

## Density Measurement of Molten Oxides of $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$ System by Aerodynamic Levitated Technique

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### Abstract

In this study, we measured the density of a molten oxide of  $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$  system using Aerodynamic Levitation (ADL) with  $\text{CO}_2$ -laser heating which can achieve container-less and non-contact conditions for measurements. For density measurements, we improved our ADL system to obtain the appropriate shape for the droplet based on the previously reported system by Langstaff *et al.* We measured the density of molten  $\text{CaAl}_2\text{O}_4$  between 1600 to 2300K. We conclude that our ADL system has the potential to precisely measure molten oxide density due to good agreement of our density of  $\text{CaAl}_2\text{O}_4$  with previous data. Using these techniques, we succeeded also to measure the density of a molten  $\text{SiO}_2\text{-CaAl}_2\text{O}_4$  oxide system and discussed the molar volume change by varying the  $\text{SiO}_2$  composition.

**Keyword(s):** Levitation, Thermophysical properties, Density, Molten oxide

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### 1. Introduction

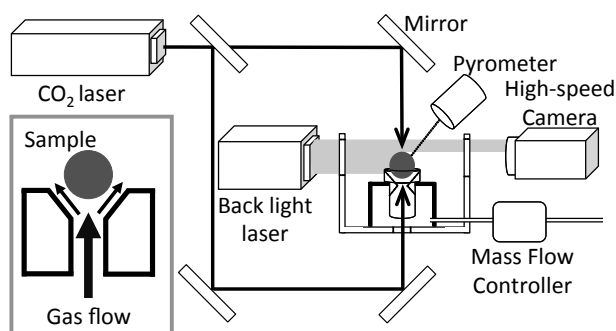
Density and its temperature dependence on liquid matter is very important for thermophysical property measurements and predictions, however it is difficult to estimate when multiple elements are involved, due to the complexity of volume changes for each individual liquid element. Especially, thermophysical properties, including density of molten oxides, are of great interest in numerous research fields including, steel making, geological science and material science. Molten oxide systems have an extensive range of components and therefore their density is correspondingly difficult to predict based on chemical composition and temperature. As a result, we must directly and precisely measure the densities of the numerous key components in the molten oxide. However, precisely measuring density of a molten oxide is very difficult due to high temperature and high chemical reactivity. Therefore, previous measurements have been limited to temperatures around 1800K<sup>1)</sup>. To achieve measurements over 1800K, we must use a container-less approach. However, electromagnetic levitation<sup>2)</sup> cannot be applied due to the low electrical conductivity of the molten oxides. The electrostatic levitation (ESL)<sup>3)</sup> technique is a possible alternative. However since ESL needs ultra-high vacuum conditions to avoid electric sparks at the electrode, we can use it on the limited molten oxide samples to measure its thermophysical properties. This limitation is the reason of changing the composition of the conditions for the molten oxide due to their high evaporation rate in a vacuum. Previously, the thermophysical properties (density, surface tension and vis-

cosity) of the molten oxides,  $\text{Al}_2\text{O}_3$ <sup>3)</sup> and  $\text{CaAl}_2\text{O}_4$ <sup>4)</sup> were successfully measured using ESL, due to their low evaporation rate even in vacuum. In order to measure the same properties of more complex compositions of molten oxides, we must use the aerodynamic levitation (ADL) technique<sup>5)</sup>. To measure the properties mentioned above using ADL, we improved the levitation gas-jet nozzle unit. For density measurements, we improved our ADL system to obtain the correct shape of the droplet based on the previously reported system by Langstaff *et al.*<sup>6)</sup>.

Using our improved conical nozzle system with ADL, we obtained the density of the molten oxides,  $\text{CaAl}_2\text{O}_4$  and  $\text{SiO}_2\text{-CaAl}_2\text{O}_4$ . We obtained the density of molten  $\text{CaAl}_2\text{O}_4$  with temperature variations from 1600 to 2250K, then using the data we examined the system and procedure using our ADL technique to measure density with temperature variations and compared this with previously reported results<sup>4)7)</sup>. We also tried to measure the multicomponent system of molten oxides using  $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$  in our ADL technique. The  $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$  is popular in many research fields, and their thermophysical properties have been obtained for limited compositions and temperature. Using our ADL technique, we succeeded to measure the density of a molten  $\text{SiO}_2\text{-CaAl}_2\text{O}_4$  system with various amounts of  $\text{SiO}_2$  and also in various temperature range from 1000 to 2600K. From density measurements for the  $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$  system, we discuss the molar volume change for  $\text{SiO}_2\text{-CaAl}_2\text{O}_4$  and  $\text{SiO}_2\text{-Ca}_3\text{Al}_2\text{O}_6$  and compare this with the previous models<sup>8)</sup> derived from measurements in a limited temperature range in a container.

