

Thermophysical properties of dense molten Al_2O_3 determined by aerodynamic levitation

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Abstract

Aerodynamic levitation (ADL) is a widely used contactless technique for evaluating the thermophysical properties of molten materials. During data analysis, these molten samples are typically assumed to be bubble-free and to adopt an oblate spheroid shape. However, these assumptions have not been empirically validated. The presence of trapped bubbles and potential deformation at the bottom of the levitated sample, which is obscured from view by the levitation nozzle, can lead to an underestimation of the sample's density. These factors could also interfere with the sample's damped oscillation, thereby affecting the reliability of the measured surface tension and viscosity. Consequently, in this study, we address these issues by using smaller alumina samples to minimize sample deformation and by inspecting each sample's cross-section after cooling for trapped bubbles. This approach enabled us to mitigate the impact of these potential distortions on the measured thermophysical properties of molten alumina. Our findings reveal that the density of dense molten alumina can be expressed as $2.917-1.228 \times 10^{-4}(T-2327)$ [g cm^{-3}] from 1978 to 2789 K. Additionally, between 2375 and 2770 K,

we determined the surface tension of alumina to be $0.632\text{--}2.310 \times 10^{-5}(\text{T}-2327)[\text{N m}^{-1}]$ and the viscosity to be $0.577\text{e}^{9.743 \times 10^3/\text{T}}[\text{m Pa s}]$.

Keywords: aerodynamic levitation, molten alumina, density, surface tension, viscosity, bubbles

1 Introduction

Evaluating the thermophysical properties of high-temperature liquids presents a significant challenge due to the need for precise measurements without sample contamination. In general, experimental methods can be divided into contact (with a container)[1–6] and contactless techniques[7–14]. Contact methods, such as the Archimedes’ method[1, 2], sessile drop method[3, 4], maximum bubble pressure method[5], and oscillating cup method[6], have been utilized to investigate molten metals and oxides. However, these techniques become increasingly challenging to implement at extremely high temperatures due to the risk of sample contamination. Therefore, contactless methods have become more desirable due to their reliability. These include electrostatic levitation (ESL)[7–9], electromagnetic levitation (EML)[10, 11], and aerodynamic levitation (ADL)[12–14] techniques that levitate the sample during measurements. As suggested by their names, ESL, EML, and ADL use electrostatic, electromagnetic, and aerodynamic forces, respectively, to levitate the sample. Due to the differences in their operating principles, EML is limited to levitating electrically-conducting materials, and while ESL is primarily used to study molten metals[15, 16], it has also been employed on molten oxides[17–20]. Among the three levitation methods, ADL is considered the most suitable technique for levitating molten oxides, such as UO_2 and ZrO_2 , because oxygen-mixed levitation gases can be used to prevent sample reduction at elevated temperatures when necessary.

Levitation techniques can be used to evaluate several key thermophysical properties, including density, surface tension, viscosity, and heat capacity[7, 13, 21–23]. Among these properties, density is a fundamental parameter required for calculating the excess volume, viscosity, or thermal conductivity of a material. Consequently, uncertainties or errors in density measurements can propagate to these other properties. To demonstrate the accuracy of a given levitation measurement system, standard materials such as platinum or alumina are often measured before other target materials[13, 17, 24–27]. For density evaluations, the major source of uncertainty arises from pixel identification along the sample’s circumference, yielding a $\frac{\Delta V}{V} = \frac{3\Delta R}{R}$ term[28], where V and R represent the sample’s volume and radius, respectively. In general, levitation studies can achieve an uncertainty of less than 1-2% in density measurements[13, 17, 26, 27]. However, in the case of alumina, its measured density via levitation experiments (ESL and ADL) at the melting point of 2327 K differs by as much as 5.3 % [17, 19], suggesting the presence of other overlooked sources of error.

A potential source of error in these measurements could be the common assumption that the levitated sample is bubble-free (i.e. 100% dense). This assumption, often made when analyzing a sample’s thermophysical properties, has not been empirically

validated through direct observation of the cross-section of the levitated sample after solidification, or by in-situ observation of bubble inclusions using methods such as X-radiography. The validity of this assumption is questioned by a recent ESL study on alumina conducted by Tamaru et al.[19], in which they reported a density deviation of approximately 2% between two molten alumina spheres measured with the same setup. They attributed this discrepancy to potential voids that are difficult to eliminate under microgravity. Although the exact formation mechanism of these bubbles remains unclear, they could potentially form during sample preparation or heating (movie showing pores within a melting alumina sample is provided in the supplementary information), leading to an underestimation of the density value. This source of error could potentially explain the observed scatter in measured density values. As a result, the impact of trapped bubbles on the measurement accuracy of contactless techniques like ADL warrants further scrutiny.

Furthermore, the sample levitated by ADL is commonly assumed to adopt an oblate spheroid shape, and its perimeter is typically fitted with an ellipse during data analysis[13, 24, 25]. However, this assumption may not always be valid. Specifically, for molten oxides with lower surface tension than liquid metals, the pressure exerted by the levitation gas on the sample’s bottom can cause its shape to deviate from a perfect oblate spheroid. This deviation can potentially influence both the fitted sample perimeter and its oscillation. Therefore, in addition to confirming the absence of trapped bubbles, it is also crucial to investigate potential sample deformation when accurately characterizing the properties of molten materials using ADL. Nevertheless, quantifying the degree of deformation during ADL experiments is particularly challenging due to the partial obscuring of the sample’s bottom half by the levitation nozzle from the camera’s perspective.

