Research article

Management of hospital radioactive liquid waste: treatment proposal for radioimmunoassay wastes

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Abstract: Radioactive liquid wastes are produced at hospitals from diagnostic and therapeutic applications of radionuclides. The most usual management of these wastes is temporary storage at the hospital for radioactivity decay and, then, discharge into sewage if not other pollutants are present in waste, always after authorization of the corresponding institution. In some cases, radioactive wastes have other hazards, such as chemical or biological ones, which can be more dangerous than radiological hazard, and do not allow direct discharge into sewage in spite of decaying activity below the clearance level. Therefore, these wastes have to be treated and condition before discharge in spite of activity decay below discharge limit. This is the case of liquid wastes from radioimmunoassay (RIA), a laboratory technique that allows to determine human substances in very low concentrations (below 10^{-12} g/mL), like hormones, using ^{125}I as radionuclide. This study summarizes the usual management of radioactive liquid wastes from hospitals, including conventional and recent treatments applied. Furthermore, based on experimental results obtained with real RIA wastes, this work exposes a proposal of treatment with ultrafiltration and reverse osmosis membranes, and determines the most suitable application of this treatment according to radiological and operational considerations.

Keywords: radioactive waste; hospital; RIA; membranes; ultrafiltration; reverse osmosis

1. Introduction

1.1. Radioactive liquid wastes from hospital

Radionuclides have a broad application in modern medicine thanks to their favourable properties, easy availability and relatively low cost. Applications of radionuclides in medicine can be divided into diagnostic and therapeutic ones:

a) Diagnostic applications of radionuclides. Radionuclides are used for diagnostic purposes both "in vitro" and "in vivo" procedures. In vitro studies use aqueous-based radionuclides to measure levels of drugs or hormones in biomedical samples, and they are performed outside the human body. The most common radionuclides used in vitro are ^{125}I, ^{57}Co, ^{58}Co and ^{14}C [1]. In the case of in vivo applications, radionuclides are administered as tracers to monitor body functions, being detected outside the body by gamma camera imaging. Among the best-known techniques of this type, the positron emission tomography (PET) figures as one of the ones which are experiencing the fastest diffusion. ^{99m}Tc is the most usual radionuclide for in vivo diagnostic applications. Other common diagnostic imaging radionuclides are: ^{67}Ga, ^{99}Mo, ^{201}Tl and ^{131}I [2].

b) Therapeutic applications of radionuclides. Radionuclides can be oral or intravenous administered in liquid or capsule form for systemic therapy. Some examples are the application of ^{131}I for thyroid cancer, or ^{89}Sr for bone metastases. Another type of radionuclide application is by introduction of colloidal suspensions into closed body cavities. An example of this kind of therapy is the application of ^{32}P for malignancies of the pleural and peritoneal cavities [3]. In comparison to diagnostic applications, therapeutic ones use radionuclides with greater activities and longer half-lives.

As a result of medical applications of radionuclides, both solid and liquid wastes are produced. Liquid radioactive wastes include contaminated water and effluents, solvents, blood or body fluids, chemical processing and decontamination solutions, urine, scintillation fluids which are immiscible with water, etc. [4].

The application of radionuclides for medical purposes is growing worldwide on account of their efficient results, therefore a subsequently growing in the amount of radioactive wastes is expected, with the consequent growing demand for adequate treatment and disposal to achieve the sustainability of the health-care system.

1.2. Management of radioactive liquid wastes from hospitals

Radioactive liquid wastes generated in hospitals require a comprehensive management system. The main principles of an effective program of waste management are: waste prevention and minimization, protection of personnel and environment, and consistency with the requirements established by the regulatory authority. This management system has to integrate all the potential additional hazards of the wastes besides the radiological ones [1].

The main strategies for radioactive liquid wastes management are described below.

A) Waste prevention and minimization. Waste minimization is fundamental in any radioactive waste management strategy. The aim of waste minimization is to reduce the activity and the volume of wastes for their storage, treatment and disposal. As a result, environmental impact and costs of management will be reduced. First actions to minimize the production of radioactive waste are source reduction strategies, including limitation of radioactive quantity, and performance of laboratory procedures that reduce the volume of wastes. When possible, long-lived radionuclides must be

replaced by shorter half-life ones or even non-radioactive substitutes [4]. Next action for waste minimization is segregation, which also facilitates the subsequent processing of wastes. Main criteria for waste segregation are [1]: radionuclide content, level of activity, half-life of radionuclides, infectious hazards, chemical hazards and organic/aqueous liquids. Scintillation liquids deserve special mention, since they cannot be diluted with water, so they cannot be discharged into the sewage system [5]. Segregation is fundamental for the economic sustainability of health-care centers. In fact, it has been estimated that implementation of this strategy can result in disposal savings around 40–70% [6].

B)   Dilution and dispersion. Dilution consists of the addition of nonradioactive waste to decrease activity levels below the clearance ones, and then release resulted waste to the environment. It is a possible practice for non-biologically and non-chemically hazardous wastes with very low level of radioactivity. Anyway, such practice must be previously authorized taking into account the impact on the environment [1]. However, this proceeding is either not practiced or specifically prohibited in most countries.

