



Surface tension convection caused by free surface deformation in $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$ melts

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ABSTRACT

Experiments on surface tension driven convection of high temperature potassium tantalate niobate ($\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$, KTN) melts have been carried out in an in-situ observation system. There is a double loop axially symmetric vortex pattern. With the temperature increases, the convection can be transferred from one stable state to oscillatory state and then to another stable state. The oscillatory surface tension convection, which is related to the free surface deformation, can be characterized by the oscillatory main trunk and two unequal branches. The branches depress the large pressure gradient and compensate the return flow to stabilize the circular current. There is a linear decreasing relationship between the deformation angle θ and the oscillatory frequency f . For KTN melts with higher Nb content, the convective pattern is not various, but a smaller degree of deformation can occur accompanied by a weaker oscillation process. The values of critical deformation coefficient S parameter for both the onset and the offset of oscillations decrease with the increase of the content of Nb. The critical values of oscillation satisfy with $S_{\text{on}} \cdot x \approx 2.3 \times 10^{-3}$ and $S_{\text{off}} \cdot x \approx 4.0 \times 10^{-3}$, respectively.

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1. Introduction

Potassium tantalate niobate ($\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$, KTN) crystals have been investigated due to their large electro-optic and photorefractive characteristics [1,2]. KTN single crystals can be prepared by several growth methods from melt/solution, such as Czochralski (CZ) method, cooling method, and top seeded solution growth (TSSG) technique [3–6]. However, many growth defects, such as striation and inclusions, can be generated [7–9]. As the convection gives an important influence to heat transport, mass transport and interface kinetics during crystal growth, growth defects can be reduced by well controlled convection [9,10]. Besides the common buoyancy convection induced by gravity, surface tension convection (or Marangoni convection) is also extremely important because of the widespread existence of free surfaces. In general, only stable convection is helpful for crystal growth with high quality. As a result, the stability/instability of Marangoni convections together with how the instability occurred has been investigated [11,12]. In effect, the stability of Marangoni convection can be enhanced by choosing reasonable matching parameters of the fluid [13]. Furthermore, solute Marangoni convection can even be used

to control the shape of crystal-melt interface without the change of temperature gradient or cooling rate [14].

However, to our knowledge, there has been seldom study on KTN melt Marangoni convection so far, which limits the improvement of crystal quality greatly. For a thin layer of melt in a loop-shape heater with horizontal temperature difference, convection would occur as a result of surface tension and temperature difference. In this work, the flow fields of surface tension convection in KTN melts were studied with the help of a high-temperature in-situ observation system. The system had been used to study the convection of KNbO_3 [15], BaB_2O_4 [16,17], $\text{Bi}_{12}\text{SiO}_{20}$ [18] oxide melts previously and some instructive results were obtained. The analysis was performed for various temperatures together with diverse components, to get more insight into the surface tension convection in KTN melts.

2. Experiment

The KTN crystal blocks were prepared with 20 wt% excess K_2CO_3 to compensate the volatilization of the K element. Then the melt convection was visualized under the condition of high temperature in an in-situ observation system. The observation section of the system is comprised of a heating chamber and a loop-shaped Pt wire heater as shown in Fig. 1. The Pt wire (Φ 0.2 mm)

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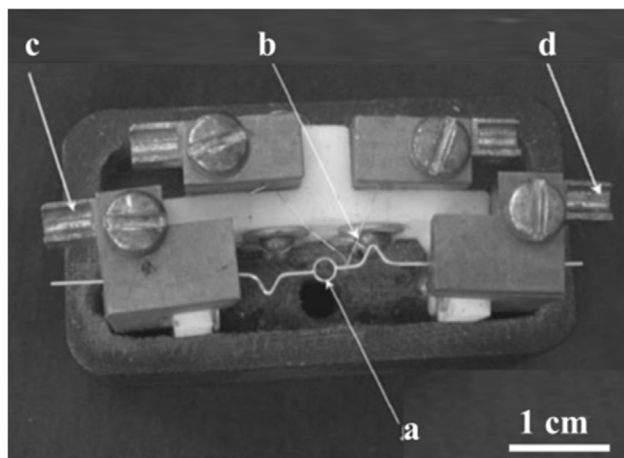


Fig. 1. The top view of the growth cell: (a) Pt wire heater, (b) Pt-10% Rh thermocouple, (c) and (d) are electrodes.

as shown by (a) is employed to heat and suspend the melt. The inner diameter of the loop is ~ 1.20 mm. A V-type of electrode is used to defend the loop-shaped heater from deformation. A Pt-10% Rh thermocouple (Φ 0.08 mm) as shown by (b) located in the right side of the loop is used to measure the temperature with the fluctuations of less than ± 2 °C above 1000 °C. Owing to the thermal conductivity of thermocouple, the lowest temperature along the loop is the side with thermocouple wire. The power is applied to both the electrodes of the wire as shown by (c) and (d) in Fig. 1. The video of the melt convection was recorded from the microscope by a camera.

