of dose rate.

#### 3.3.4 Tank 2

Decant water was pre-filtered with the same 120  $\mu$ m-filter which was used for tank 1, and transferred to a tank. The dose of the 120  $\mu$ m-filter increased to 5 mSv/h and was therefore removed to safe storage after the filtration was completed. Tank 2 contained approximately 700 L of water and was connected to the pilot plant and used for test 2.

A characterisation sample (0,8 L) was taken when the stirrer had been on for 15 minutes. 50 mL of the water was measured with HPGe for 3 h. Analysis of Boron, Silicic acid, pH and TOC were carried out according to the method instructions. The size distribution of activity and particles were evaluated with the sequence of filters according to 3.1.7 and particle counting.

#### 3.3.5 Test 2 - BPH with 10 minute intervals

In this test the BPH was in operation for the first time. The time interval was set to be 10 minutes between the pulses. Other operation parameters were set to be equal as the operation parameters in test 1A (operation pressure at 1,00 bar, driving force at 0,50 bar , difference pressure at 0,5 bar and permeate flow on 30 L/h). The test was carried out over night with a total time of approximately 16 h. Sample of the permeate and concentrate were taken during the first day according to table D3 in Appendix D. On the second day were sample taken from the permeate tank and the reaming water in the feed tank. The probes were monitoring the dose rate during the whole experiment.

Before test 2 was started, a different type of washing was tested. Water was lead to the bottom of the membrane in order to flush it ( $2 \times 20$  seconds) from beneath. The dose rate was monitored with the probes and GM before and after. A standard reverse flush  $2 \times 20$  seconds was also performed.

A week after test 2 was finished, a sample of the water which resided in the membrane was taken. The sample was analysed with HPGe (3 h) to see if any activity had been leached from the membrane.

## 3.3.7 Tank 3

Decant water was pre-filtered with a 120  $\mu$ m-filter and transferred to the feed tank. Tank 3 contained approximately 900 L of water and was connected to the pilot plant and used for test 3.

A sample of the feed tank was taken when the stirrer had been on for 15 minutes. As in the characterisation of tank 1 and 2, the sample of tank 3 was analysed with HPGe (3 h), Boron, Silicic acid, pH, TOC, conductivity, particle counter and the sequence of filtrations.

#### 3.3.8 Test 3 - BPH with 5 minute intervals

Test 3 was conducted with the BPH time interval of 5 minutes and operation parameters as in test 1B and test 2. The test lasted for approximately 23 h. Samples were taken according to table D4 in Appendix D. The membrane was exposed to reverse flush for 2×20 seconds before the test was started and when it was finished. During the test were the probes monitoring the dose rate.

## 3.3.9 Test 3 – IX experiments

Approximately 0,25 L of water from the permeate tank sample ('Permeate tank 3') was polished with Dionex cation IX resin. 20 mL of resin were transferred to a column (referred to as column 1) with inner diameter of 39 mm. 20 mL of sample was transferred to the column every minute. The water left the column with the speed of approximately 145 droplets per minute. The first 30 mL of sample which left the column was discarded and the rest collected. 50 mL of the collected sample was analysed with HPGe for 8 h.

A second test with Dionex cation IX resin was performed. Approximately 20 mL of resin were transferred to a column with inner diameter of 8,8 mm (referred to as column 2). Glass wool (approximately 3 cm in height) stopped the resin from exit the column. 0,25 L of sample from 'Permeate tank 3' was transferred to the column with the average speed of 10 mL per minute. The first 50 mL was discarded, the remaining 0,2 L was collected and used for HPGe measurement where 50 mL was measured for 8 h.

# 4. Results

This section give a summary of the results conducted by the analysis and experiments with the pilot plant. The raw data is found in Appendix E to H.

## 4.1 Characterisation of decant water

The gamma activities conducted by HPGe-analysis of the decant water were measured to hold  $3,03\cdot10^5$  (±1210) Bq/L in tank 1,  $6,76\cdot10^5$  (±6400) Bq/L in tank 2 and  $3,50\cdot10^5$  (±6000) Bq/L in tank 3, see figure 11. In all three tanks contributed Co-60 and Ag-110m to approximately 70 % of the total activity.



Figure 11. Gamma activities of the decant water conducted by HPGe-analysis.

The sequence of filtrations, based on using the Micron and Vivacell100 filters, decreased the gamma activities according to figure 12. The activity was in average (between tank 1, 2 and 3) reduced by 98 % after the 300.000 Da Vivacell100-filters.



*Figure 12. Change of gamma activities (in logarithm scale) of the decant water conducted by the sequence of filtrations.* 



From the analysis using the particle counter showed the results that a particle size of 14  $\mu$ m and smaller were dominating for the decant water. The size distribution of the three tanks of decanted water is presented in figure 13.

*Figure 13. The particle size distribution of the decant water conducted by the particle counter.* 

The average results of the analysis of Boron, Silicic acid, pH, conductivity and TOC of tank 1, 2 and 3 are given in table 2. Raw data from the analysis is given in Appendix E.

1 dole 2. 1 mary sis results of decant water.					
Analysis	Tank 1	Tank 2	Tank 3	Unit	Uncertainty
Boron	360	391	396	mg/L	2 %
Silicic acid	1968	2113	2028	μg/L	2,6 %
pН	6,97	5,62	5,53	-	±0,01
Conductivity	26,2	22,4	3	μS/cm	2 %
TOC	4,9	6,7	6,6	mg/L	4,6 %

Table 2. Analysis results of decant water

#### 4.2 Test 1A – Flux test

DF for the different permeate flows was calculated using equation 5. The activities of the feed were taken from the characterisation sample and divided by actives of sample P1.1 to P1.8. How the total DF and DF of the main nuclides differ at the different permeate flows is showed in figure 14. A permeate flow of 30 L/h was chosen for the following tests.



Figure 14. Values of DF conducted from test 1A.

A flow of 30L/h results in the flux:

$$J = \frac{30 L/h}{0.3 m^2} = 100 \frac{L}{hm^2} = 2,78 \cdot 10^{-5} \frac{m}{s}$$
(Eq. 7)

Conversion for the flow of 30 L/h can be calculated according to equation 3.

$$C = \frac{30}{1600} = 0,019 \tag{Eq. 8}$$

The increase of dose rate during test 1A is represented in figure 15. The test was executed during two shifts with a break in between. One can see how the dose rate decreases after the second shift which can be explained by the reverse flush operation performed after the second shift.



*Figure 15. Change of dose rate from the top probe during test 1A. The test started at 9.15 continued to 15.30 with a break between 11.30 and 13.30.* 

#### 4.3 Test 1B - Concentration of the feed

The total gamma activity of the permeate tank sample was  $4,91\cdot10^3$  (±51,7) Bq/L. Using equation 5 and the activity of tank 1 conducted by the characterisation, the DF for tank 1 can be calculated.

$$DF = \frac{3,03 \cdot 10^5}{4,91 \cdot 10^3} = 62$$
 (Eq. 9)

In equation 10 an activity balance is described, which is derived from equation 4. A is the activity concentration, V is the volume of feed, permeate and retentate (i.e. concentrate) and  $L_{system}$  is losses of activity due to deposit in the system (i.e. the pilot plant).

$$A_F V_F = A_P V_P + A_R V_R + L_{system}$$
(Eq. 10)

The percentage of the activity which is lost in the system can be calculated according to equation 11.

$$\Theta = \left(1 - \frac{A_P V_P + A_R V_R}{A_F V_F}\right) \cdot 100$$
 (Eq. 11)

An estimation of the volume of the reaming water in the feed tank after test 1B was done. Approximately 800 L of the water in tank 1 had been reduced to 50 L (the permeate tank was estimated to contain approximately 750 L). The volume reduction of tank 1 was therefore 93,75 %.

The activity of remaining water in the feed tank was  $3,93 \cdot 10^5$  (±3010) Bq/L. By using the estimations of the tank volumes and the measured activity concentrations, the loss of activity in the system can be calculated using equation 11:

$$\Theta = \left(1 - \frac{750 \cdot 4,91 \cdot 10^3 + 50 \cdot 3,93 \cdot 10^5}{800 \cdot 3,03 \cdot 10^3}\right) \cdot 100 = 90\%$$
(Eq. 12)

Test 1B was executed during 4 days; 16-th of March to 19-th of March. 4 steps of increased dose rate can be noted in figure 16 which shows the dose rate measured by the bottom and top probe. The dose rate measured by the bottom probe increase from 320-550  $\mu$ Sv/h and the top probe from 55 to 200  $\mu$ Sv/h.



Figure 16. Dose rate build up during test 1B.

#### 4.4 Test 2 - BPH with 10 minute intervals

Sample 'Permeate tank 2' held an total gamma activity of  $4,03 \cdot 10^2$  (±15,5) Bq/L and the sample 'Concentrate tank 2' held  $1,01 \cdot 10^7$  (±32200) Bq/L. The DF for the water used in test 2 is calculated according to equation 4:

$$DF = \frac{6.76 \cdot 10^5}{4.03 \cdot 10^2} = 1700$$
 (Eq. 13)

The volume of concentrate left in the feed was estimated to be 40 L. As in equation 11, the percentage of activity deposit can be estimated to:

$$\Theta = \left(1 - \frac{40 \cdot 1,01 \cdot 10^7 + 660 \cdot 4,03 \cdot 10^2}{700 \cdot 6,76 \cdot 10^5}\right) \cdot 100 = 15\%$$
(Eq. 14)

The change of dose rate during test 2 is represented in figure 17. At the start of the test measured the bottom probe approximately 450  $\mu$ Sv/h and the top probe 150  $\mu$ Sv/h. The test ended around 3 Am and the increase of dose rate stopped and was then stable around 800  $\mu$ Sv/h for the bottom probe and 480  $\mu$ Sv/h for the top probe. In the morning, around 8 AM, a reverse flush was preformed which correspond to the decrease of dose rate. The reverse flush was most effective to decrease the dose at the top of the membrane. It can be noted that the increase of dose rate was fastest close to the end of the test.



Figure 17. The dose rate during test 2 monitored by the probes.

The leaching sample taken after test 2 held an activity of  $3,29 \cdot 10^5 (\pm 44000)$  Bq/L.

#### 4.5 Test 3 - BPH with 5 minute intervals

The sample from the third permeate tank ('Permeate tank 3') was measured to hold  $4,42\cdot10^2 (\pm 42,6)$  Bq/L and the sample 'Concentrate tank 3' 5,89 $\cdot10^6 (\pm 20200)$  Bq/L. The DF of tank 3 is calculated according to equation 4:

$$DF = \frac{3,50 \cdot 10^5}{4,42 \cdot 10^2} = 800$$
 (Eq. 15)

Approximately 50 L of water remained in the feed tank after the test. The activity deposit in the system can be estimated according to equation 11:

$$\Theta = \left(1 - \frac{50 \cdot 5,89 \cdot 10^6 + 850 \cdot 4,42 \cdot 10^2}{900 \cdot 3,50 \cdot 10^5}\right) \cdot 100 = 6\%$$
(Eq. 16)

At the start of the test 2 (10:30 AM) the bottom probe measured approximately 700  $\mu$ Sv/h and the top 300  $\mu$ Sv/h, see figure 18. At the end of the test, the bottom probe measured 850  $\mu$ Sv/h and the top 420  $\mu$ Sv/h. The reverse flush decreased the dose rate to approximately 780  $\mu$ Sv/h for the bottom and 370  $\mu$ Sv/h for the top probe.


## 4.6 Polishing with ion exchange resin

Results of total gamma activity and pH after the IXR polish are given in table 3. The used sample 'Permeate tank 3' held an activity of  $4,42 \cdot 10^2$  ( $\pm 42,6$ ) Bq/L before the polishing. In the experiment with Column 1, the activity was reduced by approximately 50 %. Reduction of activity was approximately 40 % for Column 2.

Analysis	Column 1	Column 2
Activity (Bq/L)	$2,19\cdot10^2$ (±14,7)	$2,59\cdot10^2$ (±15,5)
Activity reduction (%)	50,4	41,4
pH before the IXR	5,	75
pH after the IXR	4,36	4,43

Table 3. IX experiment with sample 'Permeate tank 3'.

## 4.7 Decontamination

The main nuclides found in the decanted water, permeate and concentrate were Mn-54, Co-58, Co-60, Ag-110m and Sb-124. Specific values of DF for these nuclides are found in table 4 which also include their half-lives.

Nuclide	Test 1 B	Test 2	Test 3	Average of test 2 and 3	Half-life [28]
Mn-54	24	5300	1000	3100	312,2 d
Co-58	18	850	640	740	70,86 d
Co-60	120	3300	1700	2500	5,27 a
Ag-110m	54	1600	1000	1300	249,9 d
Sb-124	50	140	87	110	60,3 d
Total	62	1700	790	1200	-

Table 4. Nuclide specific DF-values.

## 5. Discussion

This section discuss the results based on the aims of the project.

## 5.1 Membrane technology

Membranes have been used in the nuclear industry for many years. Several examples of membrane based separation system exist world-wide. Their low energy consumption and range of applications makes the future look bright for membrane technology.

In the design of membrane based separation system is the characterisation of the future feed water is the alpha and omega. An extensive characterisation can save both money and time by avoiding problems which can arise by choosing a membrane which is not suitable for the application.

When choosing a membrane the demands of the separation of the separation must also be considered. The demands can involve economic considerations, energy consumption, limitation of secondary waste and environmental demands (which can involve limit of allowed activity in the permeate).

## 5.2 Measurement uncertainties and sampling

As mentioned in the delimitations has the focus of the measurements been to see the differences between feed, concentrate and permeate. No duplicates of samples were taken during the experiments with the pilot plant. Duplicates would resulted in handling of more activity which is unfavourable in respect to safety concerns (ALARA). It would also had led to a higher pressure on the resources of the Chemistry department and it was not certain that there would have been enough time to analyse all the samples.

