EXAFS Measurement of U(VI)-N,N-Di-alkylamides Complexes

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Introduction

In the nuclear spent fuel reprocessing, FBR spent fuels will coexist with LWR spent fuels for several decades until FBR cycle begins to operate. For the treatment of present LWR spent fuels, high decontamination factor for FP was required for U(VI) storage, and solvent extraction technique was selected in the nuclear fuel treatment. In our laboratory, *N*,*N*-di-alkyl monoamides have been developed as extractant based on solvent extraction technique for one of spent fuel reprocessing technology in the LF transition periods. For spent nuclear fuel treatment of LF transition periods, branched alkyl type *N*,*N*-dialkyl monoamides isolate uranium(VI) from plutonium(IV) with high separation factor[1]. We studied U(VI) complexes with these ligands in the organic solution by extended X-ray absorption fine structure(EXAFS).

Experimental

N,*N*-dialkylamides : *N*,*N*-di-hexyl(2-ethyl)butanamide (DH2EBA), N,N-di-octyl(2-ethyl)butanamide (DO2EBA) N,N-di-(2-ethyl)hexyl(2,2-dimethyl)propanamide (D2EHDMPA), *N*,*N*-di-(2-ethyl)hexyl(2-methyl) propanamide (D2EHMPA), and N,N-di-octyl-butanamide (DOBA) were synthesized in our laboratory (Fig 1). The uranium samples were made as following procedure. 1 ml of n-dodecane as an organic solvent containing 1.0 mol/dm³ N,N-dialkylamides was shaken with the same volume of the various nitric acids containing 40 mM of UO_2^{2+} . After centrifugation, 0.45 ml of the organic solution was separated to use as a sample. 0.45 ml of U(VI)-N,N-dialkylamides complex solution was enclosed in polyethylene tube. The XAFS measurement carried out at beam line BL27B. The absorption edges were 17.16 keV for U(L_{III}). The extraction data of EXAFS oscillation from absorption spectra were carried out by WinXAS Ver. 3.1[2]. The theoretical parameters for curve fittings of the EXAFS spectra were calculated using FEFF 8.20 [3].

Results and Discussion

Figure 2 shows the raw k^3 -weghted U L_{III}-edge EXAFS spectra and the corresponding Fourier transforms (FT) of U-*N*,*N*-dialkyl amide complexes in n-dodecane prepared by the solvent extraction methods. The first peak of the RSF is based on UO₂ axial 2 oxygen atoms, and the second peak is based on the carbonyl oxygen of N,Ndialkylamide and the nitrate ion which coordinated to UO₂ by bidentate and 2 nitrogen atoms of nitrate ion. The obtained bond distance between uranium(VI) and oxygen of *N*,*N*-dialkyl monoamide are 2.40 Å for DOBA; 2.36 Å for DO2EBA, 2.35 Å for DH2EBA, 2.31 Å for D2EHDMPA, and 2.33 Å for D2EHMPA, respectively. Bond distances of U-O (O=C; *N*,*N*-dialkylamides) were changed dependence on amides structure, especially carbonyl alkyl branching on α -position.





Fig.2 The raw k^3 -weighted U L_{III}-edge EXAFS spectra (left) and the corresponding FTs (right) of the U-*N*,*N*-

References

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