New X-ray spectroscopic methods for the speciation of cesium in environment

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Speciation of cesium, or radiocesium, in environment is essential for the better understanding of its behavior in environment. For this purpose, X-ray absorption fine structure spectroscopy (XAFS) in fluorescence mode is one of most effective methods, but interference of Cs signal by signals from background elements (ca. calcium (Ca) and titanium (Ti)) is a severe problem to selectively obtain the signal from Cs. To overcome the difficulties, we have employed two methods, which are using (i) high energy resolution energy dispersive detector and (ii) wavelength dispersive detection system. In this presentation, the two methods were applied to chemical state of radiocesium (RCs) using X-ray absorption near-edge structure (XANES) analysis in fluorescence mode for the RCs incorporated into cesium-bearing silica microparticles (CsMP) categorized as B-type (non-spherical, with diameter about 500 μm; RCs concentration lower than 50 μg/g) emitted by Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in 2011. It was difficult to observe X-ray fluorescence of Cs Lα emission in this CsMP using a silicon drift detector because the Cs concentration in the particle was three orders of magnitude smaller than the concentrations of calcium and titanium, which caused severe deterioration of the signal-to-background ratio of the Cs Lα emission line. In this study, a high energy resolution (ca. 5 eV FWHM) energy-dispersive transition edge sensor (TES) spectrometer was applied to XANES spectroscopy of trace elements in environmental materials for the first time, which enabled us to measure Cs LIII-edge XANES for RCs in CsMP. It was demonstrated that TES can be a powerful tool for the speciation of trace elements in environmental samples by fluorescence XANES considering its high energy resolution coupled with wide application range and simplicity of operation as energy-dispersive type detector. The results in this study revealed that RCs was dissolved into the silicate matrix. This information is consistent with the emission process that the type-B CsMP was formed by the cooling of melted materials during the FDNPP accident suggested in previous studies. We also employed another method using crystal analyzer coupled with Pilatus detector, instead of using TES. The energy resolution is even better than TES, which finally enables us to measure high energy resolution fluorescence detection (HERFD) XANES spectroscopy, which can give more information on the chemical state of Cs. The application of HERFD-XANES will be also given in this presentation.