C)   Storage for decay. Decay storage to clearance levels is usually the preferred waste management option. For wastes contaminated with radionuclides with a half-life lower than 100 days, a storage time of 10 half-life times will reduce the initial radioactivity to less than one thousandth, which in many cases means below the clearance levels for release (depending on regulatory requirements). For wastes with radionuclides with a half-life greater than 100 days, storage reduces the level of activity before transport to a centralized waste processing facility where they are conditioned for disposal [1].

D)   Discharge after decay. After storage for decay, wastes can be discharged into sewers according to local requirements. Most of the low and medium level radioactive hospital waste has a short half-life that permits this type of waste disposal [7]. However, prior to discharge, decayed waste has to meet all release criteria: radiological, chemical and biological. Sometimes, biological and/or chemical hazards make the waste unsuitable for discharge in spite of having a level of activity below the clearance levels. In these cases, waste should be sent for appropriate waste treatment, according to biological and chemical hazards.

E)   Concentrate and contain. When radioactive wastes have very high activity levels and/or long half-lives (longer than a month), the alternative of delay and decay is impractical due to extended storage period, especially if space availability is limited [7]. In this case, radioactive waste must be concentrated by a conditioning process to reduce the volume and confine the radionuclides to prevent their dispersion in the environment [8]. Concentrated wastes are then collected in suitable containers to be buried in authorized sites.

1.3. Treatments for radioactive liquid wastes from hospitals

The purpose of treating radioactive liquid wastes is to reduce the volume to manage and dispose as radioactive at the same time that a decontaminated liquid with an activity below clearance level is obtained.

The treatments usually applied to radioactive liquid wastes have been the same for conditioning any wastewater or effluent from industry. Thus, before 1990, evaporation, chemical precipitation, and ion exchange resin were extensively applied to decontaminate radioactive wastes [9]. However, these processes were not efficient in terms of energy consumption, treatment times and costs in all cases [10–12].

- Evaporation. It is the most efficient process in terms of decontamination factor with a high-volume reduction; but it is a high-energy consuming process, and it cannot be applied when radionuclides can evaporate.

- Ion exchange. It is only effective when radionuclides are in ionic form and the concentration of salts and the suspended matter are low. Main disadvantages of this process are related to periodical regeneration of resins and the production of a secondary waste.

- Chemical precipitation. It is a very efficient decontamination process when specific precipitants are used for each radionuclide. However, suitable volume reduction needs other additional treatments.

1.3.1. Pressure driven processes for radioactive effluent decontamination

After 1990, some emergent processes were used with more efficient performance in radioactive decontamination. The more relevant within these processes were membrane technologies; especially pressure-driven ones: microfiltration, ultrafiltration and reverse osmosis.

In comparison with conventional processes, membrane technologies show as main advantages: lower energy consumption, lack of chemicals' addition, operation at low temperatures and ease of scaling-up. Furthermore, suitable combination of different processes can remove radiological, organic and biological substances in the same treatment. These processes have been applied with excellent results in the treatment of different radioactive effluents [13].

On the other hand, one of the main problems of membrane processes is fouling which can be caused by inorganic, organic or even biological substances. Membrane fouling decreases the flux that permeates through membrane and worsens permeate quality, thus reducing membrane life and so increasing operating costs. Membrane fouling can be minimized by applying suitable pretreatment operating conditions.

At present, researchers are working in developing some innovations to improve water and waste management process with promising results in the near future. Within these innovations, the following ones can be highlighted [14]:

- Composite membranes. They contain inorganic nanomaterial, such as TiO_2, ZnO and graphene oxide, among others, incorporated to membrane as nanoparticles. The presence of this inorganic material improves the physicochemical properties of organic membranes. This new type of membranes have better mechanical, thermal, physical and chemical stability, as well as higher biofouling resistance.

- Graphene oxides membranes. These membranes have nanoscale capillaries, which produce a higher water flux and better selectivity of membranes. Another advantage is s that they generate less secondary waste than conventional membrane processes.

- Mixed matrix membranes. They are made by incorporating inorganic fillers into the organic matrix of the membrane. The main advantages are: low manufacturing cost, long-term stability, high mechanical strength and the possibility of regeneration of ceramic materials.

Although these new membranes are in development phase and there are not many applications tested with radioactive liquid wastes, all the advantages commented and successful performance at lab scale tests [15] indicate promising prospects for application in radioactive waste treatment in near future.

1.4. Environmental impact of the hospital liquid wastes discharge

The most usual option for disposal of radioactive liquid wastes from hospitals is discharge into sewage after decay. This practice has to be strictly controlled through an authorized process [8], since the discharge of radionuclides into the environment can be the cause of indirect contamination through the food chain [16]. For example, in the case of ^{131}I, which is the most used radionuclide in diagnostic and therapeutic medical applications; its daily discharge into sewage near medical centers has been estimated in the range of 130–370 MBq [3]. Because of its half-life (8.04 days), ^{131}I not removed in the wastewater treatment plant can reach marine environment and then accumulate in marine algae that are later used in food, cosmetics and medical products [16].

Conventional wastewater treatments have different influence on radionuclide concentration. The treatments that mostly reduce radioactivity are biological and sludge ones, mainly because residence times of these stages favors radionuclide decay [17,18]. In fact, it has been shown that sludge acts as a concentrator of some radionuclides of the influent of the wastewater treatment plants. Thus concentrated sludges can be used to estimate the activity concentration in wastewater, which sometimes is difficult to measure due to low concentration values [19].