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**Fig. 1** Schematics of aerodynamic levitation system for density measurements of molten oxides.

## 2. Aerodynamic Levitation (ADL) System for Density Measurement of Molten Oxides

The aerodynamic levitation (ADL) technique is a simple method for floating the samples in a gas-jet flow from conical nozzles, which maintains the sample position and is containerless. For levitation of the molten oxide system, we used Ar or dehydrated air for the levitation gas-jet. The gas was selected for stability of the sample and to reduce evaporation. **Figure 1** shows a schematic of our ADL system for measuring thermo-physical properties based on the previously reported system by Langstaff *et al.*<sup>6)</sup>. A mass-flow controller was used to precisely control the levitation gas-jet flow and to prevent the shape of the drop deviating from a perfect sphere. Spherical solid oxide samples, about 2mm in diameter, were levitated in the gas-jet flow, and then they were melted under the container-less conditions by irradiation from two CO<sub>2</sub> lasers. The sample temperature was measured using a monochromatic pyrometer at a wavelength of 0.9 $\mu$ m. In the ADL system, we installed an acoustic oscillation system to generate surface oscillations on the levitated droplets in order to measure surface tension and viscosity by the oscillating drop method<sup>6)</sup>. Since this report focus the density measurement, the surface oscillation excitation by the acoustic speaker system described in detail in elsewhere<sup>9)</sup>.

The density of the molten sample by levitation is obtained by dividing its mass by its volume. The mass can be easily obtained precisely using a balance before and after levitation. The volume of levitated molten sample was obtained from its shape using images of its shadow as lit from the side using a laser system at 532nm, which eliminates radiation from the molten oxide. The shape of ADL droplet is modified along the vertical direction due to gravity and the pressure of the levitation gas-jet flow. We need to improve the nozzle unit for observing half of the full droplet to obtain the volume when it is vertically elongated. Another reason for improving the nozzle unit is for passing a laser beam through the nozzle to the levitated samples from below. This is required to make a homogeneous temperature in the levitated molten oxide using two laser beams from above and below. Conventional

nozzles did not allow the laser beam from below to efficiently penetrated through the nozzle, therefore the laser power was reduced to 70%. To compensate for this, we improved the nozzle unit by implementing a conical shape and modifying the gas-flow path. Due to these improvements, we succeeded in observing the droplet shape over half of the full droplet and homogenized the temperature distribution of the sample by applying the laser from below at 94% power.

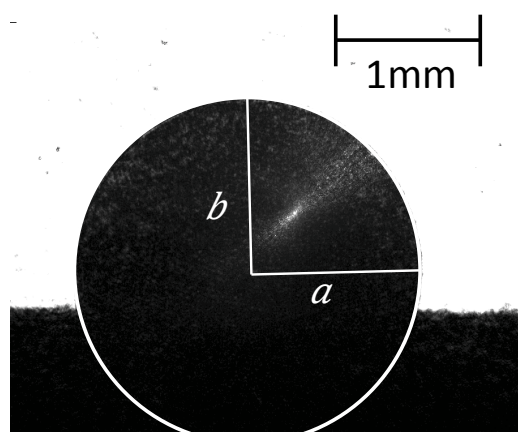
## 3. Observation of Static Shape of ADL Droplet and Density Measurement of Molten CaAl<sub>2</sub>O<sub>4</sub>

On the ground, the static shape of a 2mm diameter ADL droplet is slightly distorted from a perfect sphere due to gravity. Therefore, we must observe the droplet from the side, using back-lighting, to obtain the volume of the droplet from the cross sectional image. **Fig. 2** shows cross sectional images of the ADL molten Al<sub>2</sub>O<sub>3</sub> droplet. From **Fig. 2**, we found that the droplet is slightly ellipsoidal in shape due to gravitational distortion. From the shadow images we determined the outline of the droplet and fitted the following ellipsoid equation,

