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(Original Paper)

Surface Oscillation Phenomena of Aerodynamically Levitated Molten Al_2O_3

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Abstract

We propose viscosity measurements of molten oxides using Aerodynamic Levitation (ADL) with CO_2-laser heating which can achieve container-less and non-contact conditions for measurements. For viscosity measurements, we used a unique vibration method that causes oscillations in a sample and levitates a gas via acoustic speakers. Using these techniques, we clarified the surface oscillation excitation mechanism and then, based on this understanding, we measured the viscosity of molten Al_2O_3 and its temperature dependence.

Keyword(s): Levitation, Thermophysical properties, Molten oxide, Surface oscillation
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1. Introduction

Understanding the thermophysical properties of molten oxides is very important in many research fields, such as materials science, material process engineering, and geological science. Molten oxide systems have an extensive range of components, and therefore there is a corresponding difficulty in predicting their properties based on chemical composition and temperature. For this reason, we must directly and precisely measure the thermophysical properties of several key components of molten oxide. However, precisely measuring the thermophysical properties of high temperature a molten oxide is hampered due to the high temperature and high chemical reactivity. Therefore, previous measurements have been limited to temperatures around 1800K [1]. For measurements exceeding this temperature, we must use a container-less approach. However, electromagnetic levitation [2] cannot be implemented due to the low electrical conductivity of the molten oxides. The electrostatic levitation (ESL) [3] technique is a possible alternative. However since ESL requires an ultra-high vacuum to avoid sparking at the electrode, the types of samples that can be used with this system on the ground are limited due to their evaporation rate in a vacuum. The density, surface tension and viscosity of molten Al_2O_3 can be measured using ESL because of its low evaporation rate in a vacuum [4]. However, the high vacuum evaporation rates of other oxide materials make measuring the thermophysical properties of their melts difficult in an ESL system on the ground.

For these reasons, we are now preparing to measure the thermophysical properties of a molten oxide composed of SiO_2-CaO-Al_2O_3, which readily evaporates in a vacuum, using the electrostatic levitation furnace (ELF) facility on the "KIBO" module of the International Space Station (ISS). The microgravity conditions on the ISS negate the need for a high vacuum in ESL since the sample can be levitated using a very small electrostatic force. However, due to the limited time allocated on the ISS, we are unable conduct a sufficient number of experiments to measure the thermophysical properties of molten oxides. Due to the above, we will use a ground based, container-less technique to measure the thermophysical properties of molten oxides. We selected the aerodynamic levitation (ADL) [5], with drop oscillation technique to take the measurements. Using the drop oscillation technique, we can simultaneously obtain surface tension and viscosity of liquid samples from the oscillation frequencies and their decay times. To generate oscillations of the levitated droplet, Langstaff *et al.* [6] and Kargl *et al.* [7] applied acoustic excitation using audio speakers. However, the excitation mechanism of droplets in ADL using an acoustically oscillating gas-jet flow for levitation is not clearly understood. Also we should confirm whether or not the drop oscillations formed using this method are the natural surface oscillations. This is because an analysis by Rayleigh [8], relates the surface tension to the normal mode of the natural oscillation of a free droplet with no external forces, and also analysis by Lamb [9], relates the viscosity to the damping time constant of the normal mode of the natural surface oscillations. For surface tension and viscosity measurements of molten oxides, the natural surface oscillation of the ADL droplet of molten oxides should therefore be induced.

In this study, in order to clarify the excitation mechanism of the surface oscillation of ADL droplets and also to confirm the oscillation type, we focused on observing oscillation phenomena of a droplet of molten Al_2O_3 in ADL using a surface oscillation generation system under several conditions.

