



## Effects of solidification rate on pore shape in solid



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### ABSTRACT

The effects of solidification rate on the shape of a pore, resulting from a bubble entrapped by a solidification front, are investigated in this study. Solidification rate plays an important role in the pore shape due to its strong influence on solute concentration, nucleation rate, solute transport across the bubble cap, morphology of the solidification front, etc. Understanding and controlling porosity in solid is important for manufacturing porosity-free materials, and functional materials such as lotus-type porous materials in nanotechnologies and scaffolds in tissue engineering, etc. In addition to self-consistent determination of the cap shape at which balance of pressures and physico-chemical equilibrium are satisfied, this model accounts for solute transport across the cap in three different cases. Case 1 is subject to solute transport from the pore across an emerged cap in the concentration boundary layer along the solidification front in the early stage. Cases 2a and 2b indicate opposite directions of solute transport across a submerged cap in the concentration boundary layer. In contrast to Case 2a, expansion of pore volume in Case 2b exhibits a stronger effect on solute gas pressure in the pore than solute transport in the late stage. The results show that an increase in solidification rate in Case 1 decreases and increases pore radius in the early and late stage, respectively. The bubble is entrapped earlier by increasing solidification rate. The cap cannot be entrapped, because solute concentration increases and decreases rapidly in the late stage in Cases 2a and 2b, respectively. Predicted and measured pore shapes in solid are in good agreement. The pore shape can be controlled via selecting solidification rate to change directions and magnitudes of solute transport.

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### 1. Introduction

Porosity in solids has been well-known to play an important role in biological, engineering, geophysical and climate change sciences, etc. [1–12]. Understanding of pore formation is thus critical. In view of gas solubility in solid usually much less than that in liquid, gas is accumulated ahead of the solidification front [13–16]. Supersaturation nucleates bubbles on the solidification front [15–21]. The emanated gas bubbles may remain at the solidification front where they may be trapped in the solid as discrete pores, chains of pores, or gas tubes [22,23]. Pore formation can be divided into different stages: nucleation on the solidification front, spherical growth, solidification rate-controlled elongation, disappearance of the bubbles, and formation of the pores in solid [24].

Solidification rate strongly influences different stages of pore formation. Bubble nucleation on a solidification front is affected by

solidification rate. Carte [25] observed that bubble were formed at the ice-water interface, when concentration of dissolved air reached a critical value which, for solidification rates greater than 2 mm/min, corresponded to a supersaturation ratio of 30. Park et al. [18] and Drenchev [19], respectively, showed that an increase in solidification rate increases pore nucleation rates due to an increased difference between pressures in the pore and the ambient, and supersaturation ratio in the melt. The free-energy barrier and supersaturation ratio also increase with solidification rate. The solidification front is planar when the solidification rate is low. On increasing solidification rate, the solidification front becomes cellular, dendritic and equiaxed [1,6,26]. The pores nucleate preferentially at the grain boundary grooves on the solidification front. In this case, the pores nucleate just behind the tips of the cells [21,23].

The pore size usually decreases as solidification rate increases [18,27,28]. During fast growth there was less time for diffusion and the bubble decreased in cross section based on solute gas conservation viewpoint. However, another interpretation is that an increase in solidification rate enhances mass transport to the pore and increases gas pressure in the pore. Pore radius therefore

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decreases in order to satisfy balance of pressures at the cap [27,28]. Murakami and Nakajima [23] observed pore formation of water-carbon dioxide solutions. Columnar pores were formed at low growth rates. The length of the columnar pores became shorter and approached spherical pores or few pores as the freezing rate increased. Similar results were observed by Park et al. [18] and Yoshimura et al. [29] for lotus-type porous metals containing aligned long columnar pores.

Morphology of a pore is also affected by solidification rate. Bari and Hallett [30] observed that bubbles grew as cylinders below 5  $\mu\text{m/s}$ , becoming egg shaped, the narrow ends toward the solidification front at higher solidification rates. Yoshimura et al. [29] also observed features of oxygen gas bubbles incorporated into a growing ice crystal at various ice growth rates. A decrease in growth rate results in four patterns of the bubble shapes from (a) egg-shaped bubbles, (b) egg-shaped bubbles and cylindrical bubbles, (c) cylindrical bubbles, to (d) bifurcated cylindrical bubbles.

Solidification rate is responsible for entrapment of a bubble by a solidification front. A bubble will be entrapped by a solidification front if the ratio between bubble growth rate-to-solidification rate is less than unity [21], or negligibly small [31,32], or solidification rate is greater than a critical velocity determined by a force balance between a bubble and solidification front [33].

