



Removal of Strontium Ions in Aqueous Solution by Polymer

Enhanced Ultra-filtration

A Major Qualifying Project Report

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Ву

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Abstract

It is shown in many laboratories and pilot plants that membrane techniques are more effective than other traditional treatments for low level radioactive liquid waste. By applying membrane techniques, energy consumption can be reduced and better removal rate can be achieved. In this project, research on a polymer enhanced ultra-filtration process for strontium ions removal was conducted. A dead end ultra-filtration system was used. Operating parameters such as feed pH values and polymer loading ratio were varied in order to optimize the system. Up to 10% of strontium ions in the feed were removed and the system can be further improved.

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Introduction

As a clean and promising energy source, nuclear energy is getting more and more widely used around the world nowadays. Radioactive wastes generated from the nuclear power plant therefore become an environmental concern. Due to the hazard of the radioactive waste to the environment and public health, the waste need to be treated before discharged to the environment.¹

Traditional treatments on radioactive liquid waste usually are direct evaporation, conventional filtration, chemical precipitation, activated carbon adsorption and electrodialysis, or combinations of these processes. However, these traditional processes generate large amount of radioactive solid waste, and the treated liquid usually is not safe enough to be discharged directly to the environment.²

In the past five to ten years, membrane technology has been applied to radioactive liquid waste treatment in order to get a better solution to radioactive liquid waste.² In the case of this major qualifying project (MQP), a polymer-enhanced ultra-filtration (PEUF) process for low level radioactive waste water treatment is studied. In this project, treatments for radioactive waste containing ⁹⁰Sr were focused. In the experiments, non-radioactive solution of strontium nitrate was used instead of ⁹⁰Sr for simplification and safety concerns. The membrane used in the process is polyethersulfone (PES) due to its resistance to radioactivity. Polymer used to enhance the ultra-filtration process was water soluble Chitosan, a polymer that can be obtained from sea food waste.

The main objective of this project is to find the optimal operating conditions of the PEUF process for strontium ion removal. Operating parameters that affect the effluent concentrations significantly are pH values, concentration and polymer to metal ratio in the feed stream, as well as complexation reaction time and stirring rate of the continuous stirred reactor. In this project, pH values and polymer to metal ratio of the feed stream were varied aiming to find the optimal conditions. Also, fouling of membranes and cleaning techniques were investigated.

Background

Nuclear Energy

Nuclear energy is an energy obtained from a process called fission, in which uranium atoms split. The energy released from the fission process can be used to heat up steam, which can be put through a turbine and generate electricity.³ Compared to traditional power plants, in which fossil fuels are burned to generate electricity, nuclear power plants are much less harmless to the environment if operated correctly. There is no carbon dioxide, sulfur dioxide or nitrogen oxides emitted from a nuclear power plant.³ Even though fossil fuels are needed in the uranium mining and enrichment process, the amount of fossil fuels used is still much less than the amount used in a fossil fuel based power plant. Uranium is a non-renewable resource. However, a nuclear power plant is much more efficient than a traditional fossil fuel based power plant. The heat produced by the fission of a uranium ion is 10 million times of the heat produced by combustion of a carbon ion from coal. Studies show that the existing uranium resource is abundant for the present nuclear reactors for hundreds of years. Also, except for the conventional uranium resources, there exist many other cheap uranium resources, such as uranium in sea water and uranium impurity in coal. Such resources will not be exhausted in some thousands of years.⁴ As fossil fuels resources are getting scarce and other clean energy alternatives such as solar energy are not mature enough to be used, nuclear energy is very likely to be the most certain future source.

After the first commercial nuclear energy station started operation in the 1950s, many nuclear power plants were built every year all around the world. Up until today, there are more than 400 commercial nuclear reactors under operation in 30 countries, and provide about 14% of the world's electricity as shown in figure 1.⁵⁻⁶



Figure 1 World Electricity Generation6

Radioactive wastes

However, due to the wide spread of nuclear power plants, public raised concerns over nuclear safety. Radioactive materials used and generated during nuclear reaction are highly hazardous to the environment and public health, and therefore need to be carefully handled, recycled or disposed.¹ A nuclear accident in a nuclear power plant will of course release radioactive materials to the environment, ⁷ but wastes generated from nuclear reactors are also radioactive.¹ It can be very dangerous If such wastes are not well treated before discharged to the environment. In United States, radioactive wastes are categorized into high-level, transuranic and low-level waste.⁸ In this project, treatments for low level radioactive liquid waste (LLRLW) are focused.

Uranium fuel cycle

The nuclear fuel cycle is a set of processes that involve producing electricity by uranium fission reaction in nuclear reactors. ⁹The cycle is divided into two parts, the front end and the back end. The front end of the cycle involves mining, milling, conversion, enrichment and fuel fabrication, while the back end refers to procedures after uranium is spent, such as temporary storage, reprocessing, recycling and waste disposal.¹⁰

Figure 2 is a block diagram indicating different processes of the uranium fuel cycle.



Figure 2 The Nuclear Fuel Cycle⁹

Sources and characteristics of LLRLW

LLRLW is generated from both parts of the fuel cycle, but a large proportion of the waste is waste water from cooling and cleansing of the nuclear reactor. ¹¹ According to the regulations given by Chinese government, LLRLW is defined as radioactive liquid waste that has radioactivity below 4×10^7 Becquerel per meter cube. In this project, the concentration of the waste stream was calculated according to this definition. In the LLRLW from a nuclear power plant, the majority of nucleotides are fission products. These fission products can be categorized into three different groups: long-lived members such as ⁹⁰Sr and ¹³⁷Cs; medium-

lived members such as ⁹⁵Zr, ⁹⁵Nb, ¹⁴⁴Ce, ¹⁴⁴Pr, ¹⁰⁶Ru, ¹⁰⁶Rh, ¹⁴⁷Pm and ¹⁵⁴Sm; and short-lived nuclides with half-lives ranging from a few seconds to days that can be neglected. ¹² In this project, only ⁹⁰Sr was considered for simplification.

Treatments for LLRLW

Although the radioactivity of LLRLW is not high, the amount produced is enormous. The total volume of low level waste generated in United States is larger than the volumes of high level waste and transuranic waste combined.¹³ Therefore, the main objective of LLRLW treatment is to reduce the waste volume. The radioactivity of the waste will increase as concentrated, and therefore can be further treated as a high level radioactive waste. Due to the complexity of the components in LLRLW, the wastes usually have to go through a combination of several different treatments before discharged. There are currently five well-established and widely used treatments for LLRLW: chemical precipitation, evaporation, electrodialysis, activated carbon adsorption and ion-exchange.

