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In Situ Temperature Measurement of Oxide Melt in an Arc Imaging Furnace by a Monochromatic Radiation Pyrometer

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放射温度計を用いたアークイメージ炉中の酸化物融液のその場温度測定

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An arc imaging furnace has various advantages as an apparatus for ceramic synthesis. However, the disadvantage of this furnace is the difficulty of *in situ* temperature measurement of the sample, because of a very small heating spot in the furnace and the effect of infrared radiation from the light source on the measurement, a monochromatic radiation pyrometer is used. In this paper, a technique for *in situ* temperature measurement by monochromatic radiation pyrometer is proposed and applied to the melts of a CaO-SiO_2 system above 1700°C. Heating up was performed in an arc imaging furnace equipped with a Xenon lamp. The monochromatic radiation pyrometer was proven to work appropriately for *in situ* temperature measurement of the melting samples above 1750°C, because it can focus on a small melting drop 2-3 mm in diameter, and also because it can avoid the effect of infrared radiation from the Xe lamp, by using a 5 μm wavelength to detect the radiation from the sample melts. [Received June 4, 1999; Accepted July 21, 1999]

Key-words : *Temperature measurement, Arc imaging furnace, Monochromatic radiation pyrometer, Radiation, Emissivity, Oxide melt, CaO-SiO_2 system*

1. Introduction

An arc imaging furnace with a Xenon (Xe) or a halogen lamp is widely utilized in the process of ceramic synthesis, for example, growth of single crystals by floating zone method and preparation of novel glasses by rapid-quenching method with twin-roller. Because an arc imaging furnace has various advantages as the apparatus for ceramic synthesis, i.e., the attainment of high temperature over 1800°C with ease, super-rapid heating and quenching, a sample melting with no container, easy control of a surrounding atmosphere of a sample. However, striking disadvantage of this furnace is the difficulty of temperature measurement of a sample because of a very small focal point of the arc image, in other words, a very small heating spot in the furnace. When a thermocouple is applied to measure a temperature of a sample in an arc imaging furnace, it needs to contact the sample directly because only the sample is heated by the infrared radiation. It is sure that such an operation will cause contamination of the sample with impurity from a thermocouple, especially when the sample is necessary to be melted at high temperature for a long time.

A radiation pyrometer is one of conventional apparatuses for the measurement of high temperature in ceramic processing such as glass and refractory manufacturing. A radiation pyrometer is not only utilized in manufacturing processes, but also applied to various investigations at high temperature recently, for example, dilatometry of ZrO_2 and SiC, absorption coefficient measurements of molten Al_2O_3, surface temperature measurements of small areas during laser materials processing and spectral emissivity measurements of ceramic composites.[1)-4)]

Thus, a radiation pyrometer becomes a candidate as an apparatus for the temperature measurement of melting samples in the arc imaging furnace. However, there are some problems in utilization of a conventional radiation pyrometer for *in situ* and continuous temperature measurement of samples in the arc imaging furnace. One is based on the principle of the temperature measurement by use of the radiation from the sample; that is, it is necessary that the obtained temperature have to be corrected by using the emissivity of respective samples and temperatures. Emissivity of pure substance is determined by its nature, temperature and wavelength of radiation, when the surface of the substance is large, flat and smooth. Otherwise, the shape and roughness of the sample surface affect its emissivity, considerably.

Another problem is the effect of the radiation from a light source of the furnace. In order to heat a sample over 1800°C, a high power Xe lamp is usually used as an infrared radiation source. The region of radiation from the Xe lamp with silica glass tube is from ca. 0.2 to 4.5 μm according to the specification of the Xe lamp used in this study. On the other hand, a conventional monochromatic radiation pyrometer with Si or InGaAs photodetector utilizes the radiation from the sample in the region from 0.6 to 1.6 μm. This region of radiation overlaps with that of radiation from a Xe lamp.

