

1-7 Estimating Radioactive Waste Inventory

— Sorption Behavior of Actinides on Zeolite —

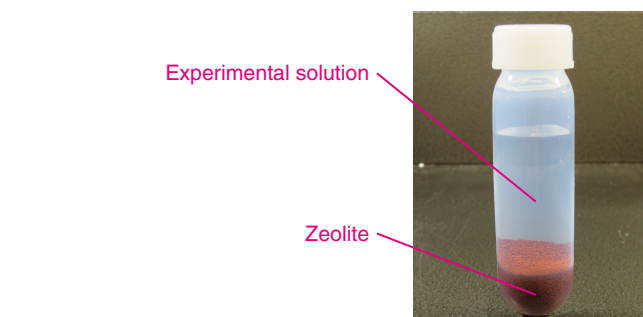


Fig.1-16 A test tube used in the sorption experiment of U and Np

U or Np is added to the experimental solution containing zeolite. The sorption behavior of U and Np is observed by the change of U and Np concentration in solution with time.

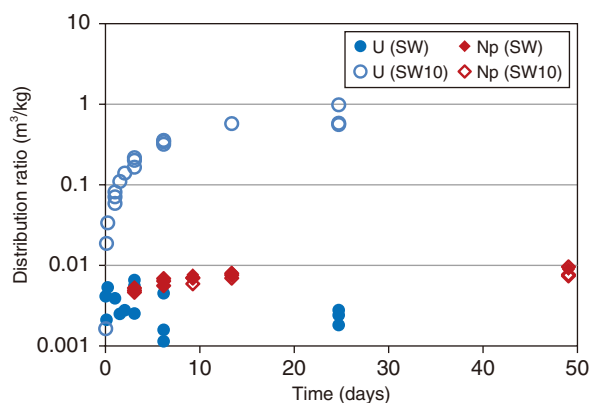


Fig.1-17 Change in distribution ratio of U and Np with time

The distribution ratio of U in simulated seawater diluted ten times by deionized water (SW10) was higher than that of Np and U in simulated seawater (SW).

At the TEPCO's Fukushima Daiichi NPS, contaminated water has been processed by water treatment apparatuses to remove radionuclides. Spent zeolite, used as an absorbing material in these apparatuses, is disposed of as radioactive waste. An estimation of the kinds and amount of the radionuclides present in this spent zeolite is necessary for selecting a suitable disposal concept and safety assessment. However, estimation of the inventory via sampling of zeolite from spent zeolite vessels is difficult because of the high dose rate and vessel structure. Therefore, sorption experiments of U and Np on zeolite (IONSIV™ IE-96, UOP), which is used in the water treatment apparatus SARRY, were conducted to develop an inventory estimation method based on the sorption behavior of radionuclides on zeolite (Fig.1-16).

The resulting distribution ratios using simulated seawater (SW) and simulated seawater diluted ten times by deionized water (SW10) are shown in Fig.1-17. Here, the distribution ratio was calculated by dividing the amount of U or Np sorption on zeolite (i.e., solid phase concentration) by the concentration of U or Np in the solution. A high distribution ratio indicates that a large amount of U or Np was sorbed onto the zeolite. The results indicated that a low distribution ratio of U was obtained in SW, whereas the distribution ratio was significantly increased in SW10. Alternatively, the distribution ratio of Np was low and independent of seawater concentration.

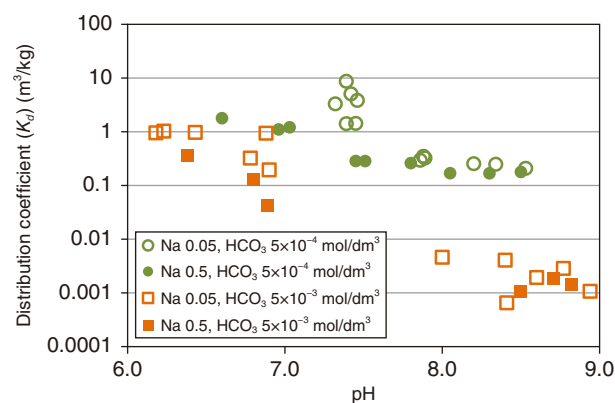


Fig.1-18 Dependency of uranium distribution coefficient on pH

The distribution coefficient of U decreased due to the increase in carbonate concentration when the pH was higher than 8. The distribution coefficient was independent of Na⁺ concentration.

To clarify the dominant factor causing the variation of the distribution ratio of U between SW and SW10, an experiment was carried out as functions of carbonate and Na⁺ concentration in the experimental solution. The resulting distribution coefficient (K_d) is shown in Fig.1-18 as a function of the pH, where K_d is defined as the distribution ratio at sorption equilibrium. In the pH range from 8 to 9, the K_d of U was low at a higher carbonate concentration, whereas the K_d was independent of Na⁺ concentration. This indicates that the difference in K_d of U between SW and SW10 is due to the difference of carbonate concentration in the solution. The decreasing K_d with increasing carbonate concentration may be caused by the increase in the concentration of uranium-carbonate complexes, which have a low sorption ability on zeolite. The K_d of Np was found to be independent of Na⁺ and carbonate concentration. This was consistent with the trend of the distribution ratio of Np observed in the experiment using SW and SW10.

Thus, the carbonate concentration may be a dominant factor in controlling the inventory of radionuclides in spent zeolite.

This work was part of the result conducted “Development of technology for treatment and disposal of accident waste”, and supported by the Agency of Natural Resources and Energy (ANRE), the Ministry of Economy, Trade and Industry (METI), Japan.

Reference

Ishidera, T. et al., Sorption Behavior of U and Np on Zeolite, Progress in Nuclear Science and Technology, vol.5, 2018, p.221–224.