

N. Miyata, M. Nogami and N. Sato

Faculty of Science and Engineering, Kinki University  
<sup>1</sup>Research Reactor Institute, Kyoto University

**INTRODUCTION:** Pyrrolidone derivatives (NRPs) are promising as selective compounds for actinoid(IV, VI) species in  $\text{HNO}_3$  media. We have been studying the stability of NRPs against  $\gamma$ -ray irradiation in  $\text{HNO}_3$  and have found that the degradation of NRPs start from the cleavage of the pyrrolidone ring by the addition of oxygen atom originating from  $\text{HNO}_3$ , followed by the formation of chain monoamides and C4 compounds by the continuous addition of oxygen[1]. These compounds have multiple coordinative oxygen atoms and it is expected that they may be used as novel types of chelating agents. In the present study, adsorptivities of polyvinylpyrrolidone (PVPP), a solid adsorbent categorized into NRPs, irradiated by  $\gamma$ -Ray in  $\text{HNO}_3$  to some metal ions was examined in  $\text{HNO}_3$  solutions.

**EXPERIMENTS:** 10 g PVPP purchased from Sigma and  $50\text{cm}^3$   $\text{HNO}_3$  (6, 9 mol/dm<sup>3</sup> (= M)) were mixed in a Pyrex sample tube and used as the samples for  $\gamma$ -ray irradiation. Irradiation by the <sup>60</sup>Co source was performed at maximum 13.0 kGy/h up to 1 MGy at room temperature under ambient atmosphere. The irradiated PVPP was separated from the supernatant liquid and washed using distilled water to avoid further degradation of generated chain monoamides by  $\text{HNO}_3$ . Adsorptivities of the irradiated PVPP to metal ions at equilibrium were obtained by a batch method solution using Mo(VI) and Nd(III). Samples of the conditioned adsorbent (wet 0.25 g) and  $5\text{cm}^3$  of 0.1 - 3 M  $\text{HNO}_3$  solutions containing 10 mM Mo(VI) or Nd(III) were shaken at 298K in a thermostatic shaking bath for 24 h. After shaking, samples of the supernatant were taken and the concentrations of Mo and Nd were measured using ICP-AES. 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 IR spectra of PVPP irradiated in 6 and 9 M  $\text{HNO}_3$  were found almost identical at the same dose, where a new peak generated at around  $1800\text{cm}^{-1}$ . The peak would be attributed to a carbonyl group existing next to a carbonyl or carboxyl group. This indicates that the degradation of PVPP proceeds to a similar extent un-

like the cases for the samples irradiated in lower concentration of  $\text{HNO}_3$ , where the degradation decreases with decreasing concentration of  $\text{HNO}_3$ [1].

Adsorptivities of PVPP irradiated in 6M  $\text{HNO}_3$  at various dose to Zr(IV) and Nd(III) are shown in Fig. 1. For Zr(IV), it can be seen that the  $K_d$  values increase with decreasing concentration of  $\text{HNO}_3$  and increasing dose. Neat PVPP has no adsorptivity to metal ions except U(VI), Re(VII), and Pd(II) in  $\text{HNO}_3$  media[2]. Conventional chelating resins such as those with iminodiacetate as the functional group have no adsorptivity to metal ions at pH = 1 (= 0.1 M  $\text{HNO}_3$ ). This means that the irradiated PVPP may be a promising resin to Zr(IV) in acidic  $\text{HNO}_3$  solution. It is highly likely that the higher adsorptivity results from the plural carbonyl groups in the irradiated PVPP. The adsorptivity to Nd(III) shows the same tendency, but the  $K_d$  values are much lower compared with those of Zr(IV). This may mean that more coordinative oxygen atoms are necessary to adsorb Nd(III).

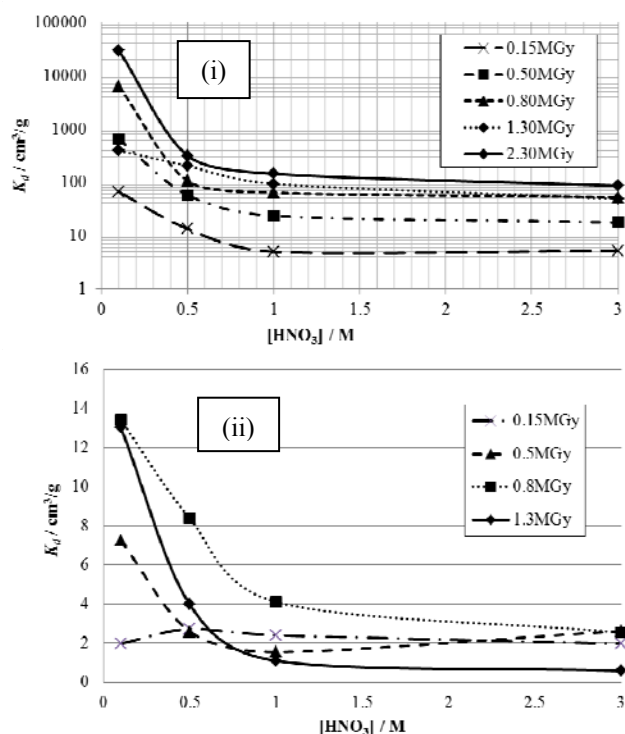


Fig. 1. Adsorptivity of PVPP irradiated in 6M  $\text{HNO}_3$  at various dose to metal ions in  $\text{HNO}_3$ ; (i) Zr(IV), (ii) Nd(III).

#### REFERENCES:

- [1] M. Nogami *et al.*, JAEA-Review 2009-041 (2008) 25.
- [2] M. Nogami *et al.*, J. Radioanal. Nucl. Chem. **284** (2010) 195.