In response to the issues outlined above, this study primarily aims to confirm the presence of trapped bubbles in the levitated samples by observing the cross-sections of solidified alumina samples via microscopy after levitation. To mitigate the effect of deformation, we used smaller starting samples, which require less levitation force and exhibit higher internal pressure (Young-Laplace), making them more resistant to deformation. We selected molten alumina as our target material due to its high-temperature stability and the extensive documentation of its thermophysical properties in both ADL [13, 24, 25, 29–31] and ESL [17, 19, 32] studies. Consequently, we report the measured density, surface tension, and viscosity of dense molten alumina samples with minimal deformation. We further compare these results with values from the literature to assess the impact of bubble inclusion and deformation on the accuracy of thermophysical measurements using ADL.

2 Methods

2.1 Sample Preparation

The alumina samples used in the ADL experiments were prepared by spark plasma sintering alumina powder (4N, Kojundo Chemicals) under an argon flow. The bulk samples were sintered at 1673 K under a pressure of 100 MPa and held for 5 minutes. Two alumina pellets with relative densities of 98.3% and 99.1% were fabricated; the

theoretical density of the sample was calculated from its X-ray diffraction patterns. These pellets were then crushed into smaller pieces for the ADL experiments. Figure 1 shows images of a prepared bulk alumina sample and a solidified spherical sample post-ADL. The cross-sections of the samples after the ADL experiments were analyzed using scanning electron microscopy (SEM, JSM-7500F, JEOL).

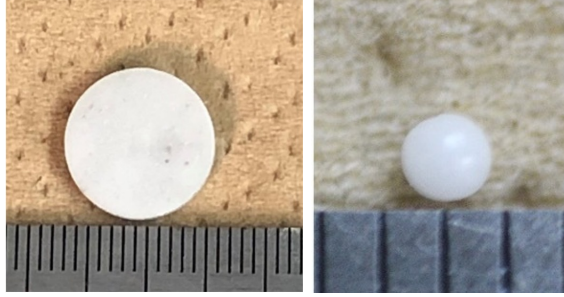


Fig. 1 (a) Bulk alumina sample prepared via spark plasma sintering (b) Spherical alumina sample after ADL experiments

2.2 Aerodynamic Levitator

Figure 2 presents a simplified schematic of the aerodynamic levitator used in this study. To prevent the reduction of Al_2O_3 at high temperatures, the sample was levitated using Ar and O_2 gases at flow rates of 550 and 220 mL min^{-1} , respectively. The sample was melted using a top and bottom CO_2 laser (10.6 μm , maximum output of 100W, GEM100 Coherent), and its temperature was monitored using a single-color pyrometer (0.9 μm , IR-CAI8CNL, CHINO). The pyrometer's signal was collected every 1 ms using a data logger (midi LOGGER, GRAPHTEC). The levitated sample was illuminated with a UV background light source (LC-L1V5, Hamamatsu), and shadowgraphs were captured using a high-speed camera (MEMRECAM HX-7s, nac Image Technology) equipped with a bandpass filter (365 ± 10 nm, Edmund optics) and a telecentric lens (1.0X, 2/3", Gold TL, Edmund optics). The shadowgraphs were recorded at a rate of 4000 fps, with a resolution of 1024×928 and a shutter speed of 1/50000. During the oscillating drop experiments, a sinusoidal wave function was generated by a function generator (AWG1005, As One), amplified (A-10 Pioneer), and transmitted to the levitation gas via a speaker (PLS-P830985, Tymphany) to induce the $l = 2$ mode oscillations in the levitated sample. For additional information about the aerodynamic levitator setup, refer to our previous works[24, 33, 34].

2.3 Temperature Analysis

The temperature curve measured directly from the experiment is corrected using the following equation derived from the Wien approximation[35]:

$$\frac{1}{T_{\text{corrected}}} - \frac{1}{T_{\text{measured}}} = \frac{1}{T_{\text{m,real}}} - \frac{1}{T_{\text{m,measured}}} \quad (1)$$

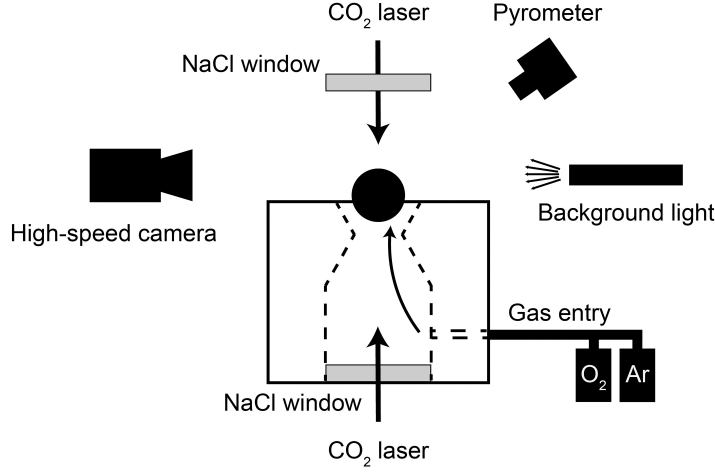


Fig. 2 Schematic of the aerodynamic levitator used in this work.

where $T_{\text{corrected}}$ is the corrected temperature, T_{measured} is the directly measured temperature by the pyrometer, $T_{\text{m,real}}$ is the sample's melting point, and $T_{\text{m,measured}}$ is the measured temperature by the pyrometer at the sample's melting point. $T_{\text{m,measured}}$ is determined by finding the measured temperature at the end of recalescence, as shown in Figure 3. The melting point $T_{\text{m,real}}$ of alumina is taken as 2327 K[36].

