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## **Original Article**

# NUCLIDE SEPARATION MODELING THROUGH REVERSE OSMOSIS MEMBRANES IN RADIOACTIVE LIQUID WASTE

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#### ABSTRACT

The aim of this work is to investigate the transport mechanism of radioactive nuclides through the reverse osmosis (RO) membrane and to estimate its effectiveness for nuclide separation from radioactive liquid waste. An analytical model is developed to simulate the RO separation, and a series of experiments are set up to confirm its estimated separation behavior. The model is based on the extended Nernst-Plank equation, which handles the convective flux, diffusive flux, and electromigration flux under electroneutrality and zero electric current conditions. The distribution coefficient which arises due to ion interactions with the membrane material and the electric potential jump at the membrane interface are included as boundary conditions in solving the equation. A high Peclet approximation is adopted to simplify the calculation, but the effect of concentration polarization is included for a more accurate prediction of separation. Cobalt and cesium are specifically selected for the experiments in order to check the separation mechanism from liquid waste composed of various radioactive nuclides and nonradioactive substances, and the results are compared with the estimated cobalt and cesium rejections of the RO membrane using the model. Experimental and calculated results are shown to be in excellent agreement. The proposed model will be very useful for the prediction of separation behavior of various radioactive nuclides by the RO membrane.

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#### 1. Introduction

Radioactive liquid wastes (liquid radwaste) generated in a nuclear facility arise in a wide range of concentrations of radioactive materials and in different physical and chemical forms, which require the use of a variety of processing methods for their treatment and conditioning. In other words, the selection of a treatment process for a liquid radwaste is determined by the physical, chemical, and radiological properties of the waste. It is also decided by factors such as release of radioactive materials to the environment, solid radioactive waste generation, occupational radiation exposure, reliability and maintainability of the process system, and capital and operational costs [1].

The key radioactive nuclides to be removed from the liquid radwaste are cobalt (Co) and cesium (Cs) [2]. Over the years,

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the reverse osmosis (RO) membrane has been applied to process the liquid radwaste from commercial nuclear power plants [3–5]. Most RO installations have operated efficiently, producing high-quality water at low operating costs. Even though membrane processes have long been employed successfully in nuclear power plants, the separation mechanism of the RO membrane is not yet clearly defined for the liquid in a variety of conditions. In this study, an analytical model is developed to simulate the RO separation in liquid radwaste, and the separation behavior of Co and Cs from various radioactive nuclides and nonradioactive substances is verified by a series of experiments.

#### 2. Theoretical model

A general theory to govern multi-components separations through an RO membrane is developed using the extended Nernst–Planck equation [6] with electroneutrality and zero electric current conditions. It is also assumed that the magnitude of the membrane charge varies with ion concentration and pH inside the membrane as well as the specific interaction potential between the ions and the membrane material.

All working equations are placed in nondimensional forms. Concentrations are presented dimensionless using the transformation  $c = C/\sqrt{K_w}$ , where  $K_w = 10^{-14} \text{ (mol/1)}^2 = 10^{-20} \text{ (mol/cm}^3)^2$  is the water ion-product. Consequently, the product of the hydrogen and hydroxyl ion concentrations in the bulk water rescales to  $k_w \equiv c_H c_{OH} = 1$ . This product, which is valid for the bulk water, is not assumed to be the same inside the membrane.

#### 2.1. Ion transport model in the membrane

The flux of ion i through an RO membrane consists of three parts: (1) convective flux in connection with the convective motion of water; (2) diffusive flux stemming from concentration gradients; and (3) electromigration flux resulting from the spontaneously arising electric field [6,7]. At the steady state, because there are no sources or sinks of salt ions, the fluxes are constant and given by:

$$J_i = a_i c_i \upsilon - D_i (c_i' + z_i c_i \varphi') = \text{constant}, \quad i = 1, \dots, N$$
(1)

where,  $J_i$  is the flux of ion i; v is the convectional velocity;  $\alpha_i$  is the factor that accounts for deviation of ion convective velocities from the solution convective velocity [8,9];  $D_i$  is the ion diffusion coefficient of ion i;  $\varphi(\chi)$  is the spontaneously arising electric potential (dimensionless,  $\varphi=\phi F/RT$ , where  $\phi$  is the potential in volts, and F, R, and T are Faraday number, gas constant, and absolute temperature, respectively); and  $z_i$  is the charge of ion i. Finally, the prime indicates the differentiation with respect to position (d/dx).

If we introduce dimensionless flux, j = J/v; Peclet number, Pe = vh/D, where D is the characteristic diffusion coefficient and h is the thickness of the membrane layer; dimensionless diffusion coefficients,  $d_i = D_i/D$ ; dimensionless position coordinate, y = x/h; and dimensionless electric field,  $E = (d\varphi/dy)/Pe$ , Eq. (1) can be rewritten in a dimensionless form as follows:

$$j_i = \alpha_i c_i - d_i (c_i'/Pe + z_i c_i E)$$
<sup>(2)</sup>

where the prime indicates the differentiation with respect to dimensionless position (d/dy).

In the solution, or in the membrane material, a given volume of material must be electroneutral and N different types of ions are present. The condition of electroneutrality results in:

$$\sum_{1}^{N} (z_i c_i + r_i) = 0$$
(3)

where  $r_i$  is the fixed charge density in the membrane, which is 0 in bulk solutions (e.g., in the feed and permeate). There are two methods to estimate the fixed charge density in the membrane: the analytical approach using selected membrane values [10,11], and the calculation approach using experimental results, i.e., measured ion retentions at a certain flux [12]. In this study, the latter method is used to estimate  $r_i$  to realistically incorporate actual measured data into the model. Experimental measurements of the fixed charge density in a polyamide membrane have shown that the isoelectric point of charged groups is at pH 3–4 [11], which means above pH 4 the membrane is negatively charged and below pH 4 it is positively charged.

Although the motion of an individual charged ion creates an electrical current, the summation of all ion fluxes must be 0 if the feed and permeate are electrically isolated. This condition results in:

$$\sum_{1}^{N} z_i j_i = 0 \tag{4}$$

#### 2.2. Theory for the single-layer membrane

Fig. 1 schematically explains the RO rejection process at the single layer membrane. By examining the behavior of rejection and potential, it is seen that the concentrations tend to be asymptotic at high Peclet numbers (Pe > 1). For the working conditions of experiments, the data approach the asymptotic behavior expected under high Peclet number conditions. Thus, for the remainder of this work, the high Peclet number approximation is used. A large Pe in the membrane results from high water-velocity through the membrane, which would tend to increase the concentration polarization.

In the experiments, even though the concentration polarization is minimized using high cross-flow velocities, its effect



Fig. 1 – Schematic representation of RO process in different zones: (0) for feed solution; (1) for concentration polarization; (2) for RO membrane; and (3) for permeate. RO, reverse osmosis.

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