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XAFS data analysis of uranium dioxide by using MD simulation and FEFF

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Introduction

We had proposed the new simulation procedure to calculate the XAFS function $k^3\chi(k)$ of highly disordered system like high-temperature molten salts[1]. In the simulation, the output from the molecular dynamics (MD) calculation was directly used as input data in the XAFS simulation code FEFF8 (MD-FEFF method). The XAFS function was obtained by averaging results of the FEFF8 calculation. Many XAFS functions of molten salt systems like RbCl, YCl_3-LiCl-KCl and UCl_3-LiCl-KCl[2] were successfully reproduced by the MD-FEFF method.

From many experiences of the simulation, we confirmed that averaging over 10000 FEFF computations was required to obtain the saturated XAFS function. In addition, judging a saturation of the averaging procedure was very difficult especially in the XAFS simulation of room temperature crystalline systems. In the present work, we tried to simulate XAFS function of room temperature solid UO_2 by using the MD-FEFF method.

Experimental and XAFS simulation

XAFS measurement

The XAFS measurements were performed at the BL-27B station in the KEK-PF. The U L_3-edge (E_0=17.167keV) XAFS for solid UO_2 were measured in the transmission method. The UO_2 sample was adjusted to O/U=2.00 by heating under reduction atmosphere. Details of the XAFS measurement and data processing of molten salts are described in ref.[3].

XAFS simulation

The molecular dynamics simulation using rigid ionic model was performed to simulate XAFS functions of molten UO_2. The potential model proposed by Karakasidis and Lindan[4] was used in the MD simulation. The XAFS function was obtained by averaging FEFF computations from position data of the MD simulation[1].

Results and discussion

The experimental and simulated XAFS functions $k^3\chi(k)$ of solid UO_2 is shown in Fig.1, respectively. Maximum k-vector value is 18.5 for the experimental function. The XAFS simulation was saturated between 10000 and 15000 FEFF computations. The simulated XAFS function in the figure is averaged value of 20000 FEFF computations. The XAFS function is reproduced by the simulation, though oscillation of the function is slightly overestimated at higher k-vector region. Fourier transform magnitude functions |FT| are shown in Fig.2. The 1st peak corresponding to the nearest U^{4+}-O^{2-} correlation is in good agreement with the experimental one. On the other hand, the 2nd peak assigned to the 1st U^{4+}-U^{4+} correlation is overestimated by the simulation. It can be seen that the simulated function reproduces all peaks found in the experimental |FT| function. It means that the MD-FEFF method is effective also for room temperature solid system.

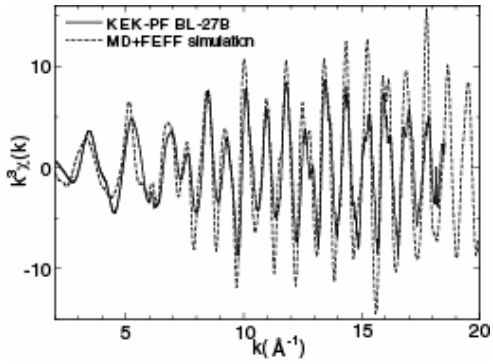

Fig.1 XAFS functions$k^3\chi(k)$ of solid UO_2

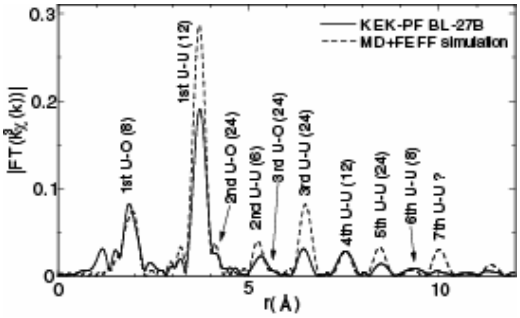

Fig.2 Fourier transform magnitude functions |FT| of solid UO_2

References

[1] Y. Okamoto, NIM-A, **526**, 572(2004)
[2] Y. Okamoto et al., KEK-PF Activity Report, 2005, vol.23, Part B, 13
[3] Y. Okamoto et al., NIM-A, **487**, 605 (2002)
[4] T. Karakasidis and P. J. D. Lindan, J. Phys.: Condens. Matter, **6**, 965(1994)

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