Polymer-Assisted Ultrafiltration for Metal Ion Removal from Aqueous Solutions

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ABSTRACT

Water treatment and drinking are two businesses that employ the membrane technique of ultrafiltration. Ultrafiltration removes actinoids in colloidal or pseudo-colloidal form from wastewater for use in nuclear research and energy. The goal of the research was to investigate if polyacrylic acid might be used as an organic polymer to extract dissolved metals from a solution, especially iron and aluminium. Three mixtures including the target metals and boric acid, which is regularly present in the LVR-15 reactor's liquid nuclear waste, were put to the test. The results shown that Al3+ and Fe3+ could be effectively removed from the solution using a carefully designed ultrafiltration membrane, pH, and organic polymer.

Keywords: Liquid radioactive waste, Metal removal, Membrane technology, Organic polymer; Polymer-assisted ultrafiltration

INTRODUCTION

Over the years, several kinds of membrane separation methods have been created. These techniques were first used in the creation of drinking water and thereafter in the cleanup of different processes and effluent. The increased usage of membrane technologies has given rise to a wide variety of commercial items, including membranes and sophisticated technical solutions. Membrane technologies provide an alternative to conventional techniques, such as sorption in the nuclear sector, for treating liquid radioactive waste because of the membranes' considerable durability and their shown functioning in conventional water treatment technologies. In situations when current technologies are insufficiently effective or cost-effective, membranes may provide assistance rather than necessarily replacing them entirely.

To recycle the flow or enhance the value of downstream products, ultrafiltration is utilised in a variety of sectors, from drinking water and wastewater treatment to the chemical, food, and pharmaceutical industries. Wastewater contaminated with alpha radiation is often treated using ultrafiltration in the nuclear industry. Actinoids are often found in colloidal or pseudo-colloidal forms in wastewater and may be successfully removed by ultrafiltration. Ultrafiltration may be used as a pre-treatment step before reverse osmosis in addition to eliminating colloidal particles from solutions.

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Ultrafiltration has been used to remediate radioactive waste, according to a number of published research. Ultrafiltration often takes place following the addition of a chemical agent. For these procedures, surface-active materials are used, such as the so-called Micellar Enhanced Ultrafiltration (MEUF) or, in the case of seeded ultrafiltration/Polymer-Assisted Ultrafiltration (PAUF), polymerization agents. In the first variant, it was explained how to employ Sodium Dodecyl Sulphate (SDS) as a surface-active chemical. The treated solution was then mixed for many hours with SDS, and this mixture was then purified via a membrane. The majority of radioactive metals, with the exception of Cs ions, were virtually entirely eliminated in pH 8 to 11, which also had the maximum retention of radioactive metals.

In the instance of PAUF, radionuclides Co-60 and Cr-51 were detected using reagents such Polyethyleneimine (PEI) and Microcrystalline Chitosan (MCH). The steps are the same as in MEUF: adding a reagent, mixing, reacting, and then filtering through a membrane. For the laboratory equipment and the pilot unit, the decontamination factors were 52 to 68 and 37 to 62, respectively. The removal of Co-60 and Sr-90 using Polyacrylic Acid (PAA) across a regenerated cellulose membrane was disclosed in addition to PEI. Retention values surpassed those of low-pressure RO. The removal of Cu, Ni, and Zn using PEI in conjunction with a cellulose acetate membrane was also studied. Metal retention rose as pH increased and somewhat reduced when salts (NaCl, K2SO4) were present. Cs-137 was not appreciably removed by the aforementioned reagents. Cs-137 was eliminated from the solution using Copper Hexacyanoferrate (CuFC) at an alkaline pH and Nickel Hexacyanoferrate (NiFC) in conjunction with a composite ultrafiltration membrane. the inclusion of SDS together with NiFC increases Cs retention. Real liquid radioactive waste (RAW) was used in the study's testing [7]. Co-60, Zn-65, Ba-133, Cs-134, Cs-137, Eu-152, and Am-241 were all present in the solution. Sodium salt of polyacrylic acid (NaPAA), reagent INSTAR AS (copolymer of macromolecular acrylamide and sodium polyacrylate), and cobalt hexacyanoferrate (CoCF) were utilised for ultrafiltration via a ceramic membrane. RAW processing was done in a manner similar to that shown in the examples above. The research found that combining all three of the compounds indicated had the greatest outcomes. Using a ceramic membrane, NaPAA, and CoCF reagents, ultrafiltration was carried out in tests using a genuine RAW solution. This investigation showed that adding both agents at once reduced the decontamination factors. A suggested two-phase system, CoCF will be introduced in the first stage, then filtered through the membrane. NaPAA will be added to the resulting permeate and filtrated once more through the membrane in the second phase.