2.4 Image and Data Analysis

The recorded shadowgraphs were analyzed using the DIPP-Macro II software. Given that the nozzle blocked the bottom half of the levitated sample during ADL, an elliptical fitting was applied to the sample's visible perimeter to extrapolate its major and minor axes. An example of this fitting process is shown in Figure 4.

To determine the sample's volume in SI units, we first established a relationship between pixels and millimeters by measuring the diameters of levitated stainless steel balls with known diameters, as depicted in Figure 5. The temperature dependence of the sample's density was obtained by analyzing each frame of the sample's shadowgraphs as it cooled during levitation, as later illustrated in Figure 7.

To calculate the surface tension at a specific temperature, we first performed a Fast Fourier Transformation (FFT) on the oscillation pattern to extract information in the frequency domain. Subsequently, we applied a Lorentzian fit to the dominant resonant peak to precisely locate the resonance frequency ν_R of the $l = 2$, $m = 0$ mode. Additionally, for viscosity calculations, the oscillation pattern was fitted with a damped sine function to determine the damping constant, τ .

$$r = Ae^{-t/\tau}\sin(2\pi\nu_R(t - t_0)) + r_0 \quad (2)$$

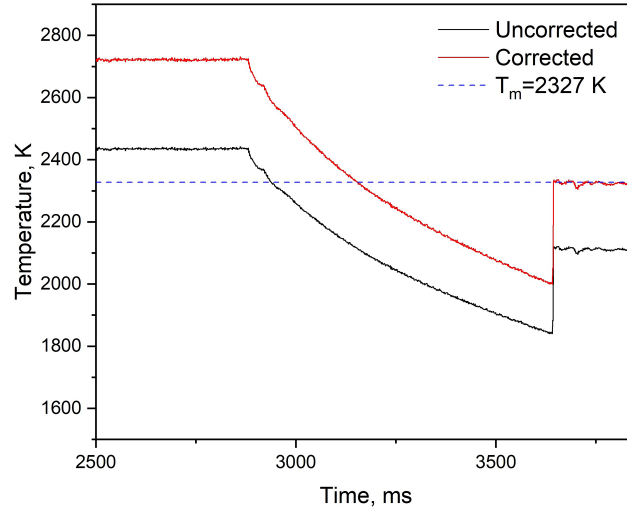


Fig. 3 Temperature correction based on Wien approximation. The corrected temperature curve $T_{\text{corrected}}$ is shown in red, and the uncorrected, directly measured temperature curve T_{measured} is shown in black.

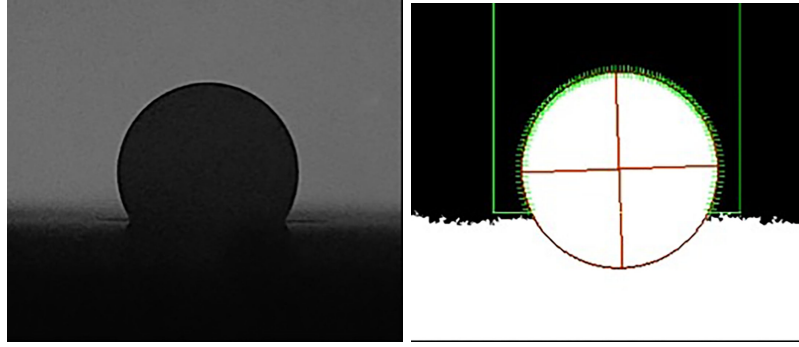


Fig. 4 Shadowgraph analysis process. (Left) Original captured shadowgraph image of the sample. (Right) Image post-analysis using DIPP-Macro II.

In this equation, r represents the length of the horizontal semi-axis, A is the amplitude, t denotes time, τ is the damping constant, ν_R is the fitted resonant frequency, $t - t_0$ accounts for the phase shift, and r_0 is the sample's radius before oscillation.

Unlike density measurements, which occur continuously as the sample cools, the temperature dependence of surface tension and viscosity are investigated by observing the sample's oscillation at several selected temperatures. For a spherical sample undergoing slight damped small-amplitude oscillations, its surface tension and viscosity can be expressed using Rayleigh's and Lamb's equations[37, 38] for an $l = 2$ mode oscillation:

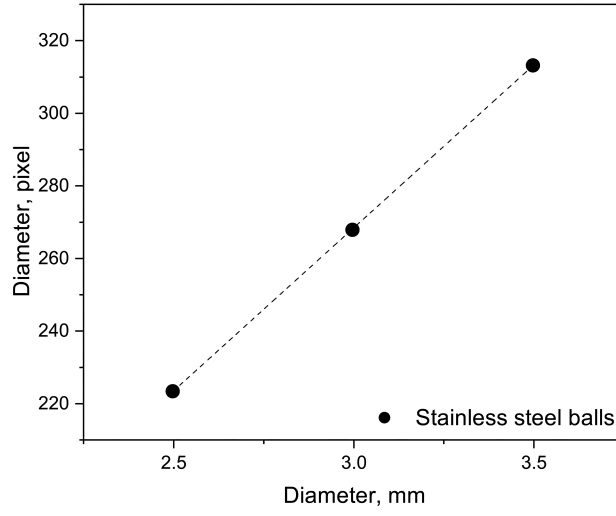


Fig. 5 Relationship between pixel and millimeter determined using stainless steel balls

