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Study on the response of dendritic growth to periodic increase–decrease pressure in solidification via in situ observation using succinonitrile

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ABSTRACT

A novel experimental apparatus for in situ observation of dendritic growth in directional solidification under dynamic pressure and thermal conditions was built, based on the principles of direct squeeze-casting, and the response of dendritic growth to an increase–hold–decrease pressure and a periodic increase–decrease pressure was investigated using succinonitrile. The results showed that, when continuous increase–hold–decrease pressure was applied, the dendrites underwent a growing and re-melting process where the dendritic re-melting phenomenon with fish-bone-like dendrites in the pressure decline stage was observed for the first time. When periodic increase–decrease pressure was applied, dendrites re-grew and were re-melted periodically owing to the change in effective undercooling, which is the difference between the equilibrium melting temperature under pressure T_m^P and the temperature at the dendrite tip T_{tip}, as expressed by $\Delta T_{eff} = T_m^P - T_{tip}$, whereas secondary dendrite arm spacing increased with the re-growth and re-melting cycle. The relationship of the velocity of the dendrite tip v with the effective undercooling ΔT_{eff} and pressure change ΔP was derived and expressed as $v = A\Delta T_{eff} + B\Delta P^2 + C$ where the effective undercooling plays a primary role in determining the tip velocity.

1. Introduction

Dendritic growth is a critical phenomenon in solidification and has been investigated for decades, as dendrite microstructure exerts a significant influence on the final physical properties of castings for engineering interest [1].

Squeeze casting is a special casting process in which molten metal solidifies under applied pressure, producing high-integrity castings with excellent mechanical properties [2–5]. Although it has been well established that pressure has a pronounced effect on solidification [6–13], the dendritic growth in squeeze casting is not well understood.

As it is difficult to observe dendritic growth evolution of metals—especially when solidified under pressure—using conventional metallographic techniques or even using various synchrotron X-ray imaging techniques [14–16], transparent model materials have been selected as a substitute in many studies for in situ observations of solidification under pressures [17–27]. Sawada et al. [18,19] developed an experimental system to investigate the influence of pressure on dendritic growth of NH_4Cl solution. A stainless-steel pressure cell with optical view windows was fabricated, and an assembled inner cell in which dendrites can grow freely was mounted inside the pressure cell. The pressure was applied to the solution via pressure-transmitting oil, and it determined dendrite growth by affecting the supersaturation of the solution. It was demonstrated that tip growth velocity can be quickly changed by a pressure jump. Koss et al. [26,27] established an experimental setup wherein a stainless-steel growth chamber was mounted inside a bath with uniform temperature and pressure. In this setup, a single leading dendrite of succinonitrile (SCN) was allowed to grow in the growth chamber to study the transient behavior of equiaxed dendritic growth under pressure. It was observed that tip velocity started changing almost immediately after the change in pressure, as the step change in pressure caused a corresponding change in the equilibrium melting temperature according to the Clapeyron effect [23], and thus changed the thermal driving force of dendritic growth. In addition, the pressure was expected to destabilize the solid/liquid (S/L) interface, leading to the initiation of side branches. Börzsönyi et al. [10,28] investigated the effects of spatially homogeneous time-periodic external forcing on dendritic solidification. It was indicated that the frequency of dendritic sidebranching can be tuned by oscillating pressure or heating.

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Although experimental apparatuses to study dendrite growth under pressure have been developed [5,18,19,26,27], these experiments only focused on equiaxed dendrite owing to the limitation of these apparatuses. In order to reduce the time required for dendrite tip to achieve steady-state conditions, a single dendrite is allowed to grow in these growth chambers, and the temperature bath adopted by Koss et al. [26,27] is uniform, all of which render it difficult to realize the directional solidification of multiple grains. Accordingly, it is necessary to build a new experimental setup for the in-situ observation of the evolution of columnar dendrites in directional solidification under complex pressures, such as periodic increase–decrease pressure.

Most of the investigations on pressurized solidification of transparent materials are focused on dendrite growth with the increase in pressure, but the re-melting of dendrite during pressure declining stage and the dendritic re-growth and re-melting process under periodic increase–decrease pressure have not been investigated. The change of dendrite behavior with pressure has been explored in previous experiments [5,18,19,26,27]; however, the quantitative relationship of tip growth velocity with dynamic pressure and thermal conditions has not been established yet.

In this work, a novel experimental apparatus was built to realize the in situ observation of dendritic growth and re-melting of multiple grains in directional solidification under combined dynamic pressure and thermal conditions, and the dendritic growth and re-melting under continuous increase–hold–decrease pressure and periodic increase–decrease pressure were investigated by using SCN. The correlation among tip velocity, effective undercooling—as expressed by $\Delta T_{eff} = T_m^P - T_{tip}$—and pressure was calculated from experiments and analyzed graphically. The relationship of tip velocity with the effective undercooling and pressure was derived and formulated.