Chemical precipitation

Chemical precipitation techniques are particularly suitable for radioactive waste that comes in large volumes and contains small amount of radioactive elements.¹⁴ Nucleotides in LLRLW, which usually exist as heavy metal ions, can react with soluble chemicals to form precipitation. Radioactive nucleotides then can be separated from aqueous solution via filtration. ¹⁵ Chemical precipitation methods are well established and widely used for low to intermediate level wastes treatments. Being a batch process, it is simple to operate and allows radioactive sludge to be removed periodically. There are a number of different precipitation

processes according to different compositions of radioactive liquid wastes. For radioactive wastes containing mostly strontium-90 ions, calcium phosphate, iron hydroxide or calcium carbonate precipitation methods are usually applied.¹⁵ The drawbacks of chemical precipitation are: the decontamination factors in these processes are generally low; decontamination factors are sensitive to pH, ionic strength, degree of flocculation, temperature, and the presence of suspended matter, which increases the difficulties for process control; to reduce the radioactivity of the effluent liquid to a satisfactory level, it is necessary to combine chemical precipitation process with other efficient methods.¹⁵

Evaporation

Evaporation is one of the earliest and most straightforward approaches for removal of radioactivity from LLRLW.¹⁷ It also provides good decontamination factors and volume reduction. As shown in figure3, the mechanism of evaporation process is simple. Liquid wastes is heated and distillated in a distillation column, where water is removed as vapor and non-volatile contents that contains most of the nucleotides are left behind. The vapor is cooled before discharged, and the remaining can be treated as high level radioactive waste. This method is very mature and has been applied in the nuclear power industry for many years. However, evaporation process can cause problems such as corrosion, foaming or scaling. Also, the high temperature and pressure required during the process can not only raise the cost, but also cause explosion.¹⁵



Electrodialysis

Electrodialysis is a process in which salts ions are transferred through ion-exchange membranes according to the electric potential difference in the solution. LLRLW discharged from a nuclear power plant usually has high electric conductivity due to the existence of a large amount of inactive ions. ¹⁸ Thus radioactive heavy metal elements can be separated and recovered from the aqueous solution via electrodialysis method. It is simple to operate instruments used in these processes and no regeneration processes are necessary.¹⁸ Also, the interference by other coexistent ions is insignificant, which is suitable for waste streams that have complex compositions. However, there exist equilibrium concentrations for heavy metal ions, below which electrodialysis process will slow down. ¹⁸ This method is very suitable for high level radioactive waste with a large amount of radioactive heavy metal ions but not for dilute LLRLW. The concentration of heavy metal ions in LLRLW is already so low that may not be brought down further by electrodialysis process.

Activated carbon adsorption

Adsorption techniques are generally utilized in waste water treatments for removal of color, odor and organic maters. Adsorbents, which are usually porous solid materials, are used to adsorb impurities in waste water. The most commonly used adsorbent is activated carbon. Oxygenated functional groupings, which can be found on the entire surface of activated carbon, are capable of adsorbing metal ions from aqueous solution. Activated carbon adsorption techniques are also applied in LLRLW treatments. This method is very effective and gives good decontamination factors.¹⁹ However, the cost of applying activated carbon is also relatively high due to the high cost of activated carbon and the difficulty of adsorbent recovering.

Ion-exchange

In ion-exchange processes, ions exchange between two electrolytes or between an electrolyte solution and a complex. Radioactive heavy metal ions in LLRLW can be removed by putting the contaminated stream through a fixed bed reactor packed with ion-exchange resins. Ion-exchange methods have been widely applied in the treatments for waste liquid streams from nuclear power plants for many years, but these methods can only treat liquid wastes that meet the following criteria: there should be small amount of suspended solid in the liquid; total salt contents in the wastes should be low; the radioactive elements in the wastes should be in ionic form. In addition, ion-exchange resins need to be regenerated when the saturation of the active groups is reached, or they will be incapable of ions exchanging. Therefore, an ion-exchange process requires a great effort in maintenance such as flushing, regeneration, rinsing and refilling. ¹⁵ Also, during the regeneration of ion-exchange resins, chemicals used can be toxic and expensive, resulting in the increase of operating cost.

Application of membrane techniques in LLRLW treatments

As membrane separation technologies develop, their applications are expanding rapidly all over the world among variety of industries including liquid radioactive waste processing. During the last five to ten years, membrane techniques have been gradually introduced to nuclear power plants.²Compared to traditional LLRLW treatments, membrane methods are more energy-saving and effective.²⁰ Traditional LLRLW treatments processes usually generate large amount of radioactive solid waste that needs to be disposed. In addition, the treated liquid does not have a low enough radioactivity to be discharged to the environment.² Membrane techniques can also be combined with other treatment methods such as evaporation and ion-exchange to achieve better decontamination result.²⁰

Variety of membrane techniques

Membrane techniques application in LLRLW treatment needs to be customized for different sites according to the local conditions such as chemical and radiochemical compositions, radioactivity and total salinity of the effluents.² Different membranes, membrane modules and other related instruments need to be selected accordingly.²⁰

Currently, there are two main membrane techniques that are tested in pilot plants all over the world: reverse osmosis (RO) and membrane distillation (MD).²⁰ In this project, a polymer enhanced ultra-filtration process was studied.

Reverse osmosis

In a RO process, contaminated liquid is pressed to a porous membrane that only allow small molecules to pass through. Radioactive elements thus can be blocked and remain in the

concentrate. According to the different size of membrane pores and permeable particles sizes, RO process can be categorized into reverse osmosis, nano-filtration, ultra-filtration, micron-filtration and particle filtration as seen in figure 4.²¹

Figure 4 Membrane Seperation Processes²¹

Results from pilot plants all around the world showed that RO process gave good volume reduction and decontamination factors if multistage process was applied. The productivity is between 0.05 and 4 m³/h, which is sufficiently big for the application of this method in a commercial scale with low cost. However, the RO process require a relatively high pressure, ranging from 4 to 5 MPa, and the permeate flux is greatly influenced by the concentration of contaminated liquid being treated.²⁰

Overall, RO processes are effective for low to intermediate level radioactive waste treatments. The radioactivity of the effluent stream is significantly lowered after the treatment with a sufficiently big contamination factor.

Membrane Distillation

Although RO processes were proved to be efficient, there still exist drawbacks and limitations: membrane fouling occurs and cleaning is needed, resulting in interruption of the processes and extra waste generated from membrane cleaning process; high pressure is required in the processes, which can increase the cost and risk; pre-treatment may be necessary due to existence of complexants. By applying thermal evaporation method involving hydrophobic porous membranes, also known as membrane distillation, such limitations can be overcome.²⁰

Membrane distillation is achieved by putting a hot concentrated stream on one side of a membrane and cold clean water stream on the other side. Vapor pressure difference due to temperature difference will drive molecules from the hot side evaporate and permeate to the cold side. Big particles will be blocked by the small pores on the membrane and left in the concentrate. However, there are requirements for membranes used in MD processes. These membranes need to be hydrophobic, thin, with low thermal conductivity and have high surface tension with the feed stream.²³

MD techniques were also tested in a few pilot plants. Data shows that retention of radioactive ion was complete that the effluent stream was safe to be discharged to the environment directly. No high pressure was required in the process and the high concentration of the feed did not influence the process. However, MD methods are still premature and have not yet been utilized in nuclear industry. The process time is too long that productivity is as low as 0.01 to 0.06 m³/h. Also, the large amount of energy consumed in heating up the feed can result in high cost if cheap energy sources are not available.^{20, 22}

Polymer enhanced ultra-filtration (PEUF) using Chitosan

Since most of metal ions mostly have molecular weight below 200, which are even too small for reverse osmosis. Pre-treatments are needed before the contaminated liquid going into membrane filtration processes. By letting heavy metal ions in the waste streams react with large organic molecules, large complexes can be formed and filtered by membranes.²⁴ Such process is called polymer enhanced ultra-filtration. Polymer used in these processes usually required to have large molecular weight, good solubility in water, good binding with metal ions, good stability in complexes formed and no toxicity, and to be available in low price.²⁴In this project, Chitosan is used as the ligand for soluble strontium ions.