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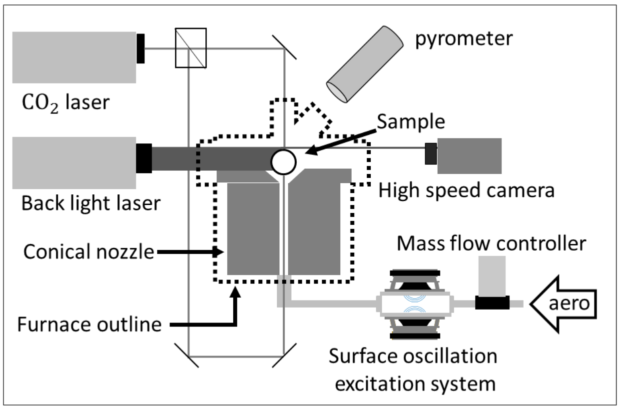

Fig. 1 Schematic of aerodynamic levitation system for measuring thermophysical properties of molten oxides.

2. Aerodynamic Levitation (ADL) System for Surface Oscillation Excitation of Molten Oxides

The aerodynamic levitation (ADL) technique is fundamentally a simple, container-less method for floating a sample in a fixed position using a gas-jet flow from conical nozzles[5)]. We applied a new acoustic oscillation system to the ADL technique to generate surface oscillations in the levitated droplets. **Figure 1** shows a schematic of our ADL system for measuring the thermophysical properties of molten oxides. Spherical solid oxide samples about 2mm in diameter were levitated in the gas-jet flow from conical nozzle, and then melted under containerless conditions by CO_2 lasers. For the thermophysical property measurements, we irradiated the samples with two CO_2 lasers from above and below to reduce the temperature gradient. The sample temperature was measured using a monochromatic pyrometer at a wavelength of 1.45μm. In order to induce surface oscillations in the ADL droplet, we applied an acoustic oscillation system to the levitation gas-jet[8)]. Two phase-matched audio speakers were set in a small chamber inserted into the gas flow path between the mass-flow controller and the conical nozzle for levitation, and a single-wavelength signal was applied to the speakers. In order to excite the surface oscillation into ADL droplet, we applied the acoustic oscillation system into the gas flow path for the gas-jet for levitation. Two phase-matched audio speakers were set in a small chamber inserted into the gas flow path between the mass-flow controller and the conical nozzle for levitation, and a single-wavelength signal was applied to the speakers. Using this system, we were able to generate surface oscillations in the ADL molten oxide droplet. The surface oscillations generated by this system were observed using a high-speed camera at 1000fps and spatial resolution of 5.3×10^{-3}mm/pix to record the shadows produced by a 539nm backlight laser system, which reduced the effects of radiation from the molten Al_2O_3.

3. Normal Mode Frequency of Natural Surface Oscillation of ADL Droplet

The shape of a levitated droplet without any external forces using time dependent droplet radius,r, is described by the flowing equation using spherical harmonics,Y_l^m: The shape of levitated droplet without any external forces using time dependent radius of droplet r is described the flowing equations using spherical harmonics Y_l^m:

$$r(\theta,\phi,t) = R + a(t)Y_l^m(\theta,\phi) \tag{1}$$

where R is the radius of an undeformed sphere and the time dependent radius of the drop,r, also depends on polar and azimuthal angles,θ and ϕ, respectively. $a(t)$ shows time dependent deformation, l and m are integers charactering the oscillation mode. From Rayleigh, the normal mode frequencies of the natural surface oscillation without viscous effects of the drop described in eq.(1) are,

$$\omega_{l,m}^2 = l(l+2)(l-1)\frac{\sigma}{\rho R^3} \tag{2}$$

here σ, r, R are the surface tension, density and undeformed radius of the droplet, respectively[8)]. The frequencies described in eq.(2) do not depend on the oscillation mode index m for small deformation from the perfect sphere. For the ADL droplet, its shape, without acoustic excitation of oscillation in the gas flow is an undeformed sphere. For the surface tension measurements, the fundamental mode of $l = 2$ is important. $l = 2$ mode is five-fold degenerate and the lowest possible surface mode. Therefore, the fundamental mode frequencies of the natural surface oscillation, $\omega_{2,0}$, or Rayleigh frequencies, are described as,

$$\omega_{\mathrm{R}}^2 = \frac{4\sigma}{\rho R^3}\,. \tag{3}$$

ω_{R} is used to obtain the surface tension from the drop oscillation method. We observed the drop oscillation in ADL droplets with molten Al_2O_3 samples. The materials properties in eq.(3) for a sample with a radius of 2.00mm are $\rho = 2.9 \times 10^3$kg/m^3 and $\sigma = 670 \times 10^{-3}$N/m. This gives an estimated surface oscillation frequency, $\omega_{\mathrm{R}}/2\pi$, of 175Hz. If we can excite a natural surface oscillation with a normal mode frequency of 175Hz in the ADL droplet of molten Al_2O_3 using the system shown in **Fig. 1**, we can apply the acoustic oscillation generator for surface tension and viscosity measurements.

