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Technical Note

Experimental study on marangoni convection and solidification in BaB_2O_4 melt

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Abstract

The technique of in situ optical observation is applied to visualize the marangoni convection (MC) in a high temperature melt. The transition from steady to oscillatory marangoni convection in the BaB_2O_4(77 wt%)–Na_2O(23 wt%) mixture melt has been visualized. The oscillatory convection, accompanied with a temperature oscillation, is connected with the free surface deformation of the melt. Moreover, two different kinds of solidifications from the highly undercooling melt are speculated; one is the common solidification, while the other is an unusual solidification of lacunose structure caused by spontaneous nucleation. The diameters of the alveolus are uneven with the largest about 30 μm and the smallest 8 μm. It is pointed out that the change in the temperature distribution due to the distortion of the loop-like heater gives rise to the discrepancy in the solidification process.

Keywords: Marangoni convection; Heat transfer; Fluid flow; Solidification

1. Introduction

The marangoni convection (MC) is driven by the surface tension gradient generated by the temperature gradient along the free surface. It has been shown for floating zones that MC becomes unsteady (oscillatory or turbulent) at higher marangoni numbers, which is accompanied by temperature oscillations in the melt. The temperature oscillations are normally undesirable because they can cause oscillatory growth speed, thus generating impure striations. Therefore, it is important to understand how and when the transition occurs. Moreover, the investigation of the transitions from steady to oscillatory motion of high temperature oxide melt as well as the mechanism study will help to optimize the technology of the crystal growth. In this regard, the flow visualization picture is very helpful.

However, it is difficult to get the image of the marangoni convection and solidification process during the melt crystal growth due to the high temperature environment. Most of the marangoni convection studies have been carried out in low temperature melt like $NaNO_3$ [1], $C_{20}H_{42}$ [2] and silicone oil [3]. Meanwhile, numerical simulations have also widely been taken to interpret the mechanism of marangoni convection. On the other hand, few results were reported about the solidification process during high temperature melt crystal growth. Although great progress has been achieved in the past decades, there still exists unclear problems about the marangoni convection. This paper introduces the in situ observation system and its application in high temperature oxide crystal mechanism study, especially on the melt flow pattern and solidification process. This technique provides a chance to discover the mechanism of the marangoni convection experimentally.

2. Experimental procedure

The test section of the in situ observation system is comprised of a heating chamber and a loop-shaped Pt wire

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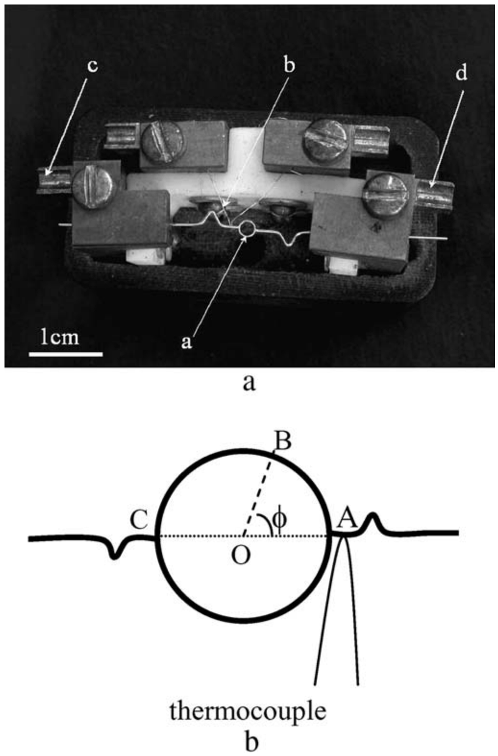

Fig. 1. (a) Picture of the heater: Pt-loop and thermocouple. (b) Schematic diagram of the heater.

heater. As shown in Fig. 1(a), the Pt wire (Φ 0.20 mm) is employed to heat and suspend the melt during the in situ observation experiment. A Pt-10% Rh thermocouple (Φ 0.08 mm) is used to measure the temperature of the loop. The inner diameter of the loop is about 1.80 mm. A V-shaped electrode is used to prevent the deformation of the loop-shaped heater.

Schlieren technique coupled with differential interference microscope (DIM) was applied to visualize the whole solidification process. Major changes were carried out in its signal acquisition system. The former video cassette record (VCR) method [4,5] had been substituted by a digital signal acquisition system with a computer, thus much clearer video could be obtained, which enabled us to make an in-depth research of the marangoni convection and the defect formation in the solidification process. During the experiments, the object lens of the microscope was vertical to the horizontal heater, so the motion of the melt and solidification could be observed directly. The image from the microscope was recorded by using CCD.

A typical experimental procedure was carried out in the following manner. To observe the solidification, the sample was melted and a thin film was formed in the loop as a result of the surface tension, then the power was adjusted gradually until the solidification was formed. It was observed that the melt shape looks like a concave mirror with the thin flat film in the middle. The free surface of the liquid zone was open to ambient air and thus "naturally" cooled.