The LVR-15 reactor, part of the Research Centre Rez's research infrastructure, generates a variety of liquid waste kinds (different radionuclide compositions, organic material concentrations, etc.). Pure water to salt solutions comprising boric acid, iron, and aluminium make up the waste spectrum. It is challenging to treat these pollutants because radionuclides in water make purification difficult. Due to this, either basic filtration with periodic membrane flushing (for deadend) or ultrafiltration, in which all undesirable chemicals are concentrated in one stream and cleansed water in the other (for crossflow), are the best options. In this paper, the removal of iron and aluminium ions from the simulated liquid radioactive waste was tested using a PAUF.

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METHOD AND MATERIALS

Chemicals

In a non-active aqueous solution that mimicked feed waste, boric acid (1 g/l), ferric nitrate nonahydrate (Fe3+; Fisher Chemical), and aluminium nitrate nonahydrate (Fe3+; Fisher Chemical) were all present. As an organic polymer, Polyacrylic Acid PAA (Sigma Aldrich) with a molecular weight (Mw) of 100 000 and 250 000 was utilised. Potassium hydroxide (Lachner) was used to modify the pH. Analytical grade was used for all compounds. The tested solution was mixed with a predetermined quantity of PAA to produce a PAA to M3+ ratio that ranged from 5 to 50.

Ultrafiltration system

A high-pressure pump and filtration chamber were used in the experiments' ultrafiltration laboratory apparatus for evaluating flat sheet membranes (Figure 1). The tested solution was added to, stirred with a magnetic stirrer (IKA) for 30 minutes at a speed of 100 rpm, and then allowed to react for a further 60 minutes. The filtering group used a pre-dried and weighed polysulfone membrane, GHPS Media 0.02 A Micron 47 MM disc. The unit was working at a flow rate of 20 ml/min, and the temperature was 22 2 oC. Three different combinations of boric acid and trivalent metals were tested: boric acid and Fe3+, boric acid and Al3+, and boric acid and both trivalent metals. Both an acidic and an alkaline pH were used to examine each experimental solution.

To determine the metal content, pH, and conductivity, samples of the feed waste solution and permeate were taken.



Figure 1. Ultrafiltration laboratory unit.

Analysis

Using a ContrAA 700 Highresolution continuum source atomic absorption spectrometer (Analytik Jena, Germany), a UV/Vis Spectrophotometer Jenway 6850 (Cole-Parmer Instrument Company, USA), and a WTW Series Photolab S12 (Xylem Analytics, Germany), the concentration of the metal ions in the feed water and permeate was determined. A WTW pH/Cond 3320 (Xylem Analytics, Germany)

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was used to test the pH and conductivity of all solutions. The following formula was used to determine the removal effectiveness of each metal:

Removal efficiency (%) = $(1 - c0 / c1) \cdot 100...$

Where c0 represents the metal ion concentration in the feed waste solution and c1 represents the metal ion concentration in the permeate.

CONCLUSION AND RESULTS

Removal of ferric ions

With the solution containing boric acid and Fe3+, it was first examined how an additional organic polymer affected the effectiveness of metal removal using an ultrafiltration membrane. The PAA:Fe3+ ratio enhanced the removal effectiveness of Mw 100 000 using PAA at an alkaline pH. At the ratios between 20 and 50, there was a considerable rejection of Fe3+ of more than 90%. An opposite pattern was seen in the case of PAA Mw 250 000 in an alkaline environment.

With a lower PAA: Fe3+ ratio (10 and 20), higher Fe3+ rejection effectiveness of up to 95% was attained. A larger PAA: Fe3+ ratio was associated with a trend in declining rejection efficiency. Low Fe3+ removal was obtained for both molecular weights of PAA in the acidic environment compared to alkaline settings. With a rising PAA: Fe3+ ratio, the rejection efficiency of PAA Mw reduced in both circumstances. In Figure 2, the removal efficiency comparison is shown.



