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BUBBLE DEPARTURE RADII AT SOLIDIFICATION INTERFACES

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Abstract – A model has been developed for the prediction of bubble departure radii from flat solidification interfaces, including the effect of thermocapillary forces. Under normal gravity conditions, the necessary gas bubble radius required for departure from a CBr_4 solidification interface is predicted to be approximately 1/2 mm in agreement with measured values. Under microgravity conditions, however, where surface forces predominate, the model predicts a seemingly prohibitive value of 40 mm. This result is at least in agreement with the microgravity tests conducted on the NASA SPAR I and SPAR III sounding rockets [1] where the bubbles were not larger than 2 mm in radius and no bubble detachment was observed.

NOMENCLATURE

F, force [N];
$f(\beta)$, geometric function defined by equation (22);
g, gravitational acceleration [m s^{-2}];
$\hat{k}$, unit vector in z-direction [m];
K_e, constant defined by equation (16);
M, molecular weight;
P, pressure [Pa];
R, radius [m];
T, temperature [K];
t, time [s];
V, volume [m^3];
v, velocity [m s^{-1}];
z, coordinate direction [m].

Greek symbols

β, contact angle, defined in Fig. 1;
θ, coordinate angle, defined in Fig. 2;
ρ, density [kg m^{-3}];
σ, surface tension [N m^{-1}];
ϕ, coordinate angle, defined in Fig. 1.

Subscripts

dep, departure;
G, gas;
g, gravity;
L, liquid;
n, normal;
s, surface;
t, tangential;
z, coordinate direction.

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INTRODUCTION

THE PRESENCE and behavior of bubbles in a melt is of great concern in many areas of materials processing. In such operations as metals casting, glass founding and crystal growth, the presence of bubbles is undesirable; their size and distribution negatively affecting properties and appearance.

Bubbles are normally removed from a melt by buoyancy forces which cause them to rise to the surface. If they originate at the solidification interface, sufficiently large removal forces must be available to prevent incorporation into the solid.

Under microgravity conditions (such as encountered in the NASA space processing program) the environment is unique in that surface, rather than gravity forces, predominate. Thus the major removal force (buoyancy) is drastically reduced while the major attachment force (surface tension) remains largely unaffected. To compensate for the reduction in magnitude of the buoyant force, it may be possible to utilize thermocapillary convection as a removal mechanism. By imposing a temperature gradient on the gas–liquid interface, the surface tension is caused to vary and surface flows occur in the direction of increasing tension. The reaction force in turn causes the gas bubble to move in the opposite direction, i.e. in the direction of increasing temperature.

The objective of this analysis is to obtain an estimate of the gas bubble radius required for departure from a solidification interface under both normal and microgravity conditions in the presence of thermocapillary forces.

ANALYSIS

The model is shown in Fig. 1. It is assumed that the

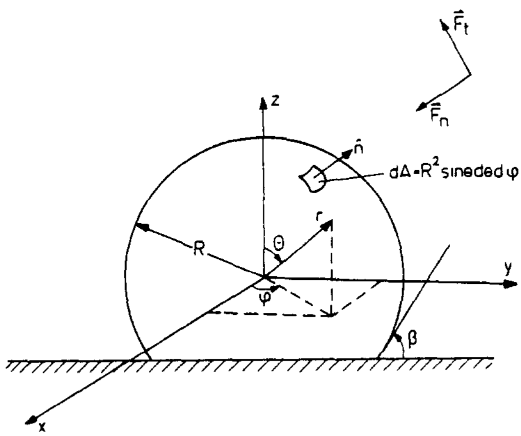

FIG. 1. Model of bubble at solidification interface.

solidification surface is planar and that the gas bubble is a portion of a sphere. All symbols are defined in the nomenclature.

The equation describing the upward motion of the bubble is

$$\Sigma F_z = \frac{d}{dt}(\rho_G V_G v + 1/2\, \rho_L V_G v), \tag{1}$$

where the added mass term $1/2\ \rho_L V_G v$ represents the momentum of liquid accelerated with the gas bubble. The forces F_z acting on the bubble include the normal and tangential forces (F_{n_z} and F_{t_z}) exerted at the gas–liquid, gas–solid interface; the surface tension force (F_{s_z}) acting at the triple interface; and the gravitational force (F_{g_z}) acting on the mass of the gas bubble.