2. Experimental apparatus and procedure

A novel experimental apparatus was built to conduct in situ observation experiments, as shown in Fig. 1, which was designed following the principles of direct squeeze-casting. The experimental apparatus contains five parts, i.e., mold, pressure control system, temperature control system, data acquisition system, and observation system.

The stainless steel mold of the apparatus includes a punch (Φ15 mm in diameter and 56 mm in height) and a lower mold (40 mm × 40 mm × 85 mm), forming a cylindrical mold cavity, which contains the sample material, SCN (99.99% pure), a transparent body-centered cubic material with a nominal melting point of approximately 58 °C.

The digital pressure control system can modulate the applied pressure on liquid SCN accurately via a pressure sensor connected to the punch.

The temperature gradient in the mold cavity is formed and controlled via a temperature control system, with increasing temperature from bottom to top in the SCN.

The pressure and temperature of liquid SCN were recorded using sensors and a precise data acquisition system manufactured by Integrated Measurement Corporation (IMC, Berlin, Germany).

The dendritic growth of SCN can be observed from the view window, with bright light through the sapphire window, and captured using a high-speed microscope (NAC Memrecam HX-6). As shown in Fig. 1(b), a growth chamber of SCN, a laminar gap, was formed by the mold and two cuboid sapphires. The thickness of growth chamber is only 0.5 mm, to ensure that only one layer of dendrite grows to avoid overlapped dendrite morphology. The whole width and height of the growth chamber made by two sapphires are 15 mm and 18 mm, respectively; the view window through which dendritic growth can be observed is 8 mm in diameter, as shown in Fig. 1(c).

The novel experimental apparatus is a powerful tool to further study the effect of pressure on dendrite behavior and kinetics. The digital pressure control system has a close-loop control, which can ensure the accuracy of applied pressure on melt SCN, rendering it convenient to investigate the response of dendritic growth to different kinds of complex pressures, such as periodic increase–decrease pressure. Moreover, controllable temperature gradient in the growth chamber is a prerequisite for columnar dendritic growth in directional solidification under pressure, which has not been explored in previous experiments.

The present experimental process of crystal growth is described as follows. First, the temperature gradient was set, with the average gradient (G_L) of 0.325 °C/mm, generating a stable S/L interface at ambient pressure (P_0) where dendrite would not grow. Second, the pressure was applied at a pressure speed to the liquid SCN, and responsively, the dendrite began to grow upwards from the bottom or was re-melted according to the variation of pressure. Finally, the dendritic evolution in the center of the view window was captured, and the size of captured area was about 1.5 mm × 2 mm; the tip growth velocity was calculated, and the correlation of tip velocity with pressure and effective undercooling was analyzed.

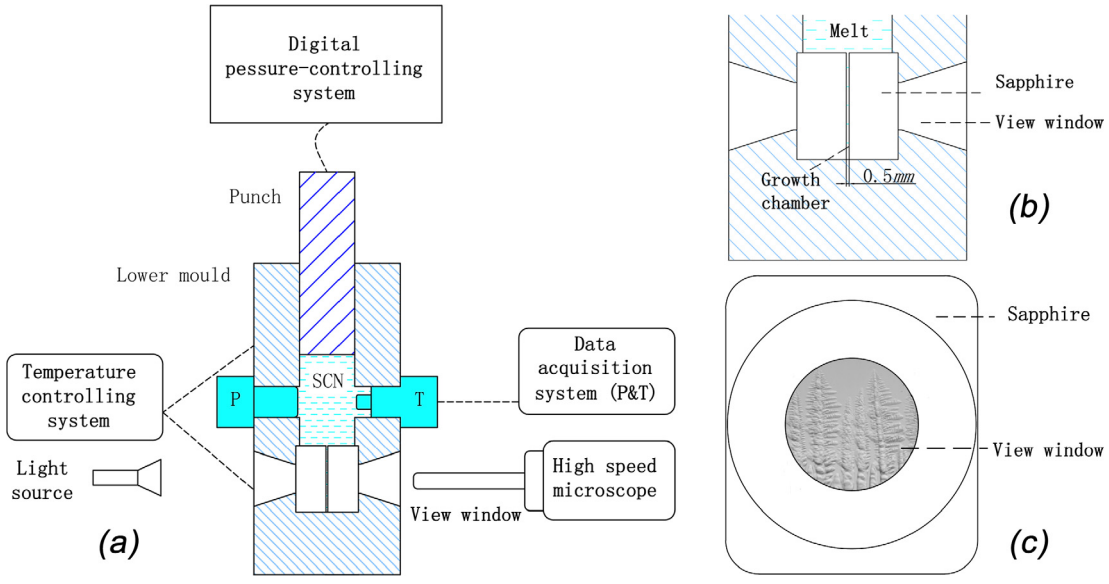

Fig. 1. (a) Schematic of the experimental setup; (b) and (c) the magnification of view window along and perpendicular to the observation direction, respectively.