Effects of initial contact angle on pore shape in solid

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ABSTRACT

The pore shape affected by initial contact angle of a bubble entrapped by a solidification front is predicted. Initial contact angle affects bubble nucleation, surface area and solute transport across the bubble cap, solid gas pressure in the pore and shape of pores in solid. Porosity influences microstructure and functional properties of materials, etc. This work accounts for solute transport across the coupled shape of cap in balance of pressures and physico-chemical equilibrium. Similar to previous work, it shows that pore shape depends on directions and magnitudes of solute across the cap. In contrast to Case 2, solute transport in Case 1 is from the pore into surrounding liquid in the early stage. The results find that a decrease in initial contact angle expedites bubble entrapment in Case 1. Solute gas pressure in the pore further exhibits three stages for initial contact angle greater than 90°. Significant drops occur in the early and late stages. In the middle stage, where contact angle is near 90°, solute concentration at the cap is about initial solute concentration in liquid, and responsible for length of the pore. Except for initial contact angle less than 90°, an isolated pore cannot be formed in Case 2. The predicted pore shape agrees with experimental data.

1. Introduction

Substances containing a number of pores have characteristics different from those of bulk substances, such as a low density and a large surface area. Therefore, they can be utilized as lightweight materials, catalysts, electrodes, vibration and acoustic energy absorption materials, impact energy absorption materials, and so on, or suffered by defects of stress concentration [1–9]. It has been known that solute gas can be rejected by the solidification front and accumulated ahead of the solidification front [10,11], leading to concentration to be high enough for bubbles to nucleate [12–18]. A pore in solid is initiated and developed from a bubble nucleated due to supersaturation ahead of the solidification front. On the other hand, complicated mechanisms and shape of forming lotus-type porous metals containing aligned long columnar pores are affected by different percentage of the mixture of hydrogen and argon imposed on the surrounding of free surface of liquid [19,20]. Zhang et al. [21] proposed that rather than retarding effect of inert gas argon, hydrogen gas can promote and retard nucleation in the lower and higher hydrogen pressure, respectively.

Pore formation is strongly affected by initial contact angle. Nucleation rate increases as initial contact angle decreases [14], as a result of a decrease in the free-energy barrier for nucleation. Increasing the nucleation sites on the solidification front decreased porosity and pore size and increased pore number per unit area [22]. A reduction in

average pore diameter in an ordered porosity structure and increase pore number per unit area were possible by decreasing surface tension of liquid via small addition of alloying elements.

Contact angle is also the dominant factor affecting the pore shape in solid [23,24]. Geometrically speaking, the pore shape in solid is delineated by the contact angle of the bubble cap [24]. Contact angle determined not only the growth of the bubble cap, but also stability and morphology of the pore shape in solid [25]. An increase in contact angle increases surface area of the bubble cap, leading to enhanced solute gas transport through the cap. Furthermore, contact angle also indicates if height of the bubble cap is smaller or greater than thickness of concentration boundary layer along the solidification front. Provided that the bubble cap subject to a high contact angle is emerged through the concentration boundary layer, solute gas transport is from the pore across bubble cap into surrounding liquid [26], especially in the early stage. On the other hand, a submerged bubble cap with a small contact angle in the concentration boundary layer results in solute transport from the surrounding liquid into the pore [23,27,28].