Chitosan is a polymer produced from chitin that usually can be found in the shells of sea crustaceans such as shrimp. Thus cheap sources of chitin such as sea food waste are usually available. Chistosan is constructed mostly by glucosamine residues. N-acetyl-glucosamine also exists but in a smaller percentage. The molecular structure of Chitosan is shown in figure 5, where part A is glucosamine and part B is N-acetyl-glucosamine. Chitosan is capable of binding with all kinds of metal ions in aqueous solution. The amine group on the glucosamine is the major binding site for metal ions. Although Chitosan has a very poor solubility in neutral water, it can however be dissolved in acid. ²⁵In this project, hydrochloric acid was used to dissolve Chitosan powder.

Figure 5 Molecular structure of Chitosan25

Currently, few researches have been done on strontium ions removal by PEUF and no publication can be found on such researches. However, related researches on Chitosan interactions with metal ions and removal ^{25, 27} of heavy metal from aqueous solution by PEUF ^{20, 24-26} show that this method is effective and more advanced than existing nuclear liquid waste treatments.

PEUF methods are proved to be effective for removal of metal ions from aqueous solution. ^{20, 24-26} A research on removal of mercury from aqueous solution showed that the retention rates were 0.97 in average and can reach 0.99 under optimal conditions for mercury.²⁶ Also, Chitosan is highly effective in binding with metal ions even in low concentration according to related researches.^{25, 27} It was also proved that Chitosan was effective in a PEUF process for removal of mercury from diluted solution.²⁶

PEUF methods are low cost and very likely can be done in an industrial scale. Although membrane distillation methods were proved to be even more effective, the high cost of the process makes it difficult to be widely applied.²⁰ On the other hand, PEUF methods have no high temperature requirement and relatively low pressure is used. In this project, the gauge pressure inside the stirred cell was 0.1 MPa. Also, chemicals required in these processes are common and inexpensive. Chitosan can be obtained in very low-price sources such as sea food wastes. Moreover, membranes used in these methods do not need to be changes frequently, since they have a relatively long life that usually lasts for two to three years.²⁰

The advantages of PEUF methods give strong incentives for conducting related researches. The researching team of Professor Shao from Shanghai Jiaotong University is one of the research groups that first study the performances of PEUF methods for strontium removal. This project was supported by Professor Shao's researching team.

The operation for PEUF processes is not complicated. Water soluble polymer and solution containing metal ions can react in batch reactors. After the feed is prepared, it can be pumped into a batch membrane separator. The parameters that have significant effect on the effluent concentrations are pH value of the feed, the proportion of amount of polymer to amount of metals in the solution and reaction time.

This project was aimed to study the performance of PEUF techniques for strontium ions removal as well as to optimize the process by finding the optimal operation conditions. The examined operation parameters were pH values and loadings of the feed. Also, fouling of membranes was also studied. Simple membrane cleaning techniques using common chemicals were investigated as well.

Methodology

Set-up of the PEUF process

Figure 6 Picture of the ultra-filtration unit

Figure 6 shows the equipments used and the set up of the ultra-filtration process. As seen in figure 6, a gas tank containing nitrogen was connected to the stirred cell membrane separator. As nitrogen flowed into the stirred cell, pressure increased. A pressure valve with a pressure gauged was used to adjust the pressure inside the stirred cell. Under the stirred cell, there was a magnetic stirrer, which allow the solution in the stirred cell be continuously stirred during the separation process.

Membrane preparation

New membrane is usually soaked in solution containing some organic chemicals to keep it from drying. The pores on the membrane might be blocked by these big molecules. Therefore, prior to be used, the membrane needs to be treated so that the pores are open. When the membrane is soaked in isopropanol, molecules of isopropanol can travel through the membrane pores and open them up.

New PES membrane was cut to fit the stirred cell. After cut, membrane was soaked with isopropanol in a plastic Petri dish with cover for one hour. Afterwards, membrane was washed with DI-water and soaked in DI-water in a plastic Petri dish for at least 9 hours.

Solution preparation

Feed solutions were prepared and stored in volumetric flask for convienience.

For Chitosan solution, 1g of Chitosan was dissolved in nitric acid in a beaker and diluted with DI-water in a 1000ml volumetric flask. The Chistosan solution with concentration of 1g/L was stored in the volumetric flask for future use.

For strontium nitrate solution, 0.9g of strontium nitrate was dissolved in DI-water in a beaker and diluted with DI-water in a 1000ml volumetric flask. The strontium nitrate solution with concentration of 0.9g/L was stored in the volumetric flask for future use.

Membrane flux measurement

Membrane flux (N_w) refers to the amount of water permeate the membrane per unit area and time. The flux needed to be measured before and after the membrane is used in order to study membrane fouling. If a membrane is fouled, its pores are blocked, thus results in a smaller membrane flux. When the membrane flux difference between before and after used is bigger than 15%, this membrane is considered fouled and needs to be cleaned.

Membrane flux is calculated by:

$$N_w = V/(t \times A)$$

Where V is the volume of water that permeates through the membrane, t is the time required for the water to permeate the membrane, and A is the effective area of the membrane.

Since it is difficult to measure the volume of water accurately, the changes in the weights of test tubes that contain the permeate water are used instead:

$$N_w = (m_a - m_b)/(t \times A \times \rho_w)$$

Where m_a is the weight of a test tube before filled with water, m_b is the weight of test tube after filled with water and ρ_w is the density of water.

Membrane was fit into the bottom of the stirred cell. About 250ml of DI-water was filled in the stirred cell. 5 test tubes were weighted and the weights were recorded as m_b . The valve on the Nitrogen tank was turned on and pressure was adjusted to 0.05 MPa by a valve on the pressure gauge. The pressure was left unchanged for 20 minutes to ensure that it was stable and the system had reached equilibrium. Water came out during this period was disposed. When the system reached equilibrium, a test tube was used to catch the water coming out from the stirred cell. Time needed to fill the test tube up to 10ml was measured by a stop watch and recorded as t. Pressure was changed to 0.075, 0.01, 0.125 and 0.15MPa, and permeated water was collected for each pressure.

After the pressure was changed each time, it was left unchanged for 10 minutes before the permeate water was collected to ensure that the system has reached equilibrium. The weights of the test tubes with water were measured and recorded as m_a. Membrane flux under each pressure was calculated with the equation mentioned above. The calculated fluxed were plotted against the pressures, and a linear line was fit to these date points. If the R² value of the line is greater than 0.9, there membrane fluxes are consider valid.

