



Effects of solute concentration in liquid on pore shape in solid



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ABSTRACT

The effects of initial solute gas concentration in the liquid on the shape of a pore, resulting from a nucleated bubble entrapped by a solidification front, are predicted in this work. Solute concentration in liquid is responsible for solute transfer across the bubble cap, gas pressure in the pore, and shapes of the cap and pore in solid. Distributions and shapes of pores in solids influence not only microstructure of materials, but also contemporary issues of biology, engineering, foods, geophysics and climate change, etc. In this work, the relevant pore shape delineated by tracing contact angle of the cap to a first approximation is determined by accounting for mass and momentum transport across a self-consistent shape of the cap whose surface is satisfied by physico-chemical equilibrium, as proposed previously. This work finds that there exist three different mechanisms for pore formation, depending on directions and magnitude of solute transfer across the cap. Case 1 is subject to solute transport from the pore into surrounding liquid as a result of the cap emerged from a thin concentration boundary layer on the solidification front. An increase in initial solute concentration in liquid decreases pore radius and times for bubble entrapment. Opposite directions of solute transport across the cap submerged into a thick concentration boundary layer along the solidification front, however, cannot result in bubble entrapment, because solute concentration at the cap increases and decreases rapidly in late stage in Cases 2a and 2b, respectively. The predicted pore shape in solid agrees with experimental data. The pore shape therefore can be controlled by initial solute concentration to change directions and magnitudes of solute transport across the cap.

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1. Introduction

Understanding and controlling of pore formation are critical [1–12]. Porosity is susceptible to stress concentration, degrading the mechanical properties and impeding efficiency or functional properties of products [1,5–9]. On the other hand, substances containing a number of pores have characteristics, such as a low density and a large surface area. They can therefore be utilized as lightweight materials, catalysts, electrodes, vibration, acoustic energy absorption, and impact energy absorption materials, and scaffold for tissue ingrowth [10–12].

Pore formation resulting from a single bubble entrapped by a solidification front is determined by dissolved solute gas concentration in liquid. In view of gas solubility in solid much less than that in liquid, solute gas is accumulated ahead of the solidification front [13,14]. Super-saturation thus induces nucleation of bubbles on the solidification front [15–20]. Bubbles nucleated preferentially at the grain boundary grooves induced by constitutional supercooling on the solidification front [21,22]. The bubble grew

because solute gas transport across the bubble cap. An increase in solute gas concentration in the liquid reduced pore sizes [23–26]. Systematically speaking, enhanced mass transport to the cap from surrounding liquid and gas pressure in the bubble led to a decrease of cap radius in order to satisfy balances of pressures at the cap [25,26]. The pores were spherical for a low degree of saturation. As degree of saturation was high, an increase in solidification rate resulted in the pore shapes to change from columnar pores, short columnar pores to few pores [22].

Solute concentration and transport rates through the bubble cap can be determined by solving transport equation of concentration in details. Zhdanov et al. [27] numerically calculated a steady-state solute concentration field ahead of the hemispherical portion of a single cylindrical gas bubble entrapped in a growing crystal. Solute concentration increased near the solidification front whereas decreased near the axisymmetric axis. Solute concentration in the liquid increased and then decreased in the direction away from the bubble cap.

On the other hand, interactions between pores in metals can be interestingly revealed by long cylindrical pores in ordered and aligned in one direction and fabricated by the so-called “metal/gas eutectic reaction” during unidirectional solidification (also called “Gasar”) under a pressurized dissolvable hydrogen or nitrogen

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Nomenclature

A_c	cap surface area $A_c = \tilde{A}_c / \tilde{R}_0^2$, given by Eq. (2)	z	cylindrical coordinate, $z \equiv \tilde{z} / \tilde{R}_0$
Bo	Bond number, $Bo \equiv \rho g \tilde{R}_0^3 / \sigma$	z_B	cap height
C_∞	initial solute concentration, $C_\infty = \tilde{C}_\infty \tilde{R}_0 \tilde{T} / \rho g \tilde{R}_0$	Greek letter	
D	solute diffusivity	σ	surface tension
h_B	liquid depth, as illustrated in Fig. 2(a)	ϕ	inclination angle
h_D	mass transfer coefficient, $h_D = \tilde{h}_D \tilde{R}_0 / \tilde{D}$	Subscripts	
K	Henry's law constant, $K = \tilde{K} / \tilde{R}_0 \tilde{T}$	a	atmosphere
k_p	equilibrium partition coefficient	B	base
L	bubble cap radius at 90 degrees, given in Eq. (14)	c	bubble cap
n	mole of solute gas	g	gas
p	pressure, $p \equiv \tilde{p} \tilde{R}_0 / \sigma$	0	initial state
r	cylindrical coordinate, $r \equiv \tilde{r} / \tilde{R}_0$	∞	location front from solidification front
R	radius, $R \equiv \tilde{R} / \tilde{R}_0$	Superscripts	
$R(0)$	apex radius of cap	\sim	dimensional quantity
\tilde{R}_u	universal gas constant		
s	solidification front location		
t	time, $t = \tilde{t} \tilde{D} / \tilde{R}_0^2$		
U	solidification rate, $U = ds/dt$		
V	volume, $V = \tilde{V} / \tilde{R}_0^3$		

