



## Sorption and solidification of iodide

Toshihiko Ohnuki (1), Naofumi Kozai (2), Kazuya Tanaka (2), Kohei Tokunaga (2), Satoshi Utsunomiya (3), Ryohei Ikehara (3), Tatsuki Komiya (3), Ayaka Takeda (3), Daniel Kaplan (4), and Peter Santschi (5)

(1) Tokyo Institute of Technology, Institute of Innovative Research, Tokyo, 152-8550, Japan (toshi.ohnuki@lane.iir.titech.ac.jp), (2) Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki, 319-1195, Japan, (3) Department of Chemistry, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka-shi, 819-0395 Japan, (4) Environmental Biogeochemistry & Technology, Savannah River National Laboratory, Aiken, SC, 29808, USA, (5) Laboratory for Oceanographic and Environmental Research, Texas A&M University, Galveston, TX 77553, USA

After the Fukushima NPP accident, radionuclides containing FPs were released from the fuel debris into the cooling water. Treatment of the contaminated cooling water by the Advanced Liquid Processing System (ALPS) facility removed most of the cationic FPs by sorption and/or co-precipitation. However, removal of anionic FPs from the cooling water remains a technical challenge. Especially problematic is iodine-129, which has a high toxicity and an extremely long half-life (16M years), and commonly exists as multiple species, iodide (I<sup>-</sup>), iodate (IO<sub>3</sub><sup>-</sup>), and/or organo-iodine (org-I). Thus, sorption and solidification of I-129 should be studied by controlling chemical species. Here we report the sorption behaviors of I<sup>-</sup> on the several sorbents involving activated carbon, CeO<sub>2</sub>-modified activated carbon, Ag-attached zeolite, titanate attached activated carbon and calcined hydrotalcite (CHT). We also studied the solidification of I<sup>-</sup> with a geopolymer formed by alkaline-activation of metakaolin. We measured distribution coefficients, K<sub>d</sub> by the sorbents, and leaching behavior from the geopolymer into a distilled water and into a sea water. We analyzed chemical species of I in the sorbents and geopolymer by XANES, and position of I and chemical components present with I in the sorbents and geopolymer were analyzed by SEM.

Sorption of I<sup>-</sup> showed that the highest K<sub>d</sub> for the activated carbon, the CeO<sub>2</sub>-modified activated carbon, the Ag-attached zeolite, and the titanate attached activated carbon were obtained at pH between 2 and 3. Increase of pH resulted in decrease of K<sub>d</sub> for the activated carbon, the CeO<sub>2</sub>-modified activated carbon, and the titanate attached activated carbon. K<sub>d</sub> was nearly 0 at alkaline solution. K<sub>d</sub> for the Ag-attached zeolite decreased gradually with increasing pH. The XANES analysis showed that the sorbed I was present as I<sup>-</sup> on the activated carbon, the CeO<sub>2</sub>-modified activated carbon, and the titanate attached activated carbon, indicating that I was not changed its oxidation state during the sorption by these sorbents. The XANES spectrum of I on the Ag-attached zeolite was different from that of I<sup>-</sup> ion, suggesting that chemical species of I was changed after association with Ag-attached zeolite. SEM analysis did not detect I on the sorbents. These results indicated that I<sup>-</sup> are adsorbed by the activated carbon, the CeO<sub>2</sub>-modified activated carbon, and the titanate attached activated carbon, and was associated by the Ag-attached zeolite.

Almost all I<sup>-</sup> was solidified with the geopolymer made from metakaolin. Approximately 80% of I was leached from the geopolymer into distilled water and sea water. SEM analysis showed homogeneous distribution of I in geopolymer. The XANES analysis showed chemical species of I in the geopolymer was I<sup>-</sup>, and was kept I<sup>-</sup> after leaching. These results suggest that I<sup>-</sup> was not strongly associated with the geopolymer, and easily contacted with distilled water to be leached out.

Therefore, iodide ions were weakly associated with the activated carbon, the CeO<sub>2</sub>-modified activated carbon, and the titanate attached activated carbon, and were associated by the Ag-attached zeolite with forming AgI. Iodide ions were weakly associated with the geopolymer made from metakaolin.