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XAFS and molecular dynamics simulation of molten RbCl

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Introduction

We have investigated the local structure of high temperature molten salts by using XAFS technique. We confirmed that it was difficult to analyze the high temperatuyre XAFS data(generally beyond 500 ºC) by a standard curve fitting technique. We have used the cumulant expansion method[1] to treat an anharmonic vibration effect in the liquid. Our XAFS data was successfully analyzed by the expansion technique. However, it contains uncertain factor because of additional fitting parameters. Recently, Filipponi[2] reported from some XAFS data analysis examples that the curve fitting analysis procedure should not be used for high temperature liquid XAFS data. In the present work, we tried to obtain calculated XAFS function from a molecular dynamics(MD) technique and the FEFF8[3]. This new analysis procedure which does not use the curve fitting was evaluated by using the XAFS data of molten RbCl.

Experimental and MD simulation

XAFS measurementst

The XAFS measurements of solid and molten RbCl (Rb K-dege) were performed in transmission method at the BL27B station in the KEK-PF. The samples were sealed off in a quartz cell under reduced pressure. Details of the XAFS measurement of molten salts are described in ref.[4]. The XAFS spectra were obtained at room temperature and 1050K. The XAFS data was analyzed by using WinXAS code[5].

Molecular dynamics simulation

MD simulations of solid and molten RbCl were performed by using NVT=constant ensemble with 1000 ions(500Rb^+ and 500Cl^-). Tosi-Fumi pair potential[6] was used in the simulation. We confirmed that simulation by using this potential resulted in good agreement with the XRD[7] of molten RbCl. Output data(position of each ion) from the simulation was directly used as input data of the FEFF8. XAFS function was calculated by accumulating the FEFF8 computations.

Results and discussions

The Experimental XAFS function $k^3\chi(k)$ of solid and molten RbCl are shown as solid lines in Fig.1(a) and (b). Amplitude of the function drastically decreases by melting. Calculated XAFS functions (dashed line in the figure) were obtained by averaging 25000 FEFF8 computations. It means that output information from 50 MD steps (this 50 MD steps were chosen from total 50000 MD steps simulation) was contained in the functions. The calculated results are in good agreement with the experimental data in both solid and molten states. It is concluded that the MD+FEFF8 computation procedure can be used for the XAFS data analysis of high temperature molten salt systems.

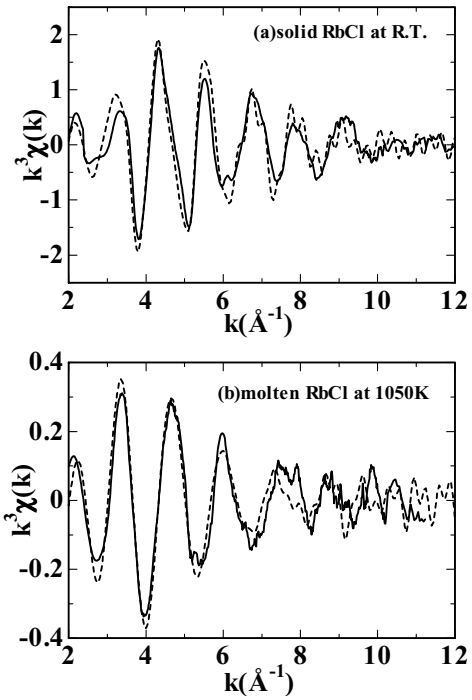

Fig.1 XAFS function $k^3\chi(k)$ of (a) solid and (b) molten RbCl.
solid: experimental dashes: MD + FEFF8

References

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