

Evaluation study on properties of a macroporous silica-based CMPO extraction resin to be used for minor actinides separation from high level liquid waste

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Abstract Basic properties of a silica-based octyl(phenyl)-N,N-diisobutylcarbamoyl-methylphosphine oxide (CMPO) extraction resin (CMPO/SiO₂-P) was investigated. Adsorption behavior for some rare earth elements (RE) which are constituents of high level liquid waste (HLLW) and the long-term stability of the extraction resin in nitric acid solution were examined. The CMPO extraction resin was significantly stable in 3 mol·L⁻¹ HNO₃ solution at 50°C. Furthermore, the RE(III) were efficiently separated from non-adsorptive fission product (FP) elements such as Sr(II) in a column experiment using a highly nitric acid solution. The separation behaviors of the elements are considered to result from the difference in their adsorption and elution selectivity based on the complex formation with CMPO. There was no strong dependency of RE(III) separation efficiency on feed solution flow rate. Only from the perspectives of the acid-resistant behavior of CMPO extraction resin and the elution kinetics for the metal ions with the extraction resin, the CMPO extraction resin can be used in the modified MAREC process for HLLW partitioning.

Key words High level liquid waste (HLLW), Minor actinide (MA), Rare earth (RE), Partitioning, Extraction chromatography, Silica-based CMPO extraction resin, Stability.

1 Introduction

As a final disposal method for the high level liquid waste (HLLW) generated by spent fuel reprocessing, the geological disposal concept of vitrified HLLW has been proposed and investigated worldwide. However, to minimize the long-term radiological risk and facilitate the management of HLLW, a partitioning of the long-lived minor actinides (MA=Am, Cm) and some specific fission products (FP) such as Cs, Sr, Tc and the platinum group elements is much more desirable. For this purpose, a number of partitioning processes (e.g., TALSPEAK^[1], TRUEX^[2], DIAMEX^[3], TRPO^[4], DIDPA^[5], SANEX^[6], CSEX^[7] and SREX^[8]) mostly by liquid-liquid extraction using various conventional or newly synthesized extractants have been developed^[9,10].

Among the extractants used in these processes, a bifunctional organophosphorous compound named CMPO, i.e. octyl(phenyl)-N,N-diisobutylcarbamoyl-methylphosphine oxide, is well known to be able to extract effectively many kinds of actinide ions from an aqueous solution containing concentrated nitric acid. Furthermore, since CMPO shows fairly excellent chemical stability in HNO₃ solution and is a commercially available product, it has been extensively studied for the actinides partitioning from HLLW^[9,11-13]. However, liquid-liquid extraction process will generate a great amount of organic waste resulted from the hydrolytic and radiolytic degradation of the solvent and diluent. In addition, many large-scale equipments are required for the multi-stage extraction, stripping and solvent-washing processes.

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Received date: 2010-10-10

These will affect the economic performance of the process. Compared to uranium and plutonium, the minor actinides are significantly less abundant in the spent fuel, so the scale of the separation process for minor actinides from HLLW should be considerably smaller than that of a main separation process such as PUREX^[10].

As an alternative technology of the liquid-liquid extraction process, we have developed a partitioning process named MAREC (Minor Actinides Recovery from HLLW by Extraction Chromatography) process^[14-16]. Compared to liquid-liquid extraction, extraction chromatography technology is advantageous in terms of (1) a minimal organic solvent utilization and less waste accumulation, (2) compact equipment, and (3) simultaneous separation of multi-components in a unit-process operation. Therefore, the backdrops in liquid-liquid extraction process mentioned above are expected to be overcome by this technology.

In the improved MAREC process as shown in Fig.1, two main columns packed with different extraction resins are utilized for the chromatographic separation of HLLW through selective adsorption and elution procedures. The first column is packed with CMPO/SiO₂-P extraction resin and the second with R-BTP/SiO₂-P (R-BTP: 2,6-bis-(5,6-dialkyl-1,2,4-triazine-3-yl) pyridine, R: alkyl group) extraction resin. The elements can be effectively separated into the following three groups in the first column depending on their different adsorption and elution behavior which results from the complexation ability of the metal ions with CMPO: (1) Cs, Sr, Rh and Ru (non-adsorptive or weakly adsorptive FPs); (2) MA and RE (rare earth elements); (3) Zr and Mo. Subsequently, the MA containing effluent is applied to the second column where the elements are separated to (1) RE, and (2) MA, respectively.