Departure criteria

Bubble departure will occur when the bubble is of such size that the detachment forces just become equal to the adherent forces. At this condition the bubble acceleration is zero and equation (1) reduces to

$$\Sigma F_z|_{dep.} = 0. \tag{2}$$

Normal force

The magnitude of the normal force acting at a differential area of the gas–liquid interface is

$$|\mathbf{F}_n| = \left(P_0 - \rho_L g R \cos\theta + \frac{2\sigma}{R}\right) R^2 \sin\theta\, d\theta\, d\phi, \tag{3}$$

where P_0 is the static pressure in the liquid at the level $z = 0$, $\rho_L g R \cos\theta$ is the hydrostatic head relative to the level $z = 0$, and $2\sigma/R$ is the normal component of the surface stress.

The component of the normal force in the upwards direction is

$$F_{n_z} = \hat{k}\cdot\mathbf{F}_n = -|\mathbf{F}_n|\cos\theta. \tag{4}$$

Thus over the entire gas–liquid interface

$$F_{n_z} = -\int_0^{2\pi}\int_0^{\pi-\beta} \times\left(P_0 - \rho_L g R\cos\theta + \frac{2\sigma}{R}\right) R^2 \sin\theta\cos\theta\, d\theta\, d\phi. \tag{5}$$

For a sphere completely surrounded by liquid, the integral involving P_0 vanishes identically. Further, for uniform temperature, the integral involving $2\sigma/R$ also vanishes. The remaining integral yields the buoyant force of the liquid on the bubble. In this problem however, the bubble is only partially surrounded by liquid and the temperature is to be assumed non-uniform. As a result neither of the above mentioned terms will vanish upon integrating.

The remaining portion of the bubble surface to be considered is that in contact with the solid. Here it is assumed that the solidification surface exerts an upward force on the bubble, equivalent to the force exerted downward at the surface by the gas, i.e.

$$F_{n_z} = P_G|_s \pi R^2 \sin^2\beta, \tag{6}$$

where

$$P_G|_s = P_L|_s + \frac{2\sigma_s}{R} = P_0 - \rho_L g R\cos\theta|_{\theta=\pi-\beta} + \frac{2\sigma_s}{R}$$
$$= P_0 + \rho_L g R\cos\beta + \frac{2\sigma_s}{R}, \tag{7}$$

so that

$$F_{n_z} = \left(P_0 + \rho_L g R\cos\beta + \frac{2\sigma_s}{R}\right)\pi R^2 \sin^2\beta \tag{8}$$

and the total z-component of the normal force exerted on the gas bubble is

$$F_{n_z} = \left(P_0 + \rho_L g R\cos\beta + \frac{2\sigma_s}{R}\right)\pi R^2 \sin^2\beta$$

$$- \int_0^{2\pi} \int_0^{\pi-\beta} \left(P_0 - \rho_L g R \cos\theta + \frac{2\sigma}{R} \right) \times R^2 \sin\theta \cos\theta \, d\theta \, d\phi. \quad (9)$$

Upon integration of equation (9), P_0 will vanish as expected; the terms involving $\rho_L g R$ will yield the buoyant force, and the terms involving the surface tension will yield an additional force acting in the direction of increasing temperature. Here it is assumed that the imposed temperature gradient is normal to the planar solidification interface.

Tangential force

The magnitude of the tangential force acting at a differential area of the gas–liquid interface is

$$|\mathbf{F}_t| = \frac{1}{R} \frac{d\sigma}{d\theta} R^2 \sin\theta \, d\theta \, d\phi, \quad (10)$$

where $(1/R)(d\sigma/d\theta)$ is the tangential component of the surface stress. In this analysis, the interface is assumed to be free of impurities so that the 'friction drag' is zero. The force $|\mathbf{F}_t|$ given by equation (10) causes a flow of liquid at the interface in the direction of increasing tension. Because there is no shear or resisting force to this interfacial flow, a reaction force is exerted on the bubble causing it to move in the opposite direction. The z-component of this force is given by

$$F_{t_z} = \hat{k} \cdot \mathbf{F}_t = |\mathbf{F}_t| \sin\theta, \quad (11)$$

so that for the entire surface

$$F_{t_z} = \int_0^{2\pi} \int_0^{\pi-\beta} \frac{1}{R} \frac{d\sigma}{d\theta} R^2 \sin^2\theta \, d\theta \, d\phi. \quad (12)$$

Surface force

The z-component of the surface force at the triple interface includes only the liquid–gas surface tension, and as illustrated in Fig. 2 is given by

$$F_{s_z} = -2\pi R \sigma_s \sin^2\beta, \quad (13)$$

where σ_s is the surface tension evaluated at the temperature of the solidifying interface.