4. Surface Oscillation Phenomena of ADL Droplet of Molten Al_2O_3

Surface oscillations of aerodynamically levitated molten oxides can be used to make surface tension and viscosity measurements in ESL, by modulating the voltage applied to the levitation electrodes to the normal mode frequency of natural surface oscillations (Rayleigh frequency, RF) predicted by eq.(3). By applying the resonant frequency, the natural surface oscillation is generated on ESL droplets since the vacuum negates the effect of ambient pressure on the surface of the droplet. However, the surface of an ADL droplet is pressurized by the levitation gas-jet and surface oscillations caused under these conditions are unclear. Therefore, we would like to clarify the surface oscillation phenomena on ADL droplets using the acoustic excitation system. Using the set up shown in **Fig. 1**, we observed surface oscillations on an ADL droplet of molten Al_2O_3 at a temperature of 2740K.

To observe the surface oscillations on an ADL droplet caused by a levitation gas-jet, we applied a sine wave signal from 0 to 250Hz via an audio speaker to a constant gas-jet levitating a molten Al_2O_3 sample, and the surface oscillation phenomena were observed using a high-speed camera. Time-varying surface oscillations were seen along the horizontal length of the droplet on the high speed footage taken from the side, and back-lit by a laser. We used a Fast Fourier Transfer (FFT) algorithm to obtain the surface oscillation frequencies from the time series of horizontal length changes in the droplet. **Figure 2** shows an example of results from the FFT analysis of surface oscillations for a signal of 100Hz. We found from **Fig. 2** that the surface oscillation frequency of the droplet was the same as the input frequency in this case. The surface oscillation frequencies were found to match the input frequencies, as shown in **Fig. 3**. Then, from these results, we identified that the surface oscillations matched the sine-wave frequency input from the speakers. The oscillating gas jet flow was not turned off for the duration of the experiment when obtaining these. From this we clarified that we can generate a forced oscillation on the surface of an ADL droplet via a sine-wave input to audio speakers. However, the forced surface oscillations are not natural surface oscillations due to surface tension, therefore we cannot use these oscillations to calculate surface tension and viscosity. For the forced oscillation case, the oscillation amplitude suddenly decreased when the signal to speakers was cut off. In the case of natural surface oscillations, the oscillation amplitude would be exponentially damped over time by the viscosity of the sample. In order to generate natural surface oscillations in the ADL droplet, we applied an input signal around the normal mode frequency of the natural surface oscillation (RF).

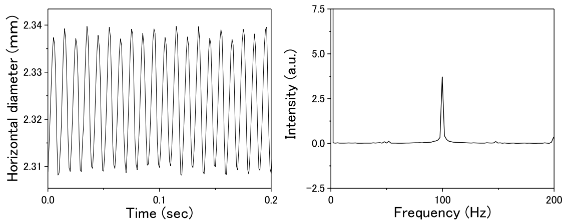

Fig. 2 Surface oscillation of ADL droplet excited by acoustic speakers at 100Hz, and its frequency, (a) surface oscillation amplitude of ADL droplet from horizontal length change with time, (b)FFT results of surface oscillation.

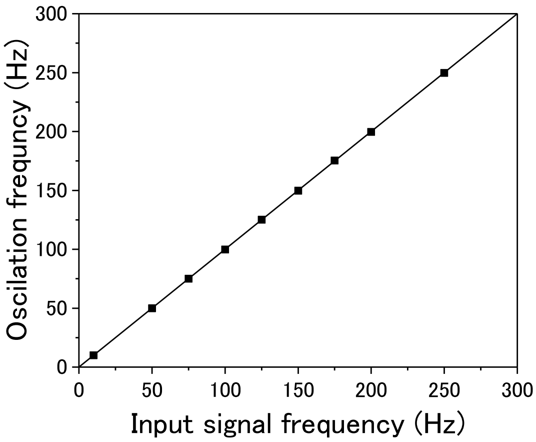

Fig. 3 Relationship between input sine wave signal frequency and surface oscillation frequency during droplet oscillation induced by acoustic speakers.

