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Experimental study on complex flow of a binary mixture in Czochralski configurations with different aspect ratios and rotation rates

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ABSTRACT

This paper reported the experimental study on complex flow stability of a binary mixture in the Czochralski configurations subjected to a radial temperature gradient. The working fluid was the *n*-decane/*n*-hexane mixture with an initial mass fraction of 50%. Results show that the rotation of crucible and crystal can promote the flow destabilization at a small aspect ratio. However, the crucible rotation can suppress the flow instability at a large aspect ratio. When the thermocapillary Reynolds number exceeds a certain threshold value, the hydrothermal waves, the bud shaped and fringes spokes appear in proper order with the increase of the aspect ratio. Furthermore, the flow patterns are also dependent on the crystal and crucible rotation rates. When the crystal and crucible rotate synchronously, the temperature fluctuation frequency in the binary mixture is nearly the same with that in the pure fluid at a small aspect ratio; however, there is a large difference at a large aspect ratio. Moreover, the temperature fluctuation amplitude in the binary mixture is greater than that in the pure fluid at a small aspect ratio, but it is contrary at a large aspect ratio.

1. Introduction

The classic physical phenomenon about thermocapillary-buoyancy convection is ubiquitous in the natural and industrial processes. For example, the coupled thermocapillary-buoyancy convection plays a key role in the Czochralski (Cz) crystal growth [1–3] and heat pipe technologies [4,5]. Generally, linear stability analysis, numerical simulations and corresponding experiments are the primary approaches to understand the underlying physical mechanism of thermocapillary-buoyancy convection. Smith and Davis [6] first conducted linear stability analysis of thermocapillary convection in an infinitely extended fluid layer and discovered two types of three-dimensional instability, which were stationary longitudinal rolls and oblique hydrothermal waves (HTWs). Considering the influence of buoyancy force on thermocapillary convection, Shi et al. [7] conducted a series of linear stability analysis and found that the buoyancy could give rise to the unsteady flow. In addition, the critical Marangoni number and the critical azimuthal wave number decrease with the increase of the layer depth. Peng et al. [8] performed the unsteady three-dimensional numerical simulations on thermocapillary-buoyancy flow in an annular pool filled with 0.65cSt silicone oil. It was found that the coexisting pattern of the HTWs and the three-dimensional oscillatory flow (3DOF), and the three-dimensional stationary flow (3DSF) appear orderly with the increase of the layer depth. Schwabe and Benz [9] carried out a series of experiments on thermocapillary-buoyancy flow of 0.65cSt silicone oil ($Pr = 6.7$) with an outer heated cylinder and an inner cooled cylinder. Via IR-images of the annual gap's free surface, they observed various flow structures under microgravity and normal gravity.

In the Czochralski growth technique, the rotation of crucible and crystal is rather necessary for growing good homogeneity crystals. Jing et al. [10] conducted a set of three-dimensional numerical simulations of $LiNbO_3$ melt flow taking into account of the crystal rotation, and predicted that the axisymmetric flow (driven by the temperature difference) without the reverse flow (driven by the crystal rotation), the axisymmetric flow with the reverse flow and the nonaxisymmetric unsteady flow appeared in order with the increase of the crystal rotation rate. Nakamura et al. [11] carried out a series of experiments on thermal convection of silicon-melt with different crucible rotation rates in a Czochralski system. It was reported that the thermal wave number increases as the crucible rotation rate increases, and the crucible rotation rate is higher than the azimuthal propagation rate of the thermal wave. Combining the crystal and crucible rotation, Wu et al. [12,13] conducted a series of unsteady three-dimensional numerical simulations to understand the flow patterns driven by the combined Marangoni

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effect and the rotation of crucible and crystal in a Czochralski configuration. It was found that the axisymmetric and steady flow with low rotation rate and small temperature difference bifurcates to a three-dimensional oscillatory flow with higher rotation rate or larger temperature difference. Shen et al. [14,15] performed a series of experimental investigations to understand the effects of crystal and crucible rotation in a Czochralski configuration. With the increase of crystal rotation rate, the hydrothermal waves and the rotation waves appear orderly in a shallow pool, and the critical thermocapillary Reynolds number decreases. In addition, the critical thermocapillary Reynolds number with the co-rotation of crystal and crucible is higher than that with the counter-rotation of crystal and crucible.

For a binary mixture solution, the flow can become more complex because of the Soret effect, which can drive a solutal-capillary convection. Shklyave et al. [16,17] performed the linear stability analysis on Marangoni convection driven by the coupled thermal-solutal force in an infinitely extended fluid layer of a binary mixture. Bergman [18] carried out two-dimensional numerical simulations of a double-diffusive convection in a rectangular cavity to investigate the thermo-solutal convection driven by concentration gradient and temperature gradient. Yu et al. [19,20] conducted a series of three-dimensional numerical simulations and experiments of thermocapillary-buoyancy flow of a binary mixture in a shallow annular pool without rotation to reveal the influence of the Soret effect on the flow instability. It was found that the Soret effect could destabilize the flow.

