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Effect of step-wise change in processing pressure on isolated pore growth during controlled directional solidification in small channels

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ABSTRACT

Directional solidification experiments were performed using succinonitrile saturated with nitrogen gas to examine the effects of *in situ* processing pressure changes on the formation, growth, and evolution of an isolated, cylindrical gaseous pore. A novel solidification facility capable of processing small cylindrical samples (I.D. $\leqslant$1.0 mm), under controlled pressure conditions, was developed for the experiments. A new experimental method for growing the isolated pore from a seed bubble is introduced. The experimental results indicate that a step-wise processing pressure change will result in either a transient change in pore diameter or a complete termination of pore growth, demonstrating that pressure changes can be used as a control parameter to influence bubble growth. During steady-state growth, however, pore size shows no dependence on processing pressure. A simple analytical model has been introduced to explain the experimental observations.

1. Introduction

The phenomenon of pore formation during directional solidification of a gas-saturated melt has been examined in a variety of materials through different processing techniques [1–3]. While early studies of pore formation focused primarily on the detrimental aspects of porosity in metal castings, more recent investigations have regarded this phenomenon as useful in the production of porous materials, which are highly regarded for their enhanced mechanical, thermal, and acoustic properties [4–6]. Furthermore, porous metals produced by directional solidification can be superior to porous metals produced by other methods (such as metal foaming, sintering, etc.) because of the ability to control the pore growth process [7,8]. However, a fundamental understanding of the relationship between pore growth and the control parameters is required before this process becomes technically viable. Although the primary parameters influencing pore growth, including solute concentration, solidification velocity, processing pressure, and temperature gradient, have been identified, no controlled studies in a real-time flow visualization context have been performed to establish the cause–effect relationship between processing pressure and pore morphology. As a first step, this study examines the effect of processing pressure changes on the evolution of an isolated pore in a cylindrical channel.

Processing pressure as an *in situ* control parameter has received limited experimental attention. Previous investigations of processing pressure and pore formation share a similar methodology. Multiple samples of porous metal have been produced under various constant pressure conditions, and then the pore characteristics evaluated by post-processing cross-sectional analysis. Using this method, a casual relationship between pore diameter and processing pressure has been developed by examining the numerous pores and showing that the mean pore diameter decreases with increasing processing pressure [9–12]. However, most of these studies have been limited to post-processing analysis because of the opaque nature of the solidification sample metal, which precludes direct observation during solidification.

Direct observation of the dynamic process of pore formation in real-time during solidification is a superior experimental

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technique for understanding the morphological response to parameter changes. Some previous investigations have directly observed pore formation during directional solidification, using both planar and bulk sample configurations. Using a thin planar configuration, Akamatsu and Faivre [13] and Jamgotchian et al. [14] observed the coupled growth of gaseous pores with a solidification interface during directional solidification of CBr_4–C_2Cl_6 and succinonitrile (SCN)–acetone systems, respectively. Also using a thin planar configuration, Han [15] observed pore formation and dynamic bubble motion in a dendritic mushy zone in real-time while studying cyclohexane. These investigations, however, did not include processing pressure as a control parameter during processing. Further, while the thin planar configuration allows excellent visualization of the solid–liquid interface and bubble motion, it is not suitable for understanding three-dimensional pore growth and termination in response to processing pressure changes. Solidification in cylindrical bulk transparent samples allows pore formation to occur in a more relevant setting, while also facilitating real-time observation of the solid–liquid interface and pore growth. In this context, Vasconcellos and Beech [16], and Wei et al. [17] observed steady-state pore formation in real-time during vertical unidirectional solidification of the H_2O–CO_2 system. However, neither of these investigations studied processing pressure as a control parameter. Lee and Hunt [18] observed *in situ* pore formation in a bulk sample of aluminum–copper alloy using an X-ray technique. Using a similar X-ray technique, Catalina et al. [19] also observed pore formation during solidification of a metal alloy; however, the effect of an *in situ* change in processing pressure was not investigated.