As examples of temperature measurements by combining a monochromatic radiation pyrometer with an arc imaging furnace, the measurements of solidification points of oxide ceramics were reported, however, the temperature of molten oxides were obtained during cooling process after cutting infrared irradiation.[5),6)] Consequently, an appropriate monochromatic radiation pyrometer, which utilizes wavelength out of the region of radiation from a Xe lamp, should be combined with a Xe arc imaging furnace for *in situ* and continuous temperature measurements of high temperature samples.

Authors are studying high-functional phase separated glasses with anisotropic properties, which are induced by distortion and orientation of phase separation texture, especially for oxide systems having two-liquid immiscibility

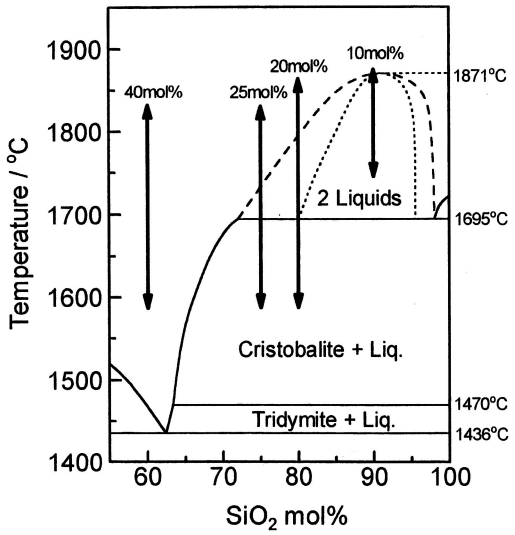

Fig. 1. Phase diagram of CaO–SiO_2 binary system, from Tewey and Hess,[8] and Phillips and Muan.[10]
The within of dashed curve is two liquids immiscibility region. Dotted curve in the immiscibility region indicates the boundary between binodal and spinodal phase separation. Arrows show regions of temperature measurements in respective samples.

region.[7] In this process, it is very important to investigate the phase separation phenomena in two-liquid immiscibility region, and *in situ* and continuous temperature measurements of the phase-separated melts is indispensable as the basic procedure for such purpose.

In this study, the temperature measurements by use of an monochromatic radiation pyrometer combined with a Xe arc imaging furnace was examined on high temperature oxide melts. As the subject of experiments, CaO–SiO_2 binary system was selected, because this system was reported to show obvious two-liquid immiscibility in the composition of CaO 2–29 mol%[8),9)] and it is very stable in the atmosphere. The phase diagram of CaO–SiO_2 binary system redrawn from reported ones[8),10)] is shown in Fig. 1.

2. Experimental

In order to melt a sample in a Xe arc imaging furnace, it is necessary to form the sample into rod shape. The preparation procedure of calcium silicate rods was as follows. $CaCO_3$ (Wako Pure Chemical Ind. Ltd., reagent grade) and silica gel (Wako Pure Chemical Ind. Ltd., Q-63) were mixed in the composition of xCaO–$(100-x)$$SiO_2$ mol% (x=10, 20, 25, 40). The samples of x=10, 20, 25 have two-liquid immiscibility region over 1695°C as shown in Fig. 1. The mixed powders were calcined at 1000°C for 1 h and formed into short rod shape by use of CIP (ca. 1000 kg · cm^{-2}).

The configuration of the arc imaging furnace with a 5.4 kW Xe lamp (Nichiden Machinery, FQ-50XS) is schematically illustrated in Fig. 2. The sample was attached to silica glass tube by partial fusion. In order to measure the temperatures of the sample melts directly as reference, thermocouple ($Pt_{80}Rh_{20}$–$Pt_{60}Rh_{40}$) was inserted into silica glass tube and was attached to the partially fused sample. Then, the calcined sample powder was stuffed in the surrounding of the thermocouple tip. As mentioned above, since the region of radiation from a Xe lamp spreads from 0.2 to 4.5 μm, the appropriate monochromatic radiation pyrometer (Japan Sensor, TSS-15G) was used in this measurement. This monochromatic radiation pyrometer detects the infrared radiation of wavelength 5±0.25 μm by thermopile and its detecting spot area is 1 mm in diameter at the focal distance; 320 mm.

