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CONCENTRATION OF LOW- AND MEDIUM-LEVEL RADIOACTIVE WASTES WITH THREE-STAGE REVERSE OSMOSIS PILOT PLANT

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ABSTRACT

A study of the application of different membrane processes to nuclear technology has been conducted at the Institute of Nuclear Chemistry and Technology (INCT) for many years. One application of membrane methods involves the purification and concentration of liquid low- and medium-level radioactive wastes (LLLRWs and LMLRWs).

The separation of radioactive model solutions as well as LLLRW and LMLRW samples has been examined with various reverse osmotic modules equipped with thin-composite polymeric membranes. On the basis of these investigations, membrane modules were selected for the pilot plant processing of liquid wastes from nuclear centers. A three-stage *JP3RO* reverse osmosis unit was constructed. The installation is described in this paper, and the results of preliminary pilot plant experiments are presented.

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INTRODUCTION

The separation of radioactive elements from various waste materials is a significant problem facing developed countries. Since there is no alternative to the nuclear power industry in many countries, the production of radioactive waste materials will continuously increase in the near future. The management of such materials is an important issue for technical, political, and social reasons.

Liquid low- and medium-level radioactive wastes (LLLRWs and LMLRWs) are generated from many sources, including nuclear reactors, isotope laboratories, and nuclear medical centers. Many of these wastes require treatment in order to reduce the total amounts of radioactive and nonradioactive contaminants to levels that would allow them to be discharged according to international conventions and national regulations. Since these wastes arise in relatively large volumes but contain low-specific-activity components, the treatment methods include a removal of radioactive compounds from the effluent and a volume reduction of the concentrate, which can then be safely stored in a radioactive "cemetery" after fossilization.

Many methods are involved to treat LLRWs and LMRWs: chemical precipitation, sedimentation, ion exchange, thermal evaporation, biological methods, and membrane permeation. However, membrane permeation is a novel technology and, therefore, has only been applied in a few nuclear centers around the world.

At the Chalk River Laboratory in Canada, membranes have been employed in a mixed aqueous waste processing plant [1,2]. This plant consists of microfiltration units, as well as spiral- wound reverse osmosis (RO) and tubular RO systems. Ultrafiltration was used in two stages in the Enhanced Actinide Removal Plant at Sellafield [3]. In this plant a primary separation stage produced a concentrate with a few weight percent solids content, which was then dewatered in a second stage. At the Paks Nuclear Power Plant, in Hungary, an ultrafiltration installation was used for cleaning and recycling contaminated boric acid solutions. The contaminated solution was treated by using polysulphone membranes to give a volume reduction factor of 45 and decontamination factors in the range 10-100. Ultrafiltration was used to separate floc and liquid at the Harwell pilot plant [3]. Low- level wastes arising from a fuel fabrication plant have been treated by ultrafiltration in Nukem, Hanau [4]. Direct ultrafiltration of the laundry and cleaning waste gave a decontamination factor of about 10 and a volume reduction 10-15 times greater than that obtained with the precipitation method. The seeded ultrafiltration procedure is being developed in the Cadarache Nuclear Research Center [5] and in Harwell [6]. Nanofiltration for the separation of ionized silica and boric acid has been examined in the Bugey Nuclear Power Plant in France [7].

Laboratory and pilot plant experiments carried out at INCT showed that RO could be applied for the treatment of LLLRWs from Polish nuclear laboratories.

However, a high decontamination level for the process should be achieved by using a multistage operation [8].

The RO method is now being implemented at Institute of Atomic Energy in Swierk, Warsaw. The wastes collected there from all users of nuclear materials in Poland have to be treated before safe disposal can be ensured. The membrane plant will be used for initial concentration of the waste before evaporation or for final cleaning of the distillate, depending on actual needs.

EXPERIMENTAL

LLLRWs and LMRWs throughout Poland are collected in a storage tank (capacity, 300 $m^{3)}$ at the Institute of Atomic Energy (Swierk). The specific activity of this stored material is not higher than 10^5 kBq/m^3. The purification/concentration procedure by membrane permeation leads to partition of the feed stream into two exit streams: permeate and retentate. The specific activities of alpha and beta emitters in the retentate and the permeate are not expected to be higher than 10^7 kBq/m^3 and 10 kBq/m^3, respectively. Actually, these values correspond to a fourfold reduction of radioactive substances concentration if the permeate is to be released to communal sewage or to be reused as process water. The total concentrations of dissolved salts in the concentrate and permeate do not exceed 250 g/dm^3 and 0.1 g/dm^3, respectively.

