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Selective reduction of actinyl ions based on electrocatalysis using platinized glassy carbon fiber column electrode

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Some actinide ions such as U, Np and Pu exist stably as trivalent to hexavalent ions in an acidic solution. The adjustment of the oxidation state of these ions to a desired one is inevitable for the efficient chemical separation or the precise determination of them, since chemical reactions such as liquid-liquid distribution and complex formation with ligands depend strongly on the oxidation state of the ions. Electrode reaction of tetravalent (An4+) and pentavalent (AnO2+) ions couples, which accompanying formation or cleavage of An-O bond, is generally irreversible and requires a large overpotential. For this reason, selective control of oxidation state of actinide ions by electrolysis is difficult. The authors have studied redox mechanisms of actinide ions, and recently clarified that electrocatalytic reaction at Pt electrode can decrease overpotential of reduction of NpO2+. In the present study, platinized carbon fiber electrodes were examined as a working electrolysis in order to develop a rapid electrolytic method for controlling oxidation state of actinide ions.

The column electrode is composed of a working electrode of a bundle of very thin glassy carbon fibers packed tightly in a cylindrical electrolytic diaphragm of porous glass, and it enables quantitative and rapid electrolysis in a flow system. Flow electrolysis was performed by passing a solution containing actinide ions through a column electrode.

For reduction of NpO2+, overpotential of the reaction was reduced, and successive reductions of NpO2+ to Np4+ and that of Np4+ to Np3+ were observed. This result indicates that the selective preparation of Np4+ by electrolysis can be achieved successfully. For reduction of Pu, overpotential of reduction of PuO2+ was also mitigated, and hence reduction of PuO22+ to Pu3+proceeded selectively at much positive potential. In contrast, the reduction of UO22+ to U4+ was little affected by working electrode materials.

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