Chitosan permeation

Chitosan obtained from natural sources such as sea food wastes usually contains molecules in different sizes. Before Chitosan reacts with strontium ions to form complexes, it is essential to ensure that the molecules of Chitosan are big enough to be blocked by the membrane pores, so that the complexes cannot permeate the membrane. If a large amount Chitosan permeates the membrane, the Chitosan solution needed to be filtered by the membrane, and only concentrate should be used to react with strontium ions.

Before filtering the Chitosan solution, the flux of the membrane used was measured. The stirred cell was rinsed with Chitosan solution with concentration of 1g/L and pH of 1.18 for three times to ensure that all the impurities were washed away. 250ml of the same Chitosan solution was filled into the stirred cell. Pressure inside the stirred cell was adjusted to 0.1 MPa and was stabilized for 10 minutes before samples were collected. 4 test tubes for collecting sample were marked as J1, J2, J3 and J4. When the pressure was stable, liquid coming out from the cell was collected by test tube J1 and a clock was started to record the total time of the process. A stop watch was used to record the time needed for the collection of approximately 10ml of permeate. When the total time of the process hit 10, 20, 30minutes, approximately 10ml of permeate were collected by test tube J2, J3 and J4 respectively. Waste permeate coming out in between was disposed. After all the samples were collected, the stirred cell was rinsed with DI-water for three times, and another group of membrane fluxes were measured. Same steps were repeated for Chitosan solution with PH values of 8 and 5.

Chitosan enhanced ultra-filtration

Chitosan can react with strontium ions in aqueous solution to form complexes with large molecular weights. These complexes are too big to go through the membrane pores so that strontium ions can be removed from the aqueous solution. The objective of this part of the project is to find the optimal operating pH value and loading.

Loading (L) is a dimensionless value that indicates the proportion of the amount of metal ions and polymer in the aqueous solution. It can be calculated using:

 $L = m_p/m_m$

Where m_p is the mass of polymer in the aqueous solution and m_m is the mass of metal salt in the solution. In this case, the polymer is Chitosan and the salt is strontium nitrate.

Experiments on different pH values:

Loading was set at 1 during these experiments. 9ml of Chitosan solution with concentration of 1g/L and 10ml of strontium nitrate solution with concentration of 0.9g/L were added to a 500ml beaker. 400ml of DI-water was added to the beaker to dilute the mixture. Similarly, 8 other beakers with 419ml of Chitosan and strontium nitrate solution were made. The pH values of the solution in these beakers were adjusted to 1, 2, 3, 4, 5, 6, 7, 8 and 9 respectively by HCl and NaOH. A pH meter was used to measure the pH values. After the pH values were adjusted, DI-water was added to each beaker so that the total volume of the solution reached 500ml. The solution in the beakers was stirred at a rate of approximately 200rad/min for 2 hours to let Chitosan and strontium ions react. The prepared solution was used as the feed to the stirred cell. Stirred cell was rinsed with the feed for three times to wash off impurities. 250ml of the solution was filled in the stirred tank. Pressure was set at 1 MPa throughout the whole process. The system was stabilized for 20 minutes first, then about 25ml of permeate was collected by a test tube as the permeate sample. The remaining liquid in the stirred cell, the concentrate, and the feed was collected by test tubes as well. All samples were sealed by plastic films for further concentration determination. Same procedure was repeated for each pH. Before the permeation experiment of each pH, membrane fluxes of the membrane were measured. If the differences of the fluxes were greater than 15%, the membrane will be replaced by a new one.

Experiments on different loadings

After the optimal pH was found, experiments on different loadings were done at this pH value. 0.72ml and 0.8ml of Chitosan solution with a concentration of 1g/L was added to two different beakers, and another beaker was left empty. 10ml of strontium nitrate solution was added to each beaker, and the mixture was diluted by 400ml of DI-water. The loadings of the solution in these beakers were then 0, 0.2 and 0.8 respectively. The three beakers of solution were adjusted to the optimal pH, and were stirred for two hours in order to let the complexation reaction occur. Same ultra-filtration step from the different pH values experiment was repeated for the feed with different loadings. Samples collected were also sealed by plastic films for future analysis.

Membrane cleaning

Membrane cleaning procedures by common chemicals such as nitric acid and sodium hydroxide were studied. The fouled membrane was first soaked in nitric acid solution with a pH value of 2 for 20 minutes. Then membrane fluxes of the membrane were measured and compared with the membrane fluxes before the membrane was treated. If no significant improvement was shown, the membrane would be soaked in sodium hydroxide solution with a pH value of 8 for 20 minutes, and the membrane fluxes would be measured and compared again.

Retention rate calculation

The most straight forward way to show the effectiveness of the separation unit is strontium concentration in the outlet stream. However, retention rate is a better way to demonstrate the results, since it indicates how much strontium is removed from the feed.

The equation for retention rate is:

$$R = 1 - \frac{c_{Sr \ permeate}}{c_{Sr \ feed}}$$

where $c_{Sr \ permeate}$ is the concentration of strontium in the permeate and $c_{Sr \ feed}$ is the concentration of strontium in the feed.

Results and Discussions

This section of the report contains 4 different parts: Chitosan permeation, ultrafiltration of feeds with different pH values, ultra-filtration of feeds with different loadings, Membrane fouling and membrane cleaning procedures.

Chitosan permeation

Since samples from the first Chitosan permeation experiments were stored for a long time before they were measured, these samples could not show accurate results. Therefore, the experiment was repeated, but with a 50kd PES membrane instead due to the exhaustion of 30 kd PES membrane. Difference of membranes used did affect the results, but not too significantly and these results can be used in the analysis. In the new experiments, the concentration of the Chitosan feed was 10mg/L and the pH values were 8 and 5.

To determine the concentration of Chitosan, the total amount of nitrogen was detected first, and then the total amount of Chitosan was calculated accordingly. This method cannot measure Chitosan in dilute aqueous solution accurately. It is chosen because this method was the most reliable method that could be found. Therefore, to minimize the inaccuracy of this method, the concentration of Chitosan in the feed was also determined using this method, and was compared to the concentrations of Chitosan in the permeated samples.

Results are shown in figure 7.

Figure 7 Percentage of Chitosan permeates from feed

From this figure, it can be seen that at pH of 8, almost 100% of chitosan could permeate through the membrane. When pH is equal to 5, less Chitosan could permeate through the membrane, and the lowest percentage of Chitosan permeated was 10%. This indicates that this Chitosan solution was not efficient for the PEUF process. Since the size of the Chitosan molecules were too small that they could go through the membrane pores, the complexes formed by these Chitosan molecules and strontium ions can possibly permeate through the membrane pores as well, leading to the failure of ultra-filtration. This Chitosan solution should be filtered by membrane first, and only the part that cannot permeate through the membrane should be used in the PEUF process.

Ultra-filtration experiments on different feed pH values

In this part of the project, the pH value of the feed to the stirred cell was varied from 1 to 11. When pH value reached 11, participation occurred. Therefore, experiments were not performed on feed with a pH value greater than 11. Results of strontium concentrations in the effluents were shown in figure 8 and table 1. Figure 8 is a plot where the concentration of strontium nitrate is plotted against the pH values, and the values are listed in table 1.

