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**INTRODUCTION:** Technetium with atomic number 43 is an artificial radioactive element that is not found in nature, and it is used as a tracer in medicine and environmental analysis. In particular, technetium-95m (half-life: 61 days) and technetium-99m (half-life: 6 hours) are frequently used as tracers. Because commercially available technetium-95m is isolated from fission products, the influence of other technetium isotopes cannot be avoided. Technetium-99m, however, is obtained as a decay product of molybdenum-99, and it does not sustain any effects from other technetium isotopes.

Technetium-99m, therefore, is superior as a tracer with its higher isotopic purity compared to technetium-95m. Nonetheless, the supply of technetium-99m from overseas has recently been unstable. Consequently, a variety of alternative efforts have been examined in Japan, such as the study of production methods using accelerators.

The present research employed molybdenum metal to production molybdenum-99/technetium-99m through reactor neutron irradiation. The handling of molybdenum metal is easy, but the generated specific radioactivity is low. Thus, methods for efficient extraction of technetium from molybdenum must be studied, and the usability of the technetium extracted as a tracer must be confirmed.

**EXPERIMENTS:** Technetium-99m was determined to be obtained through the production and subsequent decay of its parent nuclide, molybdenum-99, as a decay product. Approximately 50~100 mg of natural molybdenum metal foil (0.05 mm in thickness) was used for the production of molybdenum-99. The foil were irradiated in a Pn-2 radiation tube of the Kyoto University Research Reactor for 240 minutes at 1 MW or 50minutes at 5MW. Upon cooling for approximately 2 hours after irradiation, the foil sample was heated and dissolved in aqueous hydrogen peroxide. The solution was left to cool, followed by adsorption to an Empore filter [1,2] (made by Sumitomo 3M Ltd.), and finally elution from the filter by varying the nitric acid concentration from 1 N to 8 N.

The eluent was subjected to gamma-ray analysis at 181 keV emitted from molybdenum-99 and at 142.7 keV emitted from technetium-99m with a well-type HP-Ge detector to determine the filter adsorption and elution characteristics. The same chemical process was employed respectively for the anion-exchanged resin, activated alumina, and TEVA resin[3] as adsorbents for a column volume of 2 ml. Filter adsorption and elution characteristics for these were also determined.

**SUMMARY:** To date, the irradiation was performed on seven times, and the chemical separation on ten times experiments. Although the results are preliminary, the experimental conditions enabled the production of the requisite amount of radioactivity and decontamination of isotopes for a tracer. Experimentation work will continue in future for chemical separation in order to improve reproducibility and to determine the optimum separation conditions.

Based on the current data, chemical separation employing the TEVA resin and anion-exchanged resin yielded better results for separation and recovery rates, in comparison to the activated alumina and Empore filter.

The obtained technetium-99m as a tracer is planned for use in determining technetium-99 recovery yield from respective samples of tap water, sea water, soil, and sea weed. Quantitative analysis of the separated and collected technetium-99 is planning to use the liquid scintillation counter and ICP-MS.

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**INTRODUCTION:** It has been revealed that polyvinylpyrrolidone (PVPP), which is one of cyclic monoamide resins and has a 5-membered pyrrolidone ring, changes the chemical structure by  $\gamma$ -ray irradiation in  $\text{HNO}_3$  as shown in Fig. 1. It has also been found that the functional group (a) in Fig. 1 has uncommon chemical structures with multiple coordinative sites and is relatively stable under irradiation [1]. This means that it may be used as a novel type of chelating resin, particularly in acidic media, originated from the multiple C=O groups in the structure. In fact, the irradiated PVPP has been found to show adsorptivity to many kinds of metal ions in  $\text{HNO}_3$  of lower concentration range, and also to, *e. g.*, Zr(IV), Mo(VI), and Pd(II) from  $\text{HNO}_3$  of higher concentration range [2]. The above results indicate that a longer chain with more flexibility for capturing metal ions may be obtained by irradiation to monoamide rings with more members. In this study, therefore, another cyclic monoamide resin consisting of poly-*N*-vinyl- $\epsilon$ -caprolactam (VEC), which has a 7-membered ring, was investigated.