The temperature distribution among the Pt heater in Fig. 1(a) is important because it is directly related to the driving force of the flow. The existence of thermocouple may cause the non-uniformity of the temperature distribution. As a result, the temperature field is measured according to the methods described in the literature [19]. Fig. 2(a) shows the typical temperature distribution for KTN melt, where the point A is at the thermocouple side. The distribution of the temperature field along the radial direction is not uniform. The closer the place is to the crucible, the higher the temperature is. The temperature distribution in the two semi-circles is symmetrical with respect to the wire AC. The discrepancy of temperature between A and C is caused by heat transfer from thermocouple near position A as mentioned previously. Fig. 2(b) shows that the temperature difference $\Delta T = T_C - T_A$ increases with

rise of temperature T_A , which is measured by thermocouple (hereafter recorded as T). Here T_C and T_A are the temperature of position C and A, respectively.

A typical experimental procedure was carried out in a following way. Before each test, the oxide material is firstly heated and suspended horizontally on the loop heater to form a melt film whose thickness is 0.05 mm. The amount of the test melt, usually 4 mg, was precisely controlled so that the upper and lower planes of the liquid are parallel to each other in the center area of the melt. Both of the top and the bottom planes of the melt are free surfaces exposed to air. Then change the power and the temperature of the system was altered according to the experimental requirements. The video is recorded from the microscope through a camera. Then the convection characteristics of the KTN melt can be obtained by the video.

3. Results and discussions

With the increase of temperature, three different flow configurations having the transition from low temperature stable convection (LSC) to oscillatory convection (OC), then to high temperature stable convection (HSC) have been observed. The experiment is repeatable. At higher or lower temperatures, the OC of the melt suddenly disappears and mutates into HSC or LSC, respectively. This is supported by the numerical results in a liquid layer under microgravity conditions by Li et al. [20]. They found that convection experienced the steady convection, oscillatory convection and steady convection in sequence with increasing Marangoni number. The physical mechanism of the oscillation is associated with the deformation of the two free surfaces. Next, steady flow LSC and HSC together with oscillatory flow OC will be studied separately.

3.1. Steady surface tension convection in KTN melts

Under the thickness of 0.05 mm, the patterns of steady surface tension convection LSC and HSC in $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$ for $x = 0.90$ are shown in Fig. 3. When the temperature is lower than ~ 1145 °C, LSC stage occurs. The convection pattern is as shown in Fig. 3(a). It comes into HSC stage when the temperature is above ~ 1400 °C. The flow pattern is shown in Fig. 3(b). Both of the patterns show symmetrical double loop convection shape with respect to line AC. The difference of the color means of the various thickness near the A region as indicated by the rectangular boxes.

Buoyancy convection and surface tension-driven convection are typical melt flows on the earth. Messmer et al. found that the

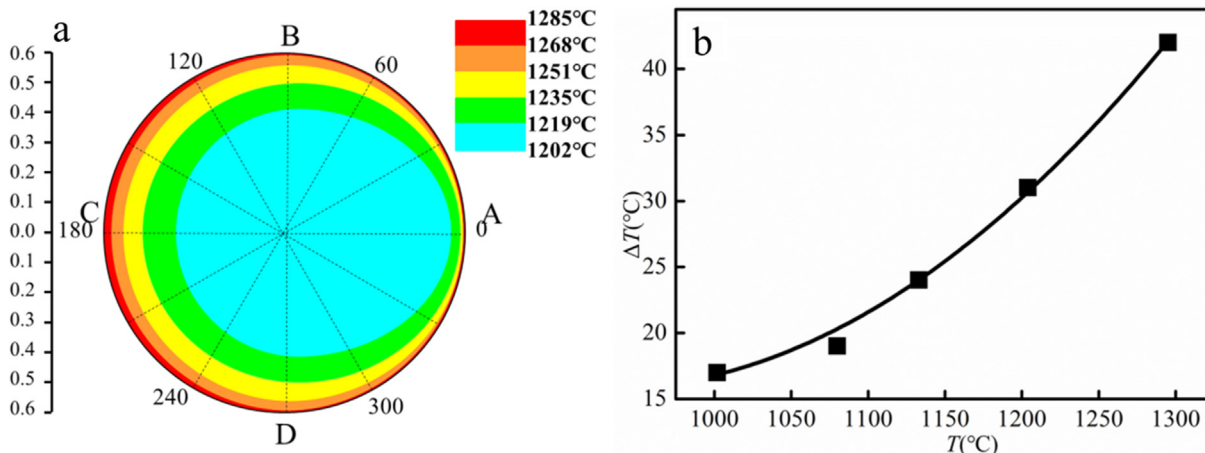


Fig. 2. (a) Temperature distribution in KTN melt at $T = 1245$ °C; (b) Temperature difference $\Delta T = T_C - T_A$ changes against the temperature.

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