In this work, the basic properties of CMPO/SiO₂-P extraction resin were studied by using highly acidic solution containing several metal ions which are constituents of HLLW. Adsorption behavior for some RE(III) and long-term stability of the extraction resin in HNO₃ solution were examined. Furthermore, column separation experiments were carried out at different flow rate using a acidic solution

containing some RE(III) and Sr(II) in 3 mol·L⁻¹ HNO₃ to evaluate the separation behavior of the elements.

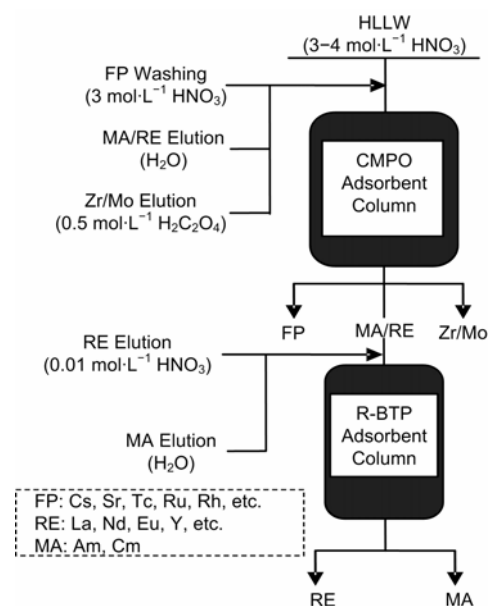


Fig.1 Conceptual flowchart of the improved MAREC partitioning process of HLLW by extraction chromatography.

2 Experimental

2.1 Materials

RE(NO₃)₃·xH₂O (RE = Nd, Eu, Gd, Dy and Y) and Sr(NO₃)₂ were of commercial reagents of analytical grade.

The purity of CMPO reagent used in this work was 99.2%. CMPO/SiO₂-P extraction resin was synthesized by impregnating CMPO into the pores of the silica/polymer composite support (SiO₂-P), which was developed in our previous work^[14,15]. SiO₂-P support contains a macroporous styrene-divinylbenzene (SDB) copolymer which is immobilized in porous silica particles with pore size of 0.6 μm and mean diameter of 60 μm. The prepared CMPO/SiO₂-P extraction resin contains 0.5 g of CMPO in 1.0 g of SiO₂-P. The chemical structure of CMPO and photographs of CMPO/SiO₂-P have been shown in the previous papers^[14,15].

Thermal decomposition properties of SiO₂-P and CMPO/SiO₂-P were measured using a TG-DTA (thermogravimetry-differential thermal analysis) apparatus (Shimadzu DTG-60) by heating from ambient temperature up to 600°C at a heating rate of 1°C·min⁻¹ in a 30 cm³·min⁻¹ flow of oxygen.

2.2 Batch adsorption

In a batch adsorption experiment, a weighed amount of CMPO/SiO₂-P (typically 0.1 g) was combined in a glass vial with Teflon stopper with a measured volume (typically 5 cm³) of 3 mol·L⁻¹ HNO₃ solution containing ~1×10⁻³ mol·L⁻¹ of RE(III). The glass vial was maintained in a thermostatic water bath shaker and shaken mechanically at 160 rpm for a determined adsorption time. The solution was separated from the extraction resin by vacuum filtration and sampled for analysis. The concentration of the metal ions and phosphorus leaked from CMPO/SiO₂-P into aqueous solution were analyzed by ICP-AES (Shimadzu ICPS-7510). The concentration of organic carbon leaked from CMPO/SiO₂-P was determined with a TOC (total organic carbon) analyzer (Shimadzu TOC-V_{CPN}). The corresponding content of CMPO contained in the aqueous solution was then calculated based on the concentration of C and P. The distribution coefficient (K_d) of the metal ions was calculated by Eq.(1) as below:

$$K_d(\text{cm}^3/\text{g}) = \frac{C_0 - C_s}{C_s} \cdot \frac{V}{W_R} \quad (1)$$

where C_0 and C_s denote the metal concentration in the aqueous phase before and after adsorption, respectively. V indicates the volume of the aqueous phase (cm³) and W_R the weight of the dry extraction resin (g).

