Shape and stability of a floating liquid zone between two solids

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The shape and stability of a floating liquid zone between two solids are analytically investigated using the principle of variation. The formulas for its shape and stability are obtained. © *1997 American Institute of Physics.* [S0021-8979(97)07324-6]

The stable surface shape of liquid such as a hanging drop has been investigated by Freund and Harkins.[1] They derived a special form applicable to the hanging drop from the familiar equation of Laplace. Through the numerical solution of it, they obtained a family of curves which express the surface shapes of the drops. Moreover, some investigations have been made into the shapes of floating liquid zones[2] between two solid rods[3,4] since the crystal growth needs the control of the geometrical configuration of the liquid zone. Keck *et al.*,[3] and Heywang and Ziegler[4] determined the surface shapes of the floating liquid zones from the curves of the surface shape of the hanging drop. According to their views, the curves of Freund and Harkins can be applied to the case of the floating liquid zone because the hanging drop and the floating liquid zone are both satisfied by the same Laplace equation. However, it is noted that the boundary conditions for the floating liquid zone are obviously different from those for the hanging drop. Hence, it is incorrect in making direct use of the curves, except for the hanging drop. In addition, Keck *et al.*[3] assumed the values of the tangent of the liquid surface at the solid–liquid interface, which are generally unknown. Using the floating zone method, we reported experimental results of bubbles in ruby single crystals[5] and recognized the importance of the control of the geometrical configuration of the liquid zone. The purpose of this article is to solve analytically the shape of the floating liquid zone between two solids and to display its stability diagram.

Figure 1 shows a schema of the floating liquid zone between two solids; here 2a and h indicate the width and height of the floating liquid zone. The problem in this study is to find the smallest free energy under a given area S. We have an interest in the case that the characteristic length of the system is larger than the capillary constant, $C=(2\gamma/\rho g)^{1/2}$ where γ is the surface tension, ρ the density of the liquid, and g the gravitational constant. The free energy, F, and the area, S, are given by

$$F=\int p dS+2\gamma\int dl, \tag{1}$$

$$S=2\int xdy, \tag{2}$$

where p is the pressure and dl the line element of the liquid surface. The integration range is given as $[0,h]$. It is convenient to modify Eqs. (1) and (2) to dimensionless forms. We shall let x, y and x' stand for the coordinates and derivative in the new unit of C, where $x'=dx/dy$. Equations (1) and (2) become

$$F/C^3\rho g=\int xydy+(1+x'^2)^{1/2}dy, \tag{3}$$

$$S/2C^2=\int xdy. \tag{4}$$

The procedure of the principle of variation leads

$$\partial f/\partial x-d(\partial f/\partial x')/dy=0, \tag{5}$$

$$f=xy+(1+x'^2)^{1/2}+\lambda x, \tag{6}$$

where λ is a constant. Integrating Eq. (5) over y, we have

$$x'=\pm(y^2/2+\lambda y+C_0)/[1-(y^2/2+\lambda y+C_0)^2]^{1/2}, \tag{7}$$

where C_0 is an integral constant. If Eq. (7) has some solution, Eq. (7) should satisfy the following condition:

$$|y^2/2+\lambda y+C_0|<1. \tag{8}$$

This equation gives the stability condition for the floating liquid zone. As seen in Fig. 1, taking the symmetry into

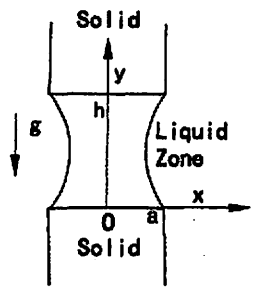

FIG. 1. Schema of floating liquid zone between two solids.

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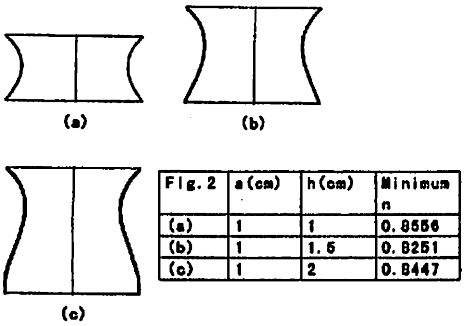

Fig. 2	a (cm)	h (cm)	Minimum n
(a)	1	1	0.8556
(b)	1	1.5	0.8251
(c)	1	2	0.8447

FIG. 2. Floating silicon shapes with three kinds of height when $n<1$.

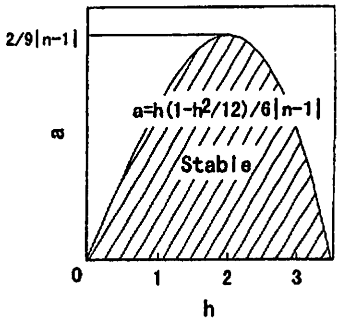

FIG. 4. Stability diagram for floating liquid zone.

consideration, we choose a plus sign of Eq. (7). Using Eq. (8) and integrating Eq. (7) over y, we have approximately

$$x \sim y^3/6+\lambda y^2/2+C_0 y+C_1. \tag{9}$$

The integral constants, C_0 and C_1, and λ are determined by the values of x at $y=0$ and $y=a$, and Eq. (4), for example, $C_0=h^2/12+6(n-1)a/h$, where n is a positive real number. Here the dimensionless area, S, is given as nah. Consequently, we have

$$x=y^3/6+\lambda y^2/2-h(y/2+h/6)y+a, \tag{10}$$

where $\lambda=-h/2+12a(1-n)/h$.