$$\sigma = \frac{3}{8}\pi\nu_R^2 M \quad (3)$$

$$\eta = \frac{\rho r^2}{5\tau} \quad (4)$$

Here, σ is the surface tension, ν_R is the resonant frequency of the $l = 2$ mode, M is the sample mass, η is the viscosity, ρ is the density, r is the sample's radius, and τ is the oscillation's damping constant. The least-squares method was used to fit the sample's temperature-dependent surface tension and viscosity to the following equations:

$$\sigma = \sigma_{T_m} + \frac{\partial\sigma}{\partial T}(T - T_m) \quad (5)$$

$$\eta = Ae^{b/T} \quad (6)$$

Here, σ_{T_m} is the surface tension at the melting point, T_m is the melting point, and A and b are fitting parameters.

3 Results and Discussion

3.1 Cross-sectional SEM Analysis

Following levitation, we examined the cross-sections of the solidified samples using SEM to identify any cavities indicative of trapped gas bubbles during levitation and shrinkage cavities during solidification. A comparison of the alumina samples' cross sections, as shown in Figure 6, revealed a prominent cavity at the center of sample no. 3, while the cavities observed in samples no. 1 and 2 were notably smaller and irregular in shape. This leads us to postulate that trapped gases led to the formation of the central cavity in sample no. 3 while the effect is negligible in samples no. 1 and 2. Interestingly, these differences emerged even though all three samples underwent nearly identical heating and levitation conditions and experienced a single heating and cooling cycle.

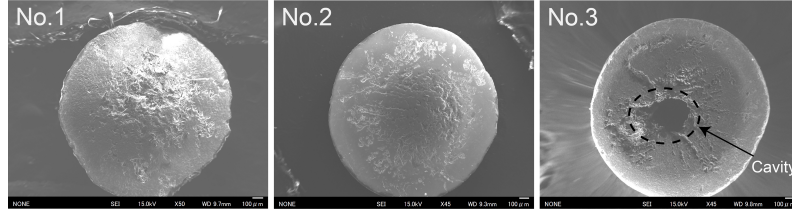


Fig. 6 Cross-sectional SEM images of alumina samples. Samples no. 1 and 2 show no noticeable cavities, while a prominent cavity is observed in sample no. 3.

3.2 Density

The calculated densities of molten alumina samples no. 1–3 are presented in Figure 7. At alumina's melting point (2327 K), the measured density of sample no. 3 is 3.5% lower than those of samples no. 1 and 2. The linear regression lines for the measured densities of samples no. 1 and 2 show minimal deviation (a maximum of 0.4% at 2789 K), a consistency we attribute to both samples being similarly sized and bubble-free, as evidenced in Figure 6. Consequently, we can express the temperature-dependent density of dense molten alumina samples from 1978 to 2789 K based on the combined density data of samples no. 1 & 2 as:

$$\rho(\text{g cm}^{-3}) = 2.917 - 1.228 \times 10^{-4}(T - 2327) \quad (7)$$

The temperature-dependent density of molten alumina reported in this study is compared with values from the literature[13, 17, 19, 24, 25, 31] in Figure 8. Our findings align closely with the data reported by Paradis et al.[17] who used ESL, exhibiting a maximum deviation of only 0.46% between 2175 and 2435 K. The density reported by Paradis et al.[17] is the highest among the existing literature, suggesting that their samples likely contained the least amount of trapped bubbles. We attribute this to the high-vacuum environment (10^{-5} Pa) used by Paradis et al.[17], which would likely have degassed the molten droplets. Additionally, the full visibility of the

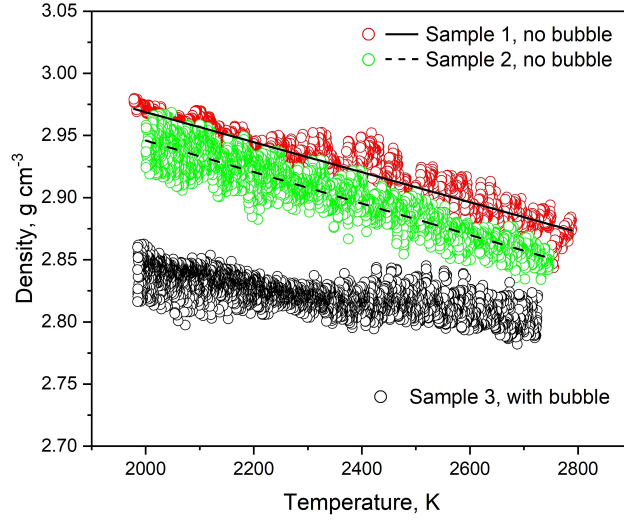


Fig. 7 Temperature-dependent densities of molten alumina samples no. 1, 2, and 3. Straight and dashed lines represent linear regressions for samples no. 1 and 2, respectively.

levitated sample in ESL allows for accurate volume determination, which likely also contributed to the high alumina density measured by Paradis et al. This agreement in the measured density of molten alumina between this work and that by Paradis et al.[17] validates our method of examining the solidified samples for bubbles and using smaller samples to avoid deformation, thereby obtaining reliable density data. Despite also using ESL, Tamaru et al.[19] conducted their experiment under microgravity which, according to their own observations, made it difficult to remove potential voids within the samples. This could potentially explain why their highest density value is lower than that reported by Paradis et al.[17].