2.3 Column separation

The schematic diagram of the column apparatus for chromatographic separation experiments has been shown in our previous papers^[14]. About 25 g of the dry CMPO/SiO₂-P extraction resin was packed into a Pyrex glass column with a dimension of 1 cm in inner-diameter and 50 cm in length, and then equilibrated with 5 BV (bed volume) of 3.0 mol·L⁻¹ HNO₃ prior to the separation experiment. The experiment was performed at 50°C by circulating thermostatic water through the jacket of the column.

After the sample solution containing ~5×10⁻³ mol·L⁻¹ of Sr(II), Y(III), Nd(III), Eu(III), Gd(III), Dy(III) and 3 mol·L⁻¹ HNO₃ was passed through the

column, the given volume of 3 mol·L⁻¹ HNO₃ (as washing solution), water (as eluent) were subsequently pumped through the column successively. The flow rate was adjusted to 1 cm³·min⁻¹ (0.76 m·h⁻¹) or 8 cm³·min⁻¹ (6.11 m·h⁻¹) using a metering pump. During the passage of all the solutions, the pressure drop inside the column was detected to be less than 0.1 MPa. The effluents from the column were collected by an auto-fractional collector in 5 cm³ aliquot. The concentrations of the metal ions in each aliquot were analyzed by ICP-AES. The pH value in each fractional solution was also measured.

3 Results and discussion

3.1 Thermal analysis of SiO₂-P and CMPO/SiO₂-P

The TGA-DTA curves of SiO₂-P and CMPO/SiO₂-P are shown in Figs.2a and 2b, respectively. As can be seen in Fig.2a, the DTA curve of SiO₂-P shows a sharp exothermic peak at 258°C which is interpreted as the burning of the SDB copolymer. The second and third flattened peaks at 348°C and 417°C observed on the diagram are presumed to correspond to a segmental mobility of crosslinked copolymer parts. The SiO₂-P finally lost 16% of its original weight by heating up to 600°C, indicating that it consists of 16 % of the SDB copolymer.

The DTA curve of CMPO/SiO₂-P in Fig.2b shows one sharp endothermic peak at 40.4°C and three sharp exothermic peaks at 173°C, 290°C and 425°C, respectively. The endothermic peak is interpreted as the melting of CMPO which is in accordance with the result of TGA-DTA measured for CMPO reagent^[17]. The first exothermic peak is interpreted as decomposition of the CMPO and the last two peaks denote burning of the SDB copolymer. The TGA result shown in Fig.2b indicates that CMPO/SiO₂-P lost 44% of its original weight finally. From these results, the mass composition of CMPO/SiO₂-P extraction resin is determined to be 33.3% of CMPO, 10.7% of SDB polymer and 56.0% of SiO₂, respectively. These values are in a good agreement with the initiative constitution of the materials used in the synthesis.

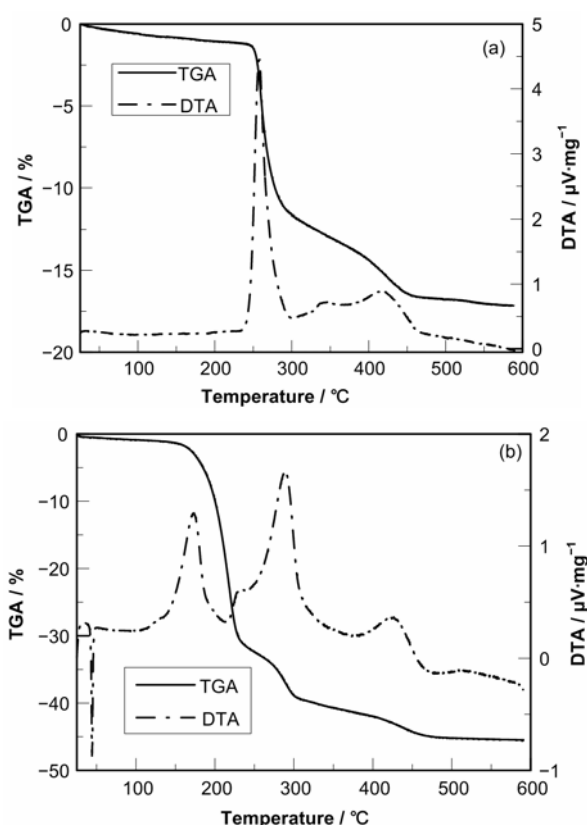


Fig.2 TGA-DTA curves of SiO₂-P (a) and CMPO/SiO₂-P (b) (Mass: 20 mg; Atmosphere: O₂; Flow rate: 30 cm³·min⁻¹; Heating rate: 1°C·min⁻¹).

