Experimental and Modelling of Aqueous Radioactive Waste Treatment by Ultrafiltration

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Ultrafiltration of untreated and pretreated aqueous radioactive wastes was conducted using a spiral-wound polysulphonamide membrane. The influence of process factors on its performances was experimental studied and predicted. Permeate volumetric flux and permeate total suspended solids (TSS) were measured at different values of feed flow rate (7 and 10 m^3/h), operating pressure (0.1-0.4 MPa), and feed TSS (15 and 60 mg/L). Permeate flux (42-200 L/(m^2·h)) increased with feed flow rate and operating pressure as well as it decreased with an increase in feed TSS, whereas permeate TSS (0.1-33.2 mg/L) exhibited an opposite trend. A 2^3 factorial plan was used to establish correlations between dependent and independent variables of ultrafiltration process.

Keywords: *aqueous radioactive waste, factorial experiment, spiral-wound membrane, total suspended solids, ultrafiltration*

Low and intermediate level radioactive wastes are mainly generated within the nuclear fuel cycle as well as during the production and application of radioisotopes [1,2]. Radioactive wastes are harmful to living organisms, natural resources, and environment [3].

Treatment is an essential phase in the management of aqueous radioactive wastes (ARW). It mainly depends on the type, radioactivity level, and chemical composition of the waste. After the treatment phase, ARW are separated into two parts, *i.e.*, a small amount of secondary waste containing the bulk of radionuclides and a large volume of aqueous solution with low radioactivity that can be discharged into the environment after achieving the regulatory requirements [1].

Treatment methods for ARW generally include chemical treatment, adsorption, filtration, ion exchange, and evaporation. However, these techniques are unable to remove all contaminants, involve high operating costs, and produce large amounts of secondary waste.

Membrane separation processes used for decontamination of radioactive water have been extensively studied in recent years [1-9]. Ultrafiltration and reverse osmosis have gained considerable attention due to their relatively low capital and operating costs. A wide range of contaminants present in untreated water, municipal and industrial wastewater discharges, *e.g.*, suspended and dissolved solids, organic matter, heavy metals, bacteria, viruses, can be removed by these processes. Membrane separation processes are often applied in the nuclear industry for removing radioisotopes and obtaining small amounts of secondary waste [9].

This paper has aimed at studying the ultrafiltration of untreated and pretreated ARW through a spiral-wound polysulphonamide membrane. The effects of process factors on membrane productivity and separation efficiency were measured and predicted.

Experimental part

ARW ultrafiltration was conducted in AQUA-EXPRESS installation (SIA “Radon”, Moscow, Russia) shown in figure 1. Ultrafiltration module of this installation contains an ERU-100-1016 membrane element (JSC STC Vladipor, Russia) (fig. 2). Main characteristics of membrane are summarized in table 1.

Membrane productivity expressed by permeate volumetric flux, j_P (L/(m^2·h)), and separation efficiency evaluated as permeate total suspended solids, TSS_P (mg/L), were measured at different values of process factors, *i.e.*, feed flow rate (G_V=7, 10 m^3/h), operating pressure (p=0.1-0.4 MPa), and feed TSS (TSS_F=15, 60 mg/L). The high value of TSS_F corresponds to untreated ARW, whereas its low level characterizes the waste pretreated by sand filtration. TSS was determined based on vacuum filtration using a 50 nm nylon membrane.

Fig. 1. AQUA-EXPRESS installation:
(1) sand filtration module, (2) ultrafiltration module, (3) reverse osmosis module

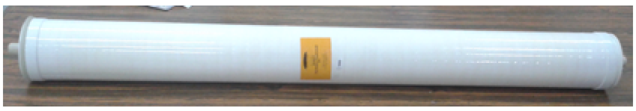

Fig. 2. ERU-100-1016 ultrafiltration membrane element

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Table 1
MEMEBRANE CHARACTERISTICS

Configuration	Spiral-wound
Polymer	Polysulphonamide
Nominal area	5 m^2
Length	1016 mm
Outer diameter	100 mm
Molecular weight cut-off	100 kDa