Among the studies performed with ADL, only Langstaff et al.[13] took additional measures to reduce bubble formation by using 2 mm-diameter ruby spheres as starting materials. When compared to the density of molten alumina reported by Langstaff et al., the density of sample no. 3 from our study is much lower, despite similarities in sample size. This suggests that the large cavity observed in sample no. 3 (Figure 6) could be partially attributed to trapped bubbles, which would lead to an overestimated sample volume during measurements. Interestingly, our smaller, bubble-free samples demonstrate higher densities than those reported by Langstaff et al., as seen in Figure 8. If we assume that both sets of samples are bubble-free, this discrepancy could result from an overestimated sample volume due to the deformation of larger samples. Unlike the ruby spheres used by Langstaff et al., our smaller samples (1.5 mm in diameter) require less force for stable levitation and exhibit a greater pressure difference compared to their surroundings because of their smaller radius. This makes the hidden bottom area of these smaller samples more resistant to deformations like

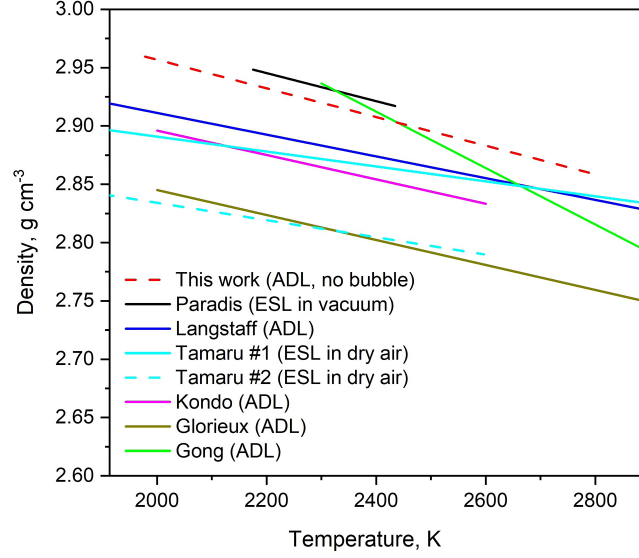


Fig. 8 Temperature-dependent density of molten alumina

flattening. Consequently, it significantly reduces the overestimation of volume that arises from assuming a non-deformed oblate spheroid shape.

Glorieux et al.[31] observed the levitated sample from the top and assumed that smaller samples (those smaller than 78 mg) were spherical. However, due to their relatively large sample size and the lack of a side-view observation, it is plausible that the samples might have been oblate spheroids rather than perfect spheres. This could have led to an overestimation of the sample's volume and, consequently, an underestimation of its density. Among the density data measured with ADL, Gong et al.[25] reported the highest values at lower temperatures. However, Gong et al. did not employ the shadowgraph imaging technique and recorded the incandescent sample against a dark background, which makes it difficult to precisely determine the sample's volume[8]. As the sample's luminosity increases with temperature, the blooming effect can make the bright sample appear larger against the dark background. This results in a steeper temperature-dependence compared to data obtained with shadowgraphs[13, 17, 19, 24], as evident in Fig 8. Consequently, at lower temperatures, where the sample's brightness is reduced, the sample might appear smaller as it is difficult to differentiate between the sample's perimeter from the dark background, leading to an overestimation of its density.

In summary, previous studies[13, 19, 24, 25, 31] may not have adequately accounted for factors such as bubble entrapment or sample deformation during levitation. This oversight could potentially explain why the reported densities are lower compared to those from our study and that of Paradis et al.[17]. To enhance the precision of

density measurements in future studies employing ADL, we recommend the implementation of post-experiment cross-sectional examinations or the use of in-situ detection techniques like X-radiography, especially for oxide samples. These measures can help identify potential bubbles that might otherwise lead to an underestimation of density. Furthermore, utilizing smaller samples could mitigate sample deformation, thus further enhancing the accuracy of density measurements with ADL.

3.3 Surface Tension and Viscosity

3.3.1 Sample Selection

For the accurate assessment of the sample’s surface tension and viscosity, we selected samples weighing less than 10 mg. This approach was adopted to minimize the impact of sample deformation on oscillation. We then identified potential bubble-containing samples by comparing the experimentally measured density of these samples with the density computed using Equation 7, which we consider as the density of bubble-free samples. Only those samples whose measured densities deviated by less than 0.5% from the value given by Equation 7 were included in our surface tension and viscosity calculations.

3.3.2 Resonance Scan

In order to reliably induce the $l = 2$, $m = 0$ mode oscillation in samples within a specified weight range (e.g., 10-15 mg) for oscillating drop experiments, we first estimated their resonance frequencies via a resonance scan. A resonance scan involves systematically varying the input signal’s frequency while capturing the sample’s horizontal oscillations, as depicted in Figure 9 (left). The derived FFT spectrum from the sample’s oscillation, shown in Figure 9 (right), features a prominent peak at approximately 270 Hz and its overtone near 540 Hz. Despite the high FFT resolution (approximately 1 Hz), there is no discernible peak splitting for the $l = 2$ mode, unlike the pronounced peak splitting observed in EML experiments due to significant sample deformations [39, 40]. The absence of peak splitting in our ADL experiments is attributed to the minimal deformation experienced by our smaller alumina samples. Their low weight requires only a small levitation force, which, along with their high internal pressure due to their small sizes, makes them less prone to deformation.