Unfortunately, several studies have proved that wastewater from treatment plants can contain radionuclides from medical applications that are not completely removed in the different stages of treatment [20]. Therefore, radioactive discharge of radionuclides from hospitals should be reduced to minimum values to prevent contamination of aquatic systems from ionizing radiation [16].

1.5. Application case: radioimmunoassay liquid wastes

1.5.1. Radioimmunoassay

In 1960, Rosalyn S. Yalow and Solomon A. Berson published an article describing a new analytical technique to determine insulin in plasma, which they denominated radioimmunoassay (RIA). This technique completely revolutionized the world of analysis since it allowed the detection of small concentrations (10^{-12} to 10^{-15} g/mL) of different substances present in biological fluids in several samples at the same time [21]. Some of these substances are: hormones, enzymes, steroids, peptides, vitamins, medications, and drugs [22]. The radioimmunoassay is performed in a laboratory with a blood sample of the patient. The most usual radionuclide is ^{125}I, a beta emitter (35 keV) with a half-life of 59.4 days [22].

1.5.2. Radioimmunoassay liquid wastes

Liquid wastes produced in radioimmunoassay contain the reagents of the measurement kit and the body sample used in the analysis. The composition of these wastes is very variable since it depends on the precise type of analysis made at each laboratory. But, in general, some common components can be identified.

From the chemical point of view, liquid wastes from radioimmunoassay are basically composed of different kinds of proteins (some of them radioactively labeled), some organic solutes of low molecular weight, and different mineral salts, all in aqueous solution [23]. From the radioactive point of view, liquid wastes from radioimmunoassay are within the category of low-medium activity. Finally, it is important to consider that RIA liquid wastes can contain pathogens from patients' blood, which

can carry various diseases (the most relevant hepatitis B and AIDS) [24]. So, in addition to radioactive, RIA wastes should always be considered potentially infectious [25].

The most usual treatment for hospital liquid wastes with ^{125}I is temporary storage for activity decay in the facility producing the waste. After this, wastes are managed according to their chemical and biological characteristics if activity has decayed, in a reasonable time, below the limit value. Otherwise, waste is disposed by an authorized company [26,27]. The possibility of waste activity decay below the set value is determined by the storage capacity of the facility. When the storage capacity of the facility is limited and insufficient for the amount of radioactive waste generated, or when chemical and biological characteristics of the wastes do not allow direct discharge once activity has completely decayed, the application of a treatment should be considered. The objectives of this treatment have to be, on one hand, declassification of a part of the waste for possible discharge, and on the other hand, waste volume reduction for final storage and disposal.

With regard to the treatment of liquid radioactive waste similar to those produced in RIA, some relevant studies have been found in the literature:

At the "Malaysian Institute of Nuclear Technology Research" the removal of ^{125}I of radioactive aqueous waste from health and research facilities was studied by means of coagulation-flocculation treatments and adsorption columns with different clay and mineral materials. The results showed a maximum decontamination factor of 4.8 for the coagulation-flocculation experiments [28], and values between 10 and 30 for the adsorption column treatment with a ferruginous mineral material [29].

B.E. Edwards et al. studied the volume reduction of liquid ^{125}I wastes from pharmaceutical research by three different methods: evaporation, adsorption on activated carbon and adsorption on ion exchange resins [30]. The evaporative treatment was discarded for industrial-scale application because it was too slow at room temperature. The combination of adsorption on activated carbon followed by further treatment with ion exchange resins resulted in an average reduction in specific activity of 79%.

In 2000, H. Inoue and M. Kagoshima [31], investigated the removal of ^{125}I from liquid medical or research waste with the aim of reducing its volume using an anion exchange membrane. After a study of the transport properties of the membrane with model NaI solutions, authors concluded that the experimented method was suitable for the removal of radioactive iodine.

More recently, forward osmosis (FO) has been integrated with the decay tanks of ^{131}I liquid wastes from therapeutic treatments, reducing the size of the septic tanks and allowing an increase of the number of potential patients [32].

1.6. Radioimmunoassay liquid wastes object of this study

At present, RIA wastes object of this work, which contain ^{125}I radionuclide, are temporarily stored in the radioactive facility of the hospital in recipients of 25 liters. Average production of these wastes are 150 liters per year, approximately. During storage at the hospital, radioactivity decay takes place, but chemical and infectious hazards of RIA wastes do not allow their direct discharge into the sewage in spite of achieving an activity level below the limit. Therefore, after partial decay during 1–2 years (depending on total amount of wastes), RIA wastes are disposed by the authorized company in Spain (ENRESA). All this management can cost the hospital several thousand euros, from about 2.8 $/kg [33] to 300 $/kg, depending on waste physic-chemical characteristics. This cost could be reduced if the waste would be partially concentrated and declassified prior to its disposal.

1.7. *Proposed solution for the management of the RIA wastes object of this study*

Membrane technology has been proposed for the treatment of RIA liquid wastes, based on the successful performance of some processes in the removal of radionuclides. The proposed treatment comprises two pressure driven membrane processes: ultrafiltration (UF) and reverse osmosis (RO).

- Ultrafiltration. This is a pressure driven membrane process that can remove macromolecules of different sizes, as well as bacteria and viruses from water. Some studies [34,35] have proved the viability of ultrafiltration membranes in the treatment of radioactive liquid wastes, mainly as a pretreatment stage of reverse osmosis, in a range of activity that includes the one of the RIA wastes. Because of the organic content of the RIA wastes object of this study, UF is proposed to remove high molecular organic compounds (to prevent reverse osmosis fouling), as well as possible pathogens [36].