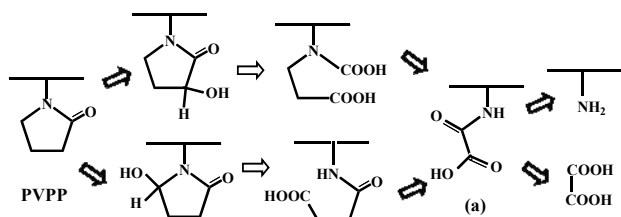


Fig. 1. Expected major route of change in chemical structure of PVPP by  $\gamma$ -ray irradiation in  $\text{HNO}_3$ .

**EXPERIMENTS:** VEC synthesized by bulk polymerization followed by grinding and sieving between 63 and 150  $\mu\text{m}$  [2] was used as the sample for irradiation. Similarly to the earlier work [3], VEC and  $\text{HNO}_3$  (6 mol/dm<sup>3</sup> (= M)) were mixed in a Pyrex sample tube, and the mixture was irradiated by the <sup>60</sup>Co source at max. 10.7 kGy/h up to 1.51 MGy at room temperature under ambient atmosphere. Adsorptivities of the irradiated VEC to various metal ions at equilibrium were examined by the batch method. Samples of the conditioned adsorbent (wet 0.15 g) and 3 cm<sup>3</sup> of 0.1 - 6 M  $\text{HNO}_3$  solutions containing a metal ion (1 mM each) were shaken at 298 K in a thermostatic shaking bath for 24 h. Adsorptivities were evaluated by the distribution ratio,  $K_d$ , defined as,

$$K_d = \frac{C_0 - C}{C} \times \frac{V}{W} \quad (\text{cm}^3/\text{g}) \quad (1)$$

where  $C_0$  and  $C$  denote the concentrations of the nuclide

in the solution before and after contact with the adsorbent, respectively.  $V$  and  $W$  represent the volume of the solution and the weight of the dry adsorbent, respectively.

**RESULTS:** The  $K_d$  values for various metal ions as a function of  $\text{HNO}_3$  concentration for VEC irradiated at 1.51 MGy are shown with those for neat VEC in Fig. 2. While only Re(VII), Pd(II) and Mo(VI) show adsorption in major metal ions, Zr(IV) and Nd(III) newly show relatively strong and weak adsorptivities, respectively, for the irradiated VEC. All  $K_d$  values for the irradiated VEC are found to increase with decreasing concentration of  $\text{HNO}_3$  except Nd(III). Pd(II) shows highest adsorptivity in the irradiated sample, which differs from the cases for the irradiated PVPP where Zr(IV) showed the highest.

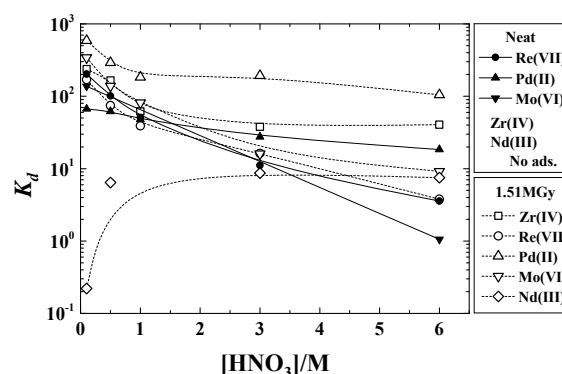


Fig. 2. Adsorptivity of VEC before and after irradiation in 6M  $\text{HNO}_3$  to various metal ions in  $\text{HNO}_3$ .

An IR spectroscopic study to VEC irradiated at 1.51 MGy revealed that there was a peak at around 1730  $\text{cm}^{-1}$  which would be attributed to carboxyl groups. Besides, interestingly enough, an adsorption at around 1830  $\text{cm}^{-1}$  observed for the irradiated PVPP which was attributed to the structure (a) in Fig. 1 was not observed for this sample. Based on the above, unlike the case for PVPP, a structure change shown in Fig. 3 may be expected for VEC irradiated in  $\text{HNO}_3$ . The reason for the difference remains unclear. Further investigations are necessary for understanding the adsorption mechanisms.

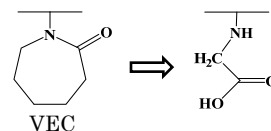


Fig. 3. Expected structure change in VEC by  $\gamma$ -ray irradiation in  $\text{HNO}_3$ .

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