3.3.3 Surface Tension

The damped oscillation pattern of a molten alumina sample and its Fourier-transformed spectrum are presented in Figure 10. A Lorentzian fit to the spectrum reveals a single dominant resonant peak at approximately 230 Hz, which we attributed to the $l = 2$, $m = 0$ oscillation mode. Unlike resonance scans that allow for extensive data collection, the FFT spectrum’s frequency resolution in Figure 10 is constrained by the rapid decay of oscillation amplitude in our smaller samples, thereby limiting the number of data available for a detailed analysis. Despite the sub-optimal frequency resolution of the damped oscillation’s FFT spectrum, our peak assignment was informed by the resonance scan results illustrated in Figure 9, because the levitation setup

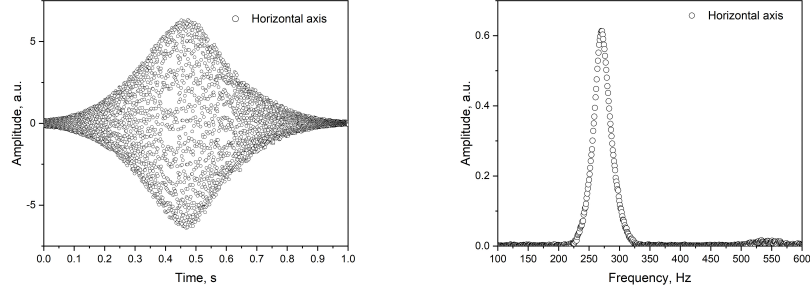


Fig. 9 Horizontal-axis oscillation pattern of a molten alumina sample (left) and the derived FFT spectrum (right) during a resonance scan.

remained identical during resonance scans and oscillating drop experiments. Furthermore, during oscillating drop experiments, we manually initiated the $l = 2, m = 0$ mode and visually confirmed its presence prior to the onset of oscillation damping. Therefore, this mode is expected to exhibit the greatest amplitude at the beginning of damping. To further enhance the frequency resolution of our FFT analysis, we plan to separate the measurement processes for surface tension and viscosity. Specifically, resonance scans will be used to accurately identify resonance peaks for surface tension measurements, while the oscillation's damping will be used for viscosity analysis.

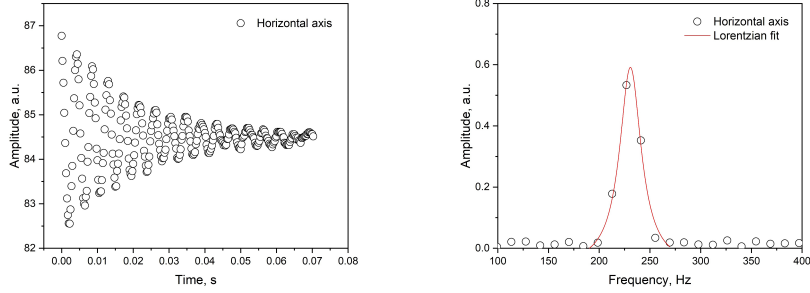


Fig. 10 Horizontal-axis oscillation pattern of a molten alumina sample (left) and the derived FFT spectrum (right) during oscillating drop experiments.

The surface tension of molten alumina obtained in this study, along with values reported in the literature, is illustrated in Figure 11. The error bars for surface tension are based on the calculations discussed in section 3.4. In contrast to the density data for liquid alumina, surface tension values show less variation across different studies. The surface tension of molten alumina measured in this study between 2375 and 2770 K exhibits a weak temperature dependence and aligns well with values from the literature[13, 18, 30, 32]. For molten alumina samples without bubbles, the temperature-dependent surface tension can be expressed as:

$$\sigma(\text{N m}^{-1}) = 0.632 - 2.310 \times 10^{-5}(T - 2327) \quad (8)$$

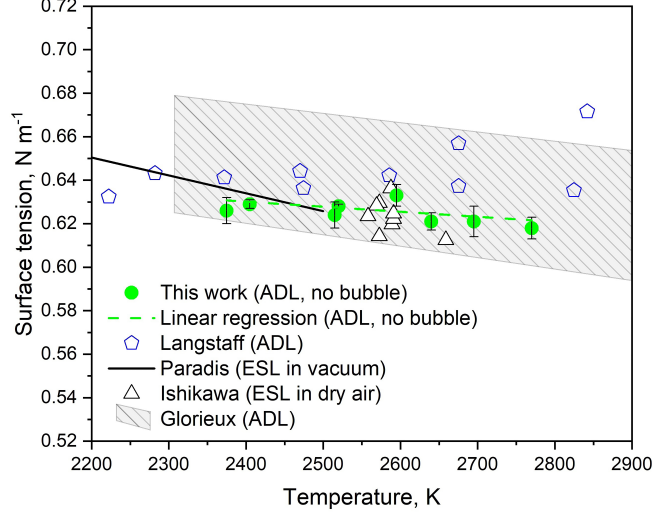


Fig. 11 Temperature-dependent surface tension of molten alumina.

