Development of levitation technique for the liquid structure analysis

by

Tadahiko Masaki[1], Takehiko Ishikawa[1], Paul-Francois Paradis[1] and Toshio Itami[1]

Abstract: The containerless technique is one of the latest methods for the handling of matters at high temperature and in undercooled liquid states. In this research project, the electrostatic levitation technique has been applied to the structural analysis of liquids by using the neutron and X-ray scattering techniques. The current status of this subject is described briefly.

1. Background

Nowadays, many attentions have been focused on the containerless techniques for the experimental research of high temperature melts or deeply undercooling liquids, especially for the structure analysis by neutron or X-ray scattering method. Recently, JAXA has been developing a droplet levitation technique due to the electro-static force either for the microgravity experiments in the international space station or for the normal gravity experiments in a laboratory on the ground. Since this technique has merits for neutron scattering experiments, JAXA and Japan Atomic Energy Research Institute, JAERI, are developing the electro-static levitation furnace (ESL) for neutron scattering experiments of extremely high temperature melts and deeply undercooled liquids. In the last year, the basic model of ESL was developed and its applicability was verified through the neutron scattering measurements for sintered alumina levitated at room temperature. The levitation of sample was able to be kept for 11 hours. In this "cold" experiment, the diffraction peak of alumina was clearly observed without any other diffraction peak in the background [1] .

2. Research activity in 2003

In this year, the actual performance of ESL for high temperature melts was verified as "hot" experiment. The facility configuration was same as that of "cold" experiment except for the sample heating system. In order to heat the levitated sample the CO_2 laser was installed in the facility. The heating laser, which was irradiated to the sample through the ZnSe optical window, was placed on the top of vacuum chamber. Zirconium was selected

Figure 1 Liquid zirconium at the temperature of 2500 K.

[1]Japan Aerospace Exploration Agency, 2-1-1 Sengen, Tsukuba, Ibaraki, 305-8505 Japan

as a test material of levitation system for the "hot" experiment because zirconium is one of the most suitable materials for keeping the levitation in long duration. The "hot" experiment was succeeded in keeping the levitated molten zirconium for 11 hours. Figure 1 shows the levitated liquid zirconium at 2500K. It can be seen that the levitation method provides the nearly spherical sample for which the data correction, for example the correction of absorption, can be easily performed on the diffraction data analysis. Since the sample size of 2 mm diameter was rather small, the counted scattering intensity from levitated sample was not sufficient within the duration of levitation in this experiment. As the next step, the experiments of much longer duration of levitation will be tried in order to obtain the scattering data.

3. Analysis of correction factors of spherical samples for scattering experiments

3.1 Correction factor of absorption

The correction of absorption is one of the most important factors to obtain the S(Q) from the intensities of scattered neutron. The exact value of correction factors can be evaluated for the spherical shape such as the a sample of electro-static levitation. The correction factor is written in the following equation.

$$A = \frac{1}{V} \int \exp(-\mu T) dV, \tag{1}$$

where V is a volume of sample, μ is a absorption coefficient of sample and T is the optical pass of X-ray or neutron.

Fortunately, the correction factor of spherical sample is tabulated in "International tables for crystallography Volume C" [2] because the spherical sample is usually used for the structural analysis of single crystal. The values of data correction can be evaluated from the interpolation of this table values as a function of the product of mass absorption coefficient, μ, (or scattering cross section) and radius of spherical sample, R. However, in this research project, the correction factors were numerically evaluated directory from equation (1) because this calculation scheme can be applied also to the evaluation of correction factors of multiple scattering, which is described in the next subsection. Figure 2 shows the obtained correction factor corresponding to $\mu R=1.0$ together with the data of Ref.2. These are in good agreement with each other. The difference between them is less than 1%. Unfortunately, the table provides the correction factors whose parameter, μR, is less than 3.0. With the use of the present evaluation scheme, the correction factors corresponding to the μR greater than 3.0 can be obtained if necessary in a further research.
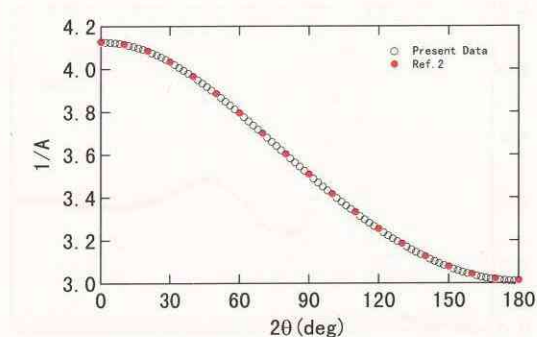

