Environmental Behaviors of Radionuclides Studied by Advanced X-ray Absorption Spectroscopy

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Keywords: Speciation, XAFS, Radionuclides, Molecular Geochemistry, Fukushima Nuclear Accident

In geochemistry and environmental chemistry, understanding the elementary processes of the behavior of elements by analyzing their chemical species (i.e. molecular geochemistry) contributes to a better understanding of the evolution of the universe, the earth, and life, and to the prediction of future environment, resources, and climate. This is basically similar to the studies of environmental radiochemistry. However, we need to consider several additional point in the environmental radiochemistry, which deals with chemistry related to radionuclides [1]: first, by gaining knowledge beyond the framework of the conventional periodic table, the behavior of all elements can be treated more systematically. The second is that the natural abundance of these elements is generally low, which needs more sensitive analysis in particular for speciation work to develop molecular geochemistry of radionuclides. Actually, tools developed in radiochemistry have often provided breakthroughs in molecular geochemistry and contributed to the understanding of the systematics of elemental behavior, especially as societal demands have made the development of such sensitive methods significant in this field. An example of such method is laser-induced fluorescence (LIF) spectroscopy, which has high-sensitivity and various information on the speciation of some elements [2], which was developed quickly in the field of environmental geochemistry in 1990s and subsequently used in trace element geochemistry afterwards. In a sense, this field is at the front of trace element geochemistry, and similar situation may be valid to the application of X-ray absorption fine structure (XAFS) spectroscopy.

Generally, XAFS at hard X-ray region including X-ray absorption near-edge structure (XANES) and extended X-ray absorption fine structure (EXAFS) are frequently employed, which provide information of (i) valence and symmpetry of the target element and (ii) local structures in the vicinity of the targe element such as interatomic distances to neighboring atoms and coordination number. To get the spectra, fluorescens mode using Si or Ge detector to detect X-ray fluorescence intensity from a target element is commonly used to get the XAFS spectra of trace elements. To conduct more sensitive analysis, high energy resolution fluorescence detection (HERFD) using both in energy-dispersive or wavelength-dispersive detector is very powerful to decrease the signal of the target element to background signal (XRF of other elements and scattered X-ray).

In this presentation, as examples of their applications to environmental radiochemistry two topics, (i) studies on the migration of radionuclides emitted by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in 2011 [3,4] and (ii) the development of advanced XAFS, which consists XANES and EXAFS. The former topic must be summarized from various scientific and engineering perspectives, and the various geochemical processes and migration behavior of radionuclides learned from these studies are important for predicting the behavior of radionuclides in the terrestrial environment [5].

The latter topic is important for a comprehensive understanding of the environmental behavior of all elements, which is also related to the first topic, because the highly sensitive analytical tool for the speciation of trace elements is necessary in all fields of environmental geochemistry. The high sensitivity method can be achieved by high energy resolution detection of fluorescent X-rays using either a crystal analyzer or a high energy resolution detector such as a transition edge sensor (TES)[6]. In particular, high energy resolution fluorescence detection XANES (HERFD-XANES) provides more speciation information than conventional XANES [7], which needs to be applied in a wide range of geochemistry in the future. Among the major radionuclides, radiocesium and U were the focus in this part. Radiocesium, which is abundant and volatile in the reactor, is the main radionuclide emitted from the FDNPP accident, and adsorption on phyllosilicates is the main reaction controlling radiocesium behavior [8].

References

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