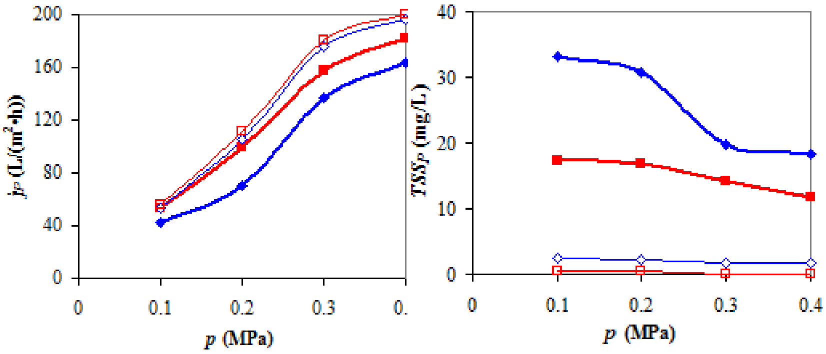

Fig. 3. Permeate flux and permeate TSS *vs.* operating pressure for different feed flow rates of untreated (TSS_F=60 mg/L) and pretreated (TSS_F=15 mg/L) ARW (untreated: ♦ G_V=7 m³/h, ■ G_V=10 m³/h; pretreated: ◇ G_V=7 m³/h, □ G_V=10 m³/h)

Table 2
EXPERIMENTAL MATRIX OF 2^3 FACTORIAL EXPERIMENT

Exp.	G_V (m³/h)	p (MPa)	TSS_F (mg/L)	x_1	x_2	x_3	j_P (L/(m²·h))	TSS_P (mg/L)
1	7	0.1	15	-1	-1	-1	53	2.4
2	7	0.1	60	-1	-1	1	42	33.2
3	7	0.4	15	-1	1	-1	196	1.8
4	7	0.4	60	-1	1	1	164	18.4
5	10	0.1	15	1	-1	-1	56	0.6
6	10	0.1	60	1	-1	1	53	17.2
7	10	0.4	15	1	1	-1	200	0.1
8	10	0.4	60	1	1	1	182	11.6

Results and discussions

Experimental data

The effects of process factors on permeate performances in terms of its volumetric flux (j_p=42-200 L/(m²·h)) and TSS (TSS_p=0.1-33.2 mg/L) are highlighted by the experimental results represented in figure 3. Depicted data emphasize the following issues: (i) j_P increases with *operation pressure* (p=0.1-0.4 MPa) up to about 4 times, whereas TSS_p decreases with an increase in p (up to 1.8 times for untreated and up to 6 times for pretreated ARW); (ii) j_P increases with *feed flow rate* (G_V=7, 10 m³/h), *i.e.*, 1.1-1.4 times for untreated and up to 1.1 times for pretreated ARW, whereas TSS_p decreases with an increase in G_V (1.4-1.9 times for untreated and 4-18 times for pretreated ARW); (iii) j_p is 1.1-1.5 higher and TSS_p is 11-141 lower (up to 15 times for G_V=7 m³/h and over 28 times for G_V=10 m³/h) for *pretreated* (TSS_F=15 mg/L) than for *untreated* (TSS_F=60 mg/L) ARW. Accordingly, j_p increases with p and G_V as well as it decreases with an increase in TSS_F, whereas TSS_p exhibits an opposite trend.

Statistical model

Permeate flux, j_p (L/(m²·h)), and permeate TSS, TSS_p (mg/L), were selected as process dependent variables (responses). Their final values, as well as dimensionless values of process factors determined by eqs. (1)-(3), are given in table 2. Tabulated data were processed using the procedure recommended for a 2^3 factorial plan resulting in eqs. (4) and (5).

$$x_1 = \frac{G_V - 8.5}{1.5} \quad (1)$$

$$x_2 = \frac{p - 0.25}{0.15} \quad (2)$$

$$x_3 = \frac{TSS_F - 37.5}{22.5} \quad (3)$$

$$j_P = 118.25 + 4.50x_1 + 67.25x_2 - 8x_3 + x_1x_2 +$$

$$+ 2.75x_1x_3 - 4.50x_2x_3 + 0.75x_1x_2x_3 \quad (4)$$

$$TSS_P = 10.662 - 3.287x_1 - 2.688x_2 + 9.438x_3 + 1.163x_1x_2 -$$

$$- 2.413x_1x_3 - 2.413x_2x_3 + 1.138x_1x_2x_3 \quad (5)$$

Statisticals models described by eqs. (6) and (7) were obtained after eliminating insignificant regression coefficients in eqs. (4) and (5) [10-13].

$$j_P = 118.25 + 4.50x_1 + 67.25x_2 - 8x_3 + 2.75x_1x_3 - 4.50x_2x_3 \quad (6)$$

$$TSS_P = 10.662 - 3.287x_1 - 2.688x_2 + 9.438x_3 -$$

$$- 2.413x_1x_3 - 2.413x_2x_3 \quad (7)$$

Regression equations (6) and (7) reveal the following issues: (i) permeate flux (j_p) increases with feed flow rate (x_1), operation pressure (x_2), and x_1x_3 interaction, decreases with an increase in feed TSS (x_3) and x_2x_3 interaction, as well as it is heavily affected by x_2 factor; (ii) permeate TSS (TSS_p) decreases with an increase in x_1, x_2, and x_1x_3 interaction, whereas it increases with x_3 and x_2x_3 interaction; moreover, x_3 factor has a more significant effect on TSS_P.

