Volume Reduction of Radioactive Liquid Wastes coming from RIA Laboratories by Membrane Techniques

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Abstract. In radio immune assay (RIA) laboratories, a great volume of radioactive liquid wastes of low-medium level are generated. These wastes are also potentially infectious since they can contain pathogens from patient blood. Usually, these liquid wastes are temporarily stored in the hospital, for partial radioactivity decay, and then they are managed by a licensed company. Membrane techniques have become more and more common in the treatment of radioactive effluents, having substituted some conventional processes. The objective of this research is to study the viability of treating RIA radioactive liquid wastes by membrane techniques in order to reduce their volume and, as a consequence, decrease management costs as well as increase radiological protection related to them. Two membrane techniques, ultrafiltration and reverse osmosis, have been tested with real wastes to assess if the combination of them is a viable way of treating RIA radioactive liquid wastes.

1. Introduction

The origin of this research work were the problems associated to radioactive liquid wastes generated in radio immune assay laboratories at hospitals. Radio immune assay (RIA) is an in vitro technique within Nuclear Medicine that is used for measuring substances in body fluids (generally blood) whose concentration are below 10 pg/mL, like certain hormones, enzymes, vitamins, etc. This technique is based on the combination of antibodies and antigens, some of them radioactively labelled with a radioisotope, to measure the interesting substance by means of a gamma counting apparatus. The most common radioisotope used in RIA is $^{125}$I, a gamma emission radioisotope with a semidecay period of 60 days.

The application of RIA techniques generates a significant volume of radioactive liquid wastes of low-medium activity level. The composition of these wastes is very variable depending on the specific determinations carried out, but it can be stated that they contain different kinds of proteins (some of them radioactively labelled), preservatives and several organic or inorganic solutes of low molecular weight. Besides radioactive, RIA wastes are potentially infectious since they can contain pathogens from patient blood (mainly hepatitis B and AIDS), and infectious risks can be even higher than radioactive ones.

At present, RIA wastes, like other radioactive wastes generated in medical applications [1,2], are temporarily stored in the hospital itself for partial radioactivity decay, and after this, they are managed by a licensed company, which is ENRESA in the case of Spain. The aim of this work is to assess the viability of treating RIA wastes by membrane techniques in order to reduce the volume to be managed and so management costs. Furthermore, volume reduction would mean an improvement from the point of view of radiological protection. This paper presents the results obtained in the experiments carried out with RIA wastes by means of the combination of two membrane processes: ultrafiltration and reverse osmosis.
2. Membrane technology

2.1. Description and classification

A semipermeable membrane is a selective barrier that separates two multicomponent systems (liquid or gaseous) and limits the transport of some components through itself. Separation takes place because some components go through the membrane in a proportion higher than others. As it can be seen in FIG. 1, in a membrane process the feed stream is divided into a permeate, which contains the components that can go through the membrane, and a concentrate or retentate, which contains the components that cannot go through the membrane. In comparison with conventional filtration, membrane processes use tangential flow to avoid membrane fouling.

![FIG. 1. Scheme of a membrane process](image)

The characteristic parameters of a membrane are the permeability, related to the permeate flow, and the selectivity, which represents the ability of the membrane to reject certain compounds.

The most important membrane processes in effluent or water treatment are the ones driven by pressure: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). The main differences between these four processes regard with membrane structure (porous or dense), membrane material, and working pressure. These different characteristics define the application range of each process, which can be seen in FIG. 2.

![FIG. 2. Application range of pressure driven membrane processes](image)
Pressure driven membrane processes are being applied more and more often in several industrial areas thanks to the following advantages:

- Continuous performance
- Low energy consumption
- Modular structure
- Ease of installation and automation

One of the areas in which membrane processes have increased their applications is the nuclear industry and the treatment of radioactive liquid wastes.

2.2. Application of membranes to radioactive waste treatment

The most common technologies applied to the treatment of radioactive liquid wastes are evaporation, chemical precipitation and ionic exchange. However, evaporation has a high energy consumption and it cannot be used when the radioisotopes are volatile; ionic exchange is only effective when radioisotopes are in ionic form and with low salt concentration; and chemical precipitation only gets a good volume reduction by means of sludge dehydration [3]. In comparison with these processes, membrane technology has the advantages of lower energy consumption and no need of chemical reagents. Furthermore, membrane techniques can remove not only the radioisotopes but also organic and biological compounds, simultaneously.