$$\frac{x^2}{a^2} + \frac{y^2}{b^2} = 1 \quad (1)$$

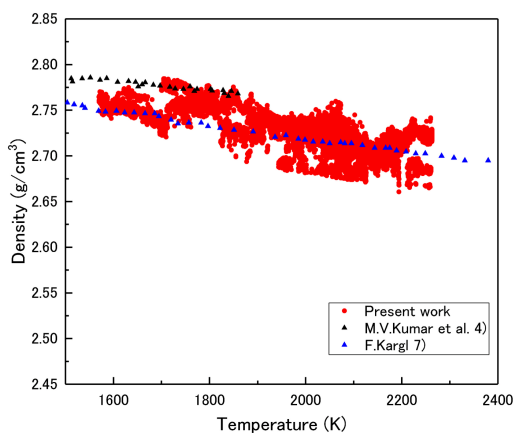
where  $a$  and  $b$  are the horizontal and vertical lengths from the center of mass. For the best fit to the equation, the values of  $a$  and  $b$  were changed to converge on the minimum of the differences between the outline and the fitting shape. Then we calculated the volume of the droplet,  $V$ , assuming the axial symmetry in the direction of gravity as;

$$V = \frac{4\pi}{3} a^2 b. \quad (2)$$



**Fig. 2** Shadow image of molten Al<sub>2</sub>O<sub>3</sub> with 2.0mm diameter by backlight laser optics system and fitting curve by ellipsoidal equations.

The density of the droplet can then be obtained by  $\rho = m/V$ , where  $m$  is the mass of the sample, and  $V$  is given by eq. (2) above. To examine the change in mass of the sample during the experiments, we measured its mass both before and after the experiments. In the present experiments, the change in mass for all samples was less than 1%. For confirmation of our density measurement system, we measured the density of molten CaAl<sub>2</sub>O<sub>4</sub>. The molar composition ratio is CaO:Al<sub>2</sub>O<sub>3</sub> = 1:1 and forms the compound CaAl<sub>2</sub>O<sub>4</sub> which has a melting temperature of 1878K and is therefore stable in the liquid phase with very low evaporation rate in high temperature ranges. Due to the stability of CaAl<sub>2</sub>O<sub>4</sub>, density measurements have already been taken using ESL<sup>4)</sup> and ADL<sup>7)</sup>. Therefore, CaAl<sub>2</sub>O<sub>4</sub> is a good sample for examining the system and procedure of our ADL techniques for the density measurements. We obtain a value for the density of molten CaAl<sub>2</sub>O<sub>4</sub> in a temperature range from 1600 to 2300K. The variation of the density of molten CaAl<sub>2</sub>O<sub>4</sub> with temperature is shown in **Fig. 3**. The uncertainty in this data at each temperature per shadow image is  $\pm 23.2\text{mg}/\text{cm}^3$ , using the coverage factor,  $k = 2$ . The uncertainty calculations were carried out in the same manner as reported in previous studies using static magnetic field applied electromagnetic levitation facilities<sup>10)</sup>, which is based on the Guide to Uncertainty of Measurement (GUM)<sup>11)</sup>. Our volume measurements of the ADL droplet used images of a partially obscured droplet, and we had to take into account the uncertainty due to the obscured portion. However, for the present measurements, we did not include the effect of the obscured portion on uncertainty of density measurements, since we confirmed that the droplet was symmetrical in both the horizontal and vertical axes after rapid solidification. However, while measuring the molten sample we were unable to discern the actual shape of the droplet, and future work should consider the accounting for the uncertainty due to the obscured portion of the droplet volume on density measurements.



**Fig. 3** Density of molten CaAl<sub>2</sub>O<sub>4</sub> with temperature variations.

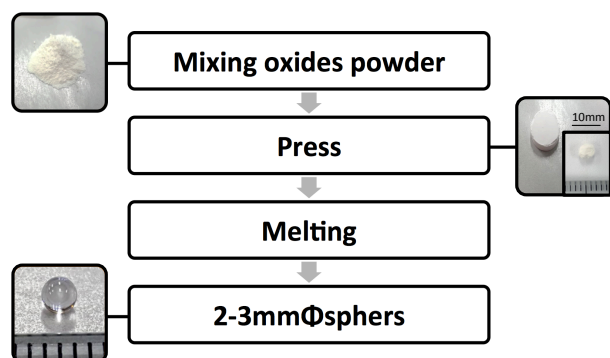
**Figure 3** shows the results of density measurements using container-less techniques by ESL<sup>4)</sup> and ADL<sup>7)</sup>. All results for density measurements from several measurements were plotted and they dispersed in the range of  $\pm 23.2\text{mg}/\text{cm}^3$  from the average density values at the corresponding temperature. From **Fig. 3**, we found that our dispersion range of measured densities includes the previous measured results by ESL and ADL. Our data are in good agreement with density values and temperature coefficients from previous results. Our results confirmed that our density measurement system can obtain values for molten oxide density in temperature ranges exceeding 1800K.