We observed the surface oscillations that meet the conditions for Rayleigh frequency (175Hz) when changing the input signal from 165Hz to 185Hz. **Figure 4** shows the results of observed surface oscillation amplitude with changing input signal frequency from 165Hz to 185Hz. The surface oscillation amplitude was normalized to the complete spherical diameter, and the data points of normalized amplitude for various input signal frequencies were fitted using the Lorentz function. From **Fig. 4** we found that the normalizes amplitude of surface oscillation was a maximum at the input signal frequency of 174Hz, which is close to the normal mode frequency of the 2.0mm diameter molten Al_2O_3 droplet at 2740K, estimated in section 3.

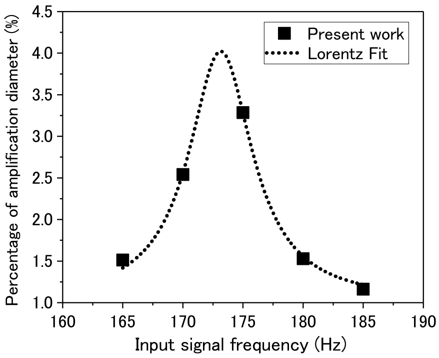

Fig. 4 Relationship between surface oscillation amplitude normalized perfect spherical diameter and input sine wave signal frequencies. Lorentz function was well fitted experiments. This case maximum peak intensity was 174Hz, which is resonant frequency of fundamental mode of natural surface oscillation. 174Hz frequency is close to the estimation frequency of surface oscillation in section3.

This change of oscillation amplitude with the applied signal frequency is characteristic behavior of the resonance forced oscillations. By generating this resonance forced oscillation, the natural surface oscillation of ADL droplet will have a resonance frequency corresponding to the normal mode frequency (Rayleigh frequency). In the present conditions, we identified that the frequency range from 165 to 180Hz induced the natural surface oscillation in ADL droplet of molten Al_2O_3. For natural surface oscillations induced by an input signal at the Rayleigh frequency, we can obtain a damped surface oscillation caused by the viscosity of droplet under conditions with no external forces. In order to confirm the natural surface oscillation of the ADL droplet, we observed the surface oscillation for an input signal of 175Hz, which is close to the resonance frequency of the natural surface oscillation. Under the present experimental conditions, when initially applying the input signal to the speakers we observed a resonantly forced oscillation, then when the input signal was terminated, we observed amplitude damping of surface oscillation with the same frequency prior to the cut-off, as shown in **Fig. 5**. During these observations, we confirmed that the natural surface oscillation induced by the oscillating levitation gas-jet flow was the fundamental oscillation mode, $l = 2$. This was determined from the changes in the droplet shape as seen from the side. For this fundamental oscillation mode, $l = 2$, the drop was seen to vary with a constant period in both the horizontal and vertical directions. Our observations agreed with the drop shape changing according to the fundamental oscillation mode, $l = 2$. If the natural surface oscillation occurs in the free oscillation conditions after the input signal has been terminated, its frequency should be independent of the input signal. In order to confirm the natural surface oscillation induced by resonant forced oscillations, we changed the input signal frequency from 160 to 185Hz, then we terminated the input signal and observed the frequency of free, damped oscillations. For these observations, we found that the surface oscillations converged to a single frequency of 174Hz with the change of input signal frequencies from 165Hz to 180Hz as shown in **Fig. 6**. 174Hz is the average oscillation frequency for input signals from 165 to 180Hz, since we identified from the curve in **Fig. 4** that the input signal of 185Hz was not the resonance forced oscillation. We conclude from the present experiments that a natural surface oscillation with a normal mode frequency of 174Hz was induced.

