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Experimental investigation on the effect of crystal and crucible rotation on thermocapillary convection in a Czochralski configuration

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A B S T R A C T

Experimental investigation is conducted to understand the effects of crystal and crucible rotations on the thermocapillary convection in a Czochralski configuration. A transparent fluid (0.65cSt silicone oil with $Pr = 6.7$) is contained in a cylindrical model crucible with the inner diameter of 92 mm and the depth of 2 mm. A copper disc with the diameter of 46 mm is used to simulate the crystal. The rotation rates of the crystal and crucible are 0 ~ ± 60 rpm and 0 ~ ± 1 rpm, respectively. Results indicate that there is a transition from stable axisymmetric flow to a three-dimensional oscillatory flow with the increase of the thermocapillary Reynolds number. The critical thermocapillary Reynolds number for the flow pattern transition decreases with the increase of the crystal rotation rate, and it is greater for the crystal–crucible co-rotation, compared to the counter-rotation. When the crystal rotation rate is small, the thermocapillary force is dominant, and the oscillatory flow behaves as the hydrothermal waves. The crystal rotation has only a slight effect on the wave number and propagation angle of the hydrothermal waves. At a high crystal rotation rate, the hydrothermal waves will transit to rotating waves. The temperature fluctuation of the hydrothermal waves is suppressed by the crucible rotation.

1. Introduction

The Czochralski (Cz) method is the most frequently-employed crystal growth technique in the industry. In Cz crystal growth, the quality of a single crystal depends mainly on the flow of the melt [1,2], which is generally driven by the buoyancy and thermocapillary forces, centrifugal and Coriolis forces due to the crystal and crucible rotations. Therefore, many researches were devoted to the melt flow in the Cz system during the past few decades [3–7].

In the experimental research on model Czochralski-oxide melts with crystal and crucible rotation and differential heating, Jones [8] reported four types of flow patterns as the rotation rate was increased. Tanaka et al. [9] observed the similar flow patterns depending on the crucible rotation rate in the experiments of the silicon melt without crystal. Nakamura et al. [10] found that the number of thermal waves increases with the increase of crucible rotation rate in a Cz configuration with a carbon-dummy crystal. Haslavsky et al. [11,12] studied the effect of crystal rotation rate (0–5 rpm) on steady-oscillatory convection utilizing thermocouples and interferometer. They found that the critical temperature difference significantly decreases even at a low crystal rotation rate. The oscillation frequency is power dependences of Grashof numbers. When the buoyancy is strong enough, the effect of crystal rotation can be neglected. Lee and Chun [13,14] performed a set of experiments on the effect of disk rotation on oscillatory buoyancy-driven convection in a Cz model melts. They found three distinct flow regimes for a wide range of Pr number (10^{-2}–10^{3}), which are the buoyancy-driven flow, baroclinic-wave, and rotation-driven flow. The regular baroclinic thermal waves were also observed at a low thermal Rossby number. With the increase of the thermal Rossby number, these regular waves become irregular. Schwabe et al. [15,16] grew the $NaNO_3$ crystals with large convex interface deflection from the melt in the model experiments. They showed that the critical rotation rate connected with a flow pattern transition increases as the convex interface deflection of crystal-melt or the strength of buoyant-thermocapillary flow increases. The crystal rotation rate for growing flat crystal-melt interface was also measured. It was concluded that the buoyant-thermocapillary convection is stronger at a larger rotation rate [7]. Kanda et al. [17] investigated the effects of the crystal and crucible rotations on the instability behaviors and flow patterns. At

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relatively small difference between the crystal and crucible rotation rates, the barotropic instability generates multiple vertically coherent vortices, whose number decreases with the increase of the rotation rate difference. Spoke patterns on the free surface in different depths (3, 8, and 200 mm) of the silicon melt were observed by CCD camera [18]. It was found that the number of spokes depends on the depth of the silicon melt. Son et al. [19–21] investigated the thermal field of Wood's metal which had a similar Prandtl ($Pr = 0.019$) number with the silicon melt for various crucible and crystal rotation rates in a Cz system. The temperature fluctuations and the flow velocities were measured using thermocouples and incorporated magnetic probe, respectively. The results revealed that the rotations of the crucible and crystal influence the frequency and amplitude of temperature fluctuation, and the flow fields. The maximum thermal fluctuation is located under the crystal, and migrates towards the crucible sidewall with the increase of the crystal rotation rate. Near the edge of the crystal the thermal fluctuation and the azimuthal velocity decrease as the rate of crystal counter-rotation with the crucible increases. However, with the increase of the co-rotation rate of the crystal, the thermal fluctuation remains strongly, while the azimuthal velocity decreases. After that, the corresponding numerical simulations were also performed in Refs. [22,23].