Thanks to these advantages, pressure driven membrane processes are being applied more and more often to the treatment of low-medium level radioactive liquid wastes, mainly to laundry wastes at nuclear power stations [4,5]. Many applications consist of a combination of several membrane processes, usually microfiltration or ultrafiltration as pre-treatment of reverse osmosis, in order to optimise membrane performance [6]. According to the results obtained by Cmielewski and Harasimowicz, the effect of radiation in membrane performance is not a critical factor [7], and with the application of RO, around 99% of the radioactivity can be removed [8]. Other areas of membrane application, apart from nuclear industry, are medical and researching centres, as well as wastes generated in radioactive incidents. The Chemical and Nuclear Engineering Department of the Polytechnic University of Valencia (Spain) has experience in treating by reverse osmosis contaminated wastes generated in a radioactive incident, having decontaminated around the 90% of the original volume [9].

2.3 Proposed solution

The proposed solution to treat RIA wastes is presented in FIG. 3. In previous works with RIA wastes [10], it was proved that part of the radioisotope remains labelled to proteins, but there is a part of the radioisotope in ionic form, which is in agreement with Tóth results [11]. Therefore, reverse osmosis must be used in order to remove all the radioactivity, because it is the pressure driven process that can reject salts and ions. Before entering the RO stage, feed must be pretreated so as to avoid membrane fouling and degradation, as well as improve membrane performance. Pretreatment of RIA waste will consists of the following two stages:

1. Filtration with 50 micron cartridges to remove suspended solids.
2. Ultrafiltration to remove high molecular weight compounds (mainly proteins) and the radioactivity associated to them. This will lead to an improvement of RO performance. If UF were not applied, proteins and other high molecular weight compounds would block RO membrane causing a permeability and selectivity decrease.

The aim of this integral treatment is to obtain a decontaminated permeate free of organic compounds and pathogens with an activity below the legal limit.
3. Experimental

3.1. Pilot plant description

FIG. 4 shows the flow diagram of the pilot plant used in the experiments with RIA wastes. Feed solution is inside a cylindrical tank (1) of polyethylene with 80 litter of capacity, and firstly goes through a 50 micron filter (4) to remove suspended solids. After this, feed is driven by a piston pump with variable speed (5) that can reach 8,5 MPa of pressure. Before entering the membrane module (9), temperature (7) and pressure (8) of the fluid are measured. The pressure is also measured at the outlet of the module (8'), where concentrate flow is measured by a rotameter (11) with a maximum flow of 1000 L/h. Regulation valve (10) at the concentrate line is used for settling the operation pressure. Concentrate stream is returned to the feed tank to reduce the feed volume as much as possible.
3.2. Experimental procedure

The pre-treatment of RIA wastes before reverse osmosis was carried out with a polysulfone spiral wound ultrafiltration membrane, which is the most resistant polymeric material to radiation [12], with a molecular size of 100,000 Da, and a membrane area of 6.5 m². The permeate obtained in the ultrafiltration stage was later treated with a polyamide spiral wound reverse osmosis membrane with an area of 2.6 m² approximately. This membrane was previously irradiated with gamma and electronic radiation in a dose range higher than the one expected to be absorbed by the membrane in the treatment of RIA wastes. By means of experiments in a pilot plant, it was checked that the performance of the irradiated membranes was very similar to that of non-irradiated ones, therefore it can be stated that this membrane is suitable for the treatment of RIA wastes [13].

Experiments were carried out in the “Hospital Universitario La Fe” of Valencia (Spain) with wastes coming from Nuclear Medicine service, with storage periods between a month and a year. Their characteristic parameters varied within the ranges presented in Table I.

<table>
<thead>
<tr>
<th>Characteristic parameters of the RIA wastes treated in the experiments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conductivity (mS/cm)</td>
</tr>
<tr>
<td>pH</td>
</tr>
<tr>
<td>Activity (kBq/L)</td>
</tr>
</tbody>
</table>

Before carrying out the experiments, the working area and the pilot plant were conditioned from the point of view of radiological protection, with the aim of avoiding contamination due to possible radioactive liquid spill. Several experiments were performed with different feed radioactivity levels, trying to meet all the possible ranges. Each experiment consisted of an ultrafiltration stage followed by a reverse osmosis treatment of the UF permeate. In all membrane applications the waste volume in the feed tank was reduced to one litter, approximately.

3.3. Analytical

Periodical measurements of flow and conductivity of the different streams were taken to calculate the parameters that define membrane performance. Furthermore, samples of feed and permeate were periodically taken to measure their radioactivity by means of a NaI scintillation solid detector and a multichannel analyser, both from SILENA company.