- Reverse osmosis. It is also a pressure driven membrane process that can remove any type of salt from water. This technology is a typical treatment for low-medium activity wastes coming from fuel cycle [37] with activity values similar to those of radioimmunoassay liquid wastes. Results from RO application for radioactive waste have proved that this process achieves decontamination factors that allow discharge of treated flow (permeate) into the environment [38–40].

This proposed solution aims to reduce the volume of waste to be managed by the authorized company, and to obtain a treated flow, free of organic matter and pathogens, whose activity is less than the discharge limit, and which can therefore be discharged directly into the sewer, after the corresponding measurements and permissions.

2. Materials and methods

Figure 1 shows a flow diagram of the pilot plant used in the experiments with RIA liquid wastes. The system works on a total concentration mode, recirculating the concentrate flow to the feed tank in all experiments..
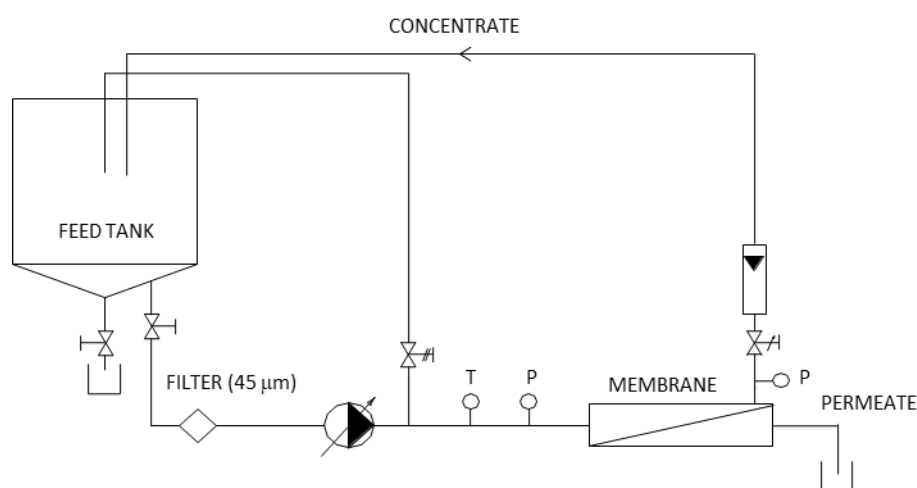

Figure 1. Flow diagram of the pilot plant used in the experiments.

The ultrafiltration membrane used in the pretreatment of RIA liquid wastes was 4040-TFV-P100 from Hydranautics, a spiral-wound membrane made of polysulphone, which is the UF polymeric

material most stable to radiation [41]. The selected membrane has a cut-off of 100 kDa and an effective area of 6.5 m^2.

After each UF test, ultrafiltrated flow (permeate) was later treated by the reverse osmosis membrane CPA2-2540 from Hydranautics. Influence of radiation (both beta and gamma) in this membrane performance had been previously studied [42], showing that permselective properties do not vary even with radiation doses much higher than the ones expected in the treatment of RIA wastes.

Due to radioactive characteristics of the wastes, it was not possible to perform a complete analysis of the composition of wastes. The main parameters measured for waste characterization were: conductivity, activity and pH.

Two complete experimental treatments were performed. In the first experiment (Exp1), containers with high storage time were selected (between 1 and 2 years). As these wastes had stayed longer in the store, they were the wastes with the lowest possible activity. In the second experiment (Exp2), containers with low storage time were selected (equal or lower than a month). In this case, the wastes were the ones with the highest possible activity, as they were recent production wastes. Table 1 shows the characteristics of feed liquid for each experiment.

Table 1. Characteristic parameters of the waste treated in each experiment.

	Average activity (kBq/L)	Conductivity (mS/cm)	pH	Storage time
Exp1	2.8	12.6	6.66	1–2 years
Exp2	240	14.3	6.73	1 month

Each experiment lasted until permeate activity was reduced as much as possible, at the same time that maximum concentration of the initial volume was achieved, reducing it to around a liter. Thus, several RO passes were applied to the successive permeates, always after a first ultrafiltration phase as pretreatment.

2.1. Analytical methods

In all the experiments, periodical measurements of permeate flow and feed and permeate concentrations were made. Volumes of initial feed, final permeate and final concentrate for each stage were estimated. Furthermore, samples of permeate and feed were taken for later measurement of activity. Treatment efficiency was determined by the following parameters:

Permeate flux (J_P). It is defined as the ratio between the permeate flow (Q_P) and membrane active area (A_{memb}), and it is usually expressed in L/(m^2·h):

$$J_P = \frac{Q_P}{A_{memb}} \tag{1}$$

This parameter depends on the permeability of the membrane to water, to the osmotic pressure of the solution and to the pressure applied. The higher the permeability and the applied pressure, and the lower the osmotic pressure a higher permeate flux is produced. Furthermore, it is very influenced by membrane fouling which causes a progressive decrease of permeate flow due to the increase of membrane resistance.

Retention index (R). It represents the percentage of solute removed by the membranes, calculated by the following expression:

$$R(\%) = \frac{C_a - C_p}{C_a} \cdot 100 \tag{2}$$

Where C_a (g/L) is the concentration of solute in the feed and C_p (g/L) is the concentration of solute in the permeate.