Figure 2 Correction factor of absorption for the spherical sample ($\mu R=1.0$)

3.2 Correction factor of multiple-scattering

In the case of neutron scattering experiments, the intensity of multiple scattering is not negligibly small though it is not so large. Some of previous researchers have published the correction factor of multiple scattering. However, those were the correction factors corresponding to a cylindrical or slab shape sample, which is popular for the conventional scattering experiment of liquid matters.

In this research project, the correction factors of spherical samples were evaluated for the preparation of further research. As the first step, the numerical simulation was performed based on the model liquid whose structure factor, $S(Q)$, was assumed to be hard sphere one .

The scattering intensity, dI, of volume element, dv, in a spherical sample is described as follows:

$$dI = s(2\theta)\, a\, dv, \qquad\qquad (2)$$

where $s(2\theta)$ is scattering factor of 2θ and a is the atomic scattering factor.

The intensity of singlet scattering, I_{single}, can be obtained from the sum of dI with the factor of sample absorption, $\exp(-\mu l)$, along the path of beam, l, as follows:

$$I_{single} = \int s(2\theta)\, a\, \exp(-\mu l)\, dv. \qquad\qquad (3)$$

In the case of doublet scattering, the intensity can be obtained from the sum of the twice of singlet scattering at the different volume element. The absorption by sample should be also taken into account similarly to the calculation of singlet scattering, as follows:

$$I_{double} = \iint s_1(2\theta)\, s_2(2\theta)\, a^2 \exp(-\mu l)\, dv_1 dv_2. \qquad (4)$$

In this model calculation, the atomic scattering factor was assumed to be same as that of zirconium and the structure factor assumed was the analytical solution of hard sphere fluid derived from Percus-Yevick model. Figure 3 shows the scattering intensity of singlet and doublet scattering obtained from this numerical analysis. The intensity of doublet scattering is small. Therefore, the scale of doublet scattering is multiplied by 100 in this figure. The angular dependence of doublet scattering is almost constant. The intensity of doublet scattering is less than 1% of coherent scattering intensity at high Q region.
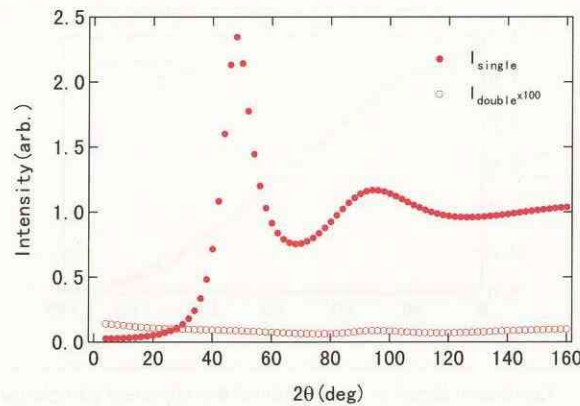

Figure 3   Intensity of singlet and doublet scattering

4. Summary

The neutron scattering experiment of liquid Zr was performed with the use of the electrostatic levitation method. The levitation of liquid Zr continued for 11 hours. However, the obtained scattering intensity was not sufficiently strong because of the small sample size. The correction factors due to respectively the absorption and multiple scattering of model fluid were evaluated in numerical way for the preparation of future further researches.

Reference

[1] H. Aoki, P.-F. Paradis, T. Ishikawa, T. Aoyama, T. Masaki, S. Yoda, Y. Ishii and T. Itami, Rev. Sci. Instrum. 74, 1147(2003).

[2] "International Tables for Crystallography" Volume C, Kluwer Academic Pub. (1999).