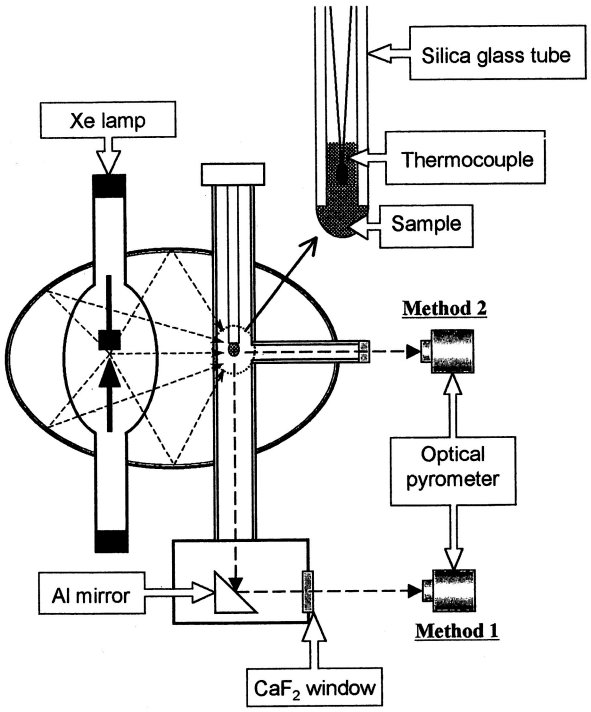

Fig. 2. Schematic illustration of the configuration of a Xe arc imaging furnace with a monochromatic radiation pyrometer.

The radiation from the sample melt was detected by the monochromatic radiation pyrometer through two routes (methods 1 and 2) as shown in Fig. 2. In method 1 applied to the samples containing 10, 20 or 25 mol% of CaO, the radiation from the bottom of a melting drop was once reflected on an aluminum mirror and it passed through a CaF_2 window before detecting by the monochromatic radiation pyrometer. In method 2 applied to the sample containing 40 mol% of CaO, the radiation from the side of a melting drop was directly detected after passing through a CaF_2 window. In both methods, the monochromatic radiation pyrometer was set on the point of its own focal length from the focal point of the arc imaging furnace, and the effects of the mirror, a CaF_2 window and a visible-light reducing filter were corrected for respective setting in advance.

The tip of sintered sample rod was once melted at high temperature by supplying large lamp power. Then, the temperatures of the respective sample melts were measured by the monochromatic radiation pyrometer and the thermocouple at the same time, as the lamp power was gradually reduced. The measured temperature regions are also shown in Fig. 1 for respective samples.

3. Results and discussions

The temperatures measured by the monochromatic radiation pyrometer and the thermocouple against supplied lamp power after melting of the sample were shown in Figs. 3(a)–(d) for the samples containing CaO 10, 20, 25 or 40 mol%, respectively. In all measurements, the temperatures obtained by the monochromatic radiation pyrometer are the values when the emissivity is supposed to be one. The size of melting drop in respective samples was ca. 2–3 mm in diameter, which was enough large for the detecting spot area, 1 mm in diameter, of the utilized monochromatic radiation pyrometer.

The temperatures detected by the monochromatic radiation pyrometer in method 1 were in good agreement with those by the thermocouple above 1750°C as shown in Figs.