In the case of removal of radioactivity, since activity is proportional to radionuclide concentration, an activity retention index (Ra) can be calculated by next equation:

$$Ra(\%) = \frac{A_a - A_p}{A_a} \cdot 100 \tag{3}$$

Where A_a (Bq/L) is the activity of feed and A_p (Bq/L) is the activity of the permeate.

Volume Reduction Factor (*VRF*). It is defined as the ratio between feed volume (V_f) and concentrate waste volume after treatment (V_c). The higher this factor, the lower the costs of transportation and storage of the final concentrate waste.

$$VRF = \frac{V_f}{V_c} \tag{4}$$

Activity measurements were carried out with a scintillation solid detector and a multichannel analyser from SILENA. The detector was isolated by lead shielding to eliminate background radiation. A solution of anti-calcitonin-^{125}I with a known activity was used as standard. Activity decay of samples was considered by the following expression:

$$A(t) = A_0 \exp(-\lambda t) \tag{5}$$

where A_0 represents the activity in the reference time, and $A(t)$ the activity in a later t time, being λ the disintegration constant of the radioisotope.

From the results of both experiments, this work aims to determine the most suitable time to apply the treatment of the proposed solution, considering radiological efficiency as well as membrane performance.

3. Results and discussion

3.1. Evolution of activity during storage time

Based on the measurements carried out in the different stored RIA wastes, and taking into account the storage time of the waste (determined from the date indicated in each recipient), the activity of the original waste was estimated using equation 5. Table 2 shows the estimated activity values.

As it can be seen in Table 2, there is a large disparity in the activity of liquid waste from RIA, which is in line with similar results found in the literature [25].

Based on the estimated values of original waste activity, and using equation 5 again, the storage time that would be required for the activity level of the waste to decay below the discharge limit, estimated at a value of 0.0175 kBq/L [38] has been estimated. Table 3 shows the calculated values for the storage times for each waste.

Table 2. Estimated values of RIA waste activity.

Waste	Storage time (months)	Average activity (kBq/L)	Estimated original activity (kBq/L)
W1	25	5.87	114000
W2	21	8.78	28250
W3	20	4.72	24870
W4	16	2.78	9908
W5	15	0.59	10290
W6	15	0.86	7824
W7	13	0.31	6846
W8	1	615.85	1021

Table 3. Estimated storage time to achieve the discharge limit.

Waste	Estimated original activity (kBq/L)	Storage time to discharge (years)
W1	114000	2.55
W2	28250	2.32
W3	24870	2.30
W4	9908	2.15
W5	10290	2.16
W6	7824	2.12
W7	6846	2.09
W8	1021	1.78

As noted in the Table 3, the storage time required to reach the discharge limit would be between 21–31 months, depending on the original activity of the waste. This time equals between 10 and 16 half-lives of the radionuclide, which is consistent with the normally recommended storage time for wastes with half-life less than 90 days [4].

In any case, the feasibility to have the waste stored during that time will be conditioned by the availability of authorized space in the hospital and the amount of RIA waste produced. Moreover, such storage would allow its declassification as radioactive waste, but not its discharge into the sewer because of the potentially infectious nature of these wastes. In addition, it should be noted that, during such storage, certain characteristics of the waste can vary, since over time a breakdown or alteration of organic matter and microbiological growth may occur.

To analyze the possible influence of storage time on the variation of organic matter, the following section examines certain parameters evolution with the storage time.

3.2. Evolution of organic matter during storage time

Since the organic load of the waste has not been directly measured due to its radioactive nature, its evolution will be analyzed indirectly, through the permeoselective parameters of the UF membrane: permeate flux (J_P) and activity rejection index (Ra), the average values of which are shown in Table 4.

Table 4. Average permeoselective characteristics of UF membrane in experimental tests.

Experiment	Storage time of the waste	$J_{P)UF}$ (L/m^{2}h)	$Ra(\%)_{UF}$
Exp1	1 month	19.5	68.57
Exp2	1–2 years	28.5	54.30

With regard of flux, it is observed that the value is significantly lower in the most recent waste, while the membrane provides a higher activity rejection rate. This may mean that the most recent waste has a larger size of proteins or other organic molecules, causing higher block of membrane pores, which causes less permeation flow. The higher value of the activity rejection rate would indicate that ^{125}I is bound to these proteins in a significant proportion. On the other hand, the higher flux value for the waste with the longest storage time may be indicative of a rupture or degradation of proteins by the effect of radiation (radiolysis) [44,45], which would cause less blocking of membrane pores and, therefore, a higher flow rate of permeation. This is consistent with a lower rejection rate to ^{125}I, because it would pass through the membrane in greater proportion by being bound in smaller portions of organic matter.

3.3. Performance of membranes in radioactivity removal

This section analyses the removal of activity at each membrane stage in each test performed, as well as the volume reduction factor achieved. Experiment 1 consisted in 4 passes of RO after the UF stage, whereas 3 passes of RO were carried out in the Experiment 2. Table 5 shows the corresponding results.

Table 5. Performance of activity removal of each membrane stage.

