Research Group for Interfacial Reaction Field Chemistry

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The research objectives of this group are to explore novel chemical reactions of aqueous species of elements at liquid-solid and liquid-liquid interfaces to contribute to environmental chemistry, actinide chemistry, and waste treatment.

More concretely, we focus our research on elucidation on environmental radionuclide migration and radioactive waste treatment. The research on elucidation on environmental radionuclide migration has two research fields, Fukushima and Ningyo-toge. Researches on the environmental aspects of the Fukushima issues include circulation of ^{137}Cs in Fukushima forest and decontamination of contaminated substances. Researches on Ningyo-toge include chemistry of uranium and radium in soil, rock and groundwater in the context with environmental safety. Research and development on radioactive waste treatment includes development of breakthrough technologies particularly for difficult-to-treat elements.

Migration 2019 conference

The 17th International Conference on the Chemistry and Migration Behavior of Actinides and Fission Products in the geosphere was held from September 15 to 20, 2019 in Kyoto. Key subjects were the geological disposal of radioactive waste in Japan and the remediation of the Fukushima Daiichi NPS site. The MIGRATION conference series is led by an international steering committee including Dr. Grambow, the GL of this group, and has provided an international forum for the timely exchange of scientific information on chemical processes controlling the migration behavior of actinides and fission products in natural aquifer systems. Experimental investigations and predictive modelling of these processes are the main topics of the conferences. Therefore, this conference is a crucial place for this group to present achievements and exchange opinions.

This group made ten first authored presentations and six co-authored presentations. Subjects presented by this group members were the formation of giant cerium(IV) clusters, the complexation of actinides with borate, the environmental issues at the uranium mine Ningyo-toge, the database for thermodynamics of actinides, the microbial effects on simulated nuclear fuel debris, the uptake of radiocesium by mushrooms, radiocesium migration in soils, the role of siderophores in radionuclide mobilization from minerals, radiocesium in sewage sludge ash, the use of chloride salts for soil decontamination at high temperature, the radium removal from uranium mining waste water, and the removal of radionuclides by coprecipitation with barite. GL Grambow presented an invited talk "Towards geological disposal in Japan and remediation of the Fukushima Daiichi NPS site: Research strategy in the global context.

Research on radioactive waste treatment

The main objective radionuclides of this research are anionic radionuclides with long half-lives. They are ^{79}Se, ^{99}Tc, and ^{129}I. Most of the other fission product radionuclides are cations in acid aqueous solution. Numerous methods have been proposed to recover or inorganically fix (solidify) cationic radionuclides since most of natural and synthetic substances have negative charges. It is, however, very difficult for those anionic radionuclides, particularly Tc and I. These two elements are present as monovalent anions ($Tc^{VII}O_4^-$, I^-, IO_3^-) under wide environmental conditions, and their stable complexes and inorganic solids are hardly known.

To obtain basic knowledge on Tc^{VII} adsorption on inorganic substances, we used Re^{VII} as analogue of Tc^{VII} and investigated adsorption on two representative materials with positive charges (hydrotalcite-like layered materials). We found that the Re^{VII} was adsorbed reversibly (anion exchange) in the form of outer-sphere complexes [1, 2]. These results are in harmony with previous knowledge and suggest that adsorption is not appropriate for inorganic fixation of anionic radionuclides when considering instability of adsorbed radionuclides toward anion exchange. Considering this fact, our group have been working on recovering anionic radionuclides from wastewater into the inside of inorganic solid (host phase) by coprecipitation and then stabilizing the host phase to internally fix anionic radionuclides. We reported a coprecipitation method for recovering Se anions in barite (barium sulfate) which has a significantly low solubility [3]. We also found coprecipitation methods for recovering iodate (IO_3^-) in barite and ettringite (hydrous calcium aluminum sulfate mineral) [4, 5]. Our current efforts are focused on post coprecipitation method to stably confine iodine in barite structure and performance evaluation of solidified bodies (waste form) containing iodine-recovered barite. Tc^{VII} coprecipitation with barite is difficult. Instead, we have been investigating an alternative method to use Tc^{VII} reduction and coprecipitation in combination.

In terms of radionuclide extraction from waste liquids, selectivity and capacity for objective element have been the critical factors in developing new extraction agents. Our group has been proposing several new extracting agents in the same approach. So-called "the third phase" formation is another crucial problem in extraction process. In the extraction process at a high metal loading, the third phase containing extractant and metal ions at high concentrations often forms at liquid-liquid interface, which obstructs stable operation. We recently have developed a new extraction system that hardly extracts nitric acid, so that the third phase does not form in the system. Because of this nature, the new system shows high metal extractability far superior to conventional extraction system in a wide nitric acid concentration range [6] (see Highlight).

References

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[5] B. Guo et al., *J. Hazard. Mater.*, in press.
[6] Y. Ueda et al., *Solvent Extraction and Ion Exchange*, **37**, 347 (2019)