Statistical models expressed by Eqs. (6) and (7) may be applied to predict the values of permeate flux and permeate TSS at levels of process factors in the ranges considered in the study, *i.e.*, G_V=7-10 m³/h, p=0.1-0.4 MPa, and TSS_F=15-60 mg/L.

Conclusions

ARW ultrafiltration was performed using an installation containing spiral-wound polysulphonamide membrane modules. Permeate volumetric flux (j_p) and permeate TSS (TSS_p) were measured at different values of feed flow rate (G_V=7, 10 m³/h), operating pressure (p=0.1-0.4 MPa), and feed TSS (TSS_F=15, 60 mg/L). Permeate flux (42-200 L/(m²·h)) increased with G_V (up to 1.4 times) and p (up to about 4 times), as well as decreased with an increase in

TSS_F (1.1-1.5 times). Permeate TSS (0.1-33.2 mg/L) exhibited an opposite trend, *i.e.*, it increased with TSS_F (11-141 times) and decreased with an increase in G_V (1.4-18 times) and p (up to about 6 times). The influence of process factors on its performances was quantified using a 2^3 factorial plan. Regression equations which were obtained could be applied to predict the values of permeate flux and TSS at levels of process factors in the ranges considered in the experimental study.

References

1. ABDEL RAHMAN, R.O., IBRAHIUM, H.A., HUNG, Y.T., Water, **3**, 2011, p. 551.
2. ZAKRZEWSKA-TRZNADEL, G., Nukleonika, **51** (S1), 2006, p. 101.
3. RANA, D., MATSUURA, T., KASSIM, M.A., ISMAIL, A.F., Desalination, **321**, 2013, p. 77.
4. MISRA, B.M., RAMACHANDRAN, V., Treatment of radioactive effluents: Introduction, fundamentals, and scope of different membrane processes, Handbook of Membrane Separations: Chemical, Pharmaceutical, and Biotechnological Applications (Chapter 29), 2008, p. 827, CRC Press, Boca Raton, FL.
5.CHMIELEWSKI, A.G., HARASIMOWICZ, M., ZAKRZEWSKA-TRZNADEL, G., Czech. J. Phys., **49** (S1), 1999, p. 979.
6. ZAKRZEWSKA-TRZNADEL, G., Radioactive waste processing: Advancement in pressure-driven processes and current world scenario, Handbook of Membrane Separations: Chemical, Pharmaceutical, and Biotechnological Applications (Chapter 30), 2008, p. 843, CRC Press, Boca Raton, FL.
7. PABBY, A.K., Membr. Technol., **11**, 2008, p. 9.
8. AMBASHTA. R.D., SILLANPAA, M.E.T., J. Environ. Radioact., **105**, 2012, p. 76.
9. SANCHO, M., ARNAL, J.M., VERDU, G., LORA, J.,VILLAESCUSA, J.I., Desalination, **201**, 2006, p. 207.
10. ARUS, A.V., NISTOR, I.D., PLATON, N., ROSU, A.M., MUNTIANU, G., JINESCU, C., Rev. Chim. (Bucharest), **66**, no. 1, 2015, p. 88.
11. CIOROIU, D.R., PARVULESCU, O.C., KONCSAG, C.I., DOBRE, T., Rev. Chim. (Bucharest), **68**, no. 10, 2017, p. 2311.
12. DOBRE, T., STOICA, A., PARVULESCU, O.C., STROESCU, M., IAVORSCHI, G., Rev. Chim. (Bucharest), **59**, no. 5, 2008, p. 590.
13. ION, V.A., PARVULESCU, O.C., DOBRE, T., Appl. Surf. Sci., **335**, 2015, p. 137.
14. ORBECI, C., PARVULESCU, O.C., ACCELEANU, E., DOBRE, T., Rev. Chim. (Bucharest), **68**, no. 10, 2017, p. 2325.

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