Samples from the feed and permeate tanks and the sampling sites on the pilot plant were taken in a similar manner. Before samples were taken from the sampling sites the valves and piping were washed by discharge of the first sample volume in order to remove possible deposits. Samples from the permeate tanks cannot be considered completely representative due to the absence of stirring. However, the permeate was considered to be homogeneous due to absence of visible sedimentation.

While performing any analysis, the methods were followed and the same set of scales and pipette were used in order to decrease the variance of uncertainties of the samples. Due to the precision of the laboratory work it can be assumed that the major contribution of uncertainties originated from the sampling process.

## 5.3 Characterisation of the decanted water

Table 2 and figure 11-13 give the results from the characterisation of the decanted water. The values vary between the three tank samples which can be explained by non-homogenous conditions in the decanted water tank.

The sequence of filtrations gave an indication of the size of particles which radioactive material were attached to. From figure 12, it can be seen that the activity of the decant water is not as affected by the 10  $\mu$ m-filter in comparison with the 0,8  $\mu$ m-filter. Only a few percent of the activity is left after the 300.000 Da and 100.000 Da filters. It appears that the main part of the activity in the decant water is attached to particles smaller than 10  $\mu$ m and larger than 300.000 Da. This is consistent between all the feed tanks.

## 5.4 Choosing operation parameters

The operation parameters (operation pressure, driving force and difference pressure) were chosen after consultation with C.H Hansson at NORDCAP who also provided the membrane.

Test 1A was performed to see how DF of different permeate flows varied. The DF were calculated by using the activity in the permeate samples taken during the test and the activity in sample from the characterisation. Calculating the DF in this way is not completely correct since the activity of the feed is assumed to be constant. Figure 14 shows the change of dose rate during test 1A and the increase is likely from deposits of active substances on the membrane which in turn change the activity of the feed.

## 5.5 Decontamination of the decant water using the SiC membrane

DFs were calculated for tests 1B, 2 and 3. The activities of the feed were in these calculations the activity conducted by the characterisations of tanks 1, 2 and 3. Since the feed water to the pilot plant is concentrated during the experiments, the value of DF will change with time since the activity of the feed is increasing. The time-dependence of DF has not been evaluated since it has not been the aim of the experiments.

The DF increased between experiments 1B and 2. One explanation could be fouling on the membrane. When the foulants attach to the membrane surface it is likely that some material will attach inside the pores. This will make the pores smaller thus more difficult to pass. The DF value of test 3 was smaller than for test 2. Both test reached approximately the same value of the activity in the permeate tank  $(4,03\cdot10^2 \text{ Bq/L} \text{ for tank 2 and } 4,42\cdot10^2 \text{ Bq/L} \text{ for tank 3})$  but tank 3 held only 50 % of the activity in the feed tank in comparison to tank 2.

The DFs of test 1B, 2 and 3 are given in table 4. Test 1B differ from test 2 and 3 because the operation of the BPH and is therefore not included in the average DFs. Ag-110m has lowest DF in test 1B, 2 and 3. One hypothesis is that Ag-110m is in ionic form or bound to small particles which can pass the membrane readily. To confirm this hypothesis, the gamma activities after the sequence of filtrations of the decanted water for Ag-110m were plotted and compared with Co-60, see figure 19. From the figure it seems like the activities of Ag-110m and Co-60 are equally decreased by the 300.000 Da and 100.000 Da filter which contradict the hypothesis. More analysis of how the radioactive nuclides are bound to the particles, especially for Ag-110m, would be needed to explain the low DF of Ag-110m.



Figure 19. Decrease of gamma activity after the sequence of filtrations of Ag-110m and Co-60.

A permeate flow of 30 L/h were used in experiments 1B, 2 and 3. Due to the fouling (i.e. increase of dose rate on the membrane) it is likely that a higher value of the permeate flow would still give a high DF of experiment 1B, 2 and 3.

## 5.6 Evaluation of the Back pulse hammer

The change of dose rate during tests 1B, 2 and 3 are given in figures 16, 17 and 18. By comparing the average starting values for the bottom and top probes with the average values after test 1B, 2 and 3, one can observe that the increase of dose rate was slowest in test 3. Since test 1B was performed during 4 days with reversed flush after the second and third day, a straight forward comparison of the increase of dose rate from test 1B with test 2 and 3 is not appropriate. It should also be noted that the experimental conditions differed between the experiments in regard to the activity in the feed water and the initial dose rate of the membrane.

From figure 16 it can be noted that the dose rate is slightly decreasing between the days at which 1B were performed. By studying figures 16 and 17 as well, it can be noted that the starting dose rates of test 3 are a little lower than the final dose rates from test 2. The sample of fluid taken from the membrane after test 2 held an activity of  $3,29 \cdot 10^5$  (±44000) Bq/L. An observation is that some of the activity attached to the membrane is leached and ends up in the stagnant fluid in the pipe work of the pilot plant while it is not running.

By looking at figure 17 it can be noted that the increase of dose rate looks linear during the first 12 h of test 2, the slope is then increasing and takes an exponential shape. This appearance is similar to the increase of dose rate during test 3 (figure 18) which is fairly linear the first 13 h. It seem as if the BPH manages to keep the dose rate increase constant to a certain point and then the concentration of the feed is too large. It would therefore be interesting to investigate at which particle size distribution of the feed this point occur. One way could be taking more samples of the concentrate during the experiment and perform the sequence of filtrations, SEM-analysis and the particle counting. Another interesting thing to investigate would be to change the time interval of the BPH during an experiment. For example, running the pilot plant as in test 3 but change the BPH time interval to 2-3 minutes the last 3-5 h of the experiment.

It was observed during experiment 2 and 3 that the dose rate increased, measured using the GM device, was lowest at the middle of the membrane. The BPH was placed in the middle which explains the observation. It would be interesting to investigate a SiC membrane with more than one BPH placed along the membrane.

The BPH time interval of test 2 was randomly chosen. In test 3, a shorter time interval was chosen in order to evaluate if the BPH could decreased the change of dose rate observed in test 2. However, a shorter time interval will increase the operation time (i.e. how long it take for the tank of decant water to be processed) which could lead problem if time is an issue. Choosing the time interval is mainly an optimisation problem.

Equations 12, 14 and 16 estimate the activity deposits in the system, which decreased from test 1B to test 3. It is assumed that the activity deposition mainly occurs on the membrane surface but deposition inside the pump is also likely. Equations 12, 14 and 16 use volumes which were estimated by measuring the dimensions of the tanks and estimate how far from the upper edge the water surface were. The decrease of activity deposit from test 1B to test 3 could be explained by the operation of the BPH. Another possibility is covering of the sites where deposit could occur.

## 5.7 Analysis of permeate and concentrate

One of the most important things to avoid was a shift in concentration of Boron in the concentrate since Boric acid can delay the curing process of concrete. By comparing concentrate tank samples to characterisation samples, no change in concentration has been observed which is a positive result.

Concentration of TOC and Silicic acid were noted. TOC increased approximately 5 times for tank 1 and 2 and 28 times for tank 3. This may cause problems in the curing process of the moulds and should therefore be further investigated. The concentration of Silicic acid was not as large, the concentrate tanks had on average 1,25 times higher concentration than the characterisation samples.

Analysis of pH and conductivity showed an increase of pH in the permeate and decrease of the concentrate, and the opposite for conductivity. Further analysis of the particles in the decanted water (perhaps by Scanning electron microscope, SEM, analysis) would be needed in order to understand the chemistry of the particles and how they interact with the membrane. In turn it could lead to an explanation of the change of pH and conductivity. According to M. Devlin at Vattenfall AB, TOC in the decanted water consists partially of organic acids with low pKa. Increasing the concentration of these acids will lower the pH but could also shift their chemical equilibrium towards dissociation which in turn will lower the pH and increase the conductivity.

Only the sample 'Permeate tank 3' was used with the IXR because too little sample volume from the other permeate tanks were available. The decrease of pH indicated that ion exchange was occurring but it does not tell if the exchange is with radioactive species. Table H4 in appendix H gives the result from the sequence of filtrations for sample 'Permeate tank 3'. One can note that the activity after 100.000 Da filter had a lower value than the activity after the IX experiments. The main part of the activity seems to be in particulate form. It is possible that the particles in the permeate sticks to the resin without binding, i.e. physically hindered by the resin. One way to reduce the activity of the permeate would be to first filtrate it with the sequence of filtrations, and then use the IXR for polishing.

## 5.8 Comparison with the projects of El Tayara and Hjelmberg

El Tayara and Hjelmberg both cleaned decant water in their projects. Hjelmberg did not characterise the feed water which makes it problematic to compare the results from this project with Hjelmbergs results. The characterisation performed by El Tayara showed similar values of total gamma activity and is given in table 5. [10, 11]

Analysis	Average result
Total gamma activity (Bq/L)	$3,1\cdot10^5 (\pm 1000)$
Boron concentration (ppm)	568,4
pH	6
Conductivity (µS/cm)	3

 Table 5. Characterisation performed by El Tayara. [11]

El Tayara evaluated two HF membranes specified in table 6. The permeate flow was 80 L/h in all experiments which corresponds to a flux of 34,8 L/hm<sup>2</sup>. The flux over the SiC membrane was almost 3 times higher than the flux over the HF-membranes. [11]

Tuble 6. The fir memorales evaluated by Er Tuyara. [11]				
	Membrane A	Membrane B		
MWCO	50.000 Da	5000 Da		
Membrane surface	$2,3 \text{ m}^2$	$2,3 \text{ m}^2$		

Table 6. The HF membranes evaluated by El Tayara. [11]

The HF membranes used by El Tayara had 8 times larger surface area than the SiC membrane. Generally, a larger membrane gives a better performance and hence a larger DF. However, a larger membrane requires more chemicals (if cleaned chemically) and cost more.

Figure 20 and 21 show the average activity concentration of nuclides in the feed water and permeate conducted by El Tayara. The feed samples have a similar nuclide composition as the characterisation samples of tank 1, 2 and 3. [11]



*Figure 20. Average activity concentration of radionuclides in the feed processed by HF membrane A and permeate.* [11]



*Figure 21. Average activity concentration of radionuclides in the feed processed by HF membrane B and permeate. [11]* 

Given figures 20 and 21, the permeate samples were estimated to hold approximately 3200 Bq/L (membrane A) and 2720 Bq/L (membrane B). Using these values and the average value of the activity in the feed water given in table 5, the DF for membrane A is estimated to be 97 and 114 for membrane B. The average DF-value of the SiC membrane is approximately 12 times higher than for the HF membranes.

Specific DF values estimated by the activity concentrations in figure 20 and 21 are given in table 7. By comparing these values with the DF-values given in table 4, it seems as if the SiC membrane is better to separate Mn-54 and Co-60. The HF membrane shows a good separation of Ag-110 m. Sb-124 is the nuclide which has the lowest DF for both the HF membranes and the SiC membrane.

Nuclide	50.000 Da	5000 Da	Test 2	Test 3
Mn-54	-	133	5300	1000
Co-58	22	7	850	640
Co-60	1000	125	3300	1700
Ag-110m	1666	2000	1600	1000
Sb-124	80	13	140	87

Table 7. Nuclide specific DF-values of the HF-membranes and test 2 and 3. [11]

Hjelmberg used two Spriral-Wound modules with MWCO at 25.000 Da and 1000 Da. The average DF values are estimated to be 25 and 11 respectively which are far lower than the average DF value of the SiC membrane. [10]

Hjelmberg tested a mixed bed IX in laboratory scale and reduced the activity of a permeate sample from  $1,76 \cdot 10^3$  to 10,4 Bq/L. The sample had a conductivity of 39,3 µS/cm (before the ion exchanger). El Tayara had an average permeate activity of  $2,5 \cdot 10^3$  Bq/L which were reduced to an average of 54 Bq/L. Both mixed bed and cation IX were tested, with and without recirculation. The largest reduction of activity El Tayara reached was with a mixed bed ion exchanger with recirculation. [10]

El Tayara used reversed flush with water to clean the membrane after each batch. HF-membrane A was also cleaned with chemical cleaning with an alkaline solution (pH 12,5) followed by an acid solution (pH 2) after the final batch. It is problematic to compare the change of dose rate during the experiments performed by El Tayara since no probes were used and that the GM-device was only used at one position on the HF-membranes. However, El Tayara draws the conclusion that reversed flush is an effective method for cleaning the HF-membranes. The chemical cleaning was also considered to be an effective method. Hjelmberg did not performed any cleaning between the batches which resulted in dose rates of approximately 6,8 mSv/h. The BPH has the benefit to continuously clean the membrane and could easily be combined with both reverse flush and chemical cleaning. [11]

## 6. Conclusion

Membrane technology is used world-wide for treatment of liquid radioactive waste. The low energy consumption and specific application makes it superior over other separation processes. However, their lack of flexibility and problems with fouling are drawbacks which can be difficult to manage. The choice of membrane, which is based on characterisation of the feed water, is crucial for minimizing the effects of the mention drawbacks.

Reducing the activity in the decanted water using the SiC membrane seems to be a promising method. The average DF value is 12 times higher than for the HF membranes evaluated by El Tayara and 30-70 times higher than for Spiral-Wound membranes tested by Hjelmberg.

No concentration change of Boron was observed during the experiments. TOC, Silicon acid and conductivity did slightly change concentration in the retentate stream.

The BPH was operated continuously together with the SiC membrane. When operating with a time interval of 5 minutes the increase of dose rate was fairly low in comparison with a time interval of 10 minutes and operation without the BPH. It is hard to draw any distinct conclusions since the experimental conditions were not constant. However, indications from the experimental results shows that the BPH can be used to control the increase of dose rate during experiments with the SiC membrane.

Two benefits with the BPH are that it can operate continuously and that there is no addition of water to the system. One drawback is that the pulse-sequence takes some time which prolong the total operation time.

Further prospects for continued evaluation of the BPH are to test two or more BPH devices on the same membrane and to change the time interval when concentrating the feed water. The deposit of activity in the system could be better evaluated by replace the pump.