In the works mentioned above, the buoyancy plays an important role in the flow. However, under the microgravity condition, the buoyancy effect is excluded. Schwabe et al. [24,25] investigated the thermocapillary flow of the fluid with $Pr = 6.7$ in an open annular gap with various aspect ratios and depths ($d = 2.5–20$ mm). In a shallow pool, the concentric steady convection rolls embedded into the main thermocapillary roll were observed at a small temperature difference. At a higher temperature difference, the hydrothermal waves were experimentally identified and the number of m-fold temperature patterns decreases with the decrease of the aspect ratio of the annular pool. In the deep pool, the oscillation was found to be more complicated. Under normal gravity, the HTWs only exist in the shallow pool, and the longitudinal rolls will replace it in thicker layers [26]. Shi et al. [27] analyzed the thermocapillary flow in a rotating annular pool of 0.65cst silicone oil with the depth of 1 mm by numerical simulation and linear stability analysis. Recently, Wu et al. [28,29] performed a series of numerical simulations in a shallow cylindrical pool with a disc on the free surface. The stability diagrams with various crystal and crucible rotation rates were presented, which showed the critical conditions for the onset of flow instabilities.

In this work, the effect of rotation on the thermocapillary convection was experimentally studied in a shallow Cz configuration with the depth of 2 mm. The flow patterns and temperature signals are presented at different temperature differences between the crystal and sidewall of the crucible, and rotation rates. The critical thermocapillary Reynolds numbers for the onset of flow instabilities are determined.

2. Experimental apparatus and methods

The schematic diagram of the experimental apparatus is shown in Fig. 1. A copper disc with radius of $r_s = 23 \pm 0.1$ mm is in contacted with the free surface of the test liquid that is contained in a cylindrical copper crucible with radius of $r_c = 46 \pm 0.1$ mm. The working fluid is the transparent 0.65cSt silicone oil. The physical properties are listed in Table 1. In order to prevent the formation of meniscus, a step plane at the inner rim of the crucible is processed. The upper surface of the plane is painted with FC-725 to avoid wetting by the silicone oil [25]. To form an adiabatic boundary condition, a high quality plexiglass (thickness 10 ± 0.1 mm) with small thermal conductivity is used as the bottom of the crucible. The temperature difference ΔT between the crucible sidewall (T_c) and the model crystal (T_s) is measured using T-type thermocouples with precision of ± 0.1 K. As shown in Fig. 1, four standing thermocouples are located near the corner of the model crystal and other four thermocouples in inner sidewall of the crucible uniformly. The wall temperatures T_s and T_c ($T_c > T_s$) are controlled by two thermostatic baths, respectively. The precision of the thermostatic baths is ± 0.1 K. To measure the temperature under the free surface, a thermocouple denoted as P is located at 1 mm below the free surface and 5 mm from the crystal. All temperature data are converted into a computer through a data acquisition system (HP 34970A). The rotations of model crystal and crucible are controlled by Panasonic servo motors (MSMD-100W and MSMD-400W, respectively). The resolution ratio of rotation for servo motors is 0.036°. The position of crystal is adjusted by using an automatic lifting platform with the precision of ± 3 μm. Furthermore, to avoid the influence of vibration as much as possible, the whole experiment system is installed on a vibration isolation platform.

Temperature fluctuation pattern on the free surface is observed by the schlieren method. As shown in Fig. 1, an optical fiber (diameter 1.5 mm) to transfer the light of a medical cold light source (150 W) is located below the pool as a point source. The light passes through the plexiglass and fluid layer, and then is projected on the screen. It is noted that the refractive index of liquid varies with the fluid density, which depends on the temperature. After the flow destabilization, the temperature of the liquid varies with time and space. As the light passes through the unstable convective liquid layer, the image projected on the screen can reflect the temperature fluctuation on the free surface. Then, the schlieren images on the screen are recorded using a digital camera. In order to avoid the influence of environmental light, the experiments are operated in a dark room.

The ambient temperature is controlled at 293.15 K to weaken the volatile of the silicone oil. Based on the experimental results, when the crucible rotation rate exceeds 1 rpm, the temperature fluctuation induced by the thermocapillary convection is very small so that the observation on the flow pattern by employing the Schlieren method becomes very difficult. However, this kind of fluctuation still exists even at a very high crystal rotation rate. Therefore, the ranges of the crucible and crystal rotation rates are $0 \sim \pm 1$ rpm and $0 \sim \pm 60$ rpm, respectively. The minus rotation rate means the counter-clockwise direction. For the purpose of examining the effect of crystal rotation rate on the thermocapillary convection, the silicone oil with a depth of 2 mm is adopted in the experiments. It should be pointed out that there are some differences between the present experimental model and the industrial Cz progress [30], including shallow pool, isothermal cold model crystal and thermally isolated bottom et al.

In general, the dynamic bond (Bo) number is used to evaluate the ratio of the buoyancy convection to thermocapillary convection, which is defined as follows [26]:

$$Bo = \frac{\rho g \rho_T d^2}{r_T}, \tag{1}$$

where ρ is density, g is the gravitational acceleration ($g = 9.81$ ms^{-2}), ρ_T is the thermal expansion coefficient, r_T is the temperature coefficient of surface tension. The value of Bo is equal to 0.5 at $d = 2$ mm. Therefore, the thermocapillary force is dominant, as reported by Peng et al. [26]. Without rotation, it takes approximately 6 h to completely volatilize the working fluid with 2 mm in depth when ΔT is 8 K, which corresponds to the evaporating Biot number of 2.3. In our experiment, the experiment for