From these results, we clarified that the fundamental mode ($l = 2$) of natural surface oscillations of an ADL droplet is induced by the resonance forced oscillation from an audio signal close to the Rayleigh frequency applied to the levitating gas jet flow . Therefore, we can use this technique to obtain surface tension and viscosity from the surface oscillation frequency and its damping time constant.

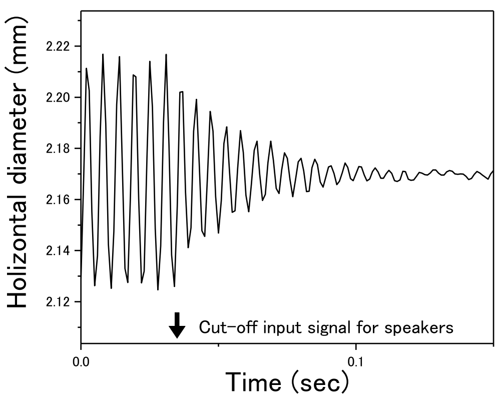

Fig. 5 Time variation of amplitude of surface oscillation before and after cut-off input sine-wave signal of 175Hz to acoustic speakers. This result shows damped surface oscillation with same frequency prior to cut-off the input signal. 175Hz is close to the resonance frequency of the natural surface oscillation.

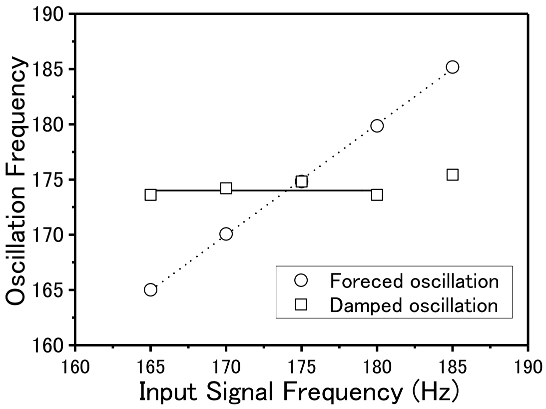

Fig. 6 Relationship between surface oscillation frequency and input signal frequency for the both case of forced oscillation during input signal applied into speakers and damped oscillation after cut-off of input signal. For damped oscillation case, surface oscillation frequencies were constant with different input signal frequencies

5. Viscosity of Molten Al_2O_3 obtained by ADL from Surface Oscillation Damping

From the above observations, we clarified the surface oscillation phenomena of an ADL molten Al_2O_3 droplet. Based on this information, we obtained the temperature dependence of viscosity for molten Al_2O_3. Surface tension was also obtained from the oscillation of the ADL droplet, but this has previously been done using the sessile drop method. We would like to discuss the details of results for the surface tension of molten Al_2O_3 in a separate paper. Here, we focused on viscosity results from the surface oscillation of the ADL droplet, because viscosity measurements with traditional methods using a container have difficulty in maintaining temperatures over 2273K. Obtaining viscosity data of molten Al_2O_3 using our method was not significant and we will report on that here. Viscosity, η, is obtained from the decay time constant, τ, of the fundamental mode $l = 2$ in the case of natural surface oscillation described as follows[9)],

$$\eta = \frac{\rho R^2}{5\tau}. \tag{4}$$

Figure 7 shows our results for the viscosity of molten Al_2O_3 together with data from studies by Langstaff *et al.*[6)] using the same method us, Paradis and Ishikawa using ESL[4)] and also the result of Rossin *et al.*[10)] using the traditional viscometer. The result of Rossin *et al.* was limited to measurements below 2500K and the viscosity value is larger than that obtained by levitation methods. The confined temperature range of Rossin's viscosity data does not allow us to describe the temperature dependence of viscosity by extrapolation to high temperature regions. On the other hand, both results for viscosity values obtained using the ADL methods with the oscillating drop technique show close values at temperatures over 2300K. Especially, in regions over 2700K our viscosity data extrapolated to higher temperatures are in good agreement with the viscosity values obtained by Langstaff *et al.*[6)] Also the viscosity values obtained by ESL with the oscillating drop method extrapolated to temperatures over 2500K agree with the viscosity values obtained by the ADL system. In the low temperature regions, dispersion of viscosity data due to measurement place and method could be accounted for by the following reasons. Since viscosity in high temperature regions is small, the decay time is considerably long. Therefore, the following damping sine curve is a good fit for the observed data;