Concentrations of strontium nitrates in the effluents were measured by spectrophotometer. However, the results were strange since some of the strontium nitrate concentrations in the effluents were greater than the concentrations in the feed. Such abnormal results may be due to the long waiting period between when the samples were taken and when the samples were measured. Chemical changes in the samples might occur during this period. The second reason for such results may be the inaccuracy of this determination method. It is not common to use a spectrophotometer to detect the amount of strontium ions in aqueous solutions, especially in dilute solutions. This method involved complicated steps of making and mixing of several types of solutions. Errors from each step would accumulate and therefore lead to the inaccuracy of the final results. Due to the limited time and budget, no other concentration determination method could be examined.

However, the main objective of this part of the experiments was to find the optimal operating pH, which would be applied in the next part of the experiments. It is sufficient to conclude from these results that the optimal operating pH for these processes was 7, since it gave the lowest concentration of strontium nitrate in the effluent.

Figure 8 Strontium nitrate concentration in the effluents versus different operating pH values

Table 1 Strontium nitrate concentration in the effluents under different operating pH values

рН	Permeate	Permeate (mg/L)	Feed
1	0.038	16.74107	18 mg/L
2	0.054	19.10714	
3	0.048	16.96429	
4	0.051	18.03571	
5	0.055	19.46429	
6	0.061	21.60714	
7	0.044	15.53571	
8	0.062	21.96429	
9	0.055	19.46429	
10	0.052	18.39286	
11	0.054	19.10714	
Ultra-filtration experiments on different feed loadings

In this part of the project, polymer loading of the feed to the stirred cell was varied in order to find the optimal values. The examined loadings were 0, 0.2 and 0.8. Results were shown in figure 9, table 2 and figure 10. Concentration of strontium nitrate is plotted against loading values of 0, 0.2 and 0.8 in figure 9, while retention rate is plotted against loadings in figure 10. Specific numbers from these two figures are listed in table 1.

As seen in the results, when polymer loading increased, concentration of strontium nitrate in the effluent decreased. The results were reasonable since when there are more polymer molecules available to react with the metal ions, more complexes will be formed. The polymer loading in this part of the experiment that gave the lowest strontium nitrate concentration in the effluent was 0.8. However, this polymer loading value could be concluded as the optimal due to small amount of data obtained. It could nevertheless be concluded that the bigger the polymer loading is the more metal ions will be removed until it reaches the equilibrium.

The results can also be presented as the retention rates under different polymer loadings. When loading was 0 and 0.2, the retention rates were 0, indicating that no metal ions were removed from the feed. When loading was increased to 0.8, retention rate also rose to 0.1, indicating that 10% of the strontium ions were removed from the feed. However, 10% removal is not impressive, showing that this system was not efficient enough. The inefficiency of the system possibly results from the small molecule sizes of the Chitosan solution. Due to limited time available, the ultra-filtration experiments were performed before the results from the Chitosan permeation experiment were available. Results from Chitosan permeation showed that large amount of Chitosan molecules were permeable, indicating that the Chitosan molecule sizes were not large enough. Therefore, complexes formed by reacting with this Chitosan solution might be too small to be blocked by the membrane pores, resulting in the inefficiency of strontium removal.



Figure 9 Strontium nitrate concentration in the effluent versus different loadings





Table 2 Data of retension rates and effuent concentration under different loadings

loading	Permeate (mg/L)	Retention	Feed (mg/L)
0	13.9	0	1.39
0.2	13.9	0	
0.8	12.5	0.10	

Membrane Fouling

Fouling of membranes was examined by comparing changes in membrane fluxes before and after used. Results were shown in figure 11, figure 12 figure 13 and table 3.

In figure 11, Chitosan solution flux through the membrane is plotted against the total

time of the process. As shown in figure 11, the flux of Chitosan permeated through the

membrane decreased along with the permeation process due to the blockage of membrane

pores by Chitosan molecules. Membrane flux under the same pressure also decreased after the membrane was used compared to before used, as can be seen in figure 12. The average difference between the membrane fluxes before and after used was 21%, indicating that this membrane was fouled and should not be used unless treatments were applied to bring the membrane flux back up.



Figure 11 Mebrane fouling in Chitosan permeation experiment



Figure 12 Membrane fluxes changes of membrane 1 for Chitosan permeation

Membrane fouling also occurred during the ultra-filtration process. In figure 13, membrane fluxes before and after used are plotted against different pressure values. As shown in figure 13, the membrane flux under the same pressure decreased after the membrane was used due to membrane fouling. Membrane 4 was used in the ultra-filtration process for a feed with loading of 1 and pH value of 2.

Same results were also obtained from ultra-filtration experiments under different operating pH values and polymer loadings, which were listed in table 3. The average membrane flux differences that were marked in red color were above 15%, indicating that membrane should be replaced. In the experiments for feeds with pH values varying from 1 to 8, membrane had to be replaced after one time used. The severe membrane fouling could be caused by the poor anti-fouling property of the PES membrane. No relation could be found between membrane fouling and operating pH values. However, results showed that membrane fouling was more severe when the polymer loading was large. Therefore, membrane fouling can also be caused by a large polymer ratio in the feed. When an operating polymer loading is chosen, its effect on membrane fouling should also be taken into account. Severe membrane fouling will require constant replacement of membranes, which results in a higher operating cost.





Table 3 Fouling of the membranes

Membrane	Operating PH	Operating	Avg. Flux %Diff.
No.		Loading	
2	1	1	20.27%
4	2	1	22.43%
5	3	1	22.85%
6	4	1	15.82%
7	5	1	31.62%
8	6	1	36.63%
9	7	1	38.92%
10	8	1	18.81%
11	9	1	2.55%
11	10	1	9.57%
11	11	1	19.40%
14	7	0.2	2.36%
14	7	0.8	11.59%

Membrane cleaning

Membrane cleaning procedures were studied in order to reactivate the fouled membranes. The objective of these procedures is to unblock the membrane pores using common and low-cost chemicals. Since it is believed that the membrane pores were blocked by Chitosan molecules, theoretically membrane needed to be treated by acid in order to dissolve the Chitosan attached to the membrane surface. Figure 14 shows the plot of membrane fluxes before and after cleaned against various pressure values. However, no significant improvement was shown in the membrane fluxes after treated as shown in figure 14. Membrane was therefore treated with base aiming to remove other organics from the surface of the membrane, but no significant improvement was shown either. As a matter of fact, membrane flux even decreased further after treated. Similar results were obtained from cleaning procedures on other membranes as seen in figure 39 attached in the appendix.

The failure in these membrane cleaning procedures can be explained by the disability to dissolve the adhered Chitosan. In fact, by soaking the membrane into acid and base solution may even help Chitosan to diffuse deeper to the membrane, causing a decrease in membrane fluxes after treated. Another reason that leads to the failure might be the chemical changes occurred on the membrane. Chitosan could undergo chemical changes by reacting with microorganisms and chemicals in the environment and form complex composite that cannot be dissolved in acid or base. In order to clean the membranes effectiveness, researches needed to be done in other chemicals that may be more effective.