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# Appendix A

## Radioactivity

In the nucleus of atoms are protons and neutrons compressed to a tiny volume. Since protons are positively charged, repulsive forces are constantly present in the nucleus. Isotopes of atoms with the "right" amount of neutrons is capable to stabilise the nucleus even though the protons acting repulsive against each other. However, some isotopes do not have the right amount of neutrons in comparison to the amount of protons. The repulsive forces in these kind of isotopes will at some point overcome the forces which holds the nucleus together and fragments nucleus are then spontaneously ejected. The nucleus undergoes decay in order to reach a stable state. [23]

A nucleus can decay in several ways and often in many steps before it reaches a stable state. Two types of decay is  $\alpha$  (alpha) and  $\beta^{-}$  (beta) decay. When radon-222 (Rn-222) decay, an  $\alpha$ -particle is ejected from the nucleus and the radon atom is transformed to a polonium-atom with mass number 218. This nuclear-reaction can be written as: [23]

$$^{222}_{86}Rn \rightarrow ^{218}_{84}Po + ^{4}_{2}\alpha$$

The radon atom is referred as the mother, and the polonium atom as the daughter. The daughter atom often ends up in an excited state, and emit photons (gamma rays) in order to reach a stable state. [23]

An example of  $\beta^{-}$  decay is when tritium, H-3, decays to He-3. The ejected particle is an electron. [23]

$$^{3}_{1}H \rightarrow ^{3}_{2}He + ^{0}_{-1}e$$

Both the radon atom and the tritium atom undergo spontaneous decay; they are said to be radioactive. Radioactivity is the physical reactions when atomic nuclei transformed to another type of nuclei by decay. The ejected fragments of the mother have enough energy to ionize material which comes in their way i.e.  $\alpha$  and  $\beta^{-}$  from the reactions above are examples of ionizing radiation. Gamma-rays is another kind of ionizing radiation. [23]

How fast an amount radioactive isotopes decay is often expressed as the half-life,  $t_{1/2}$ , of that particular substance. Half-life is the time needed for half of the initial number of atoms to decay. Some isotopes has half-life of several years whereas others less than a second. Another way to express radioactivity is with activity which is a measure of the number of decays during a time interval, divided by the length of the time interval. The SI-unit of activity is Bq (Becquerel) and 1 Bq is equal to one decay per second. Radioactive isotopes with long half-life has an lower activity than isotopes with shorter half-life. [23]

#### Dose

A material which is exposed to radiation will receive a dose. Absorbed dose, D, is a quantity representing the mean energy absorbed from the radiation per unit mass. The Si-unit is Gray (Gy) where 1 Gy equals 1J/kg. [6]

The damage of tissue caused by radiation does not only depend on the energy of the radiation. Different types of radiation (such as  $\alpha$ ,  $\beta$  and  $\gamma$ ) induce different effects in biological material. Also, different types of tissue have different sensitivity to radiation. Effective dose (H<sub>e</sub>) included these effects and is measured in Sievert (Sv) where 1 Sv=1 J/kg tissue. [6]

SSM set up dose limitations. To be able to estimate how long one can stay in a radiation field dose rate is often used to express the dose. Dose rate is the absorbed or effective dose received per unit of time and commonly expressed as mGy/h or mSv/h. [6]

#### Nuclear power

Fission is the nuclear reaction which is utilised in a NPP. When an atomic nucleus is spilt into smaller fragments, energy is released. This splitting-reaction is called fission. One isotope which can undergo fission is uranium-235 (U-235). The fission is induced by absorption of a neutron and one example of a fission reaction is given below: [23]

$${}^{235}_{92}U + {}^{1}_{0}n \rightarrow {}^{141}_{56}Ba + {}^{92}_{36}Kr + {}^{1}_{0}n + energy$$

In this particular example, 3 new neutrons and one barium and one krypton atom are created. The barium and krypton atoms are called fission-products. Examples of other fission products are Cs-137, I-131 and Sb-124. A self-sustaining chain reaction takes place if the created neutrons from the fission reaction is absorbed into other uranium atoms. [23]

A NPP consist mainly of rods with fuel, a cooling medium and turbines. One type of fuel is enriched uranium which means that amount of U-235 has increased by isotope separation from natural uranium (mainly consisting of U-235). The energy released by the fission is heating the cooling medium. In BWR, the cooling medium is water which boiling. The produced steam drives the turbines and electricity can be produced. Boiling is avoided in PWRs because of the high pressure. The energy is transferred by a heat-exchanger to another circuit of water which can boil and drive turbines. [23]

## **Appendix B**

Instruction for operation of the pilot plant given in Swedish.

#### Inledning

Riggen (pilotanläggningen) används till att göra experiment som involverar rening radioaktivt vatten med hjälp av ett membran. Syftet är att utvärdera olika membrans förmåga att avskilja radioaktivt material från vatten. Riggen består bland annat av rör och en pump som är membranet är kopplat till.

Kontaminerat vatten går från en tank in till membranet där flödet delas upp. En viss del av flödet kommer gå igenom membranet och samlas upp i en tank. Detta vatten kalls för permeat. Den del av det smutsiga vattnet som inte går igenom membranet kallas koncentrat (eller retentat). Koncentratet återcirkuleras och går tillbaka till tanken med det smutsiga vattnet.

#### Membranet

Det membran som just nu är inkopplat i Riggen är tillverkat av LiqTech. Själva membranet sitter i ett membranhus (se figur 1) tillverkar av rostfritt stål. Membranhuset har fyra öppningar för; inflöde, permeat, koncentrat och Back pulse hammer.

Membranets tvärsnittsyta visas i figur 2. Inflödet går in i kanalerna, tuberna, och pressat till viss del igenom den porösa ytan. Denna process styrs av en tryckskillnad mellan inflöde/koncentrat och permeat, dvs. tryckskillad är den drivande kraften för processen.

Membranet är tillverkat av kiselkarbid (SiC) och används vanligen för rening av vatten innehållandes suspenderade partiklar och oljedroppar.

#### Back pulse hammer

Back pulse hammer (BPH) är en apparat som pulserar permeat tillbaka genom membranet. Syftet med BPH är att ta bort material som fastnat på membranets yta. Genom inställningar i hammaren styrsystem anges hur ofta (tiden mellan pulserna) hammaren ska pulsera, hur länge pulseringen sker och i vilken frekvens. BPH styrs av tryckluft

#### Flödesschema

Ett förenklat flödesschema av riggen visas i figur 3. Inflödet (feed, ljusgrön linje) går via ventil 1 och pumpen till membranet underifrån. Efter membranet har koncentratströmmen (röd linje) tre möjligheter; att gå via ventil 8, 3 eller 9 tillbaka till inflödet. Permeatet (blå linje) går till permeat-tanken via RDV02 och ventil 11, 4 och 12. Ventil 7 på permeatlinjen är till för 'bypass', alltså när man vill köra

backspolning med 733 (svart linje) eller när ventilen för BPH-körning (RDV02) inte är öppen



Figur 1: Membranhus med öppning för inflöde (längst till vänster), koncentrat (längst till höger) och permeat (till höger riktat nedåt). På mitten av membranhuset finns en anslutning till BPH.



Figur 2: Membranets tvärsnittsyta skymtas i öppningen för koncentratet.



Figur 3: Flödesschemat

#### Ventiler

Riggen har fyra typer av ventiler; tvåvägs- och trevägsventiler, backventiler och säkerhetsventiler.

Ventil 1, 3, 4, 7, 8, 9, 13, 16 är tvåvägsventiler vilket innebär att de antingen är öppna eller stängda. Dock är ventil 4, 8, 9 och 13 strypventiler dvs. det är möjligt att minska/öka flödet genom att justera ventilen. Ventil 8 har en gul markering, när den är helt inne är ventilen stängd. Ventil 4, 9 och 13 skruvas åt höger för att strypa flödet.

Ventil 10, 11 och 12 är trevägsventiler. Dessa har pilar på handtaget som markerar hur flödet går.

Backventilerna släpper bara igenom flöde i en riktning. Dessa är markerade CV i flödesschemat och det finns två stycken; CV-09 och CV-08.

Den mörkgröna tunna linjen markerar säkerhetsventilerna som heter PSV i flödesschemat. Dessa öppnas om något skulle gå fel och trycket i riggen ökar. Säkerhetsventilerna sitter anslutna till inflödet, permeatet och koncentratet.

Flödesmätning, provtagning och temperatur

Volymsflödet kan mätas på två ställen; vid FI-02 och Fl-03.

Provuttag görs vid SP-01, SP-02 och SP-03 genom att öppna ventilen. För att få ett representativt prov ska ledningen sköljas genom att första provvolymen slaskas.

Aktuell temperatur avläses vid TI01.

#### Elsystem, tryck och BPH

Riggen har två elskåp varav det ena styr pumpen och det andra styr BPH. Pumpens elskåp heter PC01 i flödesschemat. Detta måste vara igång för att kunna starta styrningen av BPH.

Om pumpen börjar låta annorlunda är det dags att avlufta. Det sitter även en omärkt ventil direkt på pumphuset som också används för att avlufta den.

De röda ringarna i flödesschemat är olika typer av elstyrda mätanordningar. Hur elen är dragen markeras schematiskt med de tunna svarta linjerna. Vid PIT00 mäts det statiska trycket hos inflödet. Om inflödestanken är tom kommer PIT00 visa 0 i värde. OBS; på riggen står det inte PIT00 utan PS00.

PDI03 är en differentialtryckmätare och visar tryckfallet över membranet i mBar. Detta tryckfall behövs för att skapa turbulens. Vid PDI03 sitter två ventiler, V-14 och V-15. Dessa används för att avlufta PDI03.

Trycket hos inflöde/koncentrat visas vid PIT01, och permeatets tryck vid PI02 (båda visar i Bar). Skillnaden mellan PIT01 och PI02 ger tryckdifferensen mellan inflöde och permeat, alltså den drivande kraften för separationen. OBS; PIT01 är inte uppmärkt på riggen men den sitter mellan PSV01 och TT01.

När hammaren kör stängs ventilen vid RDV02. Detta görs för att hammaren ska kunna ta permeat. Vid hammaren sitter RDV03 som öppnas när hammaren dräneras. I dagsläget är inte RDV02 och RDV03 uppmärkta på riggen.

#### Cleaning tank

Cleaning tank är en extra tank som kan användas vid testkörningar.

#### Prober för aktivitetsmätning

På membranet och i rummet (bakgrundsmätning) sitter prober placerade för mätning av aktivitet. Proberna är kopplade till en "mätstation" som är anslutna till ett GM-rör som mäter aktiviten. Värderna som visas på GM-röret samlas upp och sparas på en dator med programmet PROMUX-S.

För att mäta aktivitet med proberna börjar man med att slå på strömmen till mätstationen och datorn. Slå på GM-röret på en svarta knappen uppe till vänster, se figur 4.



Figur 4

## Att köra riggen

- 1. Anslut feed-tank och permeat tanken till riggen. Var noga med packningar och slangklämmor etc. för att undvika spill och läckage. Var också observant på höjdskillnaden mellan feed-tank och inflödes-studsen på riggen!
- 2. Öppna aktuella ventiler så att inflödet kan gå genom membranet och vidare som koncentrat/permeat utan hinder. Se flödesschemat och basläggningen för aktuell körning.

## Basläggning

#### Normal drift utan BPH

## Koncentratet återcirculeras till feed-tank. Permeatet går till permeattank.

Ventilnummer	Position Läge	
-	Ventiler mellan feed-tank och V-01	Öppna
V-01	Inflöde	Öppen
V-08	Koncentratström	Justeras till önskat flöde
V-03	Koncentratström	Öppen
V-09	Koncentratström	Öppen
V-10	Inlopp till feed-tank	Pil åt vänster
V-07	Permeatström	Öppen
V-11	Permeatström	Pil åt höger
V-04	Permeatström	Justerats till önskat flöde
V-12	Inlopp till permeattank	Pil uppåt
SP-01	Provtagning, feed	Stängd
SP-02	Provtagning, permeat	Stängd
SP-03	Provtagning, koncentrat	Stängd
V-02	Flöde från cleaning-tank	Stängd
V-16	Back-flush	Stängd
V-13	Back-flusch	Stängd
V-14	Avluftning pump	Stängd
V-15	Avluftning pump	Stängd
-	Huvudströmbrytare <sup>1</sup>	På
-	Elskåp pump (PC01, SC01)²	På

 <sup>&</sup>lt;sup>1</sup> Huvudströmbrytaren sitter bakom riggen på väggen vid den tjocka elkabeln.
 <sup>2</sup> Elskåpet pumpen sitter vid membranet på riggen. Slå på det genom att vrida knappen till läge 1.

#### Normal drift med BPH

Ändringar och tillägg utifrån 'Normal drift utan BPH'. Kom ihåg att BPH behöver ett uppsamlingskärl för dränagevätska.

Ventilnummer	Position	Läge
-	Serviceluft, 753 <sup>1</sup>	Öppen
V-07	Permeatström	Stängd
-	BPH control unit <sup>2</sup>	På

#### Backspolning med vatten från 733

Ändringar och tillägg utifrån 'Normal drift utan BPH' samt 'Normal drift med BPH'

Ventilnummer	Position	Läge
-	BPH control unit	Av
-	Elskåp pump (PC01, SC01)	Av
-	Ventil vid kranen för 733	Öppen
V-16	Backspolning	Öppen
V-13	Backspolning	Justera till önskat flöde
V-07	Permeat	Öppen

<sup>2</sup> Elskåpet för BPH sitter bakom elskåpet till pumpen och är vitt. Slå på det genom att vrida ratten till läge

1.

<sup>&</sup>lt;sup>1</sup> Kranen för serviceluft finns längre in i rummet under högtalaren. Luften går till riggen via den blå slagen.

# Appendix C

Appendix C gives pictures of the membrane and pilot plant. It also include product data sheets of the membrane and its housing and the BPH.



