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P10

電磁浮遊法による酸化物融体の放射率測定

Measurement of the normal spectral emissivity of molten oxide by electromagnetic levitation method

佐藤諒平 [1], 高橋圭太 [1], 渡邉匡人 [1],
Ryohei SATO[1],Keita TAKAHASHI[1], Masahito WATANABE[1]
[1] 学習院大学，Gakushuin University,

1. Introduction

In thermophysical property measurement using a containerless levitation method is using non-contact measurement of its temperature by a pyrometer. Temperature measurement by a pyrometer needs the normal spectral emissivity of sample materials. However, there are few reports of the normal spectral emissivity of molten oxide and the investigation of the emissivity is also important for thermophysical property measurement.

Therefore, we propose to measure the normal spectral emissivity of molten oxides by utilizing the radiance difference between liquid metals and molten oxides that form immiscible compound droplets by electromagnetic levitation (EML). The normal spectral emissivity, which is defined ratio of the radiance of the sample materials R_s and the blackbody radiance R_b is expressed as:

$$\varepsilon = \frac{R_\mathrm{s}}{R_\mathrm{b}} \quad (1)$$

where R is defined by Planck's law as a function of temperature T and wavelength of radiation λ and is given by

$$R_\mathrm{b}(\lambda, T) = \left(\frac{C_1}{\lambda^5}\right)\left(\frac{1}{\exp(C_2/\lambda T) - 1}\right) \quad (2)$$

where c is the speed of light, h is Planck's constant, and k_B is Boltzmann's constant, and C_1 and C_2 are the first and second radiation constants denoted by $C_1 = 2hc^2$ and $C_2 = hc/k_\mathrm{B}$. Typically, such emissivity measurements are performed using a cavity at a known temperature instead of a blackbody[1,2]. Paradis et al. successfully determined the normal spectral emissivity of molten alumina using an electrostatic levitation (ESL) method[3]. Sample materials that can be used in the ESL method must have low evaporation rates under high vacuum conditions, so many molten oxides are not suitable for this method.

Therefore, we proposed to measurement method of the normal spectral emissivity at wavelength λ by a pyrometer[4]. In this experiment, we chose Fe as a reference material because many reports on normal spectral emissivity, and this melt temperature can use proofreading. Assume that the emissivity ε_r of the reference material is not a function of temperature, and let R_r and R_s be the radiance of the reference and sample materials. The normal spectral emissivity at the detection wavelength of pyrometer of molten oxides is expressed as:

$$\varepsilon = \varepsilon_\mathrm{r}\frac{R_\mathrm{s}(T)}{R_\mathrm{r}(T)} \quad (3)$$

Thus, ε is determined by measuring R_s and R_r at the same temperature. In this report, we describe the process of determining the normal spectral radiance of molten oxide using pyrometer and discuss the experimental characterization of the emissivity of molten oxides from the SiO_2-CaO-FeO system.

2. Experiment

Since the EML method uses electromagnetic force, it is a method that can be applied to substances with high conductivity, but it is difficult to apply to substances with low conductivity such as oxide samples. Therefore, since the oxide sample alone cannot be electromagnetically levitated, it can be levitated by combining it with Fe.

Since the molten oxide and liquid Fe that we deal with are immiscible, the molten oxide and molten Fe float while being separated during electromagnetic levitation. At this time, since the oxide melt and liquid Fe have different radiances, the state of separation can be confirmed. This is because the emissivity of molten oxide and liquid Fe are different.

The oxide sample sintered to form this composite droplet was sealed in a Fe cylindrical container with a purity of 99.9%. As an assumption for this experiment, it is important that the temperature of the molten oxide and liquid Fe be the same. If the film thickness, which is estimated from the typical thermal conductivity of the oxide melt ($1.0 Wm^{-1}K^{-1}$)[5], is greater than 5μm, the difference in surface temperature between the molten oxide and the liquid Fe occurs. Make adjustments for optimal levitation.