3.2 Effect of temperature on the adsorption of RE(III)

In partitioning of long-lived radionuclides by extraction chromatography, the quick loading and elution dynamics is advantageous for a practical process. To understand the adsorption rate, the adsorption of a few representative elements Nd(III), Eu(III) and Y(III) towards CMPO/SiO₂-P resin in 3 mol·L⁻¹ HNO₃ solution was examined at 25°C and 50°C, respectively. The results are shown in Fig.3. As can be seen in Fig.3a, Nd(III), Eu(III) and Y(III) showed very strong adsorption onto CMPO/SiO₂-P and the adsorption almost reached an equilibrium at around 30 min at 25°C, though the K_d still increased gradually in the following hours of the test. The equilibrium distribution coefficients (K_d) were over 500 cm³/g for Nd (III) and Eu(III), and higher than 160 cm³/g for Y(III). Such a high value of K_d is due to the considerably strong complex reaction between these metal ions and CMPO/SiO₂-P. It might originate from the high affinity of oxygen donor contained in CMPO molecular towards the rare earth metal ions. In

comparing with 25°C, the adsorption of Nd(III), Eu(III) and Y(III) at 50°C was much rapid and attained completely to an equilibrium state within 10 min, though the K_d values at 50°C were slightly lower than those at 25°C. Since the melting point of CMPO is around 40°C according to the result of TG-DTA as shown in Fig.2b, the significantly fast adsorption rate exhibited at 50°C may be attributed to the phase transformation, but a detailed study is needed to be investigated further. These results indicate that temperature has an obvious effect on the adsorption rate of the elements onto CMPO/SiO₂-P extraction resin and rapid loading and elution dynamics for the elemental group separation can be easily obtained at 50°C.

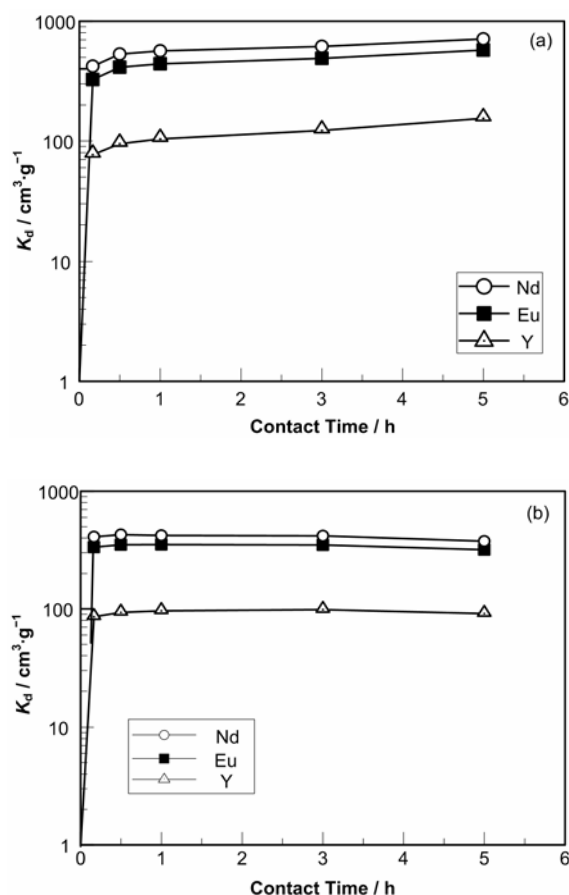


Fig.3 Effect of contact time on the adsorption of RE(III) onto CMPO/SiO₂-P at 25°C (a) and 50°C (b) ([HNO₃] = 3 mol·L⁻¹; [RE(III)] = 1×10⁻³ mol·L⁻¹; Phase ratio: 0.1g/5cm³; Shaking speed: 160 rpm).