However, the influences of trapped bubbles and sample deformation on oscillation are more complex than their impacts on density, rendering the quantitative evaluation of discrepancies in reported surface tension values across different studies a challenging task. Although a comprehensive analysis of these effects extends beyond the scope of this discussion, we can offer some insights based on the derivation of Rayleigh's equation (Equation 3). This equation, used for calculating surface tension, is established on the principle of energy conservation for a spherical droplet and describes how the kinetic energy of the liquid's oscillatory motion compensates for the potential energy change due to the creation of new surface areas [37]. Therefore, if a sample deviates significantly from the ideal spherical shape, it might necessitate adjustments to the energy terms, potentially undermining the reliability of Equation 3 in determining surface tension. Moreover, if a large bubble is trapped in the sample, it introduces a new interface and additional kinetic and potential energy terms. In the case of such a 'compound drop' with concentric fluid systems, Saffren et al. [41] demonstrated that its resonant frequencies are functions of the unperturbed Rayleigh frequency. Our FFT analysis, depicted in Figure 10, did not reveal any frequency splitting, suggesting that our samples were not subject to the complexities introduced by bubbles. This observation aligns with our assertion that the alumina samples were bubble-free. Detailed investigation into peak-splitting phenomena caused by bubbles in ADL experiments is an area for future research, necessitating higher resolution in FFT analysis.

3.3.4 Viscosity

The viscosity of molten alumina, calculated using Equation 4 and the measured densities, is compared with values from the literature in Figure 12. The error bars for viscosity are derived from the uncertainty analysis detailed in section 3.4. For bubble-free molten alumina samples, their temperature-dependent viscosity, from 2375 to 2770 K, can be expressed using Andrade’s equation[42]:

$$\eta(\text{m Pa s}) = 0.577e^{9.743 \times 10^3 / T} \quad (9)$$

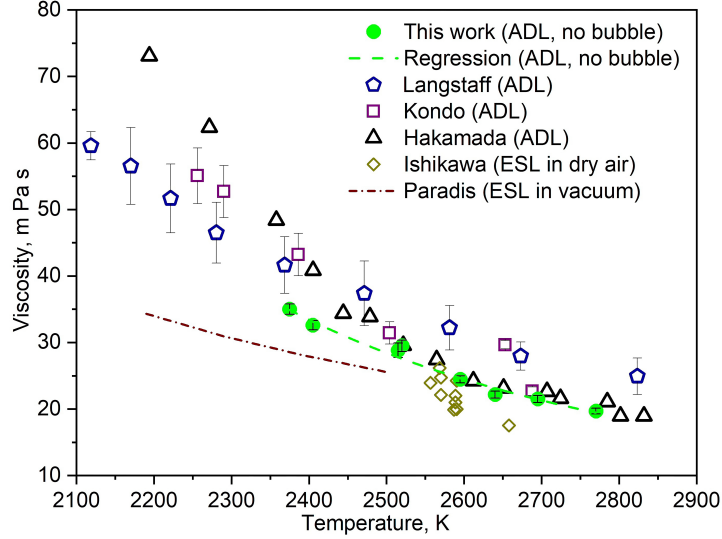


Fig. 12 Temperature-dependent viscosity of molten alumina.

In comparison with literature values, the viscosity of our dense molten alumina samples aligns closest to the minimum reported values at each temperature and is in agreement with measurements conducted using ESL by Paradis et al.[18] and Ishikawa et al.[32]. Across various studies, the reported viscosity data for molten alumina across seems to scatter more widely as temperature decreases, with dispersion exceeding 50% in undercooled samples[13, 18, 24, 29]. This increased scattering is speculated to stem from the negative correlation between viscosity and temperature. At lower temperatures, the sample’s increased viscosity leads to quicker termination of induced oscillations. This quick termination limits the amount of data available for curve fitting, thereby increasing the uncertainty in the measured data.

To gain a deeper understanding of how trapped bubbles and deformation might impact measured viscosity, we start by analyzing Equation 4, which can be rewritten as $\eta \propto M/r\tau$. In this equation, the sample’s viscosity is inversely related to its measured radius r and the damping constant τ . As discussed in section 3.2, the presence of trapped bubbles and sample deformation could result in an overestimation of the

sample's volume and thus an increase in its apparent radius. However, quantifying the influence of these factors on the damping constant is more challenging. For instance, Li et al.[43] have demonstrated that the damping constant of an oscillating liquid shell (e.g. molten alumina with a gas bubble) is determined by its Ohnesorge number and the radius ratio between the shell (molten alumina) and the core (air). Furthermore, in the event where the in-phase and out-of-phase modes[41] are both oscillatory, the observed oscillation decay during experiments would then even be a combination of their respective damping constants. Consequently, given the multitude of interacting variables, accurately quantifying the combined effects of trapped bubbles and sample deformation on measured viscosity based solely on experimental evidence presents a formidable challenge, warranting further in-depth simulation or theoretical studies.

3.4 Uncertainty Analysis

3.4.1 Density

The relative uncertainty of the measured density ($\Delta\rho/\rho$) can be expressed as:

$$\frac{\Delta\rho}{\rho} = \sqrt{\left(\frac{\Delta M}{M}\right)^2 + \left(\frac{\Delta V}{V}\right)^2} \quad (10)$$

Here, $\Delta M/M$ and $\Delta V/V$ are the relative uncertainties of the sample's measured mass and volume, respectively. For a spheroid sample, Equation 10 can be rewritten in terms of its semi-major (a) and semi-minor (c) axes as:

$$\frac{\Delta\rho}{\rho} = \frac{\Delta V}{V} = \sqrt{\left(2\frac{\Delta a}{a}\right)^2 + \left(\frac{\Delta c}{c}\right)^2} \quad (11)$$

The mass uncertainty term $\Delta M/M$ is excluded in Equation 11 due to the sample's negligible mass change ($< 0.1\%$) and its minor contribution to the overall uncertainty. Additionally, we selected the uncertainty in the detected edge points as ± 0.5 pixel (px) when calculating $\Delta a/a$ and $\Delta c/c$. Among the three samples shown in Figure 7, we only evaluated the uncertainty for samples 1 and 2, because sample 3 contained bubbles. Given the high density of data points in Figure 7, individual error bars are omitted for clarity. Consequently, the maximum $\Delta\rho/\rho$ was calculated to be 1.5% and 1.4% for these two samples, as shown in Table 1, encompassing all measured data points.