3. Result and discussion

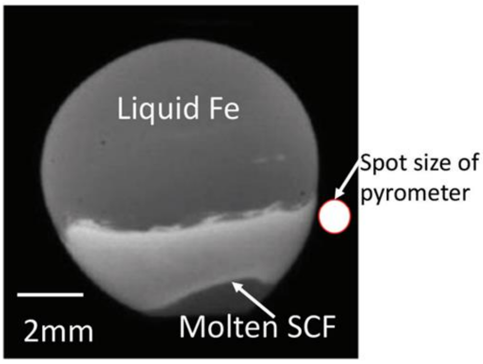

Figure 1. Photograph of complex droplet made from liquid Fe and molten SiO_2:CaO:FeO=20:20:40mass% (SCF) levitated by EML from side view.

Figure 1 is a high-speed camera image of an electromagnetically suspended molten SCF and liquid Fe. From here, the molten SCF and liquid Fe regions can be distinguished from each other. SCF is used for thermophysical property measurements using the Electrostatic Levitation Furnace (ELF) installed on the International Space Station. In ELF, sample temperature is measured with an emissivity of 1, and temperature correction using the normal spectral emissivity is necessary.

Due to the presence of FeO, the interfacial tension between the SCF and the liquid Fe is reduced, and the molten SCF spreads on the surface of the liquid Fe as shown in Fig. 1, and the shape changes randomly with time and stabilizes into a band state. The pyrometer (FTXX-ANE, JAPANENSOR Corp.) used had a spot diameter of 0.9 mm and a bandpass interference filter (900±10 nm), and the normal spectral radiance at 900 nm was incidentally obtained. The data to be acquired is measured with an emissivity of 1.

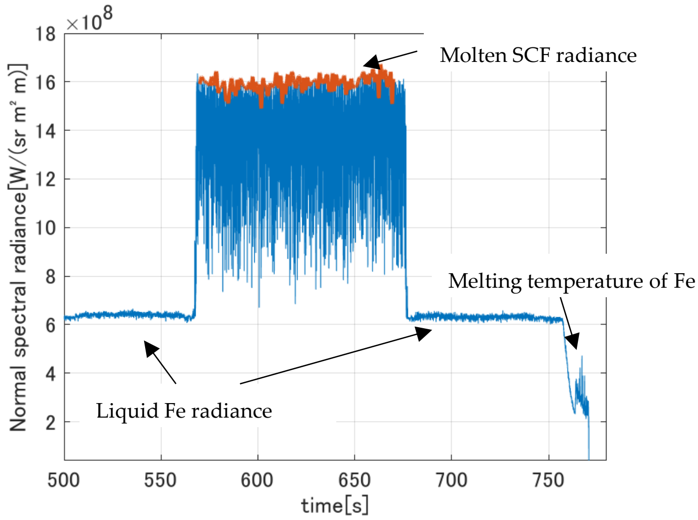

Figure 2. Normal spectral radiance of liquid Fe and molten SCF using the pyrometer set at emissivity of 1. Large fluctuation of Molten SCF radiance is caused by the half drop shape of molten SCF. Pyrometer detected the radiation from molten SCF and liquid Fe in periodically by the rotating complex droplet.

Figure 2 shows the normal spectral radiance obtained. Numerical simulations have shown that there is a temperature distribution in the entire mixed droplet of molten SCF and liquid Fe[6)] [7]. However, since the spot of the pyrometer is operated near the interface between the molten SCF and the liquid Fe, it is assumed that the temperature is uniform as assumed in the experiment. The normal spectral radiance of the liquid Fe was almost constant, indicating that the temperature of the liquid Fe was kept almost constant during the measurement. On the other hand, the normal spectral radiance of the molten SCF R_s fluctuates much more than that of liquid Fe. This is because liquid Fe alternately enters the spot of the pyrometer aligned with the molten SCF due to the flow of droplets. Therefore, it is difficult to determine the normal spectral radiance of the molten SCF. Based on this data, we evaluated R_s of the molten SCF by constructing the envelope and histogram of the normal spectral radiance data.

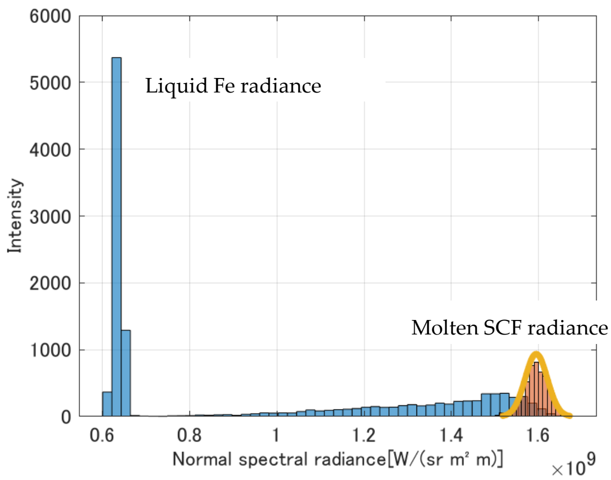

Figure 3. Histogram of the normal spectral radiance from liquid Fe and molten SCF.