Apparatus: Three-stage Reverse Osmosis Plant for Liquid Radioactive Waste Treatment

A three-stage *JP3RO* reverse osmosis (RO) unit will be included in the system for liquid radioactive waste treatment, now modernized at the Institute of Atomic Energy, Swierk. The new system will be composed of an evaporator, which cooperates with membrane installation. A schematic diagram of the membrane facility is shown in Fig. 1.

LLLRWs and LMLRWs are pretreated in a 300- m^3 storage tank. The pretreatment operations include pH regulation to 5.0-6.5, sedimentation of suspended matter, and surface removal of sediment. Clarified liquid is collected in an 8- m^3 feed reservoir. The *JP3RO* unit is composed of two purification stages (stages I and II) and one concentration stage (stage III). Each purification stage is formed by two RO 8x40 in. *SU 720R* modules (Toray) connected in series and placed in a high-pressure vessel. The concentration stage is composed of two parallel housings, with two RO 4x40 in. *SU 810* (Toray) modules in series in each. Characteristics of the modules selected for the plant are summarized in Table 1. Stages I and III are fed with a high-pressure *CRNE 2-260* (Grundfos) pump, and retentate cir-

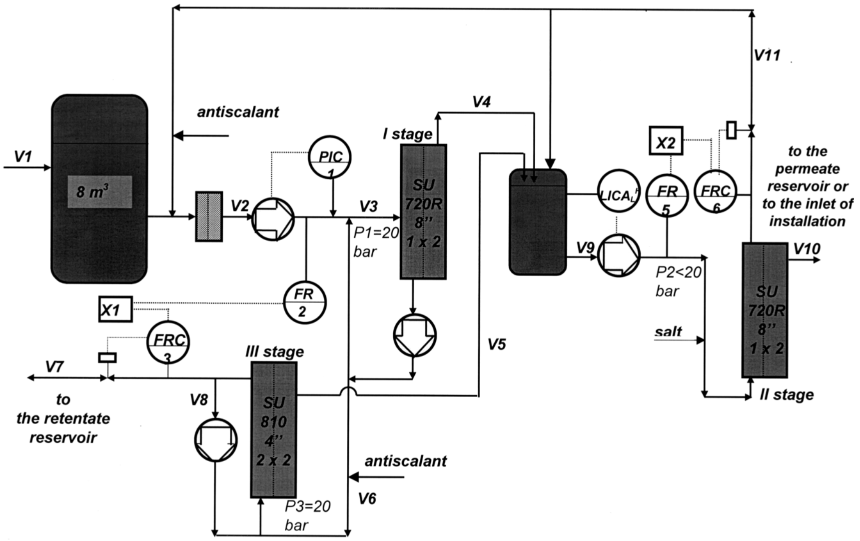

Figure 1. Schematic diagram of three-stage JP3RO plant for liquid radioactive waste treatment.

culation is maintained by an *I-NM4-32/200 BE* pump (Calpeda). Similar to that was employed in stage II. With such an arrangement the liquid pressure remains equal to 1.8-2.2 MPa in each stage, and permeate flux in stage II is approximately 1 m^3/h. Distribution of the retentate and permeate streams in the entire unit is as follows:

- retentate from stage I is directed to stage III inlet;
- retentate from stage II is directed to the stage I inlet;

Table 1. Characteristics of RO (TRAY) Modules Selected for the Pilot Plant

	Model SU-720R	Model SU-810
Membrane type	Cross- linked, fully aromatic polyamide composite	Cross- linked, fully aromatic polyamide composite
Element configuration	Spiral wound	Spiral wound
Salt rejection	99.7% [a]	99.4% [b]
Product flow rate	20.0 m^3/day [a]	4.0 m^3/day [b]

[a] *Test conditions: pressure, 15 kg/cm^2; temperature, 25^{o}C; feed concentration, 1500 ppm NaCl; brine flow rate, 80 dm^3/min; pH, 6.5.*

[b] *Test conditions: pressure 56 kg/cm^2; temperature 25^{o}C; feed concentration, 35% NaCl; brine flow rate, 20 dm^3/min; pH, 6.5.*

- retentate from stage III is directed to the concentrated- wastes reservoir; from this reservoir wastes can be directed to the evaporator;
- permeate from stages I and III is directed to intermediate tank; the liquid from this tank feeds stage II;
- permeate from stage II is directed to the pure waste tank, if the specific activity is below 10 Bq/dm3, or to an intermediate tank if the specific activity exceeds that level.