Table 1 Maximum uncertainty in the measured density of molten Al_2O_3

No.	Δa , px	a , px	Δc , px	c , px	$\Delta\rho/\rho$, %
Sample 1	0.5	77	0.5	73	1.5
Sample 2	0.5	80	0.5	79	1.4

One of the inherent challenges in ADL experiments is the obstruction of the sample's bottom by the levitation nozzles, which can introduce uncertainties in determining the sample's shape and volume. Furthermore, adjustments may be necessary for the expression of $\Delta V/V$ in error propagation to accurately reflect the sample's true shape. Currently, these sources of error are not addressed in Equation 11. To mitigate this issue, we used smaller samples for levitation, as guided by the Young-Laplace law, which states that smaller spheres are less susceptible to deformation due to their higher internal pressure in relation to surface tension. In addition, the relatively low density of molten alumina allows for levitation with minimal force, further reducing the risk of deformation. Consequently, even though the bottom portion of the sample was not directly observable, the resistance to deformation and the lower weight of smaller samples likely ensured a more accurate elliptical shape, thereby minimizing the error in density measurements that might arise from potential shape deviations.

3.4.2 Surface Tension and Viscosity

The uncertainty propagation equation for surface tension σ and viscosity η can be expressed as follows:

$$\frac{\Delta\sigma}{\sigma} = 2 \frac{\Delta\nu_R}{\nu_R} \quad (12)$$

$$\frac{\Delta\eta}{\eta} = \sqrt{\left(\frac{\Delta\rho}{\rho}\right)^2 + \left(2\frac{\Delta r}{r}\right)^2 + \left(\frac{\Delta\tau}{\tau}\right)^2} \quad (13)$$

$\Delta\nu_R$, $\Delta\rho$, Δr , $\Delta\tau$ are the standard deviations of resonant frequency, density, radius, and oscillation decay constant. The uncertainty in the sample's mass has negligible contribution to the total uncertainty and is therefore omitted in Equation 12. $\Delta\nu_R$ is determined by peak-fitting the FFT spectrum with Lorentzian functions, $\Delta\rho$ and Δr are calculated by considering the uncertainty in the sample's edge detection, and $\Delta\tau$ is obtained from fitting the damped oscillation pattern. The contribution from each error source for every data point in Figures 11 and 12 is presented in Tables 2 and 3.

Table 2 Propagated uncertainty in the measured surface tension of molten Al_2O_3

Temperature, K	ν_R , Hz	$\Delta\nu_R$, Hz	$\Delta\nu_R/\nu_R$, %	$\Delta\sigma/\sigma$, %
2595	230.2	0.9	0.4	0.8
2520	238.9	0.2	0.1	0.2
2770	237.2	0.9	0.4	0.8
2515	238.3	1.1	0.5	0.9
2640	238.2	0.8	0.3	0.7
2405	239.8	0.3	0.1	0.3
2375	239.3	1.1	0.5	0.9
2695	238.6	1.3	0.5	1.1

Table 3 Propagated uncertainty in the measured viscosity of molten Al_2O_3

Temperature, K	$\Delta\rho/\rho$, %	$\Delta r/r$, %	$\Delta\tau/\tau$, %	$\Delta\eta/\eta$, %
2595	1.3	0.59	1.2	2.1
2520	1.4	0.61	2.3	2.9
2770	1.4	0.61	1.3	2.2
2515	1.4	0.61	3.3	3.7
2640	1.4	0.61	1.6	2.4
2405	1.4	0.61	1.1	2.1
2375	1.4	0.61	1.2	2.2
2695	1.4	0.61	1.7	2.5

4 Conclusion

ADL has found extensive use in determining the density, surface tension, and viscosity of molten oxide samples. Typically, these molten samples are assumed to be free of bubbles and shaped as oblate spheroids, which may not always be accurate. In this study, we examined the cross-sectional SEM images of solidified molten alumina samples to confirm they are free of trapped bubbles. Furthermore, we used small alumina samples with a diameter of 1.5 mm to mitigate potential deformations to the sample's bottom part induced by the levitation gas. The density, surface tension, and viscosity of these small, bubble-free alumina samples were then measured and compared with values from the literature.

Our findings suggest that the presence of trapped bubbles and potential partial sample deformation can lead to an underestimation of molten sample's density. However, the exact influence of these two factors on the measured surface tension and viscosity could not be conclusively determined based on our experimental data alone. Future theoretical or simulation studies focusing on the oscillatory behavior of a droplet could provide a more in-depth quantitative evaluation. Nevertheless, our study highlights the importance of mitigating bubble formation and potential deformation in molten samples used in ADL experiments in order to precisely determine their thermophysical properties.

Statements and Declarations

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Data Availability

The datasets generated during and/or analysed during the current study are available from the corresponding author, Y. Sun, on reasonable request.

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