gases [11,17–19,28,29]. The porous structure is designated as lotus-type structure because it looks like lotus roots. It was found that the lotus-type porous metals exhibit superior mechanical properties [11]. Experimental work together with a further theoretical analysis including solute gas prediction required of lotus-type porous metals are as follow [29]: The pore diameter and interpore spacing decrease as partial pressure of hydrogen or argon increases. The porosity indicates the area ratio between the pore and its corresponding polygon. It showed that (1) the porosity decreases with increasing partial pressure of hydrogen when only hydrogen is used. (2) the porosity first increases and then decreases with increasing partial pressure of hydrogen when partial pressure of argon is kept constant, (3) the porosity decreases with increasing partial pressure of argon when partial pressure of hydrogen is kept constant; and (4) when the sum of hydrogen and argon pressures is constant, the porosity decreases with increasing partial pressure of argon.

The trends of porosity, as mentioned previously, can be relevantly confirmed by Liu et al. [17], providing analytical solute transport in the gas eutectic growth in a directional solidification subject to a flat top bubble. The dependence of porosity on partial pressures of argon and hydrogen arises, because the amount of hydrogen dissolved in the melt is proportional to the square root of hydrogen pressure, whereas density of hydrogen in the pore is proportional to the total applied pressure on the melt surface. A more complete steady state solution of concentration field around a self-consistent shape of the bubble cap in gas eutectic growth in a directional solidification was also provided by Li et al. [30] using multiple scale expansion and matching method for a small Peclet number, defined as the ratio between a half of inter-pore spacing and distance for solute diffusion, and surface tension parameter. The model accounted for Gibbs–Thomson condition at the solid–liquid interface, solute mass balances at the solid–liquid and gas–liquid interfaces, and the Young's law at the triple phase line. The computed results confirmed measured porosity as mentioned previously [29]. Morphologies of both gas–liquid and gas–solid interfaces were also presented. The height of the centers of the gas and solid phases increased as pressures of insoluble and dissolved gases beyond the liquid surface decreased.

Similar to evaluation of heat transfer rate by introducing heat transfer coefficient, solute transport across the bubble cap can be effectively evaluated by introducing a mass transfer coefficient.

Solute gas concentration in the pore can be influenced by either solute concentrations in the liquid ahead of the solidification or that away from the solidification front. Depending on which is dominant factor affecting pressure in the pore, directions and magnitudes of solute transfer rate can be different. Solute transfer is therefore from the pore into surrounding liquid in an early stage for a thin concentration boundary layer along the solidification front, as sketched in Fig. 1(a) [31]. On the other hand, for a thick thickness of solute concentration boundary layer along the solidification, solute transfer rate is determined by accumulated solute gas ahead of the solidification front and solute concentration far from the solidification front, as illustrated in Fig. 1(b) and (c) [28,32]. Directions of mass transfer is thus from the surrounding liquid into pore [25,26,32,33].

In this work, the effects of initial solute gas concentration in the liquid on the shape of a pore resulting from a bubble entrapped by a solidification front subject to different directions and magnitudes of solute transport across the cap is predicted. A realistic pore shape in solid, transient deformation of the bubble cap above the solidification front and solute gas concentration due to nucleation are included. This study provides a way to control pore shape via selecting initial solute concentration to change directions and magnitudes of solute transport across the cap.

2. System model and analysis

A pore resulting from entrapment of a bubble by a solidification front is illustrated in Fig. 2(a). It can be seen that the pore shape in solid to a first approximation is delineated by tracing the contact angle of the bubble cap [26,34,35]. The shape of the bubble cap beyond the solidification front is determined by the Young–Laplace equation, which governs normal stress balance between gas, capillary and liquid pressures. Solute gas pressure in the pore is determined by solute transport from the pore across the cap to surrounding liquid in outward and inward directions, as illustrated in Fig. 2(b) and (c), respectively, corresponding to a thin and thick concentration boundary layer on the solidification front, as discussed later. 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

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