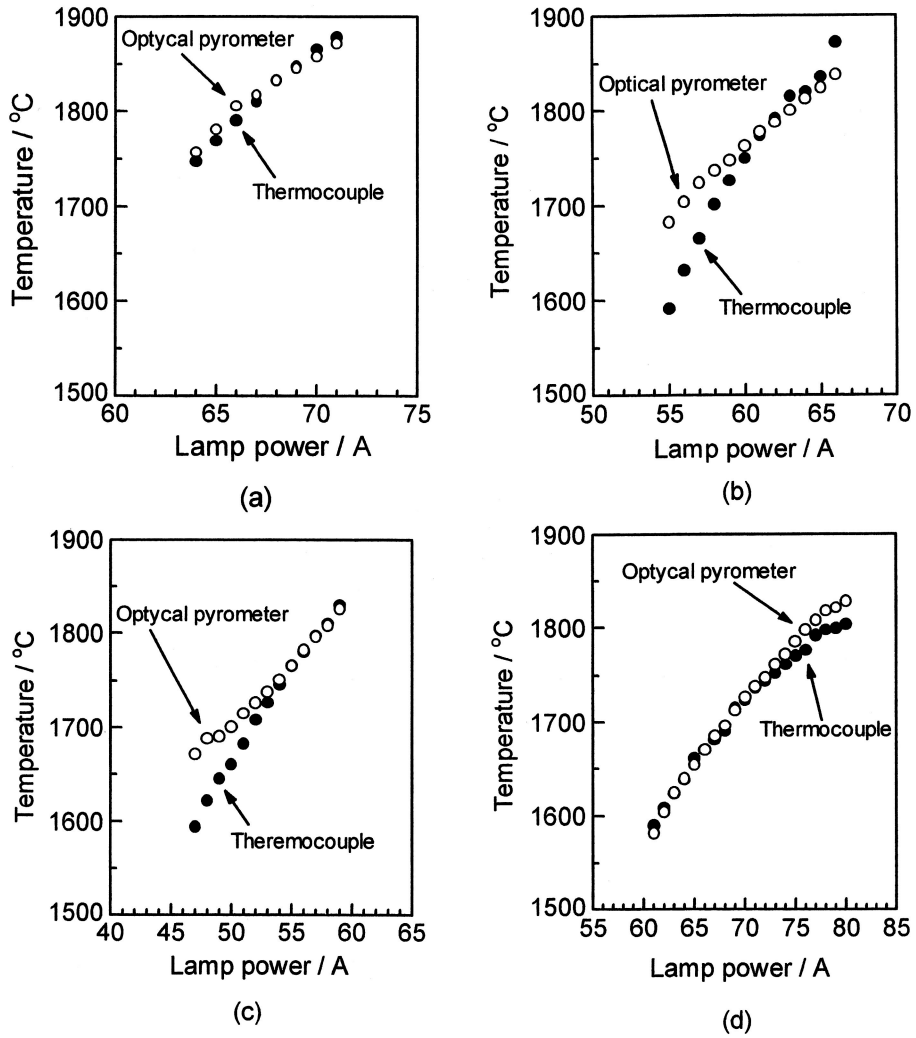

Fig. 3. Temperatures measured by the monochromatic radiation (optical) pyrometer (○) and the thermocouple (●) against supplied lamp power; (a) 10 mol%, (b) 20 mol%, (c) 25 mol%, (d) 40 mol% of CaO.

3(a)-(c). However, as the lamp power was reduced, that is, the temperature fell below 1750°C, the monochromatic radiation pyrometer showed higher temperatures than those detected by the thermocouple, though the emissivity was maintained to be one. If the emissivity of the sample changed and became less than one as falling down the temperature, the temperatures indicated by the monochromatic radiation pyrometer would become still higher than those shown in figures. That is to say, these results indicate that the temperature of the sample bottom was actually higher than that around the tip of thermocouple below 1750°C.