#### 4. Density Measurement of Molten SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> System

Density measurements of various compositions of molten SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> with made using an ADL system. In the present report we focus on the molar volume change with varying amounts of SiO<sub>2</sub> for CaAl<sub>2</sub>O<sub>4</sub> and Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>. Since CaAl<sub>2</sub>O<sub>4</sub> and Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> are stable liquids, even at high temperature, we were interested in the molar volume change from adding SiO<sub>2</sub> into these compounds. Courtial and Dingwell<sup>8)</sup> (CD) investigated the molar volume change for these systems. Using the Archimedean method, which measures density by means of weight inserted into liquid samples, they measured the density of four compositions of molten SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>. However, for molten SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> they could only measure the density of two compositions. For SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub>, using only three data points; CaAl<sub>2</sub>O<sub>4</sub>, SiO<sub>2</sub>:CaAl<sub>2</sub>O<sub>3</sub> = 1 : 1 and SiO<sub>2</sub> = 100mol%, they predicted a molar volume change from adding SiO<sub>2</sub> to CaAl<sub>2</sub>O<sub>4</sub>. (Pure SiO<sub>2</sub> data is predicted from extrapolation density data from SiO<sub>2</sub> glass by Bacon *et al.*<sup>12)</sup>, which is not direct measurement data.) Therefore, in order to confirm the molar volume change of this system predicted by CD, we tried to measure the density of many compositions of molten SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub>. For these measurements, we processed the following procedures for precisely controlling the compositions of various SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> melts. The oxide sample preparations are shown in **Fig. 4**. Initially, we dried powders of SiO<sub>2</sub>, CaO and Al<sub>2</sub>O<sub>3</sub> at 1173K in oven heaters to evaporate any H<sub>2</sub>O from the samples. Any H<sub>2</sub>O present in the sample powders will interfere with the composition adjustments when weighing them. The dried powders of each raw material, CaO, SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, were mixed in the necessary weights for each composition. Then the mixed powders were pressed into a 10mm diameter pellet. These pellets were then cut into 2mm diameter samples to be used in the ADL experiments. The pieces cut from the pellet were levitated in the conical nozzle and made spherical. The spherical samples were aerodynamically levitated and stabilized and the melted samples were used for the

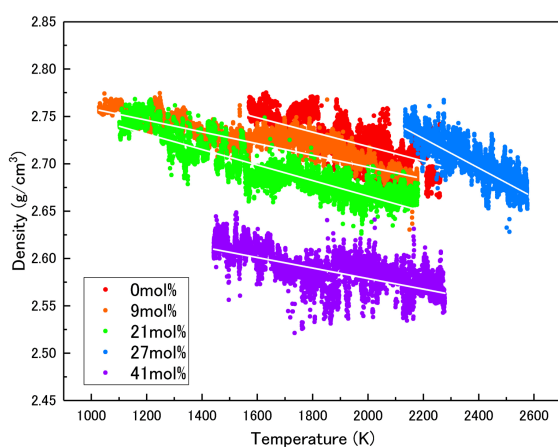
**Table 1** Compositions (mol%) of SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system for density measurements

Name	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO
0mol%	0	50.0	50.0
9mol%	9.0	45.5	45.5
21mol%	21.2	39.4	39.4
27mol%	26.6	36.7	36.7
41mol%	40.6	29.7	29.7

**Fig. 4** Procedure of oxide sample preparations for density measurements by ADL system.

density measurements. Sample SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> compositions for which we could measure density across a wide temperature range, are listed in **Table 1**.

We tried to measure the density of other SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> compositions, however for high SiO<sub>2</sub> compositions over 50mol% we could not precisely measure the density over a wide temperature range because of the high evaporation rate of SiO<sub>2</sub> at temperatures exceeding 2800K. In this experiment, for the five compositions mentioned above, we could measure their density and temperature dependence.

**Fig. 5** Temperature dependence of molten SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> density with composition variation of SiO<sub>2</sub> from 0 to 41mol%.