Gravity force

A downward directed force is exerted on the bubble

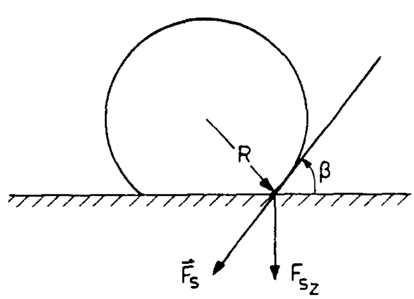

FIG. 2. Surface force at triple interface.

mass by the existing gravitational field and is given by

$$F_{g_z} = -\rho_G g V_G = -\tfrac{1}{3}\pi R^3 \rho_G g (2 + 3\cos\beta - \cos^3\beta). \quad (14)$$

Temperature field

A linear temperature profile is assumed to exist in the liquid phase, increasing in the positive z-direction from the known temperature T_s of the solidifying interface and having a prescribed temperature gradient dT/dz. Thus

$$T = T_s + R(\cos\beta + \cos\theta)\frac{dT}{dz}. \quad (15)$$

The surface tension is assumed to be a linear function of temperature having a known value of σ_s at temperature T_s and a temperature coefficient given by the Eötvös equation

$$\frac{d\sigma}{dT} = -K_e \left(\frac{\rho}{M}\right)^{2/3} = \text{constant}. \quad (16)$$

Hence

$$\sigma = \sigma_s + (T - T_s)\frac{d\sigma}{dT}. \quad (17)$$

Substituting for T from equation (15)

$$\sigma = \sigma_s + R(\cos\beta + \cos\theta)\frac{dT}{dz}\frac{d\sigma}{dT}. \quad (18)$$

With equation (18), the z-components of the normal and tangential forces may be integrated [equations (9) and (12)] to yield

$$F_{n_z} = \tfrac{1}{3}\pi R^2 \left(\rho_L g R - 2\frac{dT}{dz}\frac{d\sigma}{dT} \right)(2 + 3\cos\beta - \cos^3\beta) \quad (19)$$

and

$$F_{t_z} = -\tfrac{2}{3}\pi R^2 (2 + 3\cos\beta - \cos^3\beta)\frac{dT}{dz}\frac{d\sigma}{dT}. \quad (20)$$

It should be noted that σ decreases with increasing temperature so that $d\sigma/dT$ is numerically negative. Thus the effect of surface tension variation in both the normal and tangential terms is to provide a force tending to remove the bubble from the surface. In equation (19), the mechanism is analogous to the variation of hydrostatic head around the bubble surface producing a buoyant force; i.e. the variation in 'thermocapillary head' also produces a buoyant force. The force given by equation (20), as explained earlier, is a reaction force resulting from a surface tension driven surface flow.

Departure radius

Substituting equations (13), (14), (19) and (20) into equation (2), there results a quadratic equation for the departure radius whose solution is

$$R_{dep} = \frac{2\frac{dT}{dz}\frac{d\sigma}{dT} + \left[4\left(\frac{dT}{dz}\frac{d\sigma}{dT}\right)^2 + 6\sigma_s g(\rho_L - \rho_G)f(\beta)\right]^{1/2}}{g(\rho_L - \rho_G)}, \quad (21)$$

where

$$f(\beta) = \frac{\sin^2\beta}{2 + 3\cos\beta - \cos^3\beta}. \quad (22)$$

Under microgravity conditions, substitution into equation (2) yields a linear equation for the departure radius whose solution is

$$R_{dep}|_{g=0} = -\frac{3\sigma_s f(\beta)}{2\frac{dT}{dz}\frac{d\sigma}{dT}}. \quad (23)$$

DISCUSSION

The interaction of bubbles with solidification interfaces has been studied experimentally by Papazian and Wilcox under both normal and microgravity conditions [1]. Carbon tetrabromide, previously saturated with nitrogen was directionally solidified using a gradient freeze technique; the gases being rejected at the solidification interface. In the normal gravity tests, bubbles were observed to detach from the interface and rise to the top of the melt, yielding a specimen generally free of large voids. Bubble radii of the order of 0.5 mm at detachment, were measured [2]. In the microgravity tests conducted on the NASA SPAR I and SPAR III sounding rockets, no bubble detachment was observed and the resulting specimens contained large voids.