Each portion of the waste is analyzed chemically and radiochemically. The analytical results are used for elaboration of pretreatment procedures of the wastes before stage I of purification, for each type of the waste and each design scheme of RO installation. Such data are also helpful for the preparation of the recipes for additional conditioning of the streams directed to stages II and III. These pretreatment procedures are important for avoiding scale formation. Hydrochloric acid is injected to prevent $CaCO_3$ formation, and sulfate deposits are avoided by *RPI-2000A* and *RPI-4000* scale inhibitors; CO2 is bounded by NaOH.

Results and Discussion

Preliminary experiments were carried out at a *JP3RO* pilot plant using three samples of original radioactive wastes *A, B, and C* from storage tanks at the Institute of Atomic Energy (Table 2). The samples contained only beta and gamma emitters, primarily ^{51}Cr, ^{60}Co, ^{65}Zn, ^{90}Sr, ^{95}Zr, ^{121}Te, ^{124}Sb, ^{125}Sb, ^{131}I, ^{133}Ba, ^{137}Cs, ^{144}Ce, and ^{155}Eu. The specific activity of liquid radioactive wastes collected

Table 2. Chemical Compositions of Waste Samples from the Waste Collector in Swierk

	A	B	C
Ca, ppm	76.00	38.00	91.20
Na, ppm	124.00	62.00	148.80
Na, ppm	26.50	13.25	31.80
NH_4, ppm	70.50	35.25	84.60
Sr, ppm	14.00	7.00	16.80
Cl, ppm	55.20	27.60	66.20
SO_4, ppm	248.90	124.45	298.65
NO_3, ppm	0.11	0.05	0.12
HCO_3, ppm	0.10	0.05	0.12
SiO_2, ppm	26.00	13.00	31.20
CO_2, ppm	0.02	0.05	0.02
Total salts, ppm	641.31	320.65	769.54
A, Bq/ dm^3	2.1×10^4	2.8×10^2	2.2×10^3

in storage tanks for recent years did not exceed 7.5 x 10^4 kBq/m^3; therefore, a 10^4 –fold reduction of radioactive compounds had to be achieved. The stream partition of the installation is shown in Fig. 2. Their quantities were adjusted in each experiment in order to ensure that the permeate from stage II had a specific activity of 10 Bq/dm^3, and that the activity of the retentate from stage III to be 7-15 times higher than that of feed solution. Periodically samples of the feed solution, permeate from stage II, retentate from stage III, and solution from the intermediate tank were withdrawn for analysis. The concentration of total dissolved salts, *TDS,* and, the specific activity, $A_{s,}$ for each sample were measured. Data retention coefficients, *R,* coefficients of salt concentration reduction, K_{TDS}, decontamination factors, *DFs,* and concentration factors, *CFs,* were calculated according to the following formulae:

$$R = \frac{c_f - c_p}{c_f} * 100\%,$$

$$K_{TDS} = \frac{c_f}{c_p},$$

$$DF = \frac{A_f}{A_p}, \text{ and}$$

$$CF = \frac{A_R}{A_f},$$

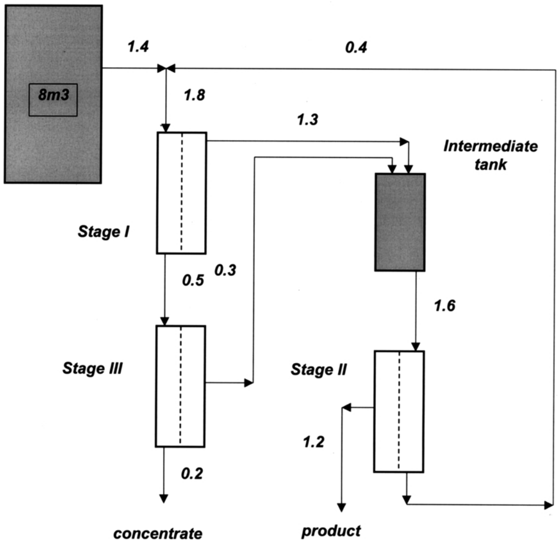

Figure 2. Distribution of the streams in JP3RO.