*Figure C1. The pilot plant with the membrane in the middle of the picture. In the bottom are two permeate samples and one concentrate sample (note the colour difference). The white cans are for drain of the pump and BPH.* 



*Figure C2. The pilot plant seen together with the feed/concentrate tank (in the middle of the picture) and the permeate tank (to the left).* 



*Figure C3. The membrane right before installed in the pilot plant.* 



## COMEM<sup>°</sup> 25mm diameter – round channels

The high flux CoMem<sup>®</sup> asymmetric silicon carbide (SiC) membrane from LiqTech is designed for removal of suspended solids as well as oil droplets and oil-emulsions from solutions. The element may be operated in cross flow mode and in dead-ended mode with fast forward flush. The CoMem<sup>®</sup> elements are designed for cross flow operation.



The OD25mm elements are available in three different lengths: 305mm, 1016mm and 1178mm. Nominal pore sizes available: 3 micron, 1 micron, 0,1 micron and 0,04 micron.

ELEMENT DATA	
Configuration	Cylindrical with round channels
Selective membrane material	Silicon carbide (SiC)
Carrier material	Silicon carbide (SiC)
Temperature tolerance	Up to 800°C
APPLICATION DATA	

APPLICATION DATA	
Operating pressure	Max 10 bar TMP; recommended below 3 bar TMP
Maximum operating temperature	Determined by system components
Maximum chlorine concentration	Unlimited
pH tolerance	0-14
Cleaning	Chlorine, acid, caustic, solvents, oxidizers
Maximum negative TMP	3 bar

SPECIFICATION SHEET 31 ROUND CHANNELS	- Comem® OD25MM			
Model	Element dimensions A (mm) x B (mm)	Channel dimensions (mm)	Membrane area (m2)	Feed flow at 2 m/s
COM0250305xxx-03	25±1 x 305±1	Ø3	0.09	1.58 m3/h
COM0251016xxx-03	25±1 x 1016±1	Ø3	0.30	1.58 m3/h
COM0251178xxx-03	25±1 x 1178±1	Ø3	0.34	1.58 m3/h



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Figure C4. Membrane data sheet.

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FLUX	
Nominal Pore size	Typical flux at 25°C - On non-fouling water @ 1bar
0,04 μm	3 m³/(m²h)
0,1 μm	4 m³/(m²h)
1 μm	10 m³/(m²h)
3 μm*	>10 m³/(m²h)

\* Carrier only. Symmetric filter. No membrane layer.

ORDERING DATA			
Nominal Pore size	OD25x305mm	OD25x1016mm	OD25x1178mm
0.04 μm	COM0250305004-03	COM0251016004-03	COM0251178004-03
0.1 μm	COM0250305010-03	COM0251016010-03	COM0251178010-03
1 μm	COM0250305100-03	COM0251016100-03	COM0251178100-03
3 μm	COM0250305300-03	COM0251016300-03	COM0251178300-03

Notice: Elements are delivered dry. Handle with care since the material is brittle. LiqTech believes the information and data contained herein to be accurate and useful. The information and data are offered in good faith, but without guarantee, as conditions and methods of use of our products are beyond our control. Liqtech assumes no liability for results obtained or damages incurred through the application of the presented information and data. It is the user's responsibility to determine the appropriateness of LiqTech's products for the user's specific end uses. Specifications are subject to change without notice. N 01/25/10

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Figure C4. Membrane data sheet (continued).



## STAINLESS STEEL HOUSINGS

#### For OD25mm elements

Housings for OD25mm × 305/1016/1178mm cross flow CoMem elements. Designed to hold and protect one element. There are four ports: feed, concentrate, permeate and port for Back Pulse Hammer (BPH). Housing includes blind for BPH port if BPH is not ordered.

For mounting instruction please refer to LiqTech's mounting manual.



SPECIFICATION SHEET	OD25X305	OD25X1016	OD25X1178
CONNECTIONS			
ISO2852:1993			
Feed port	ISO38 TriClamp	ISO38 TriClamp	ISO38 TriClamp
Concentrate port	ISO38 TriClamp	ISO38 TriClamp	ISO38 TriClamp
Permeate port	ISO38 TriClamp	ISO38 TriClamp	ISO38 TriClamp
BPH port	ISO38 TriClamp	ISO38 TriClamp	ISO38 TriClamp
OPERATING CONDITIONS			
Maximum pressure	10 bar	10 bar	10 bar
Maximum temperature	200°C	200°C	200°C
Material*	AISI 316L	AISI 316L	AISI 316L
Weight	1,7 kg	3,0 kg	3,2 kg

\*Other materials available upon request.

ORDERING DATA	OD25X305	OD25X1016	OD25X1178	
O-RING AND SEALING				
MATERIAL				
Ethylen-propylene, EPDM	HOU0250305CO1-SS	HOU0251016CO1-SS	HOU0251178C01-SS	
Nitrile, NBR	HOU0250305CO2-SS	HOU0251016CO2-SS	HOU0251178CO2-SS	
Viton, FPM	HOU0250305CO3-SS	HOU0251016CO3-SS	HOU0251178CO3-SS	
Please refer to the next pages for detailed overview of included parts and housing dimensions				

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Figure C5. Housing data sheet.

# LiqTech

## BACK PULSE HAMMER (BPH) PACKAGES

The Back Pulse Hammer (BPH) is a pulse generator, which delivers high frequency "block" pulses, from the permeate side, back through the membrane in order to keep the membrane clean and free of foulants.

The product was originally developed to remove fouling caused by humic acid from surface water, but has since proven its efficiency in many other applications and now it is a standard cleaning method. The BPH system is an excellent tool for maintaining high flux without compromising permeate production.

SPECIFICATION SHEET	BPH FOR Ø25/Ø80/Ø146 MEMBRANE ELEMENTS
Temperature tolerance	Up to 90°C
Maximum air pressure	6 bar
BPH Material	Stainless steel 316L, but also available in other steel types
BPH Diaphragm	NBR (Nitrile) or FPM (Viton)
Back pulse frequency	To be set according to the application (see operation manual)

#### PRODUCT VARIANTS

The Back Pulse Hammer can be delivered in two configurations:

BPH 1: Complete package

BPH 2: Back Pulse Hammer only



Please refer to the next pages for detailed overview of each BPH package configuration.

The BPH system is available in two different sizes: the small for OD25 and the large for OD146 membrane elements. See ordering data below.

ORDERING DATA	OD25 ELEMENTS	OD146 ELEMENTS
BPH 1: Complete Package	BPH025xxx1001	BPH146xxx1001
BPH 2: Back Pulse Hammer only	BPH025xxx1002	BPH146xxx1002

xxx = BPH Diaphragm material: NBR (Nitrile) or FPM (Viton)

For detailed information about operating and connecting the hammer please refer to the BPH manual

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Figure C6. BPH data sheet

## Appendix D

#### Samples and analysis

This Appendix lists the samples which were taken during the experiment and the analysis preformed on the samples. A summary is given in table D5.

The samples are denoted as following; the first digits correspond to the test, P and C stands for permeate and concentrate respective, and the last digit gives the order of the samples. For example: sample '1AP1' is taken during test 1A and it is the first permeate sample taken during test 1A. Samples denoted 'Permeate tank X' are samples taken directly from the water in the permeate tanks by hand. Samples of the remaining water in the feed tank are denoted 'Concentrate tank X'.

#### Test 1A – Flux test

Table D1 lists the samples taken during test 1A. When a certain permeate flow was operated, samples of the permeate (denoted as 1APX) and concentrate (denoted as 1ACX) were taken and after this was a new permeate flow tested. The pilot plant was operating for approximately 30 minutes at certain value of permeate flow before samples were taken. The first samples (1AP1/1AC1) were taken approximately 40 minutes after start up. A break in the test was taken between permeate flow 30 and 35 L/h. The samples 1AP5 and 1AC5 were taken 30 minutes after the re-start.

Tuble D1. Sumples of test 11.		
Permeate flow (L/h)	Sample title	
15	1AP1/1AC1	
20	1AP2/1AC2	
25	1AP3/1AC3	
30	1AP4/1AC4	
35	1AP5/1AC5	
40	1AP6/1AC6	
45	1AP7/1AC7	
50	1AP8/1AC8	

Table D1. Samples of test 1A

All the samples were analysed using HPGe. The permeate samples were measured for 8 h and the concentrate samples for 2 h. Sample 1AP2/1AC2, 1AP5/1AC5 and 1AP8/1AC8 were also analysed with TOC, Silicic acid, conductivity, Boron and pH.

#### Test 1B – Concentration of the feed

Samples of the permeate and concentrate were taken around 30-45 min after start up each day according to table D2. The samples from day 1 were taken with 1 h intervals while for day 2 and 3 they were taken with 2 h intervals. The final samples were taken when there was approximately 20 litres left in the feed tank. The time between sample 1BP13/1BC13 and the final samples was 70 minutes.

Day 1	Day 2	Day 3	Day 4
1BP1/1BC1	1BP5/1BC5	1BP9/1BC9	1BP13/1BC13
1BP2/1BC2	1BP6/1BC6	1BP10/1BC10	P-Final/C-Final
1BP3/1BC3	1BP7/1BC7	1BP11/1BC11	Permeate tank 1/Concentrate tank 1
1BP4/1BC4	1BP8/1BC8	1BP12/1BC12	

Table D2. Samples from test 1B.

Sample 'Permeate tank 1' and 'Feed tank 1' were analysed with HPGe, TOC, Silicic acid concentration, conductivity, Boron concentration and pH. 'Permeate tank 1' was also analysed with the particle counter and the sequence of filtrations. The remaining samples listed in table 3 were only analysed with HPGe. All permeate samples (1BP1 to 1BP13, P-Final and Permeate tank 1) were analysed during 8 h and the concentrate samples during 2 h.

#### Test 2 – BPH with 10 minutes intervals

Table D3 lists the samples taken during test 2. Samples 2P2/2C2 were taken 3 h after the first samples. The sample from the second day were taken in a similar ways as the last samples of test 1B.

Table D3. Samples from test 2.

Day 1	Day 2
2P1/2C1	Permeate tank 2/Concentrate tank 2
2P2/ 2C2	

Sample 2P1, 2C1, 2P2 and 2C2 were analysed with HPGe (permeate samples for 8 h, concentrate samples for 3h). 'Permeate tank 2' and 'Concentrate tank 2' were analysed with HPGe (8h and 3h), TOC, Silicic acid concentration, conductivity, Boron concentration, pH, particle counter and the filtration sequence.

#### Test 3 – BPH with 5 minutes intervals

The samples from test 3 are listed in table D4. During day 1 were the samples taken with 4 h in between, and on the second day it was approximately 1 h between the samples.

Table D4. Samples from test 3.

Day 1	Day 2
3P1/3C1	3P3/3C3
3P2/3C2	Permeate tank 3/Concentrate tank 3

Sample 3P1/3C1 to 3P3/3C3 were only analysed with HPGe for 3 h. Sample 'Permeate tank 3' and 'Concentrate tank 3' sample were analysed with HPGe (2h and 3h), Boron, Silicic acid, pH, TOC, conductivity, particle counter and the sequence of filtrations

Sample \Analysis	HPGe	B. acid	Si. acid	pН	Cond.	TOC	Seq.of filt.	P.c.
Tank 1	x	x	x	x	х	x	X	x
1AP1/1AC1	x					1		
1AP2/1AC2	х	х	х	х	х	х		1
1AP3/1AC3	х							
1AP4/1AC4	х							
1AP5/1AC5	х	х	х	x	х	х		
1AP6/1AC6	х							
1AP7/1AC7	х							
1AP8/1AC8	х	х	х	х	х	х		
1BP1/1BC1	х							
1BP2/1BC2	х							
1BP3/1BC3	X							
1BP4/1BC4	х							
1BP5/1BC5	х							
1BP6/1BC6	x							
1BP7/1BC7	х							
1BP8/1BC8	x							
1BP9/1BC9	х							
1BP10/1BC10	х							
1BP11/1BC11	х							
1BP12/1BC12	х							
1BP13/1BC13	х							
P-Final/C-Final	х							
Permeate tank 1	х	x	x	х	x	х	х	х
Concentrate tank 1	х	X	x	х	х	х		
Tank 2	х	х	х	x	х	x	X	X
2P1/2C1	х							
2P2/2C2	x							
Permeate tank 2	x	X	x	x	х	х	х	х
Concentrate tank 2	x	x	x	x	X	x	x	х
Leaching sample	X							
Tank 3	х	х	x	x	х	х	Х	х
3P1/3C1	x							
3P2/3C2	x							
3P3/3C3	х							
Permeate tank 3	x	x	х	x	х	x	Х	х
Concentrate tank 3	x	x	x	x	х	x	X	х
IX 1/IX2	х			х				

Table D5. Summary of analysis.

# Appendix E

Appendix E gives all results of the analysis of Boron, Silicic acid, pH, conductivity, TOC and particle counting.

Sample	Result (ppm)	Average result (ppm)
1AP2	351,1/350,2	351
1AC2	351,3/350,3	351
1AP5	349,2/350,5	350
1AC5	351,4/351,6	352
1AP8	350,3/349,6	350
1AC8	351,6/351,6	352
Tank 1	360,2/360,2	360
Permeate tank 1	342,7/342,5	343
Concentrate tank 1	346,4/345,8	346
Tank 2	390,7/391,2	391
Permeate tank 2	379,3/379,6	379
Concentrate tank 2	384,2/384,5	384
Tank 3	396,0/396,0	396
Permeate tank 3	384,1/384,0	384
Concentrate tank 3	390,1/390,0	390

Table E1. Analysis of Boron.

Table E2. Analysis of Silicic acid.

Sample	Dilution	Diluted result	Real result (µg/L)	Average result (µg/L)
1AP2	1:5	372	1860	-
1AC2	1:5	397	1985	-
1AP5	1:5	378	1890	-
1AC5	1:5	392	1960	-
1AP8	1:5	389	1945	-
1AC8	1:5	396	1980	-
Tank 1	1:5/1:5	395/392	1975/1960	1968
Permeate tank 1	1:5/1:10	388/195	1940/1950	1945
Concentrate tank 1	1:5/1:10	446/218	2230/2180	2205
Tank 2	1:5/1:10	421/212	2105/2120	2113
Permeate tank 2	1:5/1:10	402/203	2010/2030	2020
Concentrate tank 2	1:5/1:10	582/288	2910/2880	2895
Tank 3	1:5/1:10	401/205	2005/2050	2028
Permeate tank 3	1:5/1:10	384/194	1920/1940	1930
Concentrate tank 3	1:5/1:10	563/283	2815/2830	2823

Table E3. Analysis of pH.