In this work, development of the pore shape in solid during entrapment of a bubble by a solidification front for different initial contact angles of the bubble cap is predicted. Same as previous work [29–31], solute transport across the bubble cap and the shape of pore in solid are determined by contact angle of the bubble cap. Accounting for the effects of initial contact angle on different directions and magnitudes of
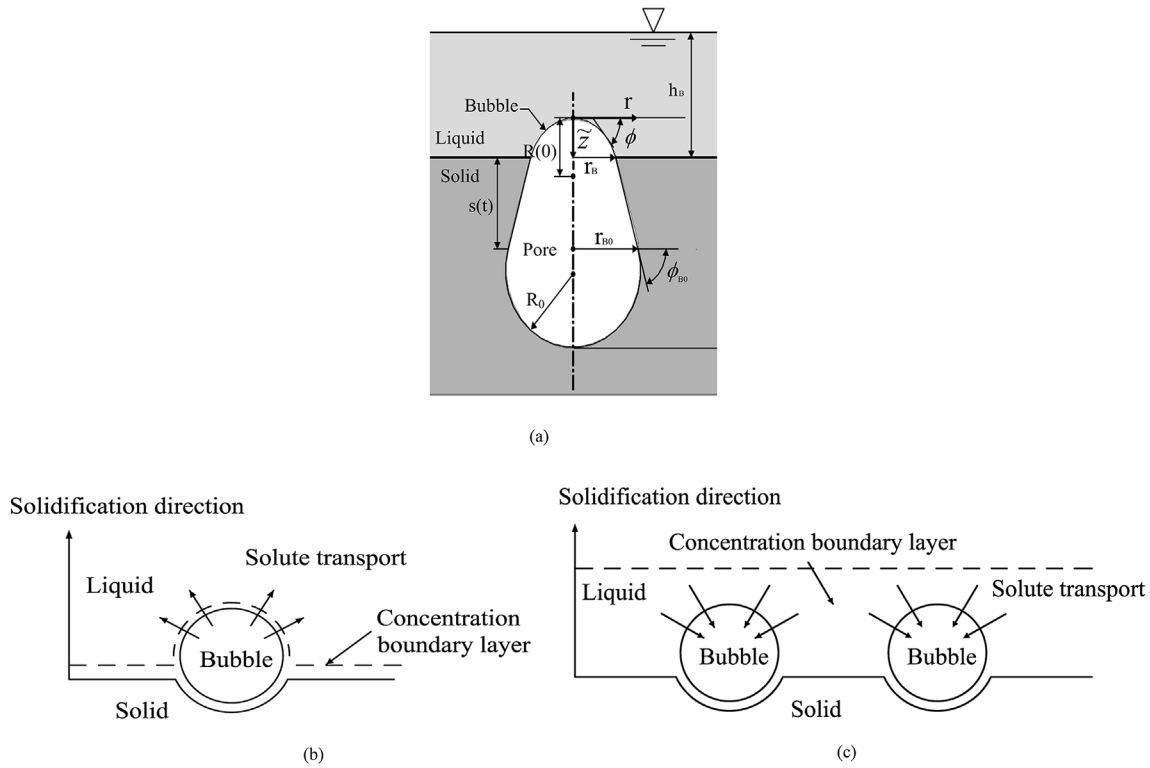

(a)

(b)

(c)

Fig. 1. Schematic sketch of (a) physical model, (a) outward solute transport across an emerged cap in a concentration boundary layer in Case 1, and (c) inward solute transport across a submerged cap in a concentration boundary layer in Case 2.

solute transfer across the bubble cap, rigorous understanding and controlling of the growth of the pore shape are obtained.

2. System model and analysis

A bubble entrapped as a pore in solid is illustrated in Fig. 1(a) [29–31]. From a geometrical viewpoint, the pore shape in solid is closely related to contact angle of the bubble cap on the solidification front [24]. The bubble cap can be determined by considering the pressure balance between gas, capillary and liquid pressures. Solute gas pressure in the pore is related to solute concentration in the liquid in physico-chemical equilibrium on the bubble cap. Solute concentration at the cap can be different for solute transport across the cap in different directions, as illustrated in Fig. 1(b) and (c). The former, Case 1, is referred to solutes transport from the pore across an emerged cap through a concentration boundary layer on the solidification front into surrounding liquid, whereas the latter, Case 2, is applicable to solute transport from the concentration boundary layer across a submerged cap into the pore in the early stage. For simplicity without loss of generality, the major assumptions made are the following:

1. The model system is axisymmetric. This is because the bubble is a spherical cap through which solute transport is of a symmetric distribution, as specified later. This model, however, can also be used to asymmetric and three-dimensional system, depending on specifications of solidification rate and mass transfer coefficient, as discussed later.
2. The tiny bubble is defined by a small Bond number, leading to apply available perturbation solutions to predict a lumped bubble in a spherical cap shape [32]. Bond number usually is much less than unity, based on a typical bubble radius of 10^{-5} m during solidification of water and liquid metals.
3. The system is static in unsteady state. Static fluid indicates that the flow of liquid can be ignored. Tangential and normal viscous stresses in the liquid are therefore neglected. Liquid pressure is hydrostatic

pressure, which is only a function of depth of liquid layer and gravitational acceleration. Unsteady state stands for processes to be time-dependent.

4. Solute transfer across the bubble cap, leading to an increase in moles of solute gas per unit time in the pore, is given by Ref. [26].

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

Since the bubble is usually initiated from heterogeneous nucleation due to solute super-saturation and solute concentration in the pore is higher than that in liquid, Eq. (1) indicates that solute transport is from the pore to surrounding liquid in the early stage. On the other hand, solute transport from surrounding liquid across the cap into pore can also be given by Refs. [23,27,28].

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

This is attributed to solute accumulation ahead of the advancing interface between solid and liquid. Cox et al. [27] and Lee et al. [28] 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.

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

$$\tilde{C}_c = \frac{\tilde{p}_g}{\tilde{K}} \tag{3}$$

which indicates solute gas pressure is proportional to solute concertation in the liquid at a liquid-gas interface.

6. Pore shape is delineated by tracing contact angle of the bubble cap beyond the solidification front [24]. Contact angle is a function of the shape of the bubble cap, determined by solute gas pressure in the pore and solute transport across the cap, as described later.