Experiment	Storage time of the waste	Stage	Average activity of permeate (kBq/L)	VRF [43]
Exp1	1 month	UF	117.5	21
		RO1	17.3	
		RO2	0.6	
		RO3	0.2	
		RO4	< background	
Exp2	1–2 years	UF	1.975	20
		RO1	0.65	
		RO2	0.12	
		RO3	< background	

As can be seen in Table 5, in the case of recent production waste, 4 RO passes are required after the UF stage to reach a level of activity below the discharge limit, while, in the case of waste with the longest storage time, this is achieved after 3 RO passes. In both cases, a volume reduction factor of about 20 has been achieved, meaning that the final volume of waste to be managed would be only 5% of the initial volume.

3.4. Fouling potential to membranes

It has been seen that, during the storage of the waste, organic matter can change in size by the

effect of radiation. Therefore, the current section analyses the variation of the characteristics of organic matter on the performance of membranes, through examination of the evolution of the permeate flux in the UF stage for each test performed. Figure 2 shows this evolution versus the test time.
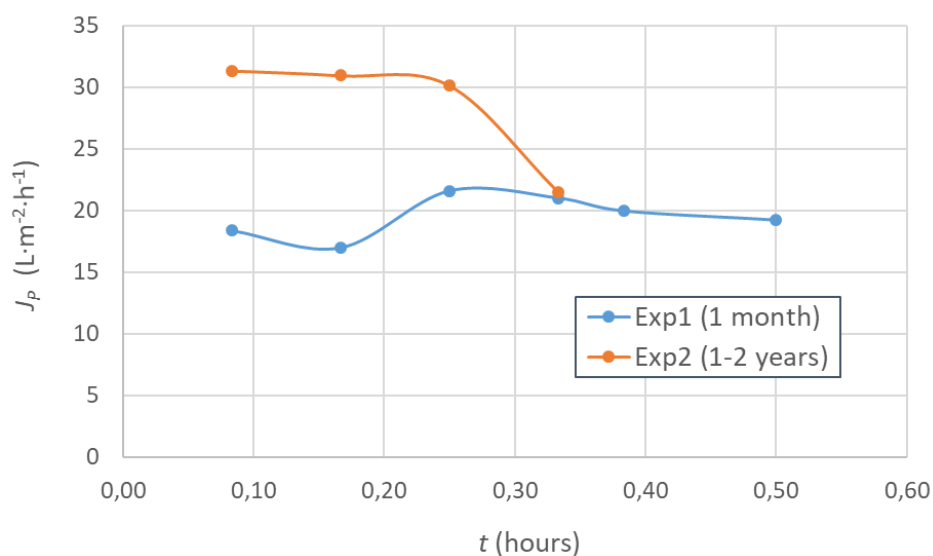

Figure 2. Evolution of permeate flux in the UF stage.

As shown in the figure, the permeate flux in the UF treatment of the longest storage time of waste was higher than the one corresponding to the most recent waste test. This confirms that the size of the molecules present in the waste with the longest storage time was smaller, as discussed above.
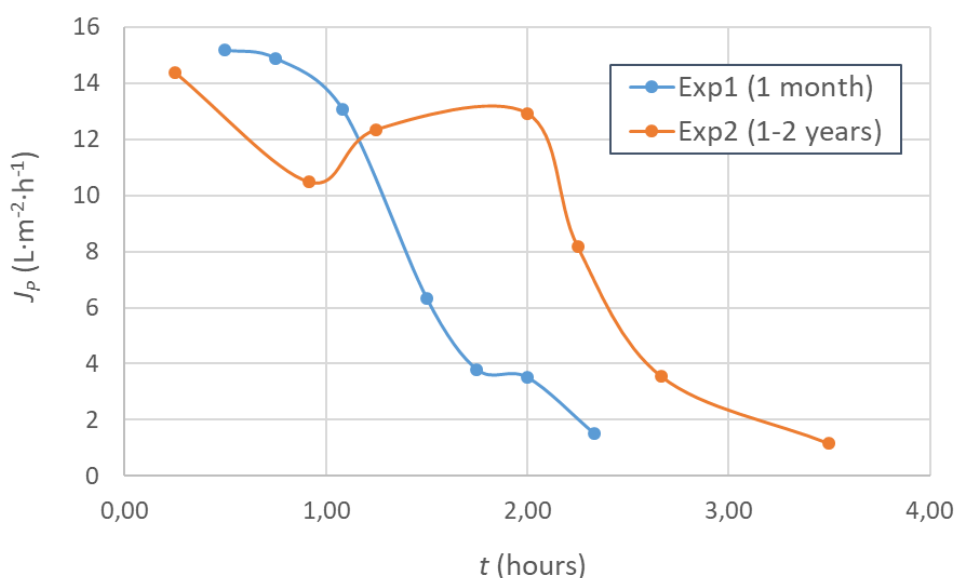

Figure 3. Evolution of permeate flux in the first RO pass.

To confirm this, the evolution of flux during the first RO pass applied to the UF permeate has also been studied. The results are shown in Figure 3. In this case, it is observed that the initial values of the permeate flux are higher for the UF permeate from the most recent waste, which would be indicative of a lower organic content, so the membrane would initially produce more permeate flow. However, it

is seen that the decrease in flux for UF permeate from recent waste is much more pronounced, indicating that, despite having a lower organic content, the size of the organic molecules present in the UF stage permeate would be larger. This can explain the sooner blocking of membrane pores in this case.

Thus, the results of RO's first pass are consistent with the conclusions inferred in the analysis of the UF stage: the most recent waste has a lower organic matter content (as the rejection of it in the UF stage has been greater), but the existing organic molecules are larger since they have not yet been affected by radiation.