Sample	Result	Average result
1AP2	6,99/6,98/6,98	6,98
1AC2	7,03/7,02/7,02	7,02
1AP5	6,99/6,99/6,98	6,99
1AC5	7,03/7,04/7,04	7,04
1AP8	7,00/7,00/6,99	7,00
1AC8	7,05/7,05/7,05	7,05
Tank 1	6,98/6,97/7,00	7,00
Permeate tank 1	7,06/7,06/7,05	7,06
Concentrate tank 1	7,24/7,24/7,24	7,24
Tank 2	5,63/5,66/5,62	5,64
Permeate tank 2	6,06/5,86/5,93	5,95
Concentrate tank 2	4,51/4,49/4,50	4,50
Tank 3	5,51/5,54/5,53	5,53
Permeate tank 3	5,76/5,77/5,71	5,75
Concentrate tank 3	4,41/4,19/4,18	4,26

Table E4. Analysis of conductivity.

Sample	Result (µS/cm)	Average result (µS/cm)
1AP2	22,00/22,70/22,70	22,47
1AC2	26,80/27,00/27,10	26,97
1AP5	21,30/22,50/21,40	21,73
1AC5	27,30/25,70/27,20	26,73
1AP8	21,50/21,70/22,60	21,93
1AC8	26,50/27,50/26,30	26,77
Tank 1	26,5/25,6/26,5	26,2
Permeate tank 1	25,5/25,6/25,6	25,6
Concentrate tank 1	39,6/40,2/40,1	40
Tank 2	22,2/22,5/22,4	22,37
Permeate tank 2	1,94/1,96/2,17	2,02
Concentrate tank 2	20,4/20,5/20,4	20,43
Tank 3	3,0/3,0/3,0	3,0
Permeate tank 3	1,8/1,8/1,8	1,8
Concentrate tank 3	34,1/34,2/33,7	34

Sample	Result (mg/L)	Average result (mg/L)
1AP2	1,61/1,61	1,61
1AC2	4,01/3,99	4,00
1AP5	1,47/1,53	1,50
1AC5	5,73/5,65	5,69
1AP8	1,52/1,56	1,54
1AC8	4,83/4,78	4,81
Tank 1	4,87/4,86	4,87
Permeate tank 1	2,39/2,49	2,44
Concentrate tank 1	24,7/24,5	24,6
Tank 2	6,92/6,48	6,70
Permeate tank 2	1,42/1,13	1,28
Concentrate tank 2	38,7/39,1	38,9
Tank 3	6,57/6,59	6,58
Permeate tank 3	2,63/2,64	2,64
Concentrate tank 3	183/185	184

Table E5. Analysis of TOC.

Table E6. Analy	vsis of	particle size	distribution (	(in %)	) with	particle of	counting.
	/	•		<b>`</b>			

Sample	2-4 μm	4-7 µm	7-14 µm	14-20 μm	20-25 μm	25-100 µm	>100 µm
Tank 1	28,5	36,7	23,0	6,8	1,3	0,6	3,2
Permeate tank 1	60,1	18,4	7,6	2,6	0,5	0,3	10,3
Tank 2	41,1	40,9	12,0	1,7	0,3	0,2	3,7
Permeate tank 2	11,4	34	39,3	13,1	1,4	0,3	0,5
Concentrate tank 2	74,7	10,7	2,2	0,6	0,1	0,1	11,6
Tank 3	55,6	36,6	6,7	0,8	0,1	0,1	5,4
Permeate tank 3	5,9	20,1	38,4	24,4	6,7	3,9	0,6
Concentrate tank 3	89,4	2,0	0,3	0,1	0,0	0,0	8,3

## Appendix F

This sections gives the data of all HPGe-analysis performed on the samples of tank 1. Table F1-F2 lists the data from the characterisation, table F3 of test 1A and table F4 of test 1B. In the end of the section lists table F5 the HPGe-data from the sequence of filtrations preformed on the sample 'Permeate tank 1'.

Nuclide	Activity (Bq/L)	Unc. (%)
Cr-51	1,95E+03	18,92
Mn-54	8,67E+03	0,85
Co-57	4,39E+02	3,7
Co-58	3,74E+04	0,29
Co-60	1,38E+05	0,13
Zn-65	1,49E+03	9,2
Nb-95	1,99E+04	0,63
Zr-95	7,72E+03	1,27
Ag-110m	7,44E+04	0,44
Sn-113	4,69E+02	10,73
Sn-117m	1,29E+03	3,57
Sb-124	8,65E+03	1,35
Sb-125	1,54E+03	7,03
Ce-144	7,02E+02	17,51
Total	3,03E+05	0,40

Table F1. Characterisation of tank 1.

10 µm			10 μm /0,8 μ	ım	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	4,11E+03	1,25	Mn-54	2,47E+03	1,24
Co-57	2,08E+02	5,44	Co-57	1,13E+02	8,88
Co-58	1,70E+04	0,45	Co-58	1,03E+04	0,59
Co-60	7,52E+04	0,17	Co-60	5,03E+04	0,20
Zr-95	1,25E+03	6,38	Zn-65	5,58E+02	13,05
Zn-65	9,54E+03	0,92	Nb-95	6,11E+03	1,25
Nb-95	3,73E+03	2,26	Zr-95	2,44E+03	2,81
Ag-110m	4,91E+04	0,49	Ag-110m	3,69E+04	0,43
Sn-113	2,85E+02	12,54	Sn-113	1,93E+02	16,32
Sn-117m	8,56E+02	3,54	Sn-117m	4,96E+02	5,27
Sb-124	5,01E+03	1,66	Sb-124	3,36E+03	1,96
Sb-125	8,49E+02	9,12	Total	1,13E+05	0,44
Ce-144	3,38E+02	24,49			
Total	1,67E+05	0,42			
10 μm /0,8 μ	10 μm /0,8 μm/300.000 Da			1m/300.000 Da	a/100.000 Da
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	6,31E+01	10,92	Mn-54	4,84E+01	9,50
Co-57	2,56E+01	11,44	Co-57	1,78E+01	12,58
Co-58	2,63E+03	0,82	Co-58	2,69E+03	0,84
Co-60	8,59E+02	1,70	Co-60	3,22E+02	2,86
Ag-110m	2,18E+03	1,86	Ag-110m	2,29E+02	7,41
Sn-117m	3,16E+01	21,11	Sn-117m	2,65E+01	18,48
Sb-124	4,27E+02	6,11	Sb-124	1,79E+02	6,81
Total	6,22E+03	1,79	Total	3,51E+03	1,87

Table F2. Filtrations of tank 1.

1 A P 1			1AC1		
Nuclide	Ba/I	Unc (%)	Nuclide	Ba/I	$\operatorname{Unc}(\%)$
Mn_54	5 51E±01	5 56	K_12	$1.44E\pm02$	29.64
Co-57	3,51E+01	14 48	Cr-51	8 98F+02	22,04
Co-58	1.13E+03	0.56	Mn-54	2.35E+03	1 37
Co-60	6.77E+02	0,30	Co-57	2,35E+03	10.29
Nh-95	7.92E+01	5 29	Co-58	1.05E+02	0.47
7r-95	4.34E+01	9.47	Co-60	3.41E+04	0,47
Δσ-110m	9.82E+02	1 29	Nh-95	4 73E+03	0,20
Sb-124	7.63E+01	4 47	7r-95	2 29E+03	2 43
Cs-137	9 53E+00	30.32	Aσ-110m	2,2>E+03	0.63
Total	3.09E+03	1.15	Sn-113	2,37E+01	18 29
Totui	5,071105	1,10	Sh-124	1.90E+03	1.50
			Ce-144	1.04E+03	27.74
			Total	8.25E+04	1.01
1AP2	I		1AC2	0,202101	1,01
Nuclide	Ba/L	Unc. (%)	Nuclide	Ba/L	Unc. (%)
Mn-54	3.92E+01	6,44	K-42	1.57E+02	24.34
Co-57	1.80E+01	6.42	Mn-54	1.76E+03	1.92
Co-58	2.09E+03	0.49	Co-57	2.95E+02	13.92
Co-60	2.89E+02	1.82	Co-58	7.35E+03	0.59
Ag-110m	1,01E+03	1,33	Co-60	2,73E+04	0,23
Sn-117m	8,51E+00	14,90	Zn-65	3,04E+02	16,93
Sb-124	1,28E+02	4,36	Nb-95	3,41E+03	1,24
Total	3,58E+03	1,05	Zr-95	1,58E+03	3,10
			Ag-110m	2,31E+04	0,62
			Sn-113	2,01E+02	18,90
			Sb-124	1,66E+03	1,69
			Sb-125	5,14E+02	16,74
			Total	6,76E+04	0,64
1AP3			1AC3		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,63E+01	7,16	Mn-54	1,65E+03	2,00
Co-57	1,74E+01	6,74	Co-57	2,65E+02	15,30
Co-58	2,07E+03	0,50	Co-58	6,61E+03	0,62
Co-60	2,66E+02	1,92	Co-60	2,61E+04	0,23
Ag-110m	8,78E+02	1,55	Zn-65	2,70E+02	18,80
Sn-117m	9,27E+00	13,98	Nb-95	3,29E+03	1,29
Sb-124	1,17E+02	4,51	Zr-95	1,53E+03	3,14
Total	3,39E+03	1,11	Ag-110m	2,30E+04	0,62
			Sn-113	2,03E+02	18,40
			Sb-124	1,62E+03	1,73
			Sb-125	6,09E+02	14,00
			Total	6,51E+04	0,65

Table F3. HPGe-data of test 1A.

1AP4			1AC4		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,67E+01	7,02	Mn-54	1,74E+03	1,90
Co-57	1,90E+01	6,76	Co-57	2,86E+02	14,20
Co-58	2,02E+03	0,51	Co-58	6,90E+03	0,57
Co-60	2,76E+02	1,87	Co-60	2,66E+04	0,23
Ag-110m	7,78E+02	1,76	Nb-95	3,31E+03	1,27
Sn-117m	1,04E+01	12,72	Zr-95	1,65E+03	3,06
Sb-124	1,25E+02	4,49	Ag-110m	2,25E+04	0,64
Total	3,27E+03	1,17	Sn-113	1,75E+02	18,43
			Sb-124	1,61E+03	1,73
			Total	6,48E+04	0,57
1AP5			1AC5		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,92E+01	6,63	K-42	1,71E+02	29,60
Co-57	2,17E+01	6,44	Mn-54	1,63E+03	2,02
Co-58	2,18E+03	0,52	Co-57	3,50E+02	11,30
Co-60	2,99E+02	1,70	Co-58	6,52E+03	0,59
Ag-110m	9,20E+02	1,64	Co-60	2,60E+04	0,23
Sn-117m	1,06E+01	13,84	Zn-65	3,22E+02	12,80
Sb-124	1,26E+02	4,11	Nb-95	3,27E+03	1,27
Total	3,60E+03	1,14	Zr-95	1,53E+03	3,15
		´	Ag-110m	2,31E+04	0,62
			Sn-113	2,09E+02	18,00
			Sb-124	1,60E+03	1,67
			Total	6,47E+04	0,61
1AP6			1AC6		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	4,35E+01	7,65	K-42	1,66E+02	31,30
Co-57	2,24E+01	6,36	Mn-54	2,10E+03	1,71
Co-58	2,16E+03	0,51	Co-57	3,55E+02	12,20
Co-60	3,65E+02	1,66	Co-58	8,68E+03	0,43
Ag-110m	8,88E+02	1,76	Co-60	3,15E+04	0,21
Sn-117m	1,04E+01	15,08	Nb-95	4,07E+03	1,09
Sb-124	1,36E+02	4,16	Zr-95	2,01E+03	2,68
Total	3,63E+03	1,17	Zr-97	2,30E+02	12,80
			Ag-110m	2,45E+04	0,57
			Sn-113	1,87E+02	18,30
			Sb-124	1,77E+03	1,64
			Sb-125	5,52E+02	16,40
			Total	7,61E+04	0,57
1AP7			1AC7		
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Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	5,75E+01	5,23	K-42	1,47E+02	29,20
Co-57	1,74E+01	7,88	Mn-54	2,32E+03	1,51
Co-58	2,02E+03	0,52	Co-57	4,18E+02	10,60
Co-60	7,64E+02	1,02	Co-58	9,53E+03	0,52
Zr-95	1,90E+01	25,10	Co-60	3,38E+04	0,20
Ag-110m	1,09E+03	1,51	Zn-65	4,77E+02	9,54
Sb-124	1,36E+02	4,44	Nb-95	4,49E+03	1,04
Total	4,10E+03	1,10	Zr-95	2,05E+03	2,62
			Ag-110m	2,55E+04	0,59
			Sn-113	2,09E+02	19,60
			Sb-124	1,89E+03	1,40
			Sb-125	6,00E+02	15,50
			Total	8,14E+04	0,56
1AP8			1AC8		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	9,20E+01	3,71	Cr-51	8,76E+02	29,70
Co-57	2,20E+01	6,54	Mn-54	2,45E+03	1,54
Co-58	2,27E+03	0,50	Co-57	4,61E+02	9,71
Co-60	1,45E+03	0,75	Co-58	9,85E+03	0,40
Nb-95	1,21E+02	5,68	Co-60	3,50E+04	0,19
Zr-95	5,27E+01	13,33	Nb-95	4,71E+03	1,00
Ag-110m	1,68E+03	1,30	Zr-95	2,13E+03	2,56
Sn-117m	1,61E+01	10,41	Ag-110m	2,59E+04	0,59
Sb-124	1,95E+02	3,85	Sn-113	2,36E+02	17,60
Total	5,90E+03	1,01	Sb-124	1,92E+03	1,72
			Total	8,35E+04	0,79