Ground-based experiments of pore formation using bulk samples are affected by gravity-driven convection in the melt. Additionally, buoyancy effects caused by gravity can affect pore growth trajectory. To overcome these limitations, Grugel et al. [20] studied pore formation in bulk samples of the transparent succinonitrile–nitrogen (SCN–N_2) system in the Pore Formation and Mobility Investigation (PFMI) on the International Space Station. They observed pore formation in real-time in the absence of gravity-driven convection and buoyancy effects; however, the use of processing pressure as a control parameter was not evaluated. Detrimental pore formation is more prevalent during Space-based materials processing because bubbles inherent in the melt do not rise to the surface as they do on Earth [21]. The current fundamental study on isolated pore growth and evolution can potentially shed insights into the microgravity porosity formation problem.

The purpose of the current investigation is to examine in real-time the effect of a processing pressure change on the formation, growth, and evolution of an isolated gaseous pore during controlled directional solidification. Using small cylindrical samples (I.D.$<$1.0 mm) of the SCN–N_2 system, in a novel horizontal directional solidification apparatus, the current study has been able to eliminate gravity-driven convection in the melt while minimizing the effects of buoyancy on pore growth trajectory, thus approximating the diffusion-limited growth conditions obtained by the PFMI in microgravity. This novel configuration also allows for the isolation of a single (isolated) pore growing near the center of the sample to study the effect of a pressure change on pore morphology.

2. Experimental setup

2.1. Experimental apparatus

A horizontal unidirectional solidification apparatus was developed for the current investigation (Fig. 1a). The apparatus was specifically designed to accommodate small cylindrical glass capillary samples (I.D.$\leqslant$1 mm) such that gravitational effects, including bulk convection in the melt and buoyancy effects on pore growth trajectory, are minimized, while a bulk solidification interface can be observed. The solidification apparatus consists of a hot zone, maintained at a uniform temperature T_H, and a cold zone, maintained at a lower uniform temperature T_c. Each zone is comprised of an upper and lower aluminum cavity through which water at the desired temperature is circulated. The hot and cold zones are separated by a 10 mm test section, and a linear temperature gradient is established in the test section. The temperature in each thermal zone and the linearity of the temperature gradient in the test section were verified by translating a melt sample with an embedded type-K thermocouple through the gradient. The test section is surrounded by thermal insulation, and viewing windows are mounted on both the horizontal and the vertical axes. A CCD camera (JAI CV-M1, Panasonic) is mounted on each axis to provide direct, real-time observation of the solidification interface during processing.

The apparatus includes a computer-based data acquisition and instrument control system. Unidirectional sample translation is controlled using a precision stepper motor (Parker) coupled to a ball-drive linear stage (Parker). The position of the sample in the test section is verified using a precision linear encoder (Heidenhahn). Processing pressure is controlled in real-time using an electronic pressure-control module (Parker VSO EP) connected to a supply of ultra-high-purity nitrogen gas. It is monitored using an independent pressure transducer (Omega).

2.2. Sample preparation

A cylindrical glass capillary tube (Drummond Scientific, ID=1 mm) is filled with a sample of high-purity succinonitrile (99.9%, T_m=331.214 K), under vacuum to minimize atmospheric contamination. The filled glass capillary sample is configured for *in situ* processing pressure control by mounting stainless-steel tubing (Popper, 22G) on each end and sealing it to the capillary tube with vacuum-grade epoxy. The steel tubing is coupled to a flexible pressure hose that is connected to the electronic pressure control module. This novel configuration (Fig. 1b), allows precise control of the processing pressure applied to the sample during isolated pore growth.

3. Experimental procedure

The experimental procedure consisted of five steps: (1) constant pressure gas saturation, (2) seed bubble initiation, (3) seed bubble attachment to the solid–liquid interface, (4) steady-state isolated pore formation, and (5) processing pressure variation. Because each of these steps is unique to this experiment, they are described in more detail below.

3.1. Constant pressure gas saturation

Pore-formation experiments require the introduction of gas into the melt. According to Henry's Law, an equilibrium gas concentration, C_o, is achieved in a melt, when a gas pressure is applied at the melt free surface, according to the equation

$$C_o = \frac{P}{K_H} \tag{1}$$

where P is the applied pressure and K_H is the Henry's constant for the melt–gas system. By controlling the applied gas pressure during the constant pressure gas saturation process, it is possible