To assess the viability of the proposed treatment, the selectivity of the membrane was measured, from the point of view of radioactivity, by means of the retention index to $^{125}$I ($R_{125I}$), which is defined by the following expression:

$$R_{125I} (%) = \frac{A_F - A_P}{A_F} \times 100$$

where

- $A_F$: feed activity (Bq/L);
- $A_P$: permeate activity (Bq/L).
4. Results and discussion

4.1. Ultrafiltration performance

In all the experiments, the ultrafiltration stage lasted half an hour, approximately, with an average permeate flow of 100 L/h. In each experiment, 60 L of waste volume were reduced to 5 L of concentrate, including the dead volume of the pilot plant (around 4 L), which represents a volume reduction factor higher than 10.

Table II shows the average activity of the feed, concentrate and permeate in two of the experiments carried out. UF membrane performance was very similar in all the experiments, in spite of having treated wastes with a very different radioactivity level.

<table>
<thead>
<tr>
<th>Stream</th>
<th>Experiment A</th>
<th>Experiment B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed</td>
<td>240 kBq/L</td>
<td>365 kBq/L</td>
</tr>
<tr>
<td>Concentrate</td>
<td>880 kBq/L</td>
<td>1000 kBq/L</td>
</tr>
<tr>
<td>Permeate</td>
<td>120 kBq/L</td>
<td>180 kBq/L</td>
</tr>
</tbody>
</table>

FIG. 5 shows the results of retention index to $^{125}$I obtained in the same two experiments. According to the results, it can be said that ultrafiltration plays an important role in the partial reduction of feed activity. Rejection of the radioisotope was higher than 50% in most of the cases, even reaching maximum values around 80%. Since UF mainly removes proteins and high molecular weight compounds, the obtained results confirm that part of the radioisotope remains labelled to that kind of species. The rest of the radioactivity is associated to ions or low molecular weight compounds that can go through UF membrane but will be removed by the reverse osmosis process.

4.2. Reverse osmosis performance

The permeate of each ultrafiltration stage was then treated by reverse osmosis. The application of RO lasted more (2-5 hours) than the UF stage, as a consequence of the high concentration reached in the tank. The effect of this high concentration on membrane permeability was higher in the case of reverse osmosis, causing a significant permeate flow decrease during the experiments. Since radioisotope activity is proportional to radioisotope concentration, the feed activity increased as volume was being
reduced and so did the permeate activity, as it can be seen in FIG. 6. Anyway, the original feed volume was reduced to 1 L of concentrate, apart from the dead volume, in all of the cases.

FIG. 6. Feed and permeate activity in one of the reverse osmosis experiments

Table III shows the average activity of the feed, concentrate and permeate in the same two experiments as before.

Table III. Activity values in the treatment of UF permeate by reverse osmosis

<table>
<thead>
<tr>
<th>Stream</th>
<th>Experiment A</th>
<th>Experiment B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed</td>
<td>120 kBq/L</td>
<td>180 kBq/L</td>
</tr>
<tr>
<td>Concentrate</td>
<td>600 kBq/L</td>
<td>850 kBq/L</td>
</tr>
<tr>
<td>Permeate</td>
<td>16 kBq/L</td>
<td>35 kBq/L</td>
</tr>
</tbody>
</table>

FIG. 7 shows the results of retention index to $^{125}\text{I}$ obtained in the reverse osmosis treatment of the two experiments. It can be seen that RO performance was very successful, since rejection to $^{125}\text{I}$ was higher than 98% during most of the experiments, and even showing values around 90% in the last moments of the application when the volume in the feed tank was very low and its concentration was very high.

FIG. 7. Retention index to $^{125}\text{I}$ of the reverse osmosis membrane
After the reverse osmosis treatment, the membrane recovered completely its permeability and selectivity, so it can be applied again to the obtained another permeate so as to reduce its radioactivity to a value close to the legal discharge limit.

5. Conclusions

According to the results of the experiments carried out, it can be stated that the combination of ultrafiltration, as pretreatment, and reverse osmosis is a suitable way of treating radioactive liquid wastes coming from radio immune assay.

The pretreatment by ultrafiltration results in a significant removal of radioactivity (50-60%), and improves reverse osmosis performance since it removes proteins and other high molecular compounds that can blocked RO membrane.

The treatment of the ultrafiltration permeate by reverse osmosis results in a removal of radioactivity around 95%, so the application of RO to the successive permeates can lead to a decontaminated liquid with an activity level below the discharge limit established by the legislation.

Besides eliminating radioactivity, high molecular weight compounds and dissolved salts, the combination of ultrafiltration and reverse osmosis removes also pathogens, which can lead to risks even more dangerous than the radioactive ones.

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References