1BP1			1BC1		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,43E+01	6,11	K-42	1,41E+02	28,97
Co-57	1,79E+01	6,64	Mn-54	1,88E+03	1,84
Co-58	1,80E+03	0,53	Co-57	3,26E+02	12,46
Co-60	2,91E+02	1,84	Co-58	7,31E+03	0,58
Sr-92	1,10E+02	34,84	Co-60	2,86E+04	0,22
Ag-110m	8,15E+02	1,66	Nb-95	3,71E+03	1,12
Sb-124	1,24E+02	4,36	Zr-95	1,84E+03	2,84
Total	3,19E+03	2,66	Ag-110m	2,31E+04	0,64
			Sn-113	1,85E+02	20,73
			Total	6,71E+04	0,58
1BP2			1BC2		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,65E+01	6,77	Mn-54	1,65E+03	1,84
Co-57	1,68E+01	6,09	Co-57	2,46E+02	16,20
Co-58	1,81E+03	0,5	Co-58	6,31E+03	0,66
Co-60	2,62E+02	1,89	Co-60	2,54E+04	0,23
Nb-97	8,84E+02	17,38	Zn-65	2,99E+02	13,50
Ag-110m	6,25E+02	1,75	Nb-95	3,28E+03	1,21
Sn-117m	8,22E+00	11,96	Zr-95	1,62E+03	3,04
Sb-124	1,17E+02	4,36	Zr-97	5,85E+02	4,49
Total	3,76E+03	8,21	Ag-110m	2,17E+04	0,62
			Total	6,11E+04	0,59
1BP3			1BC3		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,91E+01	5,03	Mn-54	1,81E+03	1,66
Co-57	1,81E+01	6,66	Co-57	3,15E+02	12,90
Co-58	1,89E+03	0,51	Co-58	6,56E+03	0,63
Co-60	2,69E+02	1,81	Co-60	2,72E+04	0,23
Ag-110m	5,69E+02	2,07	Nb-95	3,63E+03	1,17
Sn-117m	1,06E+01	9,26	Zr-95	1,71E+03	2,98
Sb-124	1,17E+02	4,25	Ag-110m	2,29E+04	0,61
Total	2,91E+03	1,16	Sn-113	1,92E+02	19,60
			Sb-124	1,53E+03	1,72
			Total	6,58E+04	0,56

Table F4. HPGe-data of test 1B.

1804			1BC4		
Nualida	P.a./I	$\operatorname{Una}(0)$	Nuclida	РаД	$\operatorname{Una}(0/4)$
Mn-54	3 99E+01	6 22	K-42	Бц/L 1 58E+02	31.06
Co-57	1 88E+01	6.41	Mn-54	1,93E+03	1 75
Co-58	1,84E+03	0.52	Co-57	3 52E+02	11.93
Co-60	2.69E+02	1.89	Co-58	7.10E+03	0.61
Ag-110m	5.34E+02	2.21	Co-60	2.96E+04	0.22
Sn-117m	1,10E+01	9,68	Zn-65	2,78E+02	17,56
Sb-124	1,23E+02	4,69	Nb-95	3,93E+03	1,12
Total	2,84E+03	1,22	Zr-95	1,89E+03	2,28
			Ag-110m	2,45E+04	0,58
			Sb-124	1,64E+03	1,82
			Total	7,14E+04	0,55
1BP5			1BC5	· .	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,78E+01	7,17	K-42	1,89E+02	20,79
Co-57	1,69E+01	6,88	Mn-54	1,86E+03	1,74
Co-58	1,86E+03	0,53	Co-57	2,99E+02	13,87
Co-60	2,93E+02	1,78	Co-58	6,71E+03	0,62
Ag-110m	7,29E+02	1,82	Co-60	2,88E+04	0,22
Sn-117m	9,13E+00	12,71	Nb-95	3,79E+03	1,15
Sb-124	1,27E+02	4,37	Zr-95	1,78E+03	2,96
Total	3,07E+03	1,20	Ag-110m	2,43E+04	0,57
			Total	6,78E+04	0,55
1BP6	T		1BC6		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	3,49E+01	7,69	Mn-54	1,95E+03	1,77
Co-57	1,73E+01	5,88	Co-57	3,42E+02	12,66
Co-58	1,79E+03	0,52	Co-58	7,11E+03	0,61
Co-60	3,18E+02	1,66	Co-60	3,06E+04	0,21
Ag-110m	5,38E+02	2,19	Nb-95	4,00E+03	1,12
Sn-117m	1,13E+01	8,7	Zr-95	1,97E+03	2,75
Sb-124	1,23E+02	4,04	Ag-110m	2,62E+04	0,55
Total	2,83E+03	1,20	Sn-113	2,28E+02	15,16
			Sb-124	1,69E+03	1,76
			Total	7,42E+04	0,52

1BP7			1BC7		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	4,96E+01	4,75	Mn-54	2,57E+03	1,49
Co-57	1,75E+01	6,83	Co-57	3,62E+02	13,18
Co-58	2,00E+03	0,49	Co-58	9,04E+03	0,55
Co-60	5,79E+02	1,14	Co-60	3,97E+04	0,19
Ag-110m	7,41E+02	2,03	Zn-65	5,26E+02	9,18
Sn-117m	1,10E+01	10,59	Nb-95	5,50E+03	0,95
Sb-124	1,51E+02	9,92	Zr-95	2,59E+03	2,40
Total	3,55E+03	1,39	Ag-110m	3,32E+04	0,51
			Sn-113	3,02E+02	14,15
			Sb-124	2,25E+03	1,65
			Total	9,60E+04	0,48
1BP8			1BC8		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	5,65E+01	6,49	K-42	2,81E+02	21,34
Co-57	2,09E+01	6,51	Mn-54	3,93E+03	0,91
Co-58	1,93E+03	0,52	Co-57	6,54E+02	8,52
Co-60	7,56E+02	1,04	Co-58	1,44E+04	0,44
Zr-95	2,54E+01	16,58	Co-60	5,79E+04	0,16
Ag-110m	8,27E+02	1,89	Zn-65	4,48E+02	13,34
Sn-117m	1,22E+01	10,05	Nb-95	7,73E+03	0,79
Sb-124	1,49E+02	9,97	Zr-95	3,66E+03	2,01
Total	3,78E+03	1,36	Ru-106	1,24E+03	29,00
			Ag-110m	4,48E+04	0,45
			Sn-113	4,00E+02	13,34
			Sb-124	3,07E+03	1,13
			Sb-125	1,13E+03	10,72
			Ce-144	1,29E+03	28,01
			Total	1,41E+05	0,67

1BP9			1B 9		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	4,58E+01	6,26	K-42	1,87E+02	27,05
Co-57	1,82E+01	6,83	Cr-51	8,86E+02	28,97
Co-58	2,07E+03	0,51	Mn-54	2,69E+03	1,58
Co-60	4,70E+02	1,39	Co-57	4,06E+02	12,44
Ag-110m	9,13E+02	1,64	Co-58	9,17E+03	0,54
Sn-117m	1,28E+01	10,15	Co-60	4,26E+04	0,18
Sb-124	1,50E+02	3,89	Cu-64	7,61E+03	34,34
Total	3,68E+03	1,12	Zn-65	4,88E+02	10,96
			Nb-95	5,74E+03	0,94
			Zr-95	2,74E+03	2,37
			Ag-110m	3,70E+04	0,50
			Sn-113	3,08E+02	13,34
			Sb-124	2,38E+03	1,61
			Sb-125	8,55E+02	12,70
			Total	1,13E+05	4,67
1BP10			1BC10		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	7,14E+01	4,11	Mn-54	2,23E+03	1,67
Co-57	2,10E+01	5,96	Co-58	7,45E+03	0,61
Co-58	2,12E+03	0,49	Co-60	3,67E+04	0,20
Co-60	1,02E+03	0,89	Zn-65	3,97E+02	16,53
Zr-95	3,01E+01	17,81	Nb-95	4,78E+03	1,05
Ag-110m	1,09E+03	1,64	Zr-95	2,26E+03	2,53
Sn-117m	1,12E+01	11,42	Ag-110m	3,36E+04	0,50
Sb-124	1,80E+02	3,84	Sn-113	2,20E+02	20,34
Total	4,54E+03	1,08	Sb-124	2,08E+03	1,56
			Ce-144	1,14E+03	28,78
			Total	9,08E+04	0,87

1BP11	l		1BC11		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	1,13E+02	4,32	K-42	3,77E+02	21,27
Co-57	1,98E+01	7,9	Cr-51	1,10E+03	28,11
Co-58	2,35E+03	0,5	Mn-54	3,75E+03	1,23
Co-60	1,90E+03	0,64	Co-57	5,26E+02	11,50
Nb-95	1,67E+02	4,23	Co-58	1,24E+04	0,50
Zr-95	8,36E+01	8,89	Co-60	6,14E+04	0,15
Ag-110m	1,84E+03	1,32	Zn-65	9,75E+02	6,56
Sn-113	1,42E+01	29,43	Nb-95	8,36E+03	0,78
Sn-117m	1,52E+01	10,53	Zr-95	3,98E+03	1,96
Sb-124	2,47E+02	3,34	Ag-108m	1,85E+02	24,53
Total	6,75E+03	0,98	Ag-110m	5,44E+04	0,41
			Sn-113	4,40E+02	12,31
			Sb-124	3,41E+03	1,06
			Total	1,51E+05	0,58
1BP12			1BC12		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Nuclide Mn-54	Bq/L 1,44E+02	Unc. (%) 4,1	Nuclide K-42	Bq/L 3,63E+02	Unc. (%) 18,47
Nuclide Mn-54 Co-57	Bq/L 1,44E+02 2,69E+01	Unc. (%) 4,1 7,1	Nuclide K-42 Mn-54	Bq/L 3,63E+02 4,83E+03	Unc. (%) 18,47 0,86
Nuclide Mn-54 Co-57 Co-58	Bq/L 1,44E+02 2,69E+01 2,47E+03	Unc. (%) 4,1 7,1 0,49	Nuclide K-42 Mn-54 Co-57	Bq/L 3,63E+02 4,83E+03 7,30E+02	Unc. (%) 18,47 0,86 9,59
Nuclide           Mn-54           Co-57           Co-58           Co-60	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03	Unc. (%) 4,1 7,1 0,49 0,54	Nuclide K-42 Mn-54 Co-57 Co-58	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04	Unc. (%) 18,47 0,86 9,59 0,44
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02	Unc. (%) 4,1 7,1 0,49 0,54 3,25	Nuclide K-42 Mn-54 Co-57 Co-58 Co-60	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04	Unc. (%) 18,47 0,86 9,59 0,44 0,13
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16	Nuclide           K-42           Mn-54           Co-57           Co-58           Co-60           Zn-65	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15	Nuclide           K-42           Mn-54           Co-57           Co-58           Co-60           Zn-65           Nb-95	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41	Nuclide           K-42           Mn-54           Co-57           Co-58           Co-60           Zn-65           Nb-95           Zr-95	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04	Nuclide           K-42           Mn-54           Co-57           Co-58           Co-60           Zn-65           Nb-95           Zr-95           Zr-97	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03 6,06E+02	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124           Total	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02 8,61E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04 <b>0,89</b>	Nuclide K-42 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Zr-97 Ag-108m	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03 6,06E+02 2,30E+02	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23 22,29
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124           Total	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02 8,61E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04 <b>0,89</b>	Nuclide           K-42           Mn-54           Co-57           Co-60           Zn-65           Nb-95           Zr-95           Zr-97           Ag-108m           Ag-110m	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03 6,06E+02 2,30E+02 7,04E+04	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23 22,29 0,35
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124           Total	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02 8,61E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04 <b>0,89</b>	Nuclide         K-42         Mn-54         Co-57         Co-60         Zn-65         Nb-95         Zr-95         Zr-97         Ag-108m         Ag-110m         Sn-113	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03 6,06E+02 2,30E+02 7,04E+04 5,72E+02	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23 22,29 0,35 11,38
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124           Total	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02 8,61E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04 <b>0,89</b>	Nuclide         K-42         Mn-54         Co-57         Co-60         Zn-65         Nb-95         Zr-95         Zr-97         Ag-108m         Ag-110m         Sn-113         Sb-124	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03 6,06E+02 2,30E+02 7,04E+04 5,72E+02 4,40E+03	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23 22,29 0,35 11,38 1,08
Nuclide           Mn-54           Co-57           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124           Total	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02 8,61E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04 <b>0,89</b>	Nuclide         K-42         Mn-54         Co-57         Co-60         Zn-65         Nb-95         Zr-95         Zr-97         Ag-108m         Ag-110m         Sn-113         Sb-124         Sb-125	Bq/L           3,63E+02           4,83E+03           7,30E+02           1,58E+04           7,99E+04           8,73E+02           1,08E+04           5,18E+03           6,06E+02           2,30E+02           7,04E+04           5,72E+02           4,40E+03           1,52E+03	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23 22,29 0,35 11,38 1,08 9,76
Nuclide           Mn-54           Co-57           Co-58           Co-60           Nb-95           Zr-95           Ag-110m           Sn-117m           Sb-124           Total	Bq/L 1,44E+02 2,69E+01 2,47E+03 2,74E+03 2,63E+02 1,20E+02 2,53E+03 2,16E+01 2,97E+02 8,61E+03	Unc. (%) 4,1 7,1 0,49 0,54 3,25 7,16 1,15 9,41 3,04 <b>0,89</b>	Nuclide         K-42         Mn-54         Co-57         Co-60         Zn-65         Nb-95         Zr-95         Zr-97         Ag-108m         Ag-110m         Sn-113         Sb-124         Sb-125         Ce-144	Bq/L 3,63E+02 4,83E+03 7,30E+02 1,58E+04 7,99E+04 8,73E+02 1,08E+04 5,18E+03 6,06E+02 2,30E+02 7,04E+04 5,72E+02 4,40E+03 1,52E+03 2,06E+03	Unc. (%) 18,47 0,86 9,59 0,44 0,13 8,10 0,69 1,70 8,23 22,29 0,35 11,38 1,08 9,76 22,11