A test of the equations developed in the analysis section is to first compare the departure radius predicted by equation (21) with the normal gravity value of 0.5 mm, using parameters equivalent to those in the actual experiments. These were

$$\frac{dT}{dz} = 500\,\mathrm{K\,m^{-1}}, \quad T_s = 363\,\mathrm{K},$$
$$g = 9.8\,\mathrm{m\,s^{-2}}, \quad \beta = \frac{\pi}{6}\,\mathrm{rad}. \quad (24)$$

It was not possible to measure the actual contact angle (β), however the chosen value seems reasonable as a melt would be expected to wet its own solid phase. Perfect wetting would yield β equal to 0 and absolute non-wetting would yield $\beta = \pi$ rad. Pertinent physical property data for carbon tetrabromide are listed below.

1. *Surface tension* [3]

$$\sigma = 62.2 - 0.109\,T,\ \mathrm{mN\,m^{-1}}, \quad (25)$$
T in K.

2. *Temperature coefficient of surface tension*
By differentiating equation (25)

$$\frac{d\sigma}{dT} = -0.109\,\mathrm{mN\,m^{-1}\,K^{-1}}.$$

3. *Liquid density* [3]

$$\rho_L = 3828 - 2.173\,T,\ \mathrm{kg\,m^{-3}}, \quad (26)$$
T in K.

The density of nitrogen gas at 373 K and 0.1016 MPa as calculated from the ideal gas law is 0.94 kg m^{-3}. Substituting into equation (21)

$$R_{dep} = \frac{-0.019\,\mathrm{kg\,m^{-1}\,s^{-2}} + [0.0119 + 255]^{1/2}\,\mathrm{kg\,m^{-1}\,s^{-2}}}{29800\,\mathrm{kg\,m^{-2}\,s^{-2}}},$$

$$R_{dep} = 0.00053\,\mathrm{m} = 0.53\,\mathrm{mm}.$$

The departure radius is tabulated in Table 1 and plotted in Fig. 3 as a function of contact angle. It is seen that any moderately non-wetting contact angle (e.g. $\pi/9$ to $2\pi/9$ rad) is in reasonable agreement with the experimental value, however the choice of $\pi/6$ radians seems to be a reasonable one.

Under microgravity conditions the departure radius is given by equation (23), which for the parameters of (24) becomes

$$R_{dep}|_{g=0} = -\frac{3 \times 22.6\,\mathrm{mN\,m^{-1}} \times 0.0633}{2 \times 500\,\mathrm{K\,m^{-1}} \times (-0.109)\,\mathrm{mN\,m^{-1}\,K^{-1}}},$$

$$R_{dep}|_{g=0} = 0.0394\,\mathrm{m} = 39.4\,\mathrm{mm}.$$

The microgravity departure radius is tabulated in Table 1 and plotted in Fig. 4 as a function of contact angle. If the contact angle of $\pi/6$ rad is a reasonable one, gas bubbles formed at a solidification interface (and in contact with that interface) would have to be much larger than experimentally observed ($R < 2$ mm [1]) in order to depart from the surface.

Increasing the temperature gradient is a possible means for reducing the departure radius, however a reduction to approximately 2 mm would require increasing the temperature gradient to 10000 K m^{-1} (100 C cm^{-1}), a value which seems prohibitively large.

It appears than, that the only reasonable means for causing bubble departure under microgravity conditions where the bubble is in actual contact with the surface, (other than introducing artificial gravity fields), is to reduce the contact angle. This is done primarily by minimizing impurities in the system.

Table 1. Departure radius as a function of contact angle

Contact angle (β) (rad)	$f(\beta)$ [equation (22)]	Departure radius at normal gravity [equation (21)] (mm)	Departure radius at zero gravity [equation (23)] (mm)
$\pi/2$	0.5000	1.504	311
$4\pi/9$	0.3855	1.320	240
$7\pi/18$	0.2957	1.156	184
$\pi/3$	0.2222	1.001	138
$5\pi/18$	0.1602	0.850	99.6
$2\pi/9$	0.1074	0.694	66.8
$\pi/6$	0.0633	0.533	39.4
$\pi/9$	0.0293	0.361	18.2
$\pi/18$	0.00754	0.182	4.7
$\pi/36$	0.00190	0.089	1.2
0	0	0	0

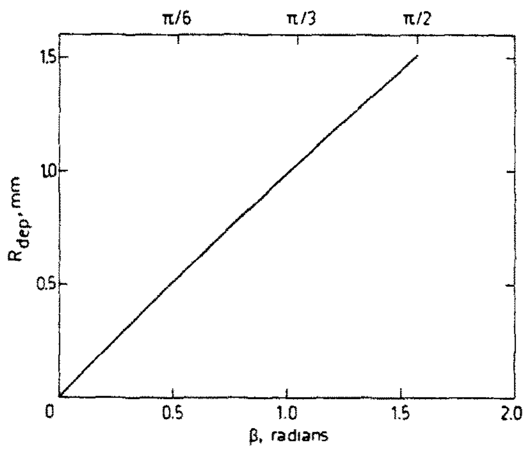

FIG. 3. Normal gravity departure radius as a function of contact angle.

