



Effects of physico-chemical interfacial equilibrium on pore shape in solid



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ABSTRACT

The shape of a pore, resulting from a bubble entrapped by a solidification front, for different Henry's law constants at the cap is predicted in this work. Henry's law, indicating an interfacial physico-chemical equilibrium, is essentially required to relate solute concentration in liquid at the cap by solute gas pressure in the pore. Pore formation and its shape in solid influence contemporary issues of biology, engineering, foods, geophysics and climate change, etc. This work applies a previous model accounting for mass and momentum transport of solute across a self-consistently determined shape of the bubble cap subject to different cases characterized by different directions and magnitude of solute transport across the cap. Case 1 is referred to solute transport from the pore across cap to surrounding liquid in the early stage. Cases 2a and 2b, corresponding to low and high Henry's law constants, respectively, indicate opposite directions of species transport across the cap. An increase in Henry's law constant decreases pore radius and time for entrapment in Case 1. The pore cannot be entrapped as a pore in solid in Cases 2. The predicted pore shape in solid agrees with experimental data. Understanding, prediction and control of the growth of the pore shape have therefore been obtained.

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1. Introduction

Porosity in solids is susceptible to stress concentration, degrading the mechanical properties and impeding efficiency or functional properties of products [1–8]. However, substances containing a number of pores can also be controlled and utilized as lightweight materials, catalysts, electrodes, vibration, acoustic energy absorption, impact energy absorption materials, food preservation and sterilization, and the scaffold for tissue ingrowth [8–10]. Pore formation in lotus-type porous metals containing aligned long columnar pores have been used for functionally materials in biological and medical technologies [11–13].

It has been well-known that a pore results from a single bubble entrapped by a solidification front. The bubble occurs due to supersaturated solute gas accumulated in liquid ahead of the solidification front [14] until concentration was high enough for bubbles to nucleate [15,16]. During fast growth there was less time for diffusion and the bubble decreased in cross section. When freezing was slow, bubbles were bigger. The formation of ice worms therefore was suppressed and the ice contained a large number of very small, round pores [17–19].

On the other hand, porous metals can be long cylindrical pores in ordered and aligned in one direction and fabricated by metal/gas unidirectional solidification (also called “Gasar”) under a pressurized hydrogen and/or nitrogen [8,11–13]. This processing technique utilizes an invariant reaction of the so-called “metal/gas eutectic reaction” in which the melt is solidified into a solid solution and a gas phase. During solidification, the gas pore and the solid metal grow in couples and finally form a regular porous structure in which the long gas pores are aligned parallel to the solidification direction. The formed 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 [8]. 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 the increasing of partial pressure of hydrogen when only hydrogen is used, (2) the porosity first increases and then decreases with the increasing of partial pressure of hydrogen when partial pressure of argon is kept constant, (3) the porosity decreases with the increasing of 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 the increasing of partial pressure of argon [20,21].

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Nomenclature

A_c	cap surface area $A_c = \tilde{A}_c / \tilde{R}_0^2 \approx 2\pi R^2(0)(1 - \cos \phi_B)$
Bo	Bond number, $Bo \equiv \rho g \tilde{R}_0^2 / \sigma$
C_∞	initial solute concentration, $C_\infty = \tilde{C}_\infty \tilde{R}_u \tilde{T} / \rho g \tilde{R}_0$
D	solute diffusivity
h_B	liquid depth, as illustrated in Fig. 1(a)
h_D	mass transfer coefficient, $h_D = h_{D0} \tilde{R}_0 / \tilde{D}$
K	Henry's law constant, $K = \tilde{K} / \tilde{R}_u \tilde{T}$
L	micro-bubble radius at $\pi/2$
p	pressure, $p \equiv \tilde{p} \tilde{R}_0 / \sigma$
r	cylindrical coordinate, $r \equiv \tilde{r} / \tilde{R}_0$
R	radius, $R \equiv \tilde{R} / \tilde{R}_0$
$R(0)$	radius of curvature at micro-bubble apex
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, $V = \tilde{V} / \tilde{R}_0^3$
z	cylindrical coordinate, $z \equiv \tilde{z} / \tilde{R}_0$