Evaluation of solute gas transport through the bubble cap as a function of solidification rate is required for controlling entrapment of a bubble into solid. Solute is not be accumulated at the bubble cap, because solubility of solute in gas is greater than that in liquid. The bubble is initiated from heterogeneous nucleation due to solute super-saturation. For a high solidification rate resulting in a thin concentration boundary layer solute gas is thus transferred from the bubble to liquid in the early stage. Mass transfer rate governed by mass transfer coefficient is based on difference in concentrations between a location far from the solidification front and bubble cap surface [34]. For a small solidification rate, the bubble cap is submerged into a thick concentration boundary layer along the solidification front. Solute gas transport across the bubble cap thus can be effectively evaluated by introducing a mass transfer coefficient due to the difference in concentrations in liquid at the solidification front and location far from the solidification front [27,28,35,36]. Lee et al. [36] describe that at the solid–melt interface away from the bubble, solute gas is rejected into the liquid as solidification progresses. When solidification occurs at the intersection of the solid–melt interface and the rim of the bubble, the solid again first rejects gas solute to the liquid side at the bubble surface. The gas is quickly released from the melt into the bubble in compliance with Henry's law.

In this work, the shapes of a pore resulting from a bubble entrapped by a solidification front with different solidification rates are predicted. Two models corresponding to an emerged and submerged cap in concentration boundary layers, leading to different directions of solute transport are studied. The bubble cap is satisfied by mass, momentum, species transport and physico-chemical equilibrium [27,28]. Controlling of solidification rate responsible for different directions and magnitudes of solute transfer across the cap and shapes of the pore is therefore provided.

### 1.1. System model and analysis

A pore in solid resulting from entrapment of a tiny bubble on a solidification front is illustrated in Fig. 1(a). From geometry viewpoint, the pore shape in solid to a zeroth approximation is delineated by tracing the contact angle of the bubble cap [28,31,32]. The shape of the bubble cap beyond the solidification front is determined by the Young-Laplace equation, governing normal stress balance between gas, capillary and liquid pressures. Solute gas

pressure in the pore is determined by solute transport across the cap of the pore in different directions, as illustrated in Fig. 1(b) and (c), corresponding to an emerged and submerged bubble cap in the concentration boundary layer along the solidification front. Thickness of concentration boundary layer is determined by solidification rate. The major assumptions made are the following:

- 1 The model system is axisymmetric.
- 2 The tiny bubble is characterized by a small Bond number, leading to apply available perturbation solutions to predict a lumped micro-bubble in a nearly spherical cap shape. In most cases, Bond number can be as small as  $10^{-5}$ , based on a typical bubble radius of  $10^{-5}$  m.
- 3 The system is static. Tangential and normal viscous stresses in the liquid are therefore neglected. Liquid pressure is gravitational force-dependent hydrostatic pressure.
- 4 Mass transfer across the bubble cap leads to unsteady variation in moles of solute gas in the pore, given by Ref. [34].

$$\frac{d\tilde{n}_g}{d\tilde{t}} = \tilde{h}_D \tilde{A}_c (\tilde{C}_\infty - \tilde{C}_c) \quad (1)$$

Since the bubble is usually initiated from heterogeneous nucleation due to solute super-saturation, a high solute concentration in the pore results in solute transfer from the pore to surrounding liquid in the early stage. On the other hand, solute transfer to the pore can be evaluated by Refs. [27,28,35,36].

$$\frac{d\tilde{n}_g}{d\tilde{t}} = \tilde{h}_D \tilde{A}_c (\tilde{C}_c - \tilde{C}_\infty) \quad (2)$$

The term on the right-hand side of Eq. (2) can be considered as a solute source. Since solute segregation is greater than initial concentration in the liquid by the factor of equilibrium partition coefficient, Cox et al. [35] and Lee et al. [36] showed that the concentration gradient between the solidification front and bubble cap results in solute transport from the side along the solidification front to the pore

$$\frac{d\tilde{n}_g}{d\tilde{t}} = \tilde{h}_D \tilde{A}_c \left( \frac{\tilde{C}_\infty}{k_p} - \tilde{C}_c \right) \quad (3)$$

which indicates solute gas concentration in the pore is affected by a solute source due to accumulated solute concentration ahead of the solidification front. The difference in concentrations between liquid on the solidification front and bubble cap to a linear approximation can be expressed by

$$\frac{\tilde{C}_\infty}{k_p} - \tilde{C}_c \sim \tilde{C}_c - \tilde{C}_\infty \quad (4)$$

Equations (3) and (4) thus indicates that solute gas is transferred from the surrounding liquid into pore [35,36]. Directions of solute transport across the bubble cap between Eqs. (1) and (2) are opposite. It is noted that solute transport across the cap can also be realistically evaluated by proposing a summation of Eqs. (1) and (2) with their weighting factors.

5. Physico-chemical equilibrium implies that Henry's law is satisfied at the bubble cap

$$\tilde{C}_c = \frac{\tilde{p}_g}{K} \quad (5)$$

which indicates that solute gas pressure is proportional to solute concentration in the liquid at the liquid-gas interface.

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