Research Group for Interfacial Reaction Field Chemistry

Group Leader: Bernd Grambow
Members: Naofumi Kozai, Hirochika Naganawa, Tetsushi Nagano, Fuminori Sakamoto, Iwao Shimoyama, Noboru Aoyagi, Kojiro Shimojo, Kazuya Tanaka, Takuya Nankawa, Hiroyuki Okamura, Tsuyoshi Sugita, Kohei Tokunaga, Yuki Ueda, Fabiola Guido-Garcia

The research objectives of this group are to explore novel chemical reactions of aqueous chemical species at liquid-solid and liquid-liquid interfaces to contribute to environmental chemistry, actinide chemistry, and waste treatment. For the year of 2018, we report progress in uranium (U) recovery technique using an emulsion flow system [1] and in environmental migration of radiocesium in Fukushima [2],[3].

Continuous removal of U from U-containing wastewater using an organic phase-refining-type emulsion flow extractor

At Ningyo-toge Environmental Engineering Center, JAEA, a large quantity of uranium-containing wastewater (UW) has been generated from the decontamination of spent centrifuges. For selective U removal from the UW, we have introduced a continuous liquid–liquid extraction method using an “emulsion flow” (EF) extractor, wherein the EF is generated by spraying fine droplets of extractant-containing organic phases into aqueous phases. However, the removal performance of U has gradually deteriorated in the conventional EF system without a mechanism of refining organic phase.

We have developed a new EF system equipped with a refining column for clean-up of used organic phase. In this apparatus, the organic phase is circulated between the extraction and the refining columns. During the circulation, U in the feed solution is extracted into the organic phase in the extraction column and stripped into the refining aqueous solution in the refining column. In this manner, the organic phase is continuously refined as it is being circulated (Fig. 1).

The new EF system was applied to the continuous removal of U from the simulated UW. The organic phase used in this system is Shellsol D70 solution containing tri-n-octylamine and 2-ethylhexanol, which is effective in selective extraction of U from dilute sulfuric acid solutions. The feed volume of the UW was 5 L, which was 10 times larger than that of the organic solution. A comparative experiment was carried out under non–refining condition using the conventional EF system. As a result, the U removal rates remained almost constant at ~90% under the refining condition while it gradually decreased from ~90% to ~36% under the non-refining condition (Fig. 1). The elemental analysis of the organic phases after the end of the experiment showed that the U in the organic phase under the refining condition decreased significantly compared to that under the non-refining condition. As a result, the newly developed organic phase–refining–type EF system is considered to be superior to the conventional one using the extraction column alone for the treatment of a large volume of UW, because the refining column works effectively in terms of preventing the deterioration of the removal performance.

Distribution and migration of Fukushima-derived radiocesium in the environment

In order to evaluate migration of radiocesium derived from the accident at Fukushima Daiichi Power Plant in environment based on a comparison of radiocesium to stable Cs, we have investigated size-fractioned samples of riverbed sediments collected at two sites, a pasture and Kuroiwa in the Abukuma River system in Fukushima Pref. The size distributions of K, Rb and stable Cs reflected the mineralogy of sediments, where primary host minerals for these alkali elements would be biotite, K-feldspar and clay minerals. Silt-size fractions contained high stable Cs and radiocesium concentrations possibly due to adsorption on clay minerals. Overall, the size distributions of stable Cs and ^{137}Cs were similar in the sediments (Fig. 2), suggesting that radiocesium released by the Fukushima accident was distributed into each particle size fraction in response to the distribution of the stable Cs that was governed by mineralogical composition [2].

Surface soil samples collected from Kawamata, Fukushima Pref. were leached with natural seawater, diluted seawaters, and ultrapure water (PW) to observe the effect of the ionic strength on leaching ratios of ^{137}Cs [3]. Overall, seawater and diluted seawater gave higher leaching ratios than PW. For long-term experiments (223 days), more than 15% of total ^{137}Cs in the soil sample was leached with seawater. While about 9% of the total ^{137}Cs was leached with 1:1 diluted seawater, less than 1% of the total ^{137}Cs was leached with PW over the 223 days. Radiocesium fixed in clay minerals of soil and sediment would be partially extracted by seawater after their transport to the ocean via rivers.

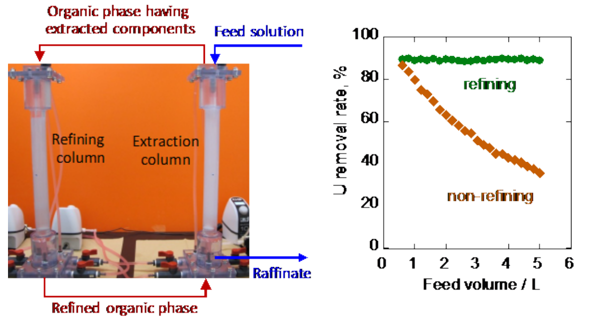

Fig. 1 Outline of organic phase-refining-type EF extractor (left) and U removal rate with and without refining organic phase (right).

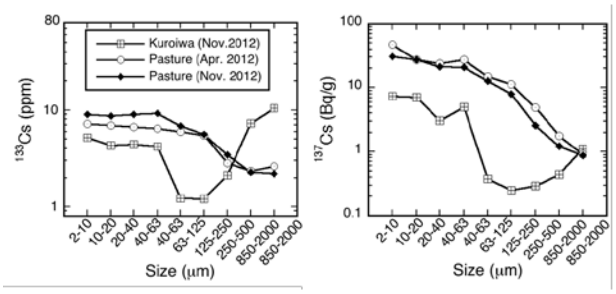

Fig. 2 Size distributions of stable Cs (left) and radiocesium (right) in riverbed sediments collected in Fukushima [2].

References

[1] T. Nagano et al., *Anal. Sci.*, **34**, 1099 (2018).
[2] K. Tanaka et al., *Geochem. J.*, **52**, 173 (2018).
[3] A. Sakaguchi et al., *Geochem. J.*, **52**, 186 (2018).