3. Results and discussion

The temperature distribution along the free surface is an important source of information because it is directly related to the driving force of the flow. The distribution of temperature along the azimuthal coordinate was measured by a newly developed non-contact method [6]. Fig. 2 shows the typical temperature distribution along the azimuthal coordinate, which is symmetrical in the two semicircles divided by the wire jointed to the loop in positions A and C. The discrepancy in temperature is caused by heat transfer from thermocouple near to position A, as shown in the schematic diagram (Fig. 1(b)). In Fig. 2, $\Delta T = T_B - T_A$, where T_A is the temperature of position A and T_B means the temperature of different positions along the azimuthal coordinate. Table 1 shows the physical properties of BBO melt, which have been measured and provided in Ref. [7].

Typical MC flow patterns for BaB_2O_4(77 wt%)–Na_2O(23 wt%) mixture melts are shown in Fig. 3. In steady convection stage, the flow occurs in an axially symmetric pattern as a result of the deformation of the free surface. The deformation of the free surface appears as the main trunk and two branches toward the low temperature area in the melt, as shown in Fig. 3(a). It was noted that the periphery of the melt is a meniscus and appears opaque as a result of the light refraction. The oscillatory MC is triggered when the temperature gradient exceeds a certain value, in this stage, the oscillation of the main trunk near

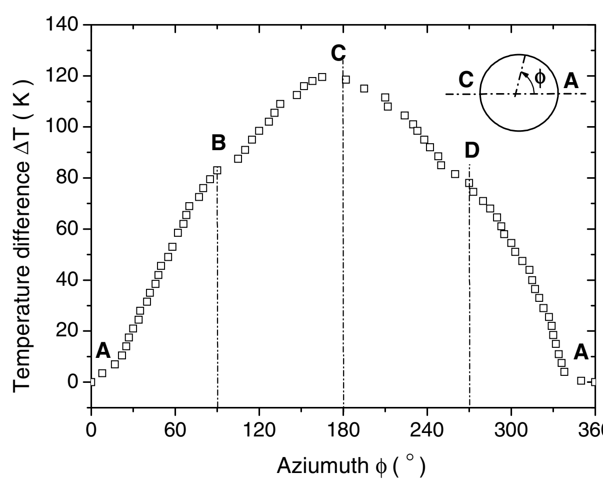

Fig. 2. Temperature difference along the azimuthal coordinate of the heater.

Table 1
The physical properties of BBO melt

Melting point (K)	1368
Density (kg/m^3)	3.64×10^3
Dynamic viscosity at 1380 K (Pa s)	0.16
Surface tension at 1380 K (N/m)	0.09
Surface tension coefficient (N/m K)	-6.0×10^{-5}

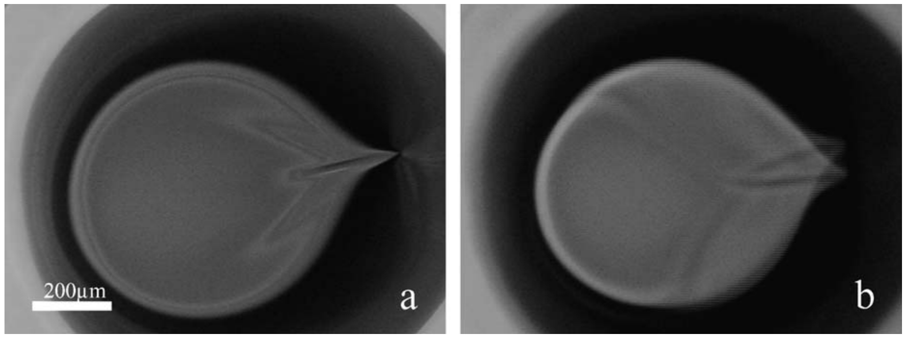

Fig. 3. (a) The steady MC flow pattern; (b) the oscillatory MC flow pattern.

position A characterizes the oscillatory flow, while the oscillatory branches affect the other area of the visualized melt and form the hydrothermal waves, which continuously sweep across the melt from position A to position C along the azimuthal coordinate of the loop, as shown in Fig. 3(b). The onset mechanism of the oscillation could be better understood when the degree of the deformation is considered. We noted that the surface tension is supposed to balance the sum of pressure and normal stress on the free surface of the melt, however, when the surface tension is not big enough to balance that, the deformation will thus form to provide extra pressure. However, the deformation will reach a maximum with the further rise of the temperature difference, and then the oscillation will be triggered. Typical temperature fluctuation accompanying the oscillatory convection was measured, as shown in Fig. 4. We also noted that similar temperature oscillation had been

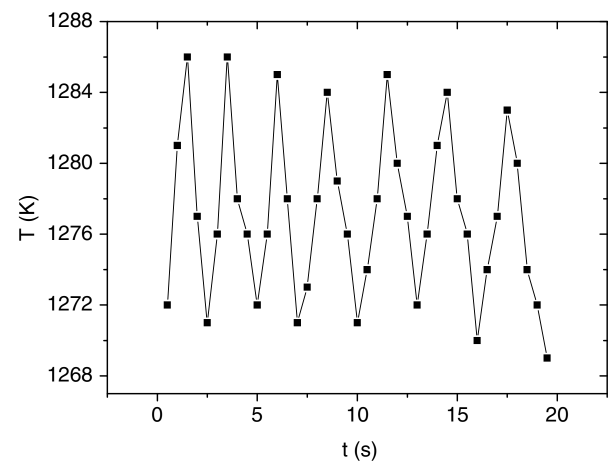

Fig. 4. The mode of the temperature perturbation accompanying the oscillatory MC.