Therefore, the shorter the waste has been stored, the greater the potential for fouling the membranes it has, due to a larger size of the organic molecules present in the waste.

3.5. Analysis of performance of the integral treatment: UF & RO

Finally, this section discusses the best alternative for treating liquid RIA wastes using an UF and RO system, based on their storage time.

From a radiological point of view, as it has been experimentally proven, it would be more convenient to treat the waste after a previous storage time between 1–2 years, since it would be possible to completely remove the radionuclide with fewer RO passes.

From the point of view of membrane performance, it has been experimentally proven that it would also be more convenient to apply the treatment to the waste after a certain storage time because radiation contributes to degrade proteins and other organic matter present in the wastes, thus decreasing the potential of membrane fouling and providing greater flow for longer.

Therefore, the recommended treatment for liquid waste of RIA will consist of a storage period for the decay of activity and degradation of organic matter, followed by a treatment through an integral system consisting in UF as pretreatment and several RO passes. The final volumes to be managed by the authorized company shall be less than 5% of the starting volume, with the consequent economic and occupancy savings of the radioactive warehouse.

4. Conclusions

Liquid radioactive wastes are produced in hospitals in several applications of radionuclides. This work focuses on liquid waste from radioimmunoassay laboratories, in which ^{125}I is the most typical radionuclide. Usually, this waste is temporary stored at the hospital for partial activity decay and then it is managed by an authorized company. This way of management of RIA waste compromises storage availability of both hospital and later radioactive centralized storage facility and means all the waste volume has to be disposed as radioactive. This work proposes a treatment by UF and RO system as an alternative. According to experimental results of treatment of real RIA waste by that system, the following conclusions can be stated: A significant reduction of waste volume can be achieved, with percentage of decontamination as radioactive waste about 95% of the initial volume; It has been proved that the best time for membrane treatment application would be after a period of storage in the hospital around 1–2 years, since membranes would perform more efficiently removing radioactivity with a fewer number of RO passes and with lower potential of fouling. In any case, treated flow should be authorized to direct discharge after proper measurements by the corresponding company.

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Conflict of interest

All authors declare no conflicts of interest in this paper.

References

1. IAEA-TECDOC-1183 (2000) Management of radioactive waste from the use of radionuclides medicine. Austria: IAEA.
2. Ravichandran R (2017) Management of Radioactive Wastes in a Hospital Environment, In: *Modelling Trends in Solid and Hazardous Waste Management*. Sengupta, Debashish, Agrahari, Sudha Editors, Singapore: 1–14.
3. Publication 94 Ann ICRP (2004) Release of patients after therapy with unsealed radionuclides. International Commission on Radiological Protection. Available from: https://journals.sagepub.com/doi/pdf/10.1177/ANIB_34_2.
4. Safe management of wastes from health-care activities (2014) Second edition, Edited by Yves Chartier, Jorge Emmanuel, Ute Pieper, Annette Prüss, Philip Rushbrook, Ruth Stringer, William Townend, Susan Wilburn and Raki Zghondi. World Health Organization. ISBN 978 92 4 154856 4.
5. Uriarte Jaureguízar J (2020) Gestión de residuos en centros hospitalarios. Available from: https://docplayer.es/17297659-Gestion-de-residuos-en-centros-hospitalarios-dr-ing-julian-uriarte-jaureguizar.html
6. Cesaro A, Belgiorno V (2017) Sustainability of medical waste management in different sized health care facilities. *Waste Biomass Valor* 8: 1819–1827.
7. Shoukat K, Syed AT, Reyaz A, et al. (2010) Radioactive waste management in a hospital. *Int J Health Sci* 4: 39–46.
8. IAEA-TECDOC-1714 (2013) Management of discharge of low level liquid radioactive waste generated in medical, educational, research and industrial facilities. Austria: IAEA.
9. Hossain F (2020) Natural and anthropogenic radionuclides in water and wastewater: Sources, treatments and recoveries. *J Environ Rad* 225: 1–18.
10. Carley-Macauly KW (1983) Options for the treatment of low and intermediate level active liquid wastes. *IAEA Radioactive Waste Management Intl Conference* 2: 15–35.
11. Koster R, Kraemer R (1989) Treatment and conditioning of liquid low and intermediate level wastes. *Management of Low and Intermediate Level Radioactive Wastes Conference* 2: 3–20.
12. Bradbury D (1989) Helping to reduce effluent generation. *Nucl Eng Int* 34: 44–46.
13. Zhang X, Gu P, Liu Y (2019) Decontamination of radioactive wastewater: State of the art and challenges forward. *Chemosphere* 215: 543–553.
14. Pabby AK, Swain B, Sonar NL, et al. (2021) Radioactive waste processing using membranes: state of the art technology, challenges and perspectives. *Sep Purif Rev* 1–23.

15. Mushtaq S, Yun SJ, Yang JE, et al. (2017) Efficient and selective removal of radioactive iodine anions using engineered nanocomposite membranes. *Environ Sci Nano* 4: 2157–2163.

16. Krawczyk E, Piñero-García F, Ferro-García MA (2013) Discharges of nuclear medicine radioisotopes in Spanish hospitals. *J Environ Rad* 116: 93–98.