1BP13			1BC13		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	8,47E+01	4,19	K-42	6,81E+02	20,63
Co-57	2,21E+01	8,28	Mn-54	6,86E+03	1,03
Co-58	2,53E+03	0,49	Co-57	9,46E+02	12,33
Co-60	1,30E+03	0,82	Co-58	2,17E+04	0,53
Ag-110m	2,14E+03	1,19	Co-60	1,16E+05	0,16
Sn-117m	1,34E+01	12,78	Zn-65	1,39E+03	8,92
Sb-124	2,15E+02	3,75	Nb-95	1,57E+04	0,80
Total	6,31E+03	1,01	Zr-95	7,66E+03	1,98
			Zr-97	1,38E+03	5,82
			Ag-110m	1,04E+05	0,40
			Sn-113	8,02E+02	13,85
			Sb-124	6,47E+03	1,27
			Sb-125	2,17E+03	11,60
			Ce-144	3,05E+03	24,94
			Total	2,89E+05	0,44
P-final	r	1	C-final	ſ	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	1,47E+02	4,73	K-42	1,07E+03	19,81
Co-57	2,77E+01	8,02	Mn-54	1,06E+04	1,23
Co-58	2,86E+03	0,47	Co-57	1,46E+03	10,95
Co-60	2,76E+03	0,54	Co-58	3,18E+04	0,50
Zn-65	3,36E+01	30,47	Co-60	1,70E+05	0,14
Nb-95	2,52E+02	3,92	Cu-64	3,08E+04	31,85
Zr-95	1,27E+02	7,94	Zn-65	2,36E+03	7,01
Ag-110m	3,45E+03	0,93	Nb-95	2,39E+04	0,73
Sn-117m	2,02E+01	11,23	Zr-95	1,13E+04	1,84
Sb-124	3,13E+02	2,92	Ag-108m	4,12E+02	24,13
Total	9,99E+03	0,87	Ag-110m	1,53E+05	0,37
			Sn-113	1,41E+03	10,17
			Sb-124	9,49E+03	1,29
			Sb-125	3,49E+03	8,88
			Ce-144	3,64E+03	28,49
			Total	4,55E+05	4,33

Permeate tank 1			Concentrate	tank 1	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	8,10E+01	5,42	Cr-51	2,93E+03	26,66
Co-57	1,89E+01	7,7	Mn-54	9,77E+03	1,34
Co-58	2,08E+03	0,5	Co-57	1,31E+03	11,72
Co-60	1,15E+03	0,78	Co-58	2,95E+04	0,52
Zr-95	4,01E+01	13,88	Co-60	1,57E+05	0,15
Ag-110m	1,37E+03	1,44	Zn-65	1,67E+03	9,85
Sb-124	1,72E+02	3,63	Nb-95	2,24E+04	0,75
Total	4,91E+03	1,05	Zr-95	1,06E+04	1,88
			Ag-110m	1,42E+05	0,39
			Sn-113	1,30E+03	11,26
			Sb-124	8,39E+03	1,19
			Sb-125	3,17E+03	10,48
			Ce-144	3,80E+03	26,32
			Total	3,93E+05	0,77

Table F5. Filtrations of Permeate tank 1.

10 µm			10 µm /0,8	μm	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	6,73E+01	8,91	Mn-54	7,89E+01	6,12
Co-57	2,20E+01	9,63	Co-57	1,85E+01	11,74
Co-58	2,05E+03	0,84	Co-58	2,07E+03	0,86
Co-60	1,12E+03	1,37	Co-60	1,14E+03	1,34
Zr-95	4,52E+01	20,08	Zr-95	4,08E+01	22,10
Ag-110m	1,33E+03	2,31	Ag-110m	1,18E+03	2,56
Sb-124	1,84E+02	6,44	Sb-124	1,79E+02	5,95
Total	4,82E+03	1,73	Total	4,71E+03	1,74
10 µm /0,8	µm/300.000 Da		10 μm /0,8 μm/300.000 Da/100.000 Da		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	4,23E+01	9,76	Mn-54	4,68E+01	8,60
Co-57	1 88E+01	10.41	0.57	1 (57) 01	
	1,001	10,41	C0-5/	1,65E+01	11,48
Co-58	1,93E+03	0,85	Co-57 Co-58	1,65E+01 1,94E+03	11,48 0,89
Co-58 Co-60	1,93E+03 3,00E+02	0,85 2,82	Co-58 Co-60	1,65E+01 1,94E+03 2,75E+02	11,48 0,89 3,00
Co-58 Co-60 Ag-110m	1,93E+03 3,00E+02 5,31E+02	0,85 2,82 3,22	Co-57 Co-58 Co-60 Ag-110m	1,65E+01 1,94E+03 2,75E+02 1,44E+02	11,48 0,89 3,00 9,24
Co-58 Co-60 Ag-110m Sn-117m	1,93E+03 3,00E+02 5,31E+02 1,25E+01	0,85 2,82 3,22 18,09	Co-57 Co-58 Co-60 Ag-110m Sn-117m	1,65E+01 1,94E+03 2,75E+02 1,44E+02 6,19E+00	11,48 0,89 3,00 9,24 34,72
Co-58 Co-60 Ag-110m Sn-117m Sb-124	1,93E+03 3,00E+02 5,31E+02 1,25E+01 1,43E+02	0,85 2,82 3,22 18,09 6,56	Co-57 Co-58 Co-60 Ag-110m Sn-117m Sb-124	1,65E+01 1,94E+03 2,75E+02 1,44E+02 6,19E+00 1,40E+02	11,48 0,89 3,00 9,24 34,72 6,72

## Appendix G

Appendix G gives the data of all HPGe-analysis performed on the samples of tank 2. Table G1-G2 lists the data from the characterisation and table G3 the data from test 2. The data from the analysis of the leaching sample is given in table G4. Table G5 and G6 gives the results from the sequences of filtrations preformed on the samples 'Permeate tank 2' and 'Concentrate tank 2'.

Nuclide	Bq/L	Unc. (%)
K-42	4,73E+03	17,91
Cr-51	2,32E+03	17,16
Mn-54	2,18E+04	0,55
Co-57	1,32E+03	1,9
Co-58	9,99E+04	0,16
Co-60	3,47E+05	0,08
Cu-64	1,50E+04	19,59
Zn-65	3,02E+03	6,21
Nb-95	2,76E+04	0,45
Zr-95	1,31E+04	1,24
Ru-106	2,50E+03	25,72
Ag-108m	3,58E+02	17,87
Ag-110m	1,18E+05	0,29
Sn-113	7,98E+02	8,72
Sn-117m	8,24E+02	3,13
Sb-124	1,28E+04	1,09
Sb-125	3,46E+03	4,77
Ce-144	1,01E+03	18,28
Total	6,76E+05	0,95

Table G1. Characterisation of tank 2.

10 µm			10 µm /0,8	μm	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	1,92E+04	0,9	Nuklid	Bq/l	Unc (%)
Co-57	1,50E+03	2,53	Mn-54	1,73E+03	2,68
Co-58	9,05E+04	0,29	Co-57	1,46E+02	9,69
Co-60	2,91E+05	0,16	Co-58	1,02E+04	0,89
Zn-65	2,62E+03	11,9	Co-60	3,00E+04	0,53
Nb-95	2,67E+04	0,81	Zn-65	3,30E+02	29,04
Zr-95	1,15E+04	2,37	Nb-95	2,38E+03	3,42
Ag-110m	9,07E+04	0,59	Zr-95	1,09E+03	6,88
Sn-113	7,70E+02	14,31	Ag-110m	1,48E+04	1,74
Sn-117m	9,32E+02	6,28	Sn-117m	1,93E+02	11,32
Sb-124	1,05E+04	2,16	Sb-124	1,37E+03	4,86
Sb-125	2,85E+03	8,82	Total	6,22E+04	1,15
Total	5,49E+05	0,36			
10 μm /0,8 μ	m/300.000 Da	a	10 μm /0,8 μm/300.000 Da/100.000 Da		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Co-58	1,10E+03	2,07	Co-57	1,58E+01	23,7
Co-60	4,05E+02	4,43	Co-58	9,85E+02	2,45
Ag-110m	1,74E+03	3,17	Co-60	1,79E+02	7,16
Sb-124	2,11E+02	8,69	Ag-110m	5,65E+02	6,13
Total	3,46E+03	3,76	Sb-124	1,95E+02	10,15
			Total	1,94E+03	5,00

Table G2. Filtrations of tank 2.

2P1			2C1		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	2,95E+01	7,22	Cr-51	1,61E+03	18,51
Co-57	4,14E+00	19,20	Mn-54	2,08E+04	0,53
Co-58	2,66E+02	1,68	Co-57	1,46E+03	1,48
Co-60	7,23E+02	1,05	Co-58	1,04E+05	0,15
Nb-95	4,71E+01	7,77	Co-60	3,12E+05	0,09
Zr-95	1,90E+01	19,76	Zn-65	2,49E+03	7,11
Ag-110m	4,59E+02	2,96	Nb-95	2,73E+04	0,40
Sn-117m	8,50E+00	11,12	Zr-95	1,28E+04	1,17
Sb-124	7,68E+01	6,43	Ru-106	1,71E+03	33,11
Total	1,63E+03	2,19	Ag-110m	9,35E+04	0,34
			Sn-113	7,43E+02	7,98
			Sn-117m	7,94E+02	2,81
			Sb-124	1,08E+03	0,90
			Sb-125	3,25E+03	4,37
			Ce-144	6,95E+01	21,40
			Total	5,84E+05	0,29
2P1			2C2		
	T				
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Nuclide Mn-54	Bq/L 6,33E+00	Unc. (%) 18,22	Nuclide 3C-42	Bq/L 2,56E+03	Unc. (%) 28,58
Nuclide Mn-54 Co-58	Bq/L 6,33E+00 1,09E+02	Unc. (%) 18,22 2,42	Nuclide 3C-42 Cr-51	Bq/L 2,56E+03 1,91E+03	Unc. (%) 28,58 20,71
Nuclide Mn-54 Co-58 Co-60	Bq/L 6,33E+00 1,09E+02 1,67E+02	Unc. (%) 18,22 2,42 2,27	Nuclide 3C-42 Cr-51 Mn-54	Bq/L 2,56E+03 1,91E+03 2,40E+04	Unc. (%) 28,58 20,71 0,49
Nuclide Mn-54 Co-58 Co-60 Ag-110m	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02	Unc. (%) 18,22 2,42 2,27 5,92	Nuclide 3C-42 Cr-51 Mn-54 Co-57	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03	Unc. (%) 28,58 20,71 0,49 1,69
Nuclide           Mn-54           Co-58           Co-60           Ag-110m           Sn-117m	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00	Unc. (%) 18,22 2,42 2,27 5,92 7,55	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05	Unc. (%) 28,58 20,71 0,49 1,69 0,14
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64
Nuclide           Mn-54           Co-58           Co-60           Ag-110m           Sn-117m           Sb-124           Cs-137           Total	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 <b>4,56E+02</b>	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 4,56E+02	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10
Nuclide           Mn-54           Co-58           Co-60           Ag-110m           Sn-117m           Sb-124           Cs-137           Total	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 <b>4,56E+02</b>	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 4,56E+02	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 <b>4,56E+02</b>	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m Ag-110m	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02 1,18E+05	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07 0,39
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 <b>4,56E+02</b>	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m Ag-110m Sn-113	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02 1,18E+05 9,25E+02	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07 0,39 7,00
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 4,56E+02	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 <b>3,95</b>	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m Ag-108m Ag-110m Sn-113	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02 1,18E+05 9,25E+02 7,91E+02	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07 0,39 7,00 3,21
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 4,56E+02	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 3,95	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m Ag-110m Sn-113 Sn-117m Sb-124	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02 1,18E+05 9,25E+02 7,91E+02 1,29E+04	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07 0,39 7,00 3,21 1,05
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 4,56E+02	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 3,95	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m Ag-110m Sn-113 Sn-117m Sb-124 Sb-125	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02 1,18E+05 9,25E+02 7,91E+02 1,29E+04 3,72E+03	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07 0,39 7,00 3,21 1,05 4,43
Nuclide Mn-54 Co-58 Co-60 Ag-110m Sn-117m Sb-124 Cs-137 <b>Total</b>	Bq/L 6,33E+00 1,09E+02 1,67E+02 1,09E+02 7,33E+00 5,19E+01 5,42E+00 4,56E+02	Unc. (%) 18,22 2,42 2,27 5,92 7,55 7,45 23,50 3,95	Nuclide 3C-42 Cr-51 Mn-54 Co-57 Co-58 Co-60 Zn-65 Nb-95 Zr-95 Ru-106 Ag-108m Ag-110m Sn-113 Sn-117m Sb-124 Sb-125 Ce-144	Bq/L 2,56E+03 1,91E+03 2,40E+04 1,51E+03 1,17E+05 3,64E+04 3,50E+03 3,12E+04 1,49E+04 2,08E+03 1,92E+02 1,18E+05 9,25E+02 7,91E+02 1,29E+04 3,72E+03 9,20E+02	Unc. (%) 28,58 20,71 0,49 1,69 0,14 0,08 5,64 0,40 1,10 30,86 32,07 0,39 7,00 3,21 1,05 4,43 20,09

Table G3. HPGe-data of test 2.

Permeate tank 2		Concentrate tank 2			
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	4,14E+00	27,85	Cr-51	1,33E+04	30,94
Co-57	1,46E+00	34,91	Mn-54	1,25E+05	0,79
Co-58	1,17E+02	2,37	Co-57	8,30E+03	3,96
Co-60	1,04E+02	2,95	Co-58	5,37E+06	0,24
Ag-110m	7,36E+01	6,24	Co-60	2,52E+06	0,09
Sn-117m	7,20E+00	8,22	Zn-65	2,26E+04	7,50
Sb-124	9,08E+01	4,73	Nb-95	2,05E+05	0,58
Cs-137	4,58E+00	25,06	Zr-95	9,25E+04	1,53
Total	4,03E+02	3,84	Ru-106	4,63E+04	14,43
			Ag-108m	2,59E+03	24,96
			Ag-110m	1,53E+06	0,28
			Sn-113	8,26E+03	9,91
			Sn-117m	8,41E+03	3,66
			Sb-124	1,10E+05	0,76
			Sb-125	2,71E+04	7,12
			Ce-144	1,12E+04	19,42
			Total	1,01E+07	0,32

Table G4. Leaching test.