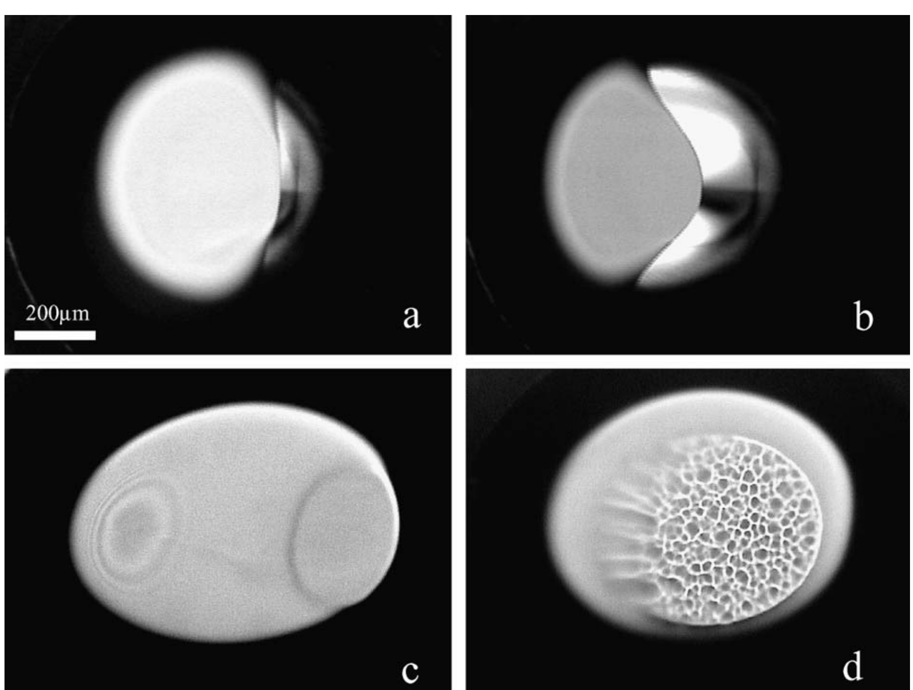

Fig. 5. (a) and (b) The common solidification process. (c) and (d) The solidification process of the lacunose structure.

observed in the $Li_2B_4O_7$(80 wt%) and $KNbO_3$(20 wt%) mixture [8].

The solidification process is also studied in the observation system. When the temperature decreases, two kinds of solidification processes are observed in the experiments. Fig. 5(a) and (b) shows the common solidification process owing to the typical temperature distribution. As shown in Fig. 2, the melt area near to position A is the lower temperature area, so the solidification first originates in this area, and then sweeps the melt along the azimuthal coordinate.

However, another totally different solidification process was observed. Fig. 5(c) and (d) shows the lacunose structure, which is formed simultaneously from the melt. The observed alveolus is uneven with the diameter varying from 8 μm to 30 μm. The discrepancy in the solidifications could be well explained by considering the temperature distribution in the loop. It is found that, before the lacunose structure happens, the loop-shaped heater turns to form an ellipse after long-term high temperature heating although the V-shaped construct is applied, so the temperature distribution is distorted in the elliptical loop. In the elliptical loop, the back flow from position A to C is affected by the flow from position C to A, and forms a deformed flow, which appears in Fig. 5(c) as the protuberance near to the colder side A, and the protuberance will expand itself when the temperature is adjusted properly and then numbers of seeds form in the melt, from which the hexagonal crystal particle grows, as shown in Fig. 5(d). The EPMA-EDS testing results show no compositional changes between the dark area and bright periphery of the hexagon-like lacunose structure. So, the lacunose structure only indicates the shape of the crystal particles as a result of growth from the spontaneous nucleation. Further efforts should be taken to study the lacunose structure.

4. Summary

The high temperature in situ observation system is suitable for the study of the marangoni convection and the solidification process in a high temperature oxide melt. The following results are obtained from the experiments:

1. Steady marangoni convection comprising the free surface deformation was observed in the thin flat melt. The deformation was sensitive to the temperature distribution in the melt.
2. Oscillatory marangoni convection would be triggered when the temperature difference on the loop exceeded a certain value, and temperature oscillations were found to accompany the oscillatory convection.
3. Two kinds of solidification processes were observed in the experiments: the common solidification and the unusual lacunose structure caused by spontaneous nucleation. No compositional difference was uncovered in the EMPA-EDS testing about the hexagonal lacunose structure.

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