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Stability of silica-supported monoamide resins with selectivity to U(IV) and U(VI) against γ-ray irradiation in HNO3

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In order to develop resins with selectivity to U(VI) in HNO3 media, we have synthesized several sili-ca-supported polymer beads with the structure of a monoamide as the functional group. The examination on their adsorptivities to various metal ions has clarified that among these resins those consisting of N,N-dimethyl-acrylamide (Silica-DMAA) and N-methyl-N-vinylbenzylacetamide(Silica-MVBAA) have selectivity to U(VI) and U(IV), respectively, where major fission product (FP) ions are found to show no or very little adsorption from HNO3 ranging up to 6 mol/dm3 (= M) [1, 2]. For wider applications of these resins including treatment of highly- radioactive solutions, it is necessary to investigate their stability under irradiation conditions. In this study, the above two resins were irradiated by γ-ray in HNO3 and adsorptivities to FP ions were examined. The structure change in the resins by irradiation was also studied.
Silica-DMAA and MVBAA resins were synthesized in a similar manner as previously reported[1, 3]. The resins and HNO3 of up to 6 M were mixed in a Pyrex sample tube at a ratio of 5 cm3/g, respectively, and used as the samples for γ-ray irradiation. Irradiation by the Co-60 source was performed at max. 16.3 kGy/h up to 1.6 MGy at room temperature under ambient atmosphere. The irradiated resins were separated from the supernatant and washed using distilled water to avoid further degradation of the resins by HNO3. The resulting resins and the supernatants were analyzed by IR and 1H NMR (solvent : D2O), respectively. Adsorptivities of the irradiated resins to FP ions were examined by the batch method similar to the previous studies[1-3].
For the IR analyses, a new peak generated at around 1700 cm-1 for DMAA irradiated in HNO3 of higher concentration. While, no clear changes in the spectra were observed in the irradiated MVBAA. 1H NMR spectra of the supernatants after irradiation showed exclusively distinguished peaks which would be attributed to N,N-dimethylamine (ca. 2.7 ppm (vs. DSS), CH3, singlet) for DMAA irradiated in 6 M HNO3 at 1.6 MGy and acetic acid (ca. 2.1 ppm (ditto), CH3, singlet) for MVBAA irradiated in 6 M HNO3 at 0.90 MGy, respectively.
Based on the above results, it would be proposed that both two resins were mainly degraded simply by the break of each amide bond, followed by the formation of polyacrylic acid from DMAA and secondary-amine-type weakly-basic anion exchange resin from MVBAA, re-spectively. These degradation properties are similar to those of chain-type monoamide extractants[4], and are simpler than that of a cyclic monoamide resin[5]. Considering the degradation properties of these monoamide compounds, the two resins are expected to be stable against irradiation in HNO3 of lower concentration such as 0.1 M.
The irradiated resins showed weak adsorptions for Zr(IV) and Pd(II) from HNO3 of lower concentration ranges and no or little adsorptions were found for other FP ions, which was in accordance with the expected adsorptivities of the degraded resins. Besides, it is expected that the two irradiated resins keep the selectivity to U species, respectively, in HNO3 of ca. 3 M and higher.

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