Using these oxide samples, we measured density using an ADL system. **Figure 5** shows the results for density of molten systems with temperature variations. The uncertainty in the density data is  $\pm 23.2 \text{ mg/cm}^3$  with the coverage factor,  $k = 2$ . This is same as that for the molten CaAl<sub>2</sub>O<sub>4</sub> density described in section 3. However, in **Fig. 5** we found larger dispersions of density for each composition and temperature. If a coverage factor of  $k = 3$  was used, we could not cover the measured density dispersions. This would be attributed to sample oscillations. Fluctuations in the levitation gas-jet flow generated oscillations in the sample which became larger than those seen in the CaAl<sub>2</sub>O<sub>4</sub> measurements. The addition of SiO<sub>2</sub> into CaAl<sub>2</sub>O<sub>4</sub> caused large fluctuations in the levitation gas-jet flow, however we were unable to ascertain the reasons for this in this experiment. Therefore, the density data in **Fig. 5** was largely dispersed and not centered on a point. For the density data of each composition, the linear temperature dependence of density,  $\rho(T) = \rho_0 + \alpha(T - T_m)$ , where  $\rho_0$  is the density at melting temperature,  $\alpha$  is the temperature coefficient, and  $T_m$  is melting temperature, was fitted to the density data to obtain the temperature dependence density for each composition.  $\rho_0$ ,  $\alpha$  and  $T_m$  for each composition are listed in **Table 2**. From the temperature dependence density we selected the density at 1973K and obtained the molar volume of each composition at this temperature. These molar volumes are plotted against SiO<sub>2</sub> compositions, as shown in **Fig. 6**. **Figure 6** shows the predicted curve for molar volume change of SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> with SiO<sub>2</sub> compositions along with data measured by CD. Only CD's data and the fitting curve is shown for the SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> system. Our data is for the SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system, and shown with CD's data and the corresponding fitting curve, which was traced from CD's paper<sup>8)</sup>. Our data is in good agreement with CD's fitting curve. Therefore, our density measurements are also in good agreement with CD's measurements. Since our data agrees with CD's data, we can now discuss the molar volume change for the SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system. CD predicted the molar volume change of the SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> system. They proposed the change in molar volume with SiO<sub>2</sub> composition,  $V(T)$ , especially for SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> and SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>,

**Table 2** Temperature dependence of density of molten SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system

Sample Name	$\rho_0$ (kg · m <sup>-3</sup> )	$\alpha$ (kg · m <sup>-3</sup> · T <sup>-1</sup> )	$T_m$ (K)
0mol%	$2.73 \times 10^{-3}$	$-7.59 \times 10^{-8}$	1878
9mol%	$2.71 \times 10^{-3}$	$-6.17 \times 10^{-8}$	1847
21mol%	$2.78 \times 10^{-3}$	$-8.21 \times 10^{-8}$	1823
27mol%	$2.80 \times 10^{-3}$	$-15.6 \times 10^{-8}$	1731
41mol%	$2.59 \times 10^{-3}$	$-5.56 \times 10^{-8}$	1714

using the following equation, which was based on the excess molar volume in a SiO<sub>2</sub>-CaO system,

$$V(T) = \sum_i X_i V_i(T) + X_{\text{SiO}_2} \cdot X_{\text{CaO}} \cdot V_{\text{SiO}_2\text{-CaO}} \quad (3)$$

where  $X_i$  and  $V_i$  are the mole fraction of oxides, and the partial molar volume of oxides, respectively.  $i$  denotes components of the system, in this case SiO<sub>2</sub> and CaAl<sub>2</sub>O<sub>4</sub>, or Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>. Also  $X_{\text{SiO}_2}$ ;  $X_{\text{CaO}}$  and  $V_{\text{SiO}_2\text{-CaO}}$  are pseudo binary-composition mole fractions of SiO<sub>2</sub> and CaO, and the molar volume of SiO<sub>2</sub>-CaO, respectively. If the molar volume of changes ideally by mixing SiO<sub>2</sub> into CaAl<sub>2</sub>O<sub>4</sub> or Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>, which implies no excess volume in eq.(3), we can add a linear line of best fit to the molar volume data for 0 to 100mol% of SiO<sub>2</sub>. However, in Fig. 6 both systems of the molar volume data did not change linearly, and we also found that they have an excess volume lower than the ideal molar volume, as the fitting line for the molar volume data is below the linear line. Our molar volume data is in good agreement with the fitting curve, similar to CD in which the molar volume of molten SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> was obtained by subtracting the molar volume of molten SiO<sub>2</sub>-CaO<sup>8)</sup> from the ideal molar volume (linear line) of SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub>. CD obtained the fitting curve from only three data points. From the agreement of our molar volume data with this curve, we consider the fitting curve correctly shows the molar volume change of the SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system.