The liquidus temperature of the samples containing CaO 10, 20 and 25 mol% is reported to be 1695°C as shown in Fig. 1. The temperature region below 1750°C is close to the liquidus temperature in those compositions. Further, the size of heating spot in the imaging furnace is very small as mentioned above. Consequently, it is considered that the above portion of a melting drop solidified first when the lamp power was reduced, that is, temperature fell down below 1750°C, as schematically illustrated in Fig. 4. At that time, the monochromatic radiation pyrometer still measured the temperature of the melts from their bottoms. It is supposed that these actual different phases of the sample between in the bottom and around the tip of thermocouple caused the difference of the temperatures measured by the monochromatic radiation pyrometer and the thermocouple near liquidus temperature in method 1.

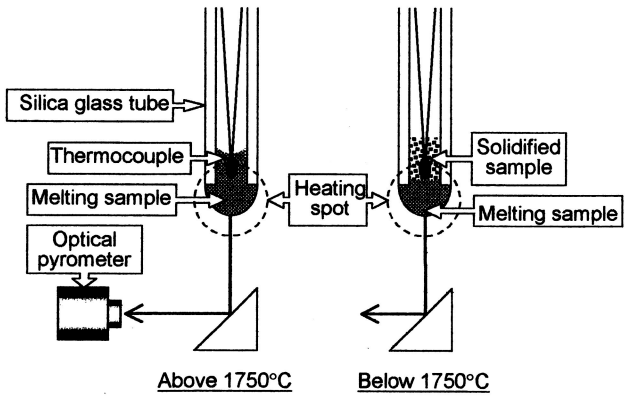

Fig. 4. Schematic illustration of the states of a sample containing 10, 20 or 25 mol% of CaO.

On the contrary, in method 2 as shown in Fig. 3(d), the monochromatic radiation pyrometer showed higher temperatures than those detected by the thermocouple above 1750°C. This result also indicates that the temperature of the side of a melting drop was actually higher than that around the tip of thermocouple above 1750°C.

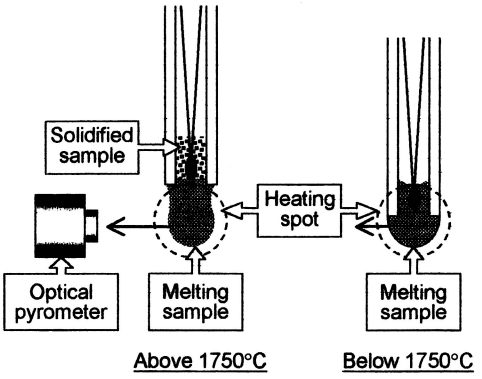

Fig. 5. Schematic illustration of the states of a sample containing 40 mol% of CaO.

The liquidus temperature of the sample containing 40 mol% of CaO is below 1500°C as shown in Fig. 1, which is much lower than the temperature region applied to this measurement, 1700–1900°C. In this case, it is anticipated that the glass tube holding the sample melt was drawn up to keep the melt in the heating spot, as the melting drop hung down because of the decrement of the melt's viscosity. Because the tip came out from the heating spot as schematically illustrated in Fig. 5, the sample around the tip of thermocouple was finally solidified. This is considered to be the reason of the difference of two measured temperatures, though the temperature of the melting drop is enough high to maintain liquid state.

4. Conclusion

From the results of the temperature measurements in method 1 and 2, it was found that the effect of infrared radiation from a Xe lamp on the temperature measurement was avoidable by use of an appropriate monochromatic radiation pyrometer, and that there is no matter even if we consider the emissivity of the oxide melts to be one. Thus, examined configurations of apparatuses have been proven to work well for *in situ* and continuous temperature measurement of the melting sample in a Xe arc imaging furnace. On the contrary, we have to take care of the emissivity of the sample so much when it is solid, and we needs so much effort to obtain the change of emissivity with the shape, the surface roughness and, of course, the temperature of the sample.

The present technique is very effective in investigating the phase separation phenomena of oxide melts in two-liquid immiscibility region, and it makes the controllability in the process of ceramic synthesis by use of the arc imaging furnace improve much finer.

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