Figure 14 Resutls of cleaning experiment of membrane 4

Conclusions and Recommendations

The objective of this project was to optimize the PEUF process for strontium ions removal. The optimal operating pH value was 7 and the optimal operating loading was 0.8. However, the removal was not effective even under these optimal operating conditions. The best retention rate found was only around 10%. The ineffectiveness of this process can be explained by the large number of permeable molecules in the Chitosan solution. Results from Chitosan permeation experiments showed that a large amount of Chitosan molecules could permeate through the membrane, which indicates that a large amount of complexes formed by Chitosan and strontium ions were also permeable. To achieve a better removal result, the Chitosan solution should be filtered by membrane first, and only the non-permeable part should be used in the PEUF process.

The PEUF process can also be further optimized by changing other operating parameters such as reaction time, reactor stirring rate and the ion strength of the feed. In addition, a multisteps system can be used to improve the performance of this process. Also, a cross-flow filtration system can be considered instead of the dead end filtration system used in this project. Membrane fouling was found to be serious. 8 out of 10 membranes used needed to be replaced after one time use. The severe fouling can be due to the poor fouling resistance of the membrane material, as well as the high concentration of polymer in the feed. Nitric acid with pH of 2 and sodium hydroxide with pH of 8 were not effective in membrane cleaning processes. The cleaning process can be improved by using other chemicals as the cleanser. Disodium is a good alternative.

References

1. Environmental Protection Agency (EPA) USA. Radioactive Waste Disposal: An

Environmental Perspective

http://www.epa.gov/radiation/docs/radwaste/ (Feb 3rd 2010)

- Anil Kumar Pabby. Membrane techniques for treatment in nuclear waste processing: global experience. Serono. Sym. 2008, November, 9-13
- 3. Environmental Protection Agency (EPA) USA. Nuclear Energy

http://www.epa.gov/RDEE/energy-and-you/affect/nuclear.html (Feb 2nd 2010)

4. John McCarthy. Frequently asked questions about nuclear energy

http://www-formal.stanford.edu/jmc/progress/nuclear-faq.html (Feb 2nd 2010)

5. ICTJ nuclear training centre. Nuclear plants in the word

http://www.icit.org/an/tech/jesvet/jesvet.htm (Feb 2nd 2010)

6. World Nuclear Association. Nuclear power in the word today

http://www.world-nuclear.org/info/inf01.html (Feb 2nd 2010)

7. Nuclear disasters and accidents

http://library.thinkquest.org/17940/texts/nuclear_disasters/nuclear_disasters.html

(Feb 3rd 2010)

8. United States Nuclear Regulation Commission. Radioactive wastes

http://www.nrc.gov/waste.html (Feb 3rd 2010)

9. World Nuclear Association. The nuclear fuel cycle

http://www.world-nuclear.org/info/inf03.html (Feb 3rd 2010)

10. Uranium USA. What is the Nuclear Fuel Cycle?

http://www.uraniumsa.org/ (Feb 3rd 2010)

11. Ohio State University. Who Creates Low-Level Radioactive Waste and what are they doing with it?

http://ohioline.osu.edu/rer-fact/rer 12.html (Feb 3rd 2010)

- 12. C.B Amphlett. Radioactive Wastes; Pergamon Press: London 1961; 9
- John F. Ahearne. Radioactive Waste: The Size of the Problem. *Phys Today* 1997, *June*, 24-29
- 14. A.S Mollah; Aleya Begum; M.M. Rahman. Removal of radionuclides from low level radioactive liquid waste by precipitation. *J Radioanal. Nuc.l Ch.* **1998**, *229*, 187-189
- V.M. Efremenkov. Radioactive waste management at nuclear power plants. *IEAE Bulletin* **1989**, *4*, 37-40
- 16. S.V.S. Rao; Biplob Paul; K.B. Lal; S.V. Narasimhan; Jaleel Ahmed. Effective removal of cesium and strontium from radioactive wastes using chemical treatment followed by ultra filtration. *J Radioanal. Nucl. Ch.* **2000**, *246*, 413-418
- G. E. McCullough. Concentration of Radioactive Liquid Waste by Evaporation. *Ind. Eng. Chem.* **1951**, *43*, 1505-1509
- Sen-ichi Sugimoto. Removal of Radioactive Ions from Nuclear Waste Solutions by Electrodialysis. J Nucl. Sci. Technol. 1978, 15, 753-759

- S. Chegrouche; A. Mellah; M. Barkat. Removal of strontium aqueous solutions by adsorption onto activated carbon: kinetic and thermodynamics studies. *Desalination* 2009, 235, 306-318
- 20. Grazyna Zakrzewska-Trznadel; Marian Harasimowicz; Andrzej G. Chmielewski. Membrane processes in nuclear technology-application for liquid radioactive waste treatment. *Sep. Purif. Technol.* **2001**, *22-23*, 617-625
- 21. KOCH membrane systems. Ultrafiltration-Filtration overview.

http://www.kochmembrane.com/sep_uf.html (Feb 16th 2010)

- 22. Grazyna Zakrzewska-Trznadel; Marian Harasimowicz; Andrzej G. Chmielewski. Concentration of radioactive components in liquid low-level radioactive waste by membrane distillation. *J Membrane Sci.* **1999**, *163*, 257-264
- 23. A. M. Alklaibi. Using Spacers in MD Channels

http://cds.comsol.com/access/dl/papers/1550/Alklaibi.pdf (Feb 16th 2010)

- 24. Grazyna Zakrzewska-Trznadel; Marian Harasimowicz. Removal of radionuclides by membrane permeation combined with complexation. *Desalination* **2002**, *144*, 207-212
- Carol L. Lasko; Matthew P. Hurst. An Investigation into the Use of Chitosan for the Removal of Soluble Silver from Industrial Wastewater. *Environ. Sci. Technol.* 1999, 33, 3622-3626
- 26. Yusuf Uludag; Hilmi Onder Ozbelge; Levent Yilmaz. Removal of mercury from aqueous solutions via polymer-enhanced ultrafiltration. *J Membrane Sci.* **1997**, *129*, 93-99
- 27. E. Guibal; E. Touraud; J. Roussy. Chitosan interactions with metal ions and dyes: dissolved-state vs. solid-state application. *World J Microb. Biot.* **2005**, *21*, 913-920

Appendix

A. Membrane Fluxes Data

Table 4 Fluxes of	membrane 1	before used
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mb (g)	ma (g)	P (Mpa)	t (s)	V (m^3)	Flux (m/s)
22.0648	32.3111	0.05	79.03	1.0246E-05	3.1017E-05
24.7807	34.7397	0.075	50.79	9.9590E-06	4.6910E-05
23.1744	33.4777	0.1	36.03	1.0303E-05	6.8413E-05
24.9858	35.1926	0.125	30.75	1.0207E-05	7.9409E-05
21.9846	33.6322	0.15	28.06	1.1648E-05	9.9305E-05