Figure 3 shows the histogram of normal spectral radiance data. Liquid Fe is concentrated in the region of $6.2 \times 10^8 Wsr^{-1}m^{-2}m^{-1}$, and the maximum normal spectral radiance of the molten SCF is $1.65 \times 10^8 Wsr^{-1}m^{-2}m^{-1}$. The value of liquid Fe is almost constant and can be easily determined. Considering the detection error of the pyrometer, we assumed the normal spectral radiance of the molten SCF is $1.6 \times 10^8 \pm 5\% Wsr^{-1}m^{-2}m^{-1}$. The normal spectral emissivity of liquid Fe has been widely reported, such as 0.40 to 0.36 at a wavelength of 780 to 920 nm7) [8] and 0.30 at a wavelength of 800 nm8) [9]. These discrepancies are believed to be caused by the purity of the samples and the effects of surface oxidation. Based on the results of Kobatake et al.7)[8] and Watanabe et al.8)[9], we extrapolated the normal spectral emissivity of liquid Fe at 900 nm to $\varepsilon_{\mathrm{Fe}} = 0.32$. Using this value, the normal spectral emissivity of the molten SCF was determined as $\varepsilon_{\mathrm{OX}} = 0.78 \pm 0.01$.

ε_{OX} and $\varepsilon_{\mathrm{Fe}}$obtained in the above procedure can be used to calibrate the sample temperature from Planck's law. However, it should be fixed using $C_s = \varepsilon_{Fe}(R_{Fe}/R_b)$. The melting temperature of Fe ($T_{\mathrm{m}} = 1536$℃8)[9]) is used for correction, and the normal spectral emissivity $\varepsilon_{\mathrm{Fe}} = 0.32$ is used for the normal spectral radiance R_{Fe}. The sample temperature corrected by C_{s} is obtained by the expression:

$$T_i = \frac{C_2}{\lambda} \frac{1}{\ln\left(\frac{1}{(C_1/\lambda^5)(\varepsilon_{\mathrm{i}}/C_{\mathrm{s}}R_{\mathrm{i}})} + 1\right)} \tag{4}$$

Figure 4 shows the calibrated sample temperature, and it can be confirmed that they match at about 1650℃.

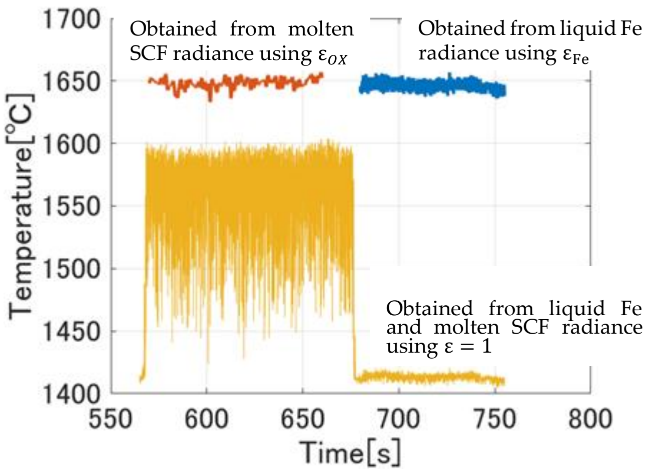

Figure 4. Temperature obtained from the normal spectral radiance from liquid Fe part and molten SCF part using the normal spectral emissivity of 0.32 for liquid Fe and 0.78 for molten SCF respectively.