3.3 Long-term stability of CMPO/SiO₂-P in HNO₃ solution

The HLLW produced in a current spent fuel reprocessing process such as Purex and its modified

processes, generally contains around $3\text{--}4\text{ mol}\cdot\text{L}^{-1}$ of HNO_3 ^[10]. To understand the resistant properties of CMPO/SiO₂-P resin against such highly acidic solution, adsorbability of the resin for Y(III), Nd(III) and Gd(III) during a long term contacting with $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 solution at 50°C was examined according to the corresponding condition of the MAREC process. The time evolution of distribution coefficients is shown in Fig.4. The analytical results of CMPO leaked from the CMPO/SiO₂-P into the HNO_3 solution are presented in Fig.5. As shown in Fig.4, the adsorbability of Y(III), Nd(III) and Gd(III) on CMPO/SiO₂-P resin kept almost the same within 30 days of contacting with $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 . In addition, the concentration of CMPO leaked into the aqueous solution, which was calculated from the quantitative analysis of P and TOC, was steady during the contacted period of time (30 days) and the values were as low as $30\text{--}60\text{ mg}\cdot\text{L}^{-1}$. It means only $0.4\%\text{--}0.8\%$ CMPO in CMPO/SiO₂-P leaked from the extraction resin. This concentration was similar to the solubility of CMPO/SiO₂-P in the corresponding acidity solutions, showing that the effect of HNO_3 on the leakage of CMPO from CMPO/SiO₂-P at 50°C was not significant^[17,18]. Due to the leaked CMPO/SiO₂-P in $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 was consistent with its solubility, namely, almost no CMPO destroyed by HNO_3 and leaked out, showing a superior stability of the CMPO/SiO₂-P extraction resin in $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 medium in a relatively long term at elevated temperature.

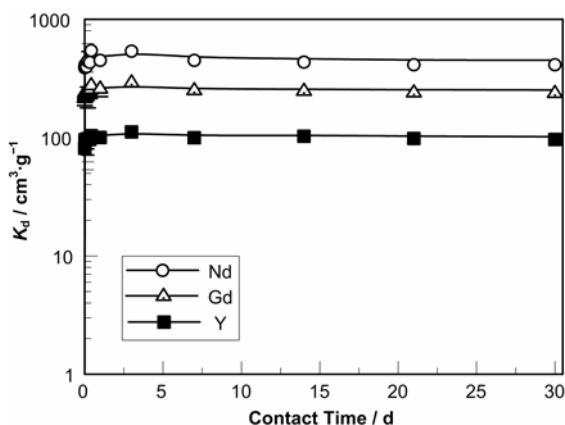


Fig.4 Effect of long term contacting with HNO_3 solution on the adsorption of RE(III) onto CMPO/SiO₂-P at 50°C ($[\text{HNO}_3]=3\text{ mol}\cdot\text{L}^{-1}$; $[\text{RE(III)}]=1\times 10^{-3}\text{ mol}\cdot\text{L}^{-1}$; Phase ratio: $0.1\text{g}/5\text{cm}^3$; Shaking speed: 160 rpm).

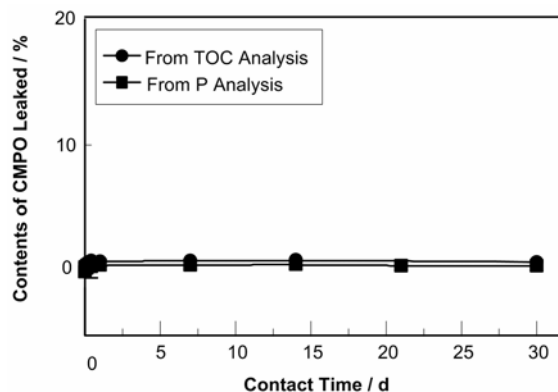


Fig.5 Leakage of CMPO from CMPO/SiO₂-P with a change in contact time in $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 at 50°C (Phase ratio: $0.1\text{g}/5\text{cm}^3$; $[\text{RE(III)}]=1\times 10^{-3}\text{ mol}\cdot\text{L}^{-1}$; Shaking speed: 160 rpm).