Figure 15 Fluxes of membrane 1 before used vs. pessure

Table 5 Fluxes of	membrane 1	after	used
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mb (g)	ma (g)	P (Mpa)	t (s)	V (m^3)	Flux (m/s)
23.8583	33.9008	0.05	93.15	1.0043E-05	2.5792E-05
25.9170	35.9451	0.075	71.28	1.0028E-05	3.3657E-05
26.7156	37.4285	0.1	48.44	1.0713E-05	5.2909E-05
23.1840	33.6260	0.125	38.59	1.0442E-05	6.4734E-05
22.2644	33.1118	0.15	33.15	1.0847E-05	7.8283E-05



Figure 16 Fluxes of membrane 1 after used vs. pressure

Membrane 2

Table 6 Fluxes of membrane 2 before used

mb	ma g	Р	t	V m^3	Flux m/s
21.8406	31.4068	0.05	82.73	9.5662E-06	2.7663E-05
23.3391	33.5146	0.075	63.74	1.0176E-05	3.8192E-05
21.8453	32.4768	0.1	52.41	1.0632E-05	4.8529E-05
23.6190	32.4695	0.125	40.73	8.8505E-06	5.1985E-05
27.6632	37.4142	0.15	31.38	9.7510E-06	7.4340E-05



Figure 17 Fluxes of membrane 2 before used vs. pressure

Table 7 Fluxes of membrane 2 after used

mb	ma g	Р	t	V m^3	Flux m/s
21.8406	31.4068	0.05	82.73	9.5662E-06	2.7663E-05
23.3391	33.5146	0.075	63.74	1.0176E-05	3.8192E-05
21.8453	32.4768	0.1	52.41	1.0632E-05	4.8529E-05
23.6190	32.4695	0.125	40.73	8.8505E-06	5.1985E-05
27.6632	37.4142	0.15	31.38	9.7510E-06	7.4340E-05



Figure 18 Fluxes of membrane 2 after used vs. pressure

Membrane 4

Table 8 Fluxes of membrane 4 before used

mb	ma g	Р	t	V m^3	Flux m/s
23.9502	34.4269	0.05	69.22	1.0477E-05	3.6209E-05
23.8972	34.1535	0.075	38.13	1.0256E-05	6.4350E-05
23.5749	36.2032	0.1	28.31	1.2628E-05	1.0672E-04
25.5452	35.1839	0.125	23.19	9.6387E-06	9.9435E-05
23.0732	34.3964	0.15	20.41	1.1323E-05	1.3272E-04



Figure 19 Fluxes of membrane 4 before used vs. Pressure

Table 9 Fluxes of membrane 4 after used

mb	ma g	Р	t	V m^3	Flux m/s
23.8428	33.8381	0.05	86.00	9.9953E-06	2.7805E-05
23.6156	34.3173	0.075	45.87	1.0702E-05	5.5815E-05
23.1779	33.7521	0.1	35.85	1.0574E-05	7.0564E-05
23.4844	33.3481	0.125	27.75	9.8637E-06	8.5036E-05
22.6565	33.7836	0.15	27.59	1.1127E-05	9.6484E-05



Figure 20 Fluxes of membrane 4 after used vs. pressure

Membrane 5

mb	ma g	Р	t	V m^3	Flux m/s
22.8313	32.5979	0.05	92.03	9.7666E-06	2.5389E-05
22.3580	32.0849	0.075	62.88	9.7269E-06	3.7007E-05
27.6600	37.3579	0.1	45.5	9.6979E-06	5.0991E-05
21.9247	31.7676	0.125	37.1	9.8429E-06	6.3471E-05
26.7052	36.1260	0.15	29.84	9.4208E-06	7.5529E-05





Figure 21 Fluxes of membrane 5 before used vs Pressure

Table 11 Fluxes of membrane 5 after used

mb	ma g	Р	t	V m^3	Flux m/s
21.9263	31.8588	0.05	125.4	9.9325E-06	1.8949E-05
21.8221	31.6046	0.075	76.21	9.7825E-06	3.0709E-05
23.3504	33.4128	0.1	63.77	1.0062E-05	3.7749E-05
22.8352	33.3867	0.125	52.85	1.0552E-05	4.7763E-05
22.8352	33.4717	0.15	42.74	1.0637E-05	5.9537E-05



Pressure (MPa)

Figure 22Fluxes of membrane 5 after used vs. Pressure

Membrane 6

Table 12 Fluxes of membrane 6 before used

mb	ma	Р	t	V m^3	Flux m/s
23.4783	33.3156	0.05	53.19	9.8373E-06	4.4246E-05
21.8365	32.0532	0.075	36.16	1.0217E-05	6.7594E-05
23.9219	33.3756	0.1	25.28	9.4537E-06	8.9464E-05
25.4948	35.5556	0.125	21.53	1.0061E-05	1.1179E-04
22.2583	31.6066	0.15	17.16	9.3483E-06	1.3033E-04



Figure 23 Fluxes of membrane 6 before used vs. pressure

Table 13	Fluxes o	f membrane	6	after	used
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mb	ma	Р	t	V m^3	Flux m/s
23.5906	33.5452	0.05	58.5	9.9546E-06	4.0709E-05
25.5323	34.6734	0.075	39.69	9.1411E-06	5.5099E-05
25.0883	34.6981	0.1	30.19	9.6098E-06	7.6151E-05
23.8447	33.7415	0.125	25.94	9.8968E-06	9.1274E-05
23.6165	34.0336	0.15	23.72	1.0417E-05	1.0506E-04



Figure 24 Fluxes of membrane 6 after used vs. pressure

Membrane 7

Table 14 Fluxes of membrane 7 before used

mb	ma	Р	t	V m^3	Flux m/s
22.3535	32.8203	0.05	47.65	1.0467E-05	5.2550E-05
27.6577	37.7713	0.075	31.09	1.0114E-05	7.7823E-05
26.7019	37.0439	0.1	24.16	1.0342E-05	1.0241E-04
22.3904	32.1255	0.125	18.31	9.7351E-06	1.2720E-04
25.4539	35.4832	0.15	15.32	1.0029E-05	1.5662E-04



Figure 25 Fluxes of membrane 7 before used vs. Pressure

Table 15 Fluxes of membrane 7 after used

mb	ma	Р	t	V m^3	Flux m/s
26.0728	35.9446	0.05	63.47	9.8718E-06	3.7209E-05
23.1681	32.9346	0.075	42.00	9.7665E-06	5.5631E-05
23.8898	34.1952	0.1	34.53	1.0305E-05	7.1399E-05
21.982	32.5151	0.125	29.29	1.0533E-05	8.6032E-05
23.2806	33.8933	0.15	26.04	1.0613E-05	9.7501E-05



Figure 26 Fluxes of membrane 7 before used vs. Pressure

Membrane 8

mb	ma	Р	t	V m^3	Flux m/s
22.3279	32.7582	0.05	51.62	1.0430E-05	4.8340E-05
24.7763	35.0707	0.075	34.22	1.0294E-05	7.1969E-05
22.8965	33.5755	0.1	26.32	1.0679E-05	9.7066E-05
22.8357	33.2982	0.125	21.15	1.0463E-05	1.1834E-04
23.5692	33.4854	0.15	16.28	9.9162E-06	1.4572E-04