In summary, thermocapillary and solute-capillary forces, rotation centrifugal force and buoyancy are the main potential forces in the crystal growth technology, and the researchers have considered mainly on one or two aspects of them. It should be noted that there are few literatures including all of the four forces. Therefore, we conducted systemic experimental studies on complex flow stability of a binary mixture in the Czochralski configurations where all the factors have been considered.

2. Experimental apparatus and techniques

Fig. 1 shows the schematic diagram of the experimental apparatus with the Czochralski configuration. The cylindrical pool and the copper disk over the pool are used to imitate the crucible and the crystal. The radius of the cylindrical pool and the disk are $r_c = 46 \pm 0.1$ mm and $r_s = 23 \pm 0.1$ mm, respectively. In order to make the light pass through the bottom easily, a transparent plexiglass with the thickness 10 ± 0.1 mm is used as the bottom of the crucible. Except that, a step plane at the inner rim of the crucible is processed to avoid the effects of the meniscus. An automatic lifting platform with the precision of ±3 μm is used to adjust the crystal to touch the free surface exactly. A platform vibration isolator is used to reduce the influence of external factors. The rotation of the crucible and the crystal is controlled by two servo motors whose resolution ratio of rotation is 0.036°.

For a good thermal conductivity, the crystal and the crucible are made of red copper. The temperature difference $\Delta T = (T_c - T_s)$ between the crucible (T_c) and the crystal (T_s) is controlled by two thermostatic baths with the precision of ±0.1 K. There are four T-type thermocouples with the precision of ±0.1 K to be arranged to measure the temperature difference ΔT of the crucible and the crystal at the inner sidewall, respectively. Besides, another T-type thermocouple is located at 1 mm below the free surface and 10 mm from the inner sidewall of the crucible to monitor the temperature fluctuation of the working fluid. All temperature data are obtained through the conversion of HP data acquisition (HP 34970A).

The working fluid are the pure n-hexane and a binary mixture of n-decane/n-hexane (mass fraction of 50%) whose physical properties are referred to Ref. [19] and listed in Table 1. The schlieren method is used to observe the temperature distribution on the free surface. A medical cold light source (150 W) is located below the bottom of the pool as a point source. The light passes through the working fluid layer and projects on the optical screen. The refractive index of the working fluid depends on the density, and

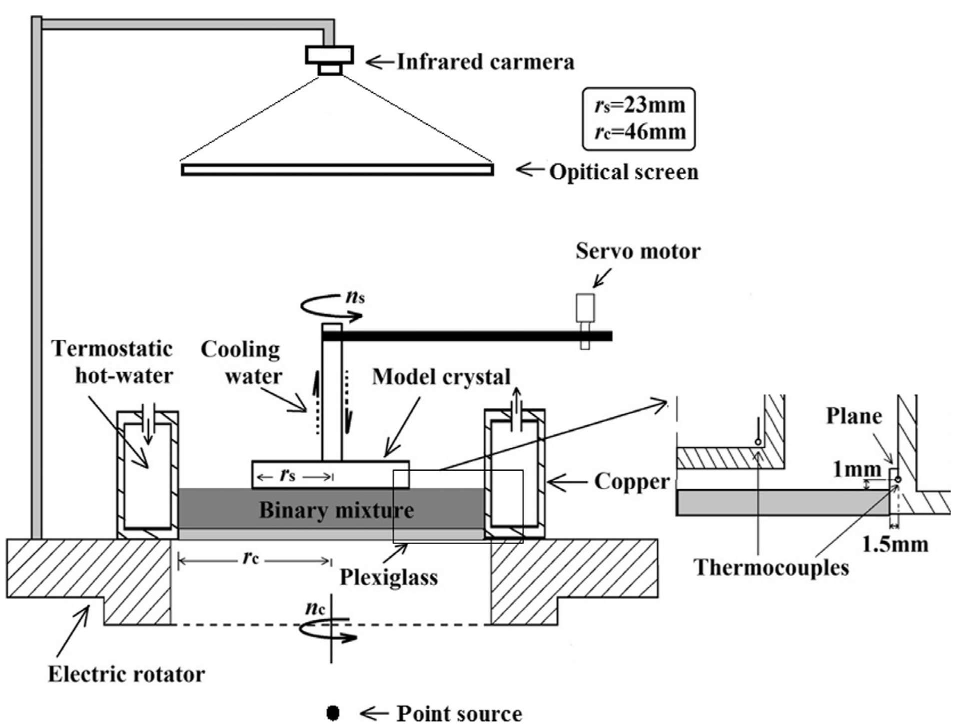

Fig. 1. The experimental apparatus.