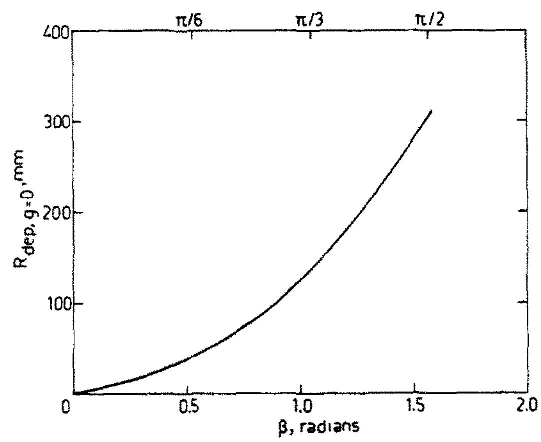

FIG. 4. Zero gravity departure radius as a function of contact angle.

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RAYON DE BULLE SE DETACHANT D'INTERFACES DE SOLIDIFICATION

Résumé – Un modèle est élaboré pour la détermination du rayon d'une bulle se détachant d'un interface plan de solidification, en tenant compte de l'effet des forces capillaires. Sous les conditions ordinaires de pesanteur, le rayon d'une bulle de gaz correspondant au détachement d'interface de solidification CBr_4 est évalué à environ 0,5 mm, en accord avec les mesures. Dans les conditions de pesanteur faible, lorsque les forces capillaires prédominent, le modèle prédit la valeur apparemment prohibitive de 40 mm. Ce résultat serait en accord avec les essais en microgravité conduits sur NASA SPAR I et SPAR III[1], pour lesquels les bulles avaient un rayon n'excédant pas 2 mm et pour lesquels aucun détachement n'a été observé.

BLASENABLÖSERADIEN AN ERSTARRUNGS-GRENZFLÄCHEN

Zusammenfassung – Es wurde ein Modell zur Berechnung der Blasenablöseradien an einer ebenen Grenzfläche beim Erstarren entwickelt, das den Einfluß von thermischen Kapillarkräften berücksichtigt. Unter dem normalen Einfluß der Schwerkraft ergibt sich der zur Ablösung erforderliche Blasenradius an einer Erstarrungsgrenzfläche aus CBr_4 in Übereinstimmung mit Meßwerten zu ungefähr 0,5 mm. Unter Bedingungen sehr geringer Schwerkraft jedoch, wenn die Oberflächenkräfte vorherrschen, liefert das Modell anscheinend einen sich verbietenden Wert von 40 mm. Immerhin ist dieses Ergebnis in Übereinstimmung mit den Versuchen bei Schwerelosigkeit, die mit den NASA-Raketen SPAR I und SPAR III durchgeführt wurden, bei denen der Blasenradius zwar nicht größer als 2 mm war, aber auch keine Blasenablösung beobachtet wurde.

РАДИУСЫ ОТРЫВА ПУЗЫРЬКОВ НА ПОВЕРХНОСТЯХ ЗАТВЕРДЕВАНИЯ

Аннотация — Разработана модель для расчёта радиусов отрыва пузырьков на плоских поверхностях затвердевания при учёте термокапиллярных сил. В результате расчёта показано, что при нормальной гравитации радиус газового пузырька, необходимый для отрыва от затвердевающей поверхности из CBr_4, составляет примерно 1/2 мм, что совпадает с измеряемыми значениями. Однако в условиях слабой гравитации, когда преобладают поверхностные силы, модель даёт нереальное значение радиуса в 40 мм. Этот результат согласуется с результатами опытов по слабой гравитации, проведенных Национальным управлением по аэронавтике и исследованию космического пространства с помощью ракет с телеметрической аппаратурой СПАР I и СПАР III. В этих опытах радиус пузырьков не превышал 2 мм, а отрыв пузырьков не наблюдался.