Table 16 Fluxes of membrane 8 before used



Figure 27 Fluxes of membrane 8 before used vs. pressure

Table 17 Fluxes of membrane 8 after used

mb	ma	Р	t	V m^3	Flux m/s
21.5834	31.7638	0.05	84.53	1.0180E-05	2.8812E-05
21.9209	32.3744	0.075	54.03	1.0454E-05	4.6286E-05
23.3462	34.278	0.1	42.19	1.0932E-05	6.1988E-05
22.7265	35.7164	0.125	40.31	1.2990E-05	7.7093E-05
22.0551	32.6063	0.15	27.10	1.0551E-05	9.3144E-05



Figure 28 Fluxes of membrane 8 after used vs. pressure

Membrane 9

Table 18 Fluxes of membrane 9 before used

mb	ma	Р	t	V m^3	Flux m/s
22.3317	32.9917	0.05	49.28	1.0660E-05	5.1750E-05
26.0523	35.7329	0.075	31.32	9.6806E-06	7.3944E-05
23.8928	33.0607	0.1	25.87	9.1679E-06	8.4781E-05
23.174	33.6274	0.125	21.91	1.0453E-05	1.1414E-04
21.5298	32.81	0.15	20.25	1.1280E-05	1.3326E-04



Figure 29 Fluxes of membrane 9 before used vs. pressure

mb	ma	Р	t	V m^3	Flux m/s
24.1444	34.0097	0.05	84.22	9.8653E-06	2.8023E-05
25.988	35.3203	0.075	57.09	9.3323E-06	3.9107E-05
24.7293	35.1134	0.1	41.66	1.0384E-05	5.9631E-05
24.0063	34.6783	0.125	35.68	1.0672E-05	7.1556E-05
23.6187	34.3346	0.15	29.44	1.0716E-05	8.7079E-05



Figure 30 Fluxes of membrane 9 after used vs. Pressure

Membrane 10

Table 20 Fluxes of membrane 10 before used

mb	ma	Р	t	V m^3	Flux m/s
23.59	33.8398	0.05	64.44	1.0250E-05	3.8053E-05
23.5691	34.1744	0.075	41.5	1.0605E-05	6.1136E-05
21.9831	33.1378	0.1	32.88	1.1155E-05	8.1161E-05
21.9264	31.9345	0.125	24.87	1.0008E-05	9.6272E-05
23.2483	34.2264	0.15	22.72	1.0978E-05	1.1560E-04



Figure 31 Fluxes of membrane 10 before used vs. Pressure

Table 21 Fluxes of membrane 10 after used

mb	ma	Р	t	V m^3	Flux m/s
25.0421	35.3524	0.05	81.26	1.0310E-05	3.0354E-05
23.8447	34.6445	0.075	53.38	1.0800E-05	4.8402E-05
25.5318	37.1056	0.1	41.50	1.1574E-05	6.6719E-05
22.8969	33.7797	0.125	32.78	1.0883E-05	7.9425E-05
26.7054	38.129	0.15	28.72	1.1424E-05	9.5157E-05



Figure 32 Fluxes of membrane 10 after used vs. pressure

Membrane 11

Table 22 Fluxes of membrane 11 before used

mb	ma	Р	t	V m^3	Flux m/s
25.0448	35.4711	0.05	58.94	1.0426E-05	4.2320E-05
23.6167	34.0461	0.075	43.22	1.0429E-05	5.7730E-05
22.3594	33.1543	0.1	30.90	1.0795E-05	8.3576E-05
21.8382	32.9707	0.125	25.60	1.1133E-05	1.0403E-04
23.4847	35.2198	0.15	23.09	1.1735E-05	1.2159E-04



Figure 33 Fluxes of membrane 11 before used vs. Pressure

mb	ma	Р	t	V m^3	Flux m/s
23.3542	33.7539	0.05	69.28	1.0400E-05	3.5912E-05
24.7777	35.4087	0.075	43.66	1.0631E-05	5.8252E-05
21.9444	32.6547	0.1	33.34	1.0710E-05	7.6853E-05
24.7279	35.5067	0.125	26.75	1.0779E-05	9.6399E-05
23.5909	34.4173	0.15	22.6	1.0826E-05	1.1460E-04

Table 23 Fluxes of membrane 11 after 1st time used



Figure 34 Fluxes of membrane 11 after 1st time used vs. Pressure

2 nd time used
2 nd time use

mb	ma	Р	t	V m^3	Flux m/s
25.2101	35.7345	0.05	71.44	1.0524E-05	3.5244E-05
21.8258	32.8561	0.075	49.06	1.1030E-05	5.3788E-05
23.5836	34.3967	0.1	37.72	1.0813E-05	6.8581E-05
23.3401	34.354	0.125	31.69	1.1014E-05	8.3146E-05
24.1505	34.3833	0.15	26.71	1.0233E-05	9.1653E-05



Figure 35 Fluxes of membrane 11 after 2nd time used vs. Pressure

Membrane 14

Table 25 Fluxes of membrane 14 before used

mb	ma	Р	t	V m^3	Flux m/s
25.0448	35.4711	0.05	58.94	1.0426E-05	4.2320E-05
23.6167	34.0461	0.075	43.22	1.0429E-05	5.7730E-05
22.3594	33.1543	0.1	30.90	1.0795E-05	8.3576E-05
21.8382	32.9707	0.125	25.60	1.1133E-05	1.0403E-04
23.4847	35.2198	0.15	23.09	1.1735E-05	1.2159E-04



Figure 36 Fluxes of membrane 14 before used vs. pressure

Table 26 Fluxes of membrane 14 after 1st time used

mb	ma	Р	t	V m^3	Flux m/s
18.0007	27.0587	0.04	103.4	9.0580E-06	2.0957E-05
17.4417	27.3915	0.06	62.79	9.9498E-06	3.7909E-05
17.8436	27.3527	0.08	45.16	9.5091E-06	5.0374E-05
17.942	28.3198	0.1	39.91	1.0378E-05	6.2208E-05
17.9983	27.7723	0.12	32.25	9.7740E-06	7.2505E-05



43.69

35.53

7.1789E-05

1.0599E-05 5.8039E-05

1.0662E-05

Figure 37 Fluxes of membrane 14 after 1st time used vs. pressure

mb	ma	Р	t	V m^3	Flux m/s
17.8659	27.4557	0.04	129.93	9.5898E-06	1.7657E-05
18.186	29.0656	0.06	76.6	1.0880E-05	3.3979E-05
17.9047	27.103	0.08	50.44	9.1983E-06	4.3627E-05

0.1

0.12

Table 27 Fluxes of membrane 14 after 2nd time used

28.8905

28.4528

18.2911

17.7911



Figure 38 Fluxes of membrane 14 after 2nd time used vs. pressure

B. Membrane Cleaning

Membrane 12 cleaned by HNO3 (PH=2)



Figure 39 Membrane cleaning results for membrane 12


C. Standard Curves for Strontium Concentration Measurement

Figure 40: 1st and 2nd measurement for standard curve



Figure 41 Final standard curve