3.4 Separation performance of RE(III) in a highly acidic solution

The above experimental results show that in $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 medium, the adsorption rates of the tested rare earth metal ions towards CMPO/SiO₂-P at 50°C were much quicker than at 25°C . Also, the quantity of CMPO leaked from CMPO/SiO₂-P at 50°C was rather low. Based on these results, separation experiments for a highly acidic solution containing some typical fission product rare earths and Sr in $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 using a column packed with CMPO/SiO₂-P were performed at different flow rates. The elution curves of the tested elements and the corresponding pH value in the effluents from the column at flow rates of $1\text{ cm}^3\cdot\text{min}^{-1}$ ($0.76\text{ m}\cdot\text{h}^{-1}$) and $8\text{ cm}^3\cdot\text{min}^{-1}$ ($6.11\text{ m}\cdot\text{h}^{-1}$) are illustrated in Figs.6 and 7, respectively.

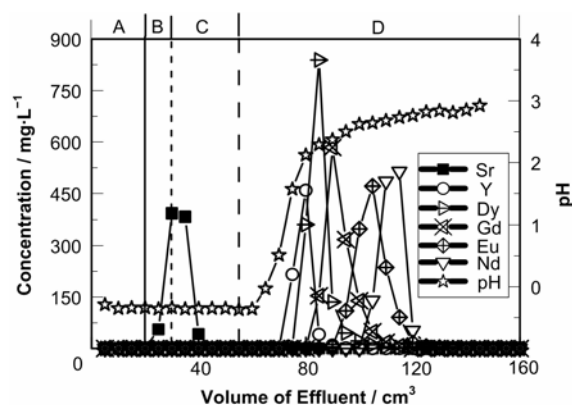


Fig.6 Results of separation experiment for a highly acidic solution containing $\sim 5.0\times 10^{-3}\text{ mol}\cdot\text{L}^{-1}$ of Sr(II), RE(III) and $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 at flow rate of $1\text{ cm}^3\cdot\text{min}^{-1}$ at 50°C (Column: $\phi 10\text{ mm}\times 500\text{ mm}$). (A) Dead volume; (B) Sample solution; (C) $3\text{ mol}\cdot\text{L}^{-1}$ HNO_3 ; (D) H_2O .

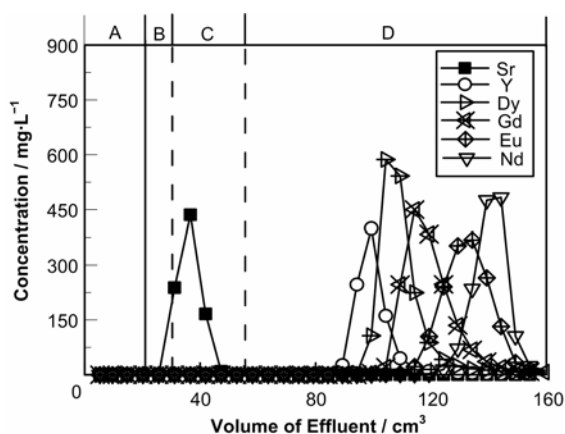
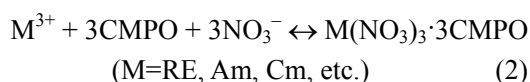
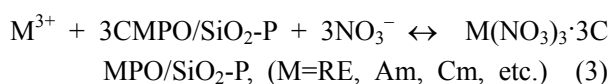


Fig.7 Results of separation experiment for a highly acidic solution containing $\sim 5.0 \times 10^{-3} \text{ mol}\cdot\text{L}^{-1}$ of Sr(II), RE(III) and 3 M HNO_3 at flow rate of $8 \text{ cm}^3\cdot\text{min}^{-1}$ at 50°C (Column: $\Phi 10 \text{ mm} \times 500 \text{ mm}$). (A) Dead volume; (B) Sample solution; (C) $3 \text{ mol}\cdot\text{L}^{-1} \text{ HNO}_3$; (D) H_2O .