Greek letter

σ	surface tension
ρ	density
ϕ	inclination angle

Subscripts

a	atmosphere
c	cap
B	base
g	gas
0	initial state

Superscripts

\sim	dimensional quantity
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Solute gas transport across the bubble cap and the free surface of the liquid require application of the Henry's law or Sievert's law governing physico-chemical equilibrium [11–13,15,20–23]. In view of different solubilities of liquid and gas, solute concentration across the bubble cap and free surface of liquid is segregated. Similar to equilibrium partition coefficient required for solute segregation between liquid and solid during solidification, Henry's law is needed to describe a relationship between gas pressure and solute gas concentration in equilibrium at a gas-liquid interface or the bubble cap. An introduction of Henry's constant can avoid complicated determination of solute concentration at the bubble cap, provided that gas pressure in the pore is found. Likewise, an introduction of equilibrium partition coefficient can avoid determination of solute concentration in solid if solute concentration in liquid at the solidification front is known.

A steady-state solute concentration field ahead of the hemispherical portion of a single cylindrical gas bubble entrapped in a growing crystal was numerically solved by Zhdanov et al. [24]. Since physico-chemical equilibrium at the bubble cap was not accounted for, time-dependent solute concentration and solute gas pressure in the pore were not presented. Time-dependent solute gas pressure is essentially required to determine growth of the bubble cap, resulting in the development of the pore shape in solid. It showed that solute concentration increased near the solidification front whereas decreased near the axisymmetric axis. Considering physico-chemical equilibrium at the free surface away from the solidification front, Liu et al. [12] provided analytical solute transport in terms of summation of different orders of Bessel's function in the gasar eutectic growth in a directional solidification subject to a flat top bubble. A more complete steady state solution of concentration field around a self-consistent shape of the bubble cap in gasar eutectic growth in a directional solidification subject to physico-chemical equilibrium at the free surface away from the solidification front was also provided by Li et al. [25] using multiple scale expansion and matching method for a small Peclet number.

Solute gas dissolved in the liquid is satisfied by a physico-chemical equilibrium governed by Sievert's law or Henry law at the free surface. On the other hand, solute gas is rejected into the liquid as the solid-liquid interface is advanced. When solidification occurs at the intersection of the solid-melt interface and the rim of the bubble cap, the solid again first rejects gas solute to the liquid side at the bubble surface. Since the melt there is slightly oversat-

urated with solute, for the pressure that is fixed by the environment, the gas is quickly released from the melt into the bubble in compliance with Henry's law. Therefore, the melt upon solidification at the bubble surface effectively releases the gas solute into the bubble instead of to the melt side, thus alleviating solute build-up. 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 [22,23].

In this work, the shape of a pore resulting from a bubble entrapped by a solidification front for different Henry's law constants is predicted. The pore shape is dominated by solute transport due to difference in solute concentrations far from the solidification front and at the bubble cap [26]. The effects of Henry's law coefficient responsible for solute concentration at the bubble cap and solute gas pressure in the pore on the pore shape are therefore important. 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. Rigorous understanding of the effects of physico-chemical equilibrium on the growth of the pore shape are obtained.

2. System model and analysis

A pore in solid resulting from an entrapped bubble by a solidification front is illustrated in Fig. 1(a). A realistic shape of the bubble cap is determined by the Young-Laplace equation governing pressure balance between gas, capillary and liquid pressures. Solute gas pressure in the pore is determined by 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 in the early stage, whereas the latter, Case 2, is applicable to solute transport from the concentration boundary layer across a submerged cap into the pore. The major assumptions made are the following:

1. The model system is axisymmetric.
2. The tiny bubble is defined by a small Bond number, leading to application of available perturbation solutions to predict a lumped bubble in a spherical cap-shape [27]. Bond number is usually much smaller than unity for a typical bubble radius smaller than 10^{-4} m.

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