Table 3. Removal of Nonradioactive Substances (Dissolved Salts)

	A	B	C	A (+NaCl)[a]
Stage I				
Feed, ppm	641.3	320.6	769.5	641.3
Permeate, ppm	3.1	1.5	3.9	3.1
Retention, R, %	99.52	99.60	99.49	99.52
TDS reduction coefficient, K_{TDS}	208	214	197	208
Stage II				
Feed, ppm	4.2	2.3	4.2	1528.4
Permeate, ppm	1.3	1.5	1.2	12.7
Retention, R %	68	34	71	99.17
TDS reduction coefficient, K_{TDS}	3.14	1.51	3.50	120
Stage III				
Feed, ppm	2344.9	1265.3	2455.9	2344.9
Permeate, ppm	8.7	5.6	7.0	8.7
Retention R, %	99.63	99.56	99.71	99.63
TDS reduction coefficient, K_{TDS}	269	226	322	269
Entire unit				
Feed, ppm	641.3	320.6	769.5	641.3
Product, ppm	1.3	1.5	1.2	12.7
Retention R, %	99.79	99.52	99.84	98
TDS reduction coefficient, K_{TDS},	478	209	635	50
ppm	6647.3	5002.3	5615.8	6647.3

[a] NaCl solution was added to the intermediate tank in quantity thus enabling a TDS value of ~1500 ppm to be achieved.

where:

c_p = permeate concentration,
c_f = feed concentration,
A_f = specific activity of the feed,
A_p = specific activity of permeate, and
A_R = specific activity of retentate.

The *R*, K_{TDS}, *DF*, and *CF* values are presented in Tables 3 and 4.

Decontamination factors in stage I are close to 200, which means that 99.5% of the radioactive compounds are retained in that stage. Stage II plays a moderate role in further purification because the concentration of dissolved salts in the solution feeding this stage is below 5 ppm. In this concentration range the retention ability of *SU720 R* membrane is low; only 1.5- to 3.5-fold reduction of *TDS* and 1.4- to 3.2 –fold reduction of specific activity are achieved. Taking into account that at the inlet of stage II the mixture of permeates from stages I and III is intro-

Table 4. Removal of Radionuclides

	A	**B**	**C**	**A (+NaCl)**
Stage I				
Feed, Bq/dm^3	1.95×10^4	280	2200	1.93×10^4
Permeate, Bq/dm^3	96	1.4	10.4	94
DF	203	200	211	205
Stage II				
Feed, Bq/dm^3	136	2.3	12.3	128
Permeate, Bq/dm^3	44	1.6	3.9	1.4
DF	3.09	1.44	3.15	91
Stage III				
Feed, Bq/dm^3	7.1×10^4	1.1×10^3	7×10^3	7×10^4
Permeate, Bq/dm^3	275	5.4	23	268
DF	258	204	304	261
CF	2.82	3.91	2.29	2.85
Entire Unit				
Feed, Bq/dm^3	1.95×10^4	280	2200	1.93×10^4
Product, Bq/dm^3	44	1.6	3.9	1.4
Comcentrate, Bq/dm^3	2×10^5	4.3×10^3	1.6×10^4	2×10^5
DF	443	175	564	13785
CF	10.25	15.36	7.27	10.36

duced, the permeate from stage II can be of poor quality as compared with the permeate from stage I (in the case of *B* solution).

It was observed that the removal efficiency of radionuclides in stage II can be increased if the TDS concentration is increased to a level that would allow NaCl retention in the *SU720 R* module to remain above 99%. The previous laboratory experiments, described below, showed an influence of the total concentration of ballast salts (TDS) on the decontamination factor for cesium isotopes.