Sr(II) showed no adsorption towards the CMPO/SiO₂-P extraction resin and leaked from the column quickly along with the sample solution and washing solution ($3 \text{ mol}\cdot\text{L}^{-1} \text{ HNO}_3$), representing the behavior of non-adsorptive fission products in HLLW. CMPO agent is a bifunctional organophosphorus compound which contains two “hard donor” oxygen atoms. It is well known that CMPO could extract effectively rare earths in HNO_3 medium according to the following Eq.(2)^[19]:



So, in the extraction chromatographic separation experiments, RE(III) were adsorbed strongly by the CMPO/SiO₂-P resin mostly as their neutral nitrate-complexes through the reaction of Eq.(3)^[14], which is basically the same with Eq.(2). The RE(III) adsorbed on the CMPO/SiO₂-P resin can be eluted out effectively by H_2O , because with supplying H_2O to the column, the NO_3^- concentration in the resin bed will be decreased and this leads the dissociation of the metal complexes according to the following dynamic interconversion reaction^[14,16].



As illustrated in Figs.6 and 7, with a successive supply of H_2O as an eluent to the column, the RE(III) adsorbed on the CMPO/SiO₂-P were efficiently eluted

out of the column, showing an elution order in the elution curves as: Y(III), Dy(III), Gd(III), Eu(III), Nd(III), i.e., the heavy RE(III) including Y(III) which always shows similar chemical behavior with heavy RE(III) firstly appeared in effluent and then the light ones. The elution order appearing in effluent is considered to result from the different stability of the complexes of these metals with CMPO.

The pH value in effluent was determined and its range was found within $-0.5 \sim 3$. With the supply of water as an eluent to the column, the pH value in effluent increased rapidly to around pH 2 and then gradually increased to about 3. From the results in Fig.6, the pH value in the effluent of MA/RE is estimated at around 2.5, which is advantageously suitable for the succeeding MA/RE separation process using R-BTP/SiO₂-P extraction resin without additional acidity adjustment^[20,21].

As illustrated in Fig.6 with flow rate of $1 \text{ cm}^3\cdot\text{min}^{-1}$, the elution curves of the adsorbed elements are in sharp peaks with very slight tailing, indicating that the elution kinetics was fairly fast. To examine the effect of flow rate of feed solution on the separation performance, a separation experiment was conducted at the flow rate of $8 \text{ cm}^3\cdot\text{min}^{-1}$, with the other conditions being identical to the experiment of Fig.6. Fig.7 shows the results of this chromatographic separation experiment. Compared to the elution curves in Fig.6, all the elements have less sharp elution peaks with little longer tails, but essentially elution performance of all the elements did not change. The water for all RE(III) elution was 75 cm^3 at flow rate of $8 \text{ cm}^3\cdot\text{min}^{-1}$. This is only 1.4 times of that at $1 \text{ cm}^3\cdot\text{min}^{-1}$. The results indicate that the effect of flow rate on the separation performance and elution rate is insignificant, exhibiting fast adsorption and elution kinetics for the metal ions with the CMPO/SiO₂-P. Therefore, the CMPO/SiO₂-P material is applicable for scaling up the separation process which can adopt a relatively fast flow rate. Furthermore, operating at a high flow rate of feed solution can shorten the contacting time between HLLW as the feed solution and the resin phase and hence decrease hydrolytic and radiolytic decomposition of the extraction resin.

4 Conclusion

The adsorption of trivalent rare earths (RE(III)) from nitric acid solution by CMPO/SiO₂-P extraction resin was strong and very rapid. The adsorption reached an equilibrium state in less than 10 min at 50°C. The adsorbability of RE(III) kept unchanged after contacting with 3 mol·L⁻¹ HNO₃ for 30 days at 50°C and the leakage of CMPO was as low as 30–60 mg·L⁻¹, indicating the extraction resin has a superior stability in relatively long term. In chromatographic experiment using a CMPO/SiO₂-P packed column, RE(III) was completely separated from the non-adsorptive FP such as Sr(II), and the effect of flow rate on the separation performance was insignificant. The extraction resin is applicable for scaling up the separation process by adopting a relatively high flow rate. Only from the perspectives of the acid-resistant behavior of CMPO extraction resin and the elution kinetics for the metal ions with the resin, the first cycle using the column packed with CMPO/SiO₂-P extraction resin in the improved MAREC process for MA partitioning from HLLW is essentially feasible, although further investigations to examine the behavior of more fission products such as Cs, Tc, Zr, Nb and Ru and some actinides such as Am, Cm, U, Pu and Np and “hot tests” using practical HLLW are needed. In addition, radiation resistance of the CMPO/SiO₂-P extraction resin needs to be investigated.

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