17. Montaña M, Camacho A, Devesa R, et al. (2013) The presence of radionuclides in wastewater treatment plants in Spain and their effect on human health. *J Clean Prod* 60: 77–82.

18. Avila R, de la Cruz I, Sundell-Bergman S, et al. (2007) Radiological consequences of radionuclide releases to sewage systems from hospitals in Sweden. SSI Rapport. Stockholm: Swedish Radiation Protection Authority.

19. Martínez A, Peñalver T, Baciua M, et al. (2018) Presence of artificial radionuclides in samples from potable water and wastewater treatment plants. *J Environ Rad* 192: 187–193.

20. Mulas D, Camacho A, Garbayo A, et al. (2019) Medically-derived radionuclides levels in seven heterogeneous urban wastewater treatment plants: The role of operating conditions and catchment area. *Sci Total Environ* 663: 818–829.

21. Ruibal Morell A (2020) Remembering our history: 60 years ago radioimmunoanalysis was discovered. *Rev Esp Med Nucl Imagen Mol* 39: 337–339.

22. Lecama- Carrasco J (1997) Radiofarmacia y radioinmunoanálisis. *Educación Química* 8: 17–21.

23. Sancho M (2002) Tratamiento de residuos líquidos hospitalarios procedentes de RIA (radioinmunoanálisis) mediante técnicas de membrana. Doctoral Thesis, Universitat Politècnica de València.

24. Peyrin JO (1992) La gestion des déchets radioactifs hospitaliers. *Radioprotection* 27: 47–53.

25. Shapilov VV, Gorsky GG, Zvonova IA (2016) Can we consider wastes generated during radioimmunoassays as a radioactive waste? *Radiacionnaâ Gigiena* 3: 67–69.

26. Efremenkov VM (1999) Management of radioactive waste from medical application of radioisotopes. *Radioactive Waste Manage Environ Remediation Conference*: 3–8.

27. Rau EH, Alaimo RJ, Ashbrook PC, et al. (2000) Minimization and management of wastes from biomedical research. *Environ Health Perspect* 108: 953–977.

28. Hakimi SS, Nik M (1995) Removal of iodine-125 from effluents by chemical and soil column methods. *J Radioanal Nucl Chem* 196: 77–87.

29. Hakimi SS (1996) Improved iodine-125 removal in anionic form of iodate by column method using laterite soil. *J Radioanal Nucl Chem* 214: 117–131.

30. Edwards BE, Couch NW, Myers KD et al. (1996) ^{125}I aqueous waste volume reduction at a pharmaceutical research laboratory. *Health Phys* 71: 379–83.

31. Inoue H, Kagoshima M (2000) Removal of 125I radioactive experimental waste with an anion exchange paper membrane. *Appl Radiat Isot* 52: 1407–1412.

32. Lee S, Kim Y, Park J, et al. (2018) Treatment of medical radioactive liquid waste using Forward Osmosis (FO) membrane process. *J Membr Sci* 556: 238–247.

33. Lee BK, Ellenbeckerb MJ, Moure-Ersaso R (2004) Alternatives for treatment and disposal cost reduction of regulated medical waste. *Waste Manag* 24: 143–151.

34. Chmielewski AG, Harasimowicz M (1992) Influence of gamma and electron irradiation on transport properties of ultrafiltration membranes. *Nukleonika* 37: 61–70.

35. Chmielewski AG, Harasimowicz M, Zakrzewska-Trznadel G (1999) Membrane technologies for liquid radioactive waste treatment. *Czech J Phys* 49: 979–985.

36. Hagen K (1998) Removal of particles, bacteria and parasites with ultrafiltration for drinking water treatment. *Desalination* 119: 85–92.

37. Petrucci C, Traino AC (2015) Focus on the legislative approach to short half life radioactive hospital waste releasing. *Phys Medica* 31: 726–732.

38. Sen Gupta SK, Slade JA, Tulk WS (1995) Integrated plant for treatment of liquid radwaste. *Ind Water Treat* 27: 33–41.

39. Chmielewski AG, Harasimowicz M, Tyminski B, Zakrzewska-Trznadel G (2001) Concentration of low level and medium level radioactive wastes with 3-stage reverse osmosis pilot plant. *Sep Sci Technol* 36: 1117–1127.

40. Abdel Rahman RO, Ibrahium HA, Hung YT (2011) Liquid radioactive wastes treatment: A review. *Water* 3: 551–565.

41. Chmielewski AG, Harasimowicz M (1997) Influence of gamma and electron irradiation on transport properties of nanofiltration and hyperfiltration membranes. *Nukleonika* 42: 857–862.

42. Arnal JM, Sancho M, Verdú G, et al. (2003) Treatment of ^{137}Cs liquid wastes by reverse osmosis. Part I. Preliminary tests. *Desalination* 154: 27–33.

43. Sancho M, Arnal JM, García-Fayos B (2013) Treatment of hospital radioactive liquid wastes from RIA (radioimmunoassay) by membrane technology. *Desalination* 321: 110–118.

44. Jayko ME, Weeks BM, Garrison WM (1960) Mechanism in the radiolysis of aqueous protein solutions. Lawrence Berkeley National Laboratory. Available from: https://escholarship.org/content/qt8rp6033g/qt8rp6033g.pdf.

45. Houée-Levina C, Bobrowskib K (2013) The use of the methods of radiolysis to explore the mechanisms of free radical modifications in proteins. *J Proteomics* 92: 51–62.