For the molar volume change of the SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system, CD proposed that SiO<sub>2</sub> highly interacts with CaO in CaAl<sub>2</sub>O<sub>4</sub> and the molar volume shrinks because of this. Their measured data of molten SiO<sub>2</sub>-CaO system showed that the addition of CaO into SiO<sub>2</sub>, the molar volume shrank drastically. From these results, CD considered that each component in the SiO<sub>2</sub>-CaO system highly interact and easily connect with each other. Then for SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub>, the molar volume would shrink due to the addition of SiO<sub>2</sub>. CD used these concepts to measure the molar volume,  $V_{\text{SiO}_2\text{-CaO}}$ , of SiO<sub>2</sub>-CaO in eq.(3). Also for the SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> system, CD applied the same concept to SiO<sub>2</sub> with CaO for fitting the molar volume data. From the agreement of the fitting curve and molar volumes from the density measurements, CD concluded the in the SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> and SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> systems, SiO<sub>2</sub>-CaO interaction is important for the molar volume change. However, the SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> interaction is not a main contributor to the molar volume change. Presently, our molar volume change in the SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> system assists CD's interaction model of SiO<sub>2</sub>-CaO in the molar volume change of CaAl<sub>2</sub>O<sub>4</sub> with added SiO<sub>2</sub>. However, CD's empirical model is based only on a limited number of measurements, therefore it is necessary to measure more compositions for the SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> system.

From our present measurements and comparisons with other density data for the same samples taken using different measurement techniques, we conclude that our density measurement

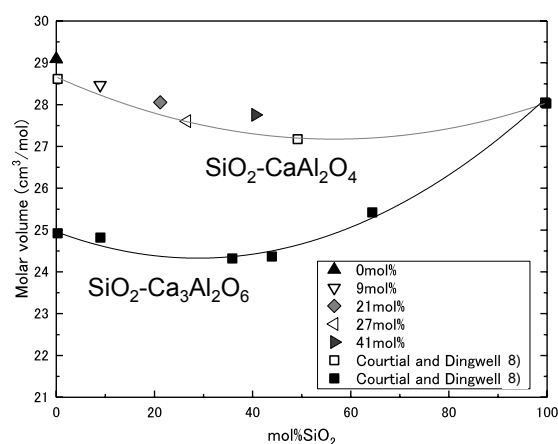


Fig. 6 Molar volume change with SiO<sub>2</sub> compositions for SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> and SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> at 1973K. Courtial and Dingwell<sup>8)</sup> results were plotted by □ (SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub>) and ■ (SiO<sub>2</sub>-Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>).

system using ADL will be effective with molten oxide systems. Therefore, for future work, we will conduct more data for density measurements of a molten SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> system, and based on these results and physical modeling, derive equations that predict molar volume change.

## 5. Conclusion

We selected the aerodynamic levitation (ADL) technique for molten oxide levitation and density measurements. For the density measurement, we obtained the volume of the ADL droplet from a three-quarters side view of the drop which was distorted due to gravity. We improved the conical nozzle shape for better observation of the images. Using the improved conical nozzle unit we successfully observed the droplet and calculated the density of molten CaAl<sub>2</sub>O<sub>4</sub> and SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> system with temperature variations. Our density data for molten CaAl<sub>2</sub>O<sub>4</sub> with temperature variations was in good agreement with previous measured data using ESL and ADL. Therefore we confirmed that our ADL measurement system can be applied to precise density measurements of molten oxide systems. Furthermore, from the density measurement results of SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> we discussed the molar volume change of SiO<sub>2</sub>-CaAl<sub>2</sub>O<sub>4</sub> with varying amounts of SiO<sub>2</sub>. Our molar volume data was in good agreement with the predictions of Courtial and Dingwell from an empirical evaluation. For future work, we will take many more density measurements of molten SiO<sub>2</sub>-CaO-Al<sub>2</sub>O<sub>3</sub> systems, and based on these results and physical modeling, derive an equation to predict the molar volume change.

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