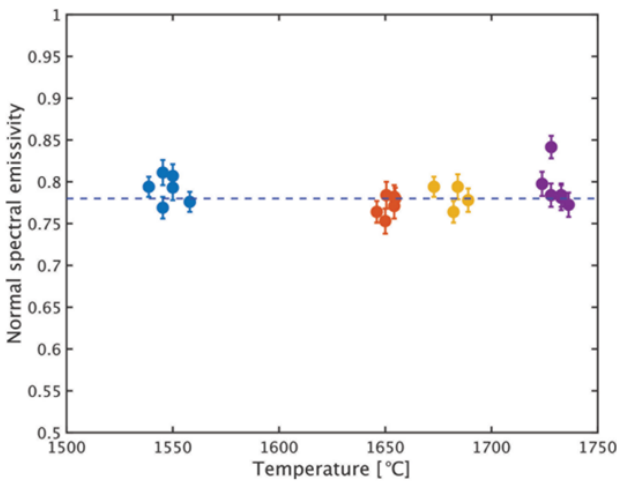

Figure 5. Normal spectral emissivity of molten SCF in the temperature range 1550°C-1725°C at the wavelength of 900nm.

Figure 5 shows the temperature dependence of the normal spectral emissivity of the SCF melt obtained by the above procedure. $\varepsilon_{OX} = 0.78$ was almost constant in the temperature range from 1550℃ to 1725℃.

The temperatures near the interface between molten Al_2O_3 and liquid Fe were measured, and the normal spectral emissivity of molten Al_2O_3 turned out to be $0.80 \pm 0.03\%$[9) [10]. Several reports reported it, and Paradis et al.[3) determined a value of 0.8 based on the change in heat capacity. Petrov et al.[11) [6] demonstrated a range of 0.87 to 0.91 using a spectrometer and pyrometer at wavelengths of 520 nm and 720 nm. These results show that the value of ε is smaller than that of ε_{AL}[12,13) [11-12]and the conduction electron density of molten SCF[12,13) is larger than that of molten Al_2O_3[14,15). This is considered to be caused by the difference in the conduction electron density of the samples.

To confirm this hypothesis, we investigated the normal spectral emissivity of the molten FeO. Since the electrical conductivity of the molten FeO[11,12) is lower than that of the A binary system, the normal spectral emissivity of the molten FeO is smaller than that of the molten SCF. **Figure 6** shows the temperature dependence of the normal spectral emissivity of molten FeO, which is almost constant at $\varepsilon_{OX} = 0.68$. This value is smaller than the normal spectral emissivity of the molten SCF, and it can be seen that the normal spectral radiance of the molten oxide is affected by the electrical conductivity. If the emissivity model of molten oxides can be created in the future, it may be possible to predict the wavelength dependence of the emissivity of molten oxides.

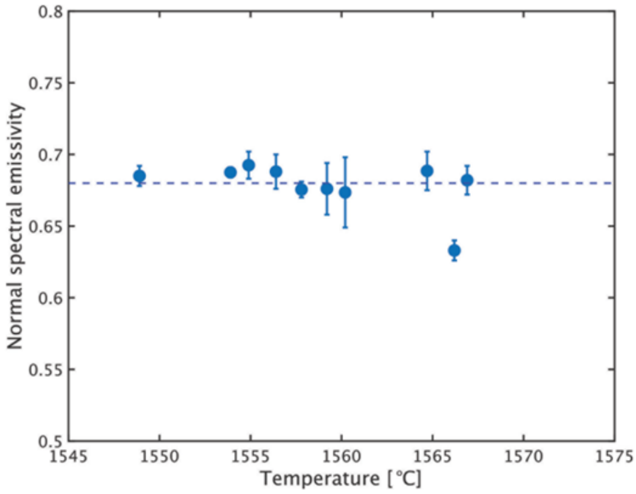

Figure 6. Normal spectral emissivity of molten FeO in the temperature range 1550℃-1565℃at the wavelength of 900nm.

4. Conclusion

Using liquid Fe as a reference material and mixed droplets of molten oxide, the procedures for obtaining the normal spectral emissivity of the molten oxide by the electromagnetic levitation method and pyrometer are shown. Our approach is based on the fact that the molten oxide and liquid Fe are immiscible and at the same temperature. The normal spectral emissivity of the molten SCF was 0.78±0.01 for the ELF sample, and that of the molten FeO was 0.68±0.03. This result indicates that the normal spectral emissivity of the molten oxide is affected by the conduction electron density. In the future, the correlation between the emissivity and conductivity of molten oxides may be investigated, and an emissivity model for molten oxides may be developed.

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