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軌道上及び地上静電浮遊炉を利用した酸化物融体の熱物性測定

Thermophysical property measurements of liquid oxides by using the electrostatic levitation furnace onboard the International Space Station and on the ground

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1. Introduction

Investigation_of liquid properties at high temperatures is very challenging particularly above about 2000 K, because chemical reactions with containers are unavoidable. In addition, crystallizations from the container makes it difficult to maintain the liquid state. However, the use of levitation techniques can overcome these problems and, in many cases, enable to measure the physical properties precisely and maintain deep undercooling owing to the elimination of contamination and extrinsic heterogeneous nucleation. Using the levitation methods, we have been recently investigating high temperature oxide melt to uncover the relationship between glass-forming ability and thermophysical properties or atomic structure of liquid. [1,2] In this talk, we report density measurements of liquid lanthanoid oxides, by using the electrostatic levitation furnace onboard the International Space Station (ISS-ELF) and viscosity measurements of liquid $Y_3Al_5O_{12}$ using an electrostatic levitation furnace on ground.

2. Experimental

2.1 Density measurement

The densities of molten lanthanoid oxides (Ln_2O_3, Ln = Er, Ho, Tb, Gd) were measured using the ISS-ELF.[3] Firstly, the oxides were levitated, and the positions were stabilized using the Coulomb force between the charged oxides and surrounding electrodes and employing a rapid feedback control process. They were subsequently melted in air at 2 atm by semiconductor lasers. The molten oxides exhibited spherical shapes and their volumes were readily calculated from magnified images. Subsequent weighing of the samples on Earth allowed the densities of the oxides to be determined.

2.2 Viscosity measurement

The viscosity of liquid $Y_3Al_5O_{12}$ was measured using the oscillating drop method and electrostatic levitator on the ground in the vacuum condition (10^{-4}Pa). The droplet oscillation was performed by applying a sinusoidal voltage to the electrode.[4] The oscillation frequency was adjusted so that mode 2 oscillation occurred. After stopping the excitation, the decay time (τ_2) was measured and then the viscosities (η) were determined by the following equation:

$$\frac{1}{\tau_2} = \frac{5\eta}{\rho {r_0}^3}. \quad (1)$$

where r_0 is radius of droplet and ρ is the density.

3. Results and discussion

3.1 Density measurement results

The densities of Er_2O_3, Ho_2O_3, Tb_2O_3, and Gd_2O_3 at their melting temperatures (T_m) were obtained over the wide temperature rage (2700-3200 K), and these density values were shown to exhibit a linear correlation with temperature. The molar volumes of these oxides at their T_m values were calculated and compared with those of other sesquioxides (Al_2O_3, Ga_2O_3, and B_2O_3). As shown in Fig.1 the molar volumes of the nonglass-forming sesquioxides (Er_2O_3, Ho_2O_3, Tb_2O_3, Gd_2O_3, Al_2O_3, and Ga_2O_3) exhibit linear correlations with the cubes of their cation radii, whereas those of the glass-forming oxide (B_2O_3, As_2O_3, and Sb_2O_3) showed different correlations.[1)]

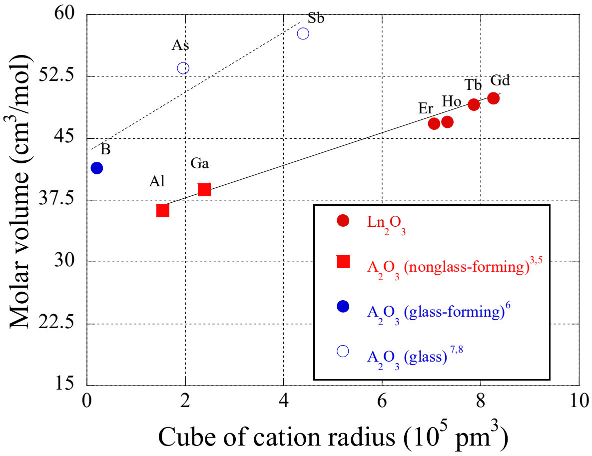

Fig. 1 (left figure) Correlation between the molar volumes of liquid Ln_2O_3 oxides (Ln = Er, Ho, Tb, Gd) at T_m and the cubes of the cation radii, including data for Al_2O_3,[3] Al_2O_3,[5] and B_2O_3,[6] vitreous As_2O_3[7] and Sb_2O_3.[8]

3.2 Viscosity measurement results

The viscosity of liquid $Y_3Al_5O_{12}$ was measured over a wider temperature range (2010 K≤T≤2430), including the undercooled region (T_m=2240 K). In this range, viscosity values were 20-40 mPa・s, and were optimized by the non-Arrhenius viscosity model equation[9)] (2)

$$\log_{10}\eta(T) = A + (12-A)\frac{T_g}{T}\left[\left(\frac{m}{12-A}\right)\left(\frac{T_g}{T}-1\right)\right] \quad (2)$$

$$A = \log_{10}\eta_\infty$$

where m and η_∞ are fitting parameters and m is fragility. The fitting shows that m is 53, which is intermediate between fragility of SiO_2, glass-forming liquid, and that of molecular liquids which are difficult to vitrify.

4. Summary

The densities of liquid Ln_2O_3 compounds were measured with the ISS-ELF and the viscosity of liquid YAG was measured with the ground-based electrostatic levitator. This process prevented contamination and crystallization such that precise density and viscosity values could be obtained over a wide temperature range. The molar volumes of Ln_2O_3 exhibited a linear relationship with the cubes of the Ln^{3+} radii in these oxides, as was also the case for other nonglass-forming sesquioxides. In contrast, glass-forming oxide showed different correlations with the cubes. Therefore, molar volumes might be a good indicator of the ability of a single component oxide to form glass.

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