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Liquid layer deformation under horizontal thermal gradient

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Résumé. — Nous étudions une couche liquide à surface libre en convection sous un gradient thermique appliqué horizontalement. Capillarité et gravité se conjuguent pour donner une légère inclinaison de la surface, sous un angle qui dépend de l'épaisseur de la couche. Ce résultat peut présenter un certain intérêt en solidification Marangoni. Surtout, il débouche sur le principe d'une mesure précise du coefficient de température de la tension de surface.

Abstract. — We consider a free-surface liquid layer convecting under an applied horizontal temperature gradient. The surface gets slightly tilted by an angle which depends on layer thickness through competing capillary and gravity effects. This result might be of some interest in Marangoni solidification set ups. It also opens up a new way of accurately measuring the surface-tension temperature coefficient.

1. Free-surface tilting.

A free-surface liquid layer, subjected to a uniform horizontal temperature gradient $G = \mathrm{d}T/\mathrm{d}x$, sets into convection. The motion is controlled by two dimensionless quantities, the Grashof and Reynolds-Marangoni numbers :

$$\mathrm{Gr} = \frac{h^4 g \beta G}{\nu^2} \; ; \; \mathrm{Re_M} = -\frac{h^2 \sigma' G}{\rho \nu^2} \tag{1}$$

associated, respectively, with bulk thermal convection and surface, thermo-capillary convection. The notations are : h, layer thickness (along vertical direction z) ; g, gravity ; $\beta = -\mathrm{d}\rho/\rho \, \mathrm{d}T$, thermal expansion ; ρ, specific mass ; $\nu = \eta/\rho$, kinematic viscosity ; and $\sigma' = \mathrm{d}\sigma/\mathrm{d}T$, temperature coefficient of surface tension (usually negative). We shall assume that the temperature variation of β and σ' themselves, and of η, can be neglected. From the outset, we exclude from the present discussion very large values of Gr and Re_M for which structures with vorticity and non-negligible advection forces $\left(\sim \left(\underset{\sim}{v}\nabla\right)\underset{\sim}{v}\right)$, are known to appear [1]. In particular, we shall keep : $\mathrm{Gr} \ll (\mathrm{Gr})_c \simeq 10^4$. In practice, for liquids with small viscosities like liquid metals, this will restrict thickness h to the millimetric range unless one works in a low-gravity environment. h is a sensitive parameter as it enters Gr with the fourth power. We do not consider very small thicknesses either ($h \lesssim 0.2$ mm, say) in order to avoid film-adhesion problems.

The problem is considerably simplified, of course, for a *thin* layer (i.e. large aspect ratios in the x and y directions) : this is the case we shall be mainly interested in here. In that case, a one-dimensional solution, ignoring any deformation of the surface, has been given by Birikh [2]. The purely Marangoni velocity profile $v_x(z)$, for instance, is parabolic. This is an excellent approximation to the velocity, but does not completely describe the pressure field. The free surface must somehow relax the stresses it is subjected to, by deforming slightly. The corresponding contribution to the vertical pressure gradient is absent from Birikh's solution.

This correction is straightforward in the case of bulk thermal convection where gravity automatically provides for the vertical pressure field : $p = p_0 + \rho g(h - z)$, p_0 being the pressure at free surface. The result (see Ref. [3]) is a small thickness gradient, parallel to grad T :

$$h^2(x) = h_0^2\,(\rho_0/\rho)^{3/4} + 3\,(\sigma - \sigma_0)\,/\rho g \tag{2}$$

where slow variations in ρ and σ have been assumed, which is hardly restrictive as long as the applied temperature gradient G does not exceed a few tens of degree cm^{-1} The