$$a(t) = a_0 \sin(\omega_R t)\exp(-t/\tau), \tag{5}$$

where a_0 is the initial sphere radius and τ is the decay time constant. From this, we can obtain precise viscosity data using the oscillating drop method for low viscosity cases. However, in low temperature regions viscosity increases and the decay time is correspondingly short. The short decay time produce large errors when fitting the observed oscillation data to eq.(5), therefore viscosity data is not accurate in the low temperature regions. We estimated the damping time under the present conditions for molten Al_2O_3 in high temperature regions to be 2.35×10^2s at 2850K and in low temperature regions to be 1.02×10^2s at 2250K. We use high-speed cameras with a frame rate of 1000fps and a spatial resolution of 5.3×10^{-3}mm/pix to obtain the surface oscillation. In low temperature regions, the damping time was 50% that of high temperature regions, therefore the number of oscillation peaks were also about 50%. Our high-speed camera can detection changes of surface oscillations even at 2250K, but the amount of data for curve fitting was reduced in low temperature, high viscosity regions. Therefore, viscosity data is more accurate in high temperature regions. In future work on viscosity measurements using the oscillating drop method, we should improve the measurement technique in low temperature, high viscosity regions. To improve the viscosity measurements, we will using the ELF installed in "KIBO" on-board the ISS. Electro-statistically levitated droplets in microgravity is ideal for obtaining accurate surface oscillations and damping without external forces. Also under microgravity, we can use samples with larger radii, which will reduce the normal mode frequency and allow us to obtain a large number of oscillating peaks and damping oscillations. We are preparing the experiment on the ISS and hope to start collecting data from autumn, 2017. After the experiments, we can compare the data obtained on the ground with that under microgravity, then we can discuss the differences between the viscosity data obtained by viscometer and that obtained by ADL with the oscillating drop method.

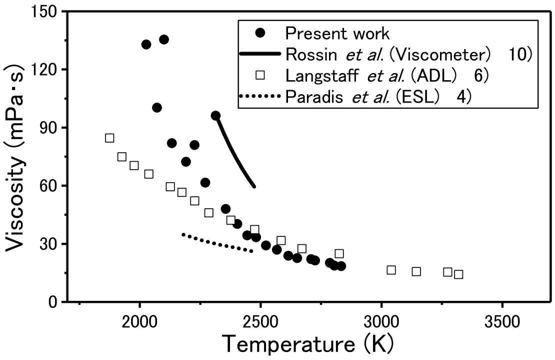

Fig. 7 Viscosity of molten Al_2O_3 with temperature variations. Previous results also showed in figure

6. conclusion

We observed the surface oscillation phenomena of ADL molten Al_2O_3. From these observation, we clarified the mechanism of surface-oscillations induced by an oscillating gas-jet flow. Based on this information, we found that the natural surface oscillation appeared after terminating the sine-wave signal input to acoustic speakers. Using the natural surface oscillation, we could obtain surface tension and viscosity. We obtained the temperature dependence of molten Al_2O_3 viscosity and decay time of the natural surface oscillations using this system. The results agreed with previous reports using the same method in high temperature low viscosity regions. This means that the decay time of the surface oscillations in low viscosity cases is sufficiently long to obtain a precise value for the viscosity, whereas the opposite is true for high viscosity cases. These viscosity data sets were smaller than those obtained using the traditional viscometer. However, the data from the traditional viscometer is limited to only low temperature regions, therefore a direct comparison and subsequent discussion cannot be made. For future work, we should clarify the reasons for differences in viscosity data obtained from the decay of surface oscillations and from the viscometer. After this consideration, we can evaluate the accuracy of molten Al_2O_3 viscosity results obtained from this measurement system. However, for surface tension we can apply the method precise measurements, and should therefore try to measure the temperature dependence on surface tension of molten oxides and also the dependence on atmospheric conditions.

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