Nuclide	Bq/L	Unc. (%)
Mn-54	8,44E+03	1,57
Co-57	3,94E+02	6,96
Co-58	2,97E+04	0,55
Co-60	1,85E+05	0,19
Zn-65	2,45E+03	8,2
Nb-95	8,78E+03	12,88
Zr-95	4,07E+03	1,64
Ag-108m	3,01E+02	4,31
Ag-110m	8,11E+04	27,11
Sn-113	3,84E+02	0,7
Sn-117m	6,84E+02	20,79
Sb-124	7,95E+03	4,43
Total	3,29E+05	13,38

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10 µm			10 μm /0,8 μm			
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Mn-54	1,19E+01	17,72	Mn-54	1,07E+01	20,95	
Co-58	1,50E+02	3,31	Co-58	1,37E+02	3,55	
Co-60	1,68E+02	3,53	Co-60	1,48E+02	3,85	
Ag-110m	1,14E+02	9,13	Ag-110m	9,54E+01	9,23	
Sn-117m	8,57E+00	19,77	Sn-117m	1,02E+01	13,84	
Sb-124	1,03E+02	7,04	Sb-124	9,10E+01	7,97	
Total	5,55E+02	5,44	Total	4,92E+02	5,65	
10 μm /0,8 μ	ım/300.000 Da		10 μm /0,8 μm/300.000 Da/100.000 Da			
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Mn-54	1,03E+01	28,79	Mn-54	8,50E+00	27,28	
Co-57	1,00E+01	15,7	Co-57	7,37E+00	19,77	
Co-58	1,17E+02	1,19	Co-58	1,37E+02	1,56	
Co-60	1,35E+02	3,82	Co-60	1,32E+02	5,1	
Ag-110m	9,54E+01	8,2	Ag-110m	8,26E+01	13,04	
Sb-124	9,10E+01	8,85	Sb-124	8,60E+01	6,39	
Total	4,59E+02	5,62	Total	4,53E+02	6,29	

Table G5. Filtrations Permeate tank 2.

10 μm			10 μm /0,8 μm		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	1,20E+05	1,03	Mn-54	9,13E+04	0,83
Co-57	6,87E+03	8,66	Co-57	4,14E+03	4,44
Co-58	5,29E+05	0,43	Co-58	3,96E+05	0,26
Co-60	2,58E+06	0,16	Co-60	1,82E+06	0,1
Zn-65	2,07E+04	14,11	Zn-65	1,64E+04	6,73
Nb-95	2,13E+05	1,1	Nb-95	1,47E+05	0,67
Zr-95	9,05E+04	2,29	Zr-95	6,18E+04	1,77
Ru-106	4,35E+04	26,75	Ru-106	2,69E+04	15,99
Ag-110m	1,44E+06	0,42	Ag-108m	1,83E+03	28,66
Sn-113	6,40E+03	20,54	Ag-110m	9,05E+05	0,38
Sn-117m	1,16E+04	7,63	Sn-113	4,29E+03	12,12
Sb-124	1,08E+05	1,39	Sn-117m	2,21E+03	13,31
Total	5,17E+06	0,57	Sb-124	6,65E+04	1,16
			Sb-125	1,47E+04	8,13
			Ce-144	5,87E+03	24,46
			Total	3,56E+06	0,35
10 μm /0,8 μ	1m/300.000 E	Da	10 μm /0,8 μm/300.000 Da/100.000 Da		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	2,72E+02	8,3	Mn-54	1,18E+02	15,35
Co-57	5,66E+02	2,82	Co-57	2,31E+02	6,08
Co-58	4,93E+04	0,29	Co-58	1,98E+04	0,7
Co-60	4,67E+03	1,11	Co-60	2,02E+03	2,65
Ag-110m	1,34E+03	6,25	Ag-110m	1,99E+02	26,62
Sb-124	1,27E+03	3,76	Sb-124	1,40E+03	5,21
Total	5,74E+04	0,63	Total	2,38E+04	1,48

Table G6. Filtrations of Concentrate tank 2.

## Appendix H

This Appendix gives the data of all HPGe-analysis performed on the samples of tank 3. In table H1 and H2 are the data from the characterisation, and table H3 lists the data from test 3. Table H4 and H5 gives the data from the sequence of filtrations and table H6 from the IX experiments.

Nuclide	Bq/L	Unc. (%)
K-42	1,05E+04	27,4
Mn-54	9,95E+03	1,36
Co-57	6,00E+02	5,85
Co-58	3,93E+04	0,48
Co-60	1,82E+05	0,2
Zn-65	2,08E+03	12,36
Nb-95	1,22E+04	1,32
Zr-95	5,41E+03	2,9
Ag-110m	8,18E+04	0,76
Sn-113	4,49E+02	19,23
Sb-124	6,01E+03	2,41
Total	3,50E+05	1,71

Table H1. Characterisation of tank 3.

## Table H2. Filtrations of tank 3.

10 µm			10 μm /0,8 μm		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	1,09E+04	1,24	Mn-54	9,30E+02	4,44
Co-57	6,32E+02	4,92	Co-57	3,81E+01	25,44
Co-58	3,98E+04	0,47	Co-58	3,34E+03	1,56
Co-60	1,91E+05	0,19	Co-60	2,15E+04	0,53
Zn-65	1,81E+03	14,47	Zr-95	7,15E+02	7,48
Nb-95	1,42E+04	1,27	Ag-110m	1,22E+04	1,30
Zr-95	6,25E+03	3,16	Sn-117m	1,20E+02	13,07
Ag-110m	8,11E+04	0,69	Sb-124	8,65E+02	6,48
Sn-113	4,69E+02	17,84	Total	3,97E+04	1,11
Sb-124	6,11E+03	2,50			
Sb-125	1,73E+03	11,93			
Ce-144	1,01E+03	22,88			
Total	3,55E+05	0,49			
10 µm /0,8	µm/300.000	Da	10 µm /0,8	µm/300.000	Da/100.000 Da
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Co-57	9,01E+00	25,99	Co-58	5,90E+01	10,18
Co-58	5,73E+02	2,85	Co-60	2,33E+01	20,79
Co-60	1,06E+02	8,71	Ag-110m	8,61E+01	15,73
Ag-110m	5,72E+02	5,29	Sb-124	8,01E+01	15,78
Sb-124	1,01E+02	13,37	Total	2,49E+02	16,15
Total	1,36E+03	5,61			

3P1	3P1			3C1		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Mn-54	7,31E+00	25,00	Mn-54	1,40E+04	1,42	
Co-58	1,98E+01	11,28	Co-57	9,41E+02	8,65	
Co-60	1,11E+02	4,55	Co-58	5,36E+04	0,65	
Ag-110m	9,46E+01	9,71	Co-60	2,61E+05	0,26	
Sn-117m	6,91E+00	14,04	Nb-95	1,61E+04	2,06	
Sb-124	3,12E+01	14,73	Zr-95	7,50E+03	4,92	
Total	2,71E+02	8,75	Ag-110m	1,19E+05	0,75	
			Sb-124	8,21E+03	2,53	
			Total	4,80E+05	0,55	
3P2			3C2			
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Co-58	6,62E+00	31,33	Mn-54	1,76E+04	1,55	
Co-60	2,58E+01	11,79	Co-57	1,12E+03	8,81	
Ag-110m	2,80E+01	19,21	Co-58	7,14E+04	0,48	
Sn-117m	7,41E+00	14,01	Co-60	3,23E+05	0,23	
Sb-124	3,29E+01	12,70	Zn-65	2,84E+03	17,51	
Total	1,01E+02	15,51	Nb-95	2,27E+04	1,63	
			Zr-95	9,82E+03	4,20	
			Ag-110m	1,45E+05	0,90	
			Sn-113	9,78E+02	21,58	
			Sh 124	$1.00E \pm 0.4$	2 27	
			30-124	1,001.704	2,21	

Table H3. HPGe-data of test 3.

3P3			3C3		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	1,59E+01	21,13	Mn-54	1,07E+05	1,12
Co-58	5,33E+01	8,24	Co-57	6,80E+03	5,25
Co-60	5,23E+02	2,01	Co-58	4,08E+05	0,38
Nb-95	2,71E+01	21,49	Co-60	2,09E+06	0,14
Ag-110m	3,71E+02	4,37	Zn-65	2,45E+04	6,81
Sn-117m	9,00E+00	20,69	Nb-95	1,38E+05	1,06
Sb-124	2,89E+02	4,55	Zr-95	6,01E+04	2,66
Sb-125	7,05E+01	12,08	Ag-110m	1,05E+06	0,47
Total	1,36E+03	3,86	Sn-113	4,91E+03	16,94
			Sn-117m	6,41E+03	7,09
			Sb-124	6,71E+04	1,38
			Ce-144	1,07E+04	25,34
			Total	3,97E+06	0,37
Permeat tank 3	3		Concentrate tank	3	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Be-7	1,09E+02	15,92	Mn-54	1,58E+05	1,00
Mn-54	9,68E+00	19,91	Co-57	9,60E+03	5,03
Co-58	6,19E+01	4,88	Co-58	6,06E+05	0,33
Co-60	1,06E+02	4,72	Co-60	3,07E+06	0,12
Ag-110m	7,83E+01	10,81	Zn-65	2,14E+04	11,74
Sn-117m	7,88E+00	13,06	Nb-95	2,06E+05	0,92
Sb-124	6,90E+01	9,45	Zr-95	9,17E+04	2,29
Total	4,42E+02	9,65	Ag-108m	5,50E+03	19,36
			Ag-110m	1,57E+06	0,43
			Sn-113	6,36E+03	17,99
			Sb-124	9,88E+04	1,21
			Sb-125	2,97E+04	8,27
			Ce-144	1,70E+04	20,54
			Total	5,89E+06	0,34

10 µm	10 μm			10 μm /0,8 μm		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Be-7	8,31E+01	22,71	Be-7	9,05E+01	18,15	
Mn-54	8,36E+00	29,55	Mn-54	8,22E+00	23,50	
Co-58	8,28E+01	4,92	Co-58	7,49E+01	5,02	
Co-60	1,16E+02	4,71	Co-60	7,74E+01	5,30	
Ag-110m	9,08E+01	11,01	Ag-110m	8,05E+01	9,72	
Sn-117m	8,75E+00	18,96	Sb-124	7,75E+01	8,06	
Sb-124	8,61E+01	8,04	Total	4,09E+02	9,84	
Total	4,76E+02	9,94				
10 μm /0,8 μ	ım/300.000 D	a	10 μm /0,8 μm/300.000 Da/100.000 Da			
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Co-58	2,55E+01	9,95	Be-7	3,91E+01	28,12	
Co-60	9,95E+00	23,60	Co-58	2,51E+01	6,91	
Ag-110m	3,68E+01	15,53	Co-60	1,04E+01	22,51	
Sb-124	6,75E+01	9,33	Sb-124	6,19E+01	9,29	
Total	1,40E+02	13,14	Total	1,37E+02	18,67	

Table H4. Filtrations of Permeate tank 3.

10			10			
10 µm			10 μm /0,8 μ	m	-	
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Mn-54	1,29E+05	1,76	Mn-54	6,42E+04	1,65	
Co-57	7,81E+03	8,71	Co-57	4,29E+03	9,29	
Co-58	4,96E+05	0,48	Co-58	2,49E+05	0,74	
Co-60	2,64E+06	0,21	Co-60	1,32E+06	0,27	
Zn-65	2,39E+04	13,1	Zn-65	1,31E+04	20,82	
Nb-95	1,71E+05	1,68	Nb-95	8,49E+04	2,21	
Zr-95	7,75E+04	3,94	Zr-95	3,75E+04	5,44	
Ag-110m	1,38E+06	0,54	Ag-110m	7,44E+05	0,68	
Sn-113	5,78E+03	27,85	Sn-113	5,23E+03	19,21	
Sb-124	8,40E+04	1,96	Sn-117m	5,45E+03	10,21	
Sb-125	2,22E+04	16,56	Sb-124	4,40E+04	2,63	
Total	5,04E+06	0,48	Total	2,57E+06	0,61	
10 µm /0,8	µm/300.000	Da	10 μm /0,8 μ	10 μm /0,8 μm/300.000 Da/100.000 Da		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)	
Mn-54	1,22E+02	13,28	Co-57	4,01E+01	12,37	
Co-57	3,27E+02	3,47	Co-58	3,31E+03	1,13	
Co-58	2,60E+04	0,4	Co-60	3,91E+02	4,11	
Co-60	2,93E+03	1,45	Ag-110m	2,40E+02	11,46	
Ag-110m	2,68E+03	2,65	Sn-117m	3,35E+01	15,74	
Sb-124	1,22E+03	3,73	Sb-124	1,32E+03	3,45	
Total	3,33E+04	0,85	Sb-125	2,37E+02	12,61	
			Total	5,57E+03	2,65	

Table H5. Filtrations of Concentrate tank 3.

Table H6. HPGe-data after the IX experiments.

Column 1			Column 2		
Nuclide	Bq/L	Unc. (%)	Nuclide	Bq/L	Unc. (%)
Mn-54	5,83E+00	21,96	Mn-54	9,83E+00	12,91
Co-58	9,07E+00	15,03	Co-58	1,78E+01	8,58
Co-60	7,64E+01	3,75	Co-60	9,53E+01	3,13
Ag-110m	4,83E+01	10,07	Ag-110m	5,49E+01	9,41
Sn-117m	6,66E+00	11,57	Sn-117m	9,91E+00	10,44
Sb-124	7,32E+01	5,75	Sb-124	7,16E+01	6,16
Total	2,19E+02	6,68	Total	2,59E+02	5,98