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ScienceDirect

Energy Procedia 131 (2017) 163–169

Energy Procedia

www.elsevier.com/locate/procedia

5th International Symposium on Innovative Nuclear Energy Systems, INES-5, 31 October – 2 November, 2016, Ookayama Campus, Tokyo Institute of Technology, JAPAN

Separation and recovery of palladium from nitric acid solution by silica based benzo-15-crown-5 ether resin

Atsunori Terashima[a,*], Toshitaka Kaneshiki[b], Masao Nomura[b], Masaki Ozawa[b]

[a]*Department of Nuclear Engineering, Graduate School of Science and Engineering, Tokyo Institute of Technology, 2-12-1-N1-9 Ookayama, Meguro-ku, Tokyo 152-8550, Japan*

[b]*Laboratory for Advanced Nuclear Energy, Institute of Inovative Research, Tokyo Institute of Technology, 2-12-1-N1-9 Ookayama, Meguro-ku, Tokyo 152-8550, Japan*

Abstract

The Aprés ORIENT research program, as a newly concept of advanced nuclear fuel cycle, was initiated in FY2011 aiming at creating stable, highly-valuable elements by nuclear transmutation from fission products (FPs). As creation of palladium (Pd) from rhodium (Rh) in FPs is one of the most important targets of the program, the research and development of highly effective separation methods between Rh and Pd are strongly required. In the present paper, an ability of silica based benzo-15-crown-5 ether (B15C5) resin for adsorption and elution of Ru(III), Rh(II), and Pd(II) was investigated in nitric acid (HNO_3) solution. As a result of the batch experiment for adsorption, Pd(II) were strongly adsorbed by B15C5 resin in HNO_3. On the other hand, Ru(III) and Rh(III) were not adsorbed by the resin. As it was difficult to elute adsorbed Pd(II) by the change of the concentration of HNO_3, ethylenediamine (EDA) was employed as an eluent for Pd(II). As a result of the column chromatography for adsorption and elution, only Ru(III) and Rh(III) were eluted in 2 mol/L of HNO_3, and then Pd(II) was clearly eluted in a mixed solution including 2 mol/L of HNO_3 and 2 mol/L of EDA. Therefore, by using EDA as the eluent, the possibility of B15C5 resin for separation and recovery of Pd created by transmutation of Rh in FPs has been found.

Peer-review under responsibility of the organizing committee of the 5th International Symposium on Innovative Nuclear Energy Systems.

Keywords: Benzo-15-crown-5 ether resin; Palladium; Nitric acid; Ethylenediamine; Extraction chromatography

* Corresponding author. Tel.: +81-3-5734-2955; fax: +81-3-5734-2959.
E-mail address: terashima.a.aa@m.titech.ac.jp

1876-6102
Peer-review under responsibility of the organizing committee of the 5th International Symposium on Innovative Nuclear Energy Systems.
10.1016/j.egypro.2017.09.423

1. Introduction

The Après ORIENT research program, which is a newly concept of advanced nuclear fuel cycle, aims at creating stable and highly-valuable elements by nuclear transmutation of fission products (FPs) separated from high level liquid waste [1]. Especially, creation of palladium (Pd) from rhodium (Rh) in FPs is one of the most important processes considered by the program because of the high efficiency of creation, the low radioactivity of product, and the high value as resources [2,3]. Then, in order to realize the advanced recycle system for Pd, the research and development of highly effective separation methods between Rh and Pd are strongly required.

It is well known that various crown ethers have an ability of forming complexes with specific metal ions [4]. Previously, our group had synthesized benzo-15-crown-5 ether (B15C5) resin supported on the surface of silica beads for isotope separation of lithium and zinc [5,6]. Then, not surprisingly, the B15C5 resin has a possibility of not only isotope- but also element-separation. Recently, the adsorptivity of various metal ions onto other crown ether resins was investigated [7]. Thus, in the present study, the performance of B15C5 resin especially for adsorption and elution of Pd was investigated in nitric acid (HNO_3) solution.

2. Experiments

B15C5 resin was synthesized in accordance with the scheme shown in Fig. 1. The detailed condition of the synthesis was same to the previous paper [6]. As a porous silica support, MST-8C (Mizusawa Industrial Chemicals, Ltd.) was used. Then, B15C5 resin was used in two batch experiments and one column chromatography to investigate the performance for adsorption and elution of Ru(III), Rh(III), Pd(II) from HNO_3 media.

Firstly, a batch experiment for adsorption was carried out. Sample solutions including 10 ppm of Ru(III), Rh(III), and Pd(II) in 0.1-9.0 mol/L (=M) of HNO_3 were prepared by diluting AAS standard solutions. After 2.0 g of B15C5 resin and 5.0 mL of each sample solution were added in a glass vial, the vial was shaken at 160 rpm for 24 hour at 25 °C. The concentrations of the metal ion before and after adsorption were measured by using ICP-MS. The performance of adsorption was evaluated by using the distribution coefficient K_d defined as

$$K_d = \frac{C_0 - C_A}{C_A} \times \frac{V}{W} \ [\mathrm{cm^3/g}] \tag{1}$$

where C_0 [M] is the initial concentration of the metal ion, C_A [M] is the concentration of the metal ion in the aqueous phase after shaking, V [cm^3] is the volume of the aqueous phase, and W [g] is the weight of the resin including silica beads. If the K_d value is higher than 1, it is roughly considered that the resin has the ability to adsorb the metal ion.

Secondly, another batch experiment for elution was also done. After 1.0 g of B15C5 resin and 2.5 mL of a sample solution (10 ppm of Pd(II) in 2.0 M of HNO_3) were added in a glass vial, the vial was shaken at 160 rpm for 12 hour at 25 °C. Then, after removing 1.0 mL from the supernatant liquid, 1.0 mL of a mixed solution including HNO_3 and ethylenediamine (EDA) was added. It should be noted that the concentration of HNO_3 was not changed by this process, and the concentration of EDA after adding the mixed solution was 0.0-2.0 M. Then, the vial was shaken again at 160 rpm for 12 hour at 25 °C. The concentrations of Pd(II) before and after adsorption and after

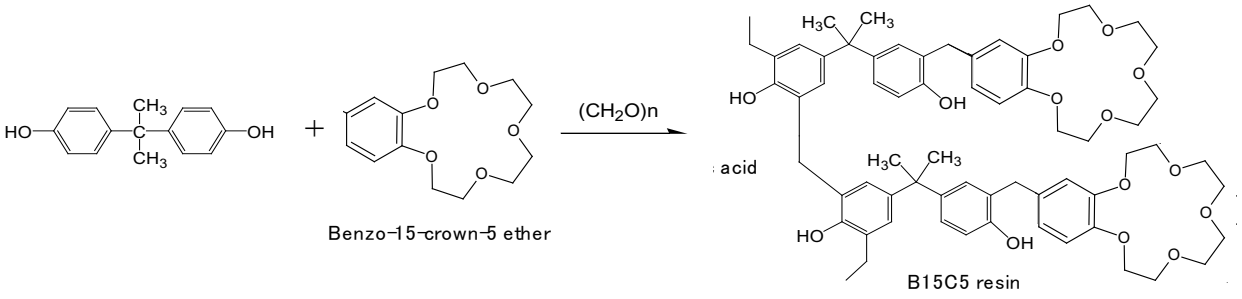

Fig. 1. Scheme of synthesis of B15C5 resin.