Figure 2 shows the surface shapes of the floating silicon zone in the case of $n<1$. We use 0.79[cm]^3 as the capillary constant of silicon. The narrow necks in Fig. 2 are seen when the floating zone is first formed between two solids in the floating zone technique.[2] If the amount of liquid is insufficient to support the liquid zone, the liquid zone will break apart. Figure 3 shows the surface shapes of the floating silicon zone in the case of $n \geq 1$. Though Keck *et al.*[3] set the tangents of the liquid surface to zero at the liquid–solid interface, these figures indicate that x' at the liquid–solid interface is always unequal to zero.

Here we consider the upward movement of the liquid zone. If one wants to grow solids with the larger width, one should bulge out the liquid zone near the lower solid. If the volume of the bulge is beyond the limit expressed by Eq. (8), the liquid zone cannot support itself and will drop.

We can derive the stability condition for the floating liquid from Eq. (8). Letting $g(y)=y^2/2+\lambda y+C_0$, in the case of $n \geq 1$, we obtain

$$C_0>0,$$

$$g(-\lambda)>-C_0 \quad \text{if } 12a(n-1)/h<h/2,$$

$$g(0)>g(h) \quad \text{if } 12a(n-1)/h \geq h/2. \tag{11}$$

Hence when $n \geq 1$, the necessary condition for Eq. (8) is $C_0<1$. This leads

$$h^2/12+6a(n-1)/h<1. \tag{12}$$

In a similar way, in the case of $n<1$, we can obtain

$$h^2/12-6a(n-1)/h<1. \tag{13}$$

Therefore, the stability condition for the floating liquid surface is given by

$$a<h(1-h^2/12)/6|n-1|. \tag{14}$$

Figure 4 shows the stability diagram for the floating liquid zone when $n \neq 1$. In the case of $n=1$, the stable condition is given as $h^2<12$. Equation (14) can be rewritten in the dimension form

$$h^2/12C^2+6a|n-1|/h<1. \tag{15}$$

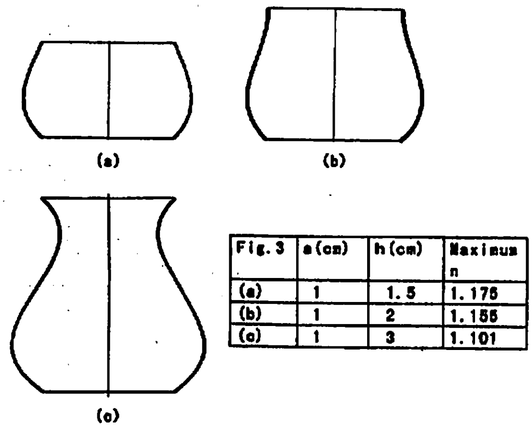

Fig. 3	a (cm)	h (cm)	Maximum n
(a)	1	1.5	1.176
(b)	1	2	1.156
(c)	1	3	1.101

FIG. 3. Floating silicon shapes with three kinds of height when $n \geq 1$.

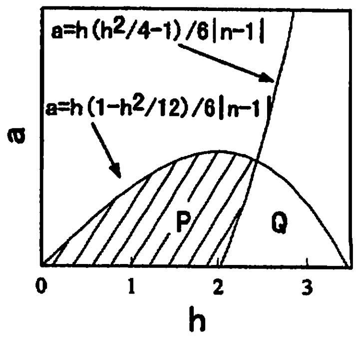

FIG. 5. Stable condition for quasi-static crystal growth. P: decrease in the width of grown crystal, Q: increase in the width of grown crystal.

Equation (15) represents the obvious conclusion that the long liquid zone holds stable if the capillary constant of the liquid has a large value.

Here we consider the quasi-static growth of the lower solid when the liquid zone moves upward. In the process, part of the upper solid melts and the liquid zone crystallizes on the lower solid side. This method for growth of single crystals is known as the floating zone technique.[2] First, consider the condition for the growth of crystals with a fixed width. We assume that the shape of crystals is dependent on the angle of the liquid at the lower liquid–solid interface. Letting x' at $y=0$ be equal to zero, we have

$$h^3/a=72(1-n) \quad \text{provided that } n<1. \qquad (16)$$

This is the condition for crystal growth with a fixed width. Second, in the case of increase in the width of the grown crystals, we can lead a condition for the stable surface shape added to Eq. (14). Using Eq. (14), we have

$$a\leqslant h(1-h^2/4)/6(n-1) \quad \text{provided that } n>1. \qquad (17)$$

On the other hand, in the case of a decrease in the width of grown crystals, in a similar way, we obtain

$$a>h(h^2/4-1)/6(1-n), \quad \text{provided that } n<1. \qquad (18)$$

Figure 5 shows the stable surface condition for the quasi-static growth. The area of the letter P or Q corresponds to Eqs. (14) and (18), or to Eqs. (14) and (17).

Thus, we obtain the formulas for the shape and stability of the floating liquid zone between two solids.

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