Influence of Total Dissolved Salts Concentration on the Retention of Radioactive Ions

The influence of the total dissolved salts concentration on the retention of radioactive ions was tested in laboratory, using a membrane module of the DOW Chemical DENMARK A/S – SW30-F type. The characteristics of the membrane module are summarized in Table 5. The investigations were carried out with water solutions composed of 100 mg/dm^3 CsCl and 0.5, 2, 5, and 10 g/dm^3 of NaCl. The results of membrane filtration of these solutions are presented in Fig. 3.

The cesium ion retention with the SW30-F membrane and with a 55% permeate recovery was lower than of the sodium ion retention over the entire exper-

Table 5. Characteristics of the Laboratory Module

SW30-F: spiral wound; surface area, 2.1 m^3	
NaCl retention, %	99.4
Operating pressure, bar	69
Membrane type	TFC
Permeate flow, dm^3/h	76.7
pH range	2-11

imental range, except for the solution with the lowest TDS (see Fig. 3). The highest retention, about 99.5%, was reached at 5 g/dm^3 of NaCl (TDS= 5100 mg/dm^3). A 190-fold concentration reduction of cesium ions was achieved in relation to the initial concentration of feed solution. The concentration of cesium ions in permeate was 0.44 g/dm^3.

For other solutions, at TDS values of 600, 2100, and 10,100 g/dm^3, the concentration reduction of cesium ions was 110, 150, and 130, respectively. The investigations showed that the highest separation (99.5%) of cesium ions was reached at a TDS of 2100-5100 g/dm^3.

Results of laboratory experiments showed an influence of TDS on cesium ion retention. At low TDS, the cesium removal efficiency for RO membranes decreased significantly. Additional solute injection improved retention, but only in some TDS ranges, when the maximum retention was reached; a further increase in TDS increase caused a decrease in cesium concentration. Therefore, in industrial installations, careful control of TDS concentration is required to achieve high cesium ion removal.

Similar experiments were carried out in the pilot plant. A sample of solution *A* waste le was used as the feed solution. The retentate stream from stage II was directed to the intermediate tank (instead of I stage inlet); then a specified amount

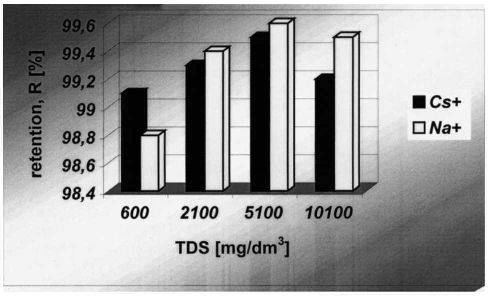

Figure 3. Effect of total dissolved salts (TDS) concentration on cesium and sodium ion removal by reverse osmosis membrane.

of NaCl solution was added before stage II in order to reach a TDS concentration of 1500 ppm. The solution prepared in such a way circulated through the modules of stage II and did not influence the working conditions of stages I and III. The calculated retention values, TDS reduction coefficients and decontamination factors are shown in Tables 3 and 4. However, the procedure described above caused an increase in the salt concentration of the permeate obtained in stage II to 12.7 ppm while the specific activity decreased to 1.4 Bq/dm^3 (this value was 44 before injection of the salt solution). The decontamination factor for stage II increased 30 times and, for the entire unit, was 1.4 x 10^4. It was expected that the specific activity of the permeate from stage II will be lower than 10 Bq/dm^3 for all the wastes with specific activities below under 10^5 Bq/dm^3 and TDS values in the range 100-1000 ppm.

CONCLUSIONS

Reverse osmosis proved to be useful for processing LLLRWs and LMLRWs. To reach a sufficient purity for permeate, the process had to be conducted as a multistage operation. Using a three-stage *JP3RO* reverse osmosis unit helped to clean up wastes with salinity of 300-800 ppm, yielding a product with a specific activity 200-14,000 times lower than that of the initial activity of the feed and a retentate with a specific activity 7-15 times higher. At very low TDS values, the efficiency of RO membrane decreased. An improvement of the efficiency and an increase of radionuclide retention were made possible by additional injection of salt solution. Since the relation between retention of radioactive ions and TDS was not linear, careful control of TDS was necessary.

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