Figure 2. Removal efficiencies with PAUF system using PAA Mw 100 000 and 250 000 at different PAA: Fe3+ ratios

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Removal of aluminium ions

Only at a lower organic polymer-to-metal ratio, 12% and 36% when employing PAA Mw 100 000 and Mw 250 000, respectively, was aluminium removed using this approach at an acidic pH. Al3+ was no longer removed from the solution as the ratio rose. Similar results were seen when PAA Mw 250 000 additions were made in an alkaline environment, where the effectiveness of removing Al3+ declined as the PAA: Al3+ ratio increased from 14% to zero values. The drop in ion removal efficiency was not as noticeable when PAA Mw 100 000 was utilised. However, the greatest values were at 18% and fell to 7% when the PAA to metal ratio rose. Figure 3 illustrates the metal removal efficiency in the PAUF system as a function of the PAA: Al3+ ratio under both acidic and alkaline environments.



Figure 3. With the PAUF system, removal efficiencies were achieved with PAA Mw 100 000 and 250 000 at various PAA: Al3+nratios. Al3+ removal effectiveness in circumstances that are acidic (A) and alkaline (B).

Removal of both ferric and aluminium ions

To evaluate the feasibility of simultaneously removing both metals with the PAUF system employing a PAA of Mw 100 000, a solution containing Fe3+, Al3+, and boric acid was used. Figure 4 compares how well both trivalent metal ions were removed from the mixture. At pH 4, a reduced rejection was seen along with an elevated PAA: M3+ ratio of both metals. At a PAA:M3+ ratio of 2.5, the removal efficiencies for Fe3+ and Al3+ were 94% and 74%, respectively, and they fell with rising ratios to 35% and 0%. This is consistent with the outcomes of the acidic pH individual metal testing. However, it can be said that Al3+ rejection at pH 4 was favourably impacted by Fe3+ presence in the solution. When Fe3+ was included in the solution, the removal efficiency increased from 9% to 74%. The removal effectiveness of Fe3+ was also affected by the presence of Al3+, reaching a maximum of 45% in the absence of Al3+ and 94% in the presence of Al3+ in the solution. The elimination efficiency of Fe3+ rose from zero values to 35% at pH 8, which is much less than when there is no other metal in

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the solution. Al3+ was left in the solution throughout this test.

Figure 4. With a PAUF system, removal efficiencies may be achieved with PAA Mw 100 000 at various PAA:M3+ ratios. Fe3+ and Al3+ removal effectiveness in acidic (A) and alkaline (B) environments.

High rejection of Fe3+ (90%) occurred in the alkaline environment between the ratios of 5 and 10, then declined.

In the presence of another metal, the total removal effectiveness of Fe3+ was somewhat reduced. On the other hand, similar to earlier experiments, the presence of Fe3+ had a favourable impact on the elimination of Al3+ at a rate of 58% as opposed to 14%. Figure 5 depicts the evolution of the efficiency of removing Fe3+ and Al3+ from the solution using ultrafiltration and PAA in combination at various PAA: M3+ ratios and various pH levels.



Figure 5. Removal efficiencies with PAUF system using PAA Mw 250 000 at different PAA: M3+ ratios. The removal efficiency of Fe3+ and Al3+ in acidic (A) and alkaline (B) conditions





CONCLUSION

Investigated was the effect of an additional organic polymer in the PAUF system on the removal of metals utilising a polysulfone flat-sheet ultrafiltration membrane. Three different solutions—one using Fe3+, one using Al3+, and one using both—were studied. All solutions included boric acid, and PAA was used as an organic polymer with two distinct molecular weights. The outcomes with the simulated wastewater show that Al3+ and Fe3+ can be effectively removed from the boric acid solution with a carefully selected ultrafiltration membrane, dosage of organic polymer, and pH.

Compared to Al3+, Fe3+ demonstrated a generally better removal efficiency. For both of the studied molecular weights of PAA, the clearance rate of Fe3+ reached 95% in an alkaline environment. On the other hand, this approach could only partially remove Al3+ from the single metal solution, with a maximum removal efficiency of 37% at pH 4. When comparing the impact of pH on a single metal solution, alkaline conditions often had better removal efficiencies for Fe3+.

However, at pH 4, both Fe3+ and Al3+ were more successfully removed from the solution simultaneously. A larger PAA:M3+ ratio was often shown to have a detrimental impact on removal efficiency.

To get the greatest outcomes, it is crucial to carefully choose the PAA dosage.

In the case of both forms of PAA, it can be said that the presence of Fe3+ in the solution had a favourable impact on Al3+ rejection at pH 4. The findings in the mixed metal solution, where the removal efficiencies were marginally lower using PAA Mw 100 000 than when using Mw 250 000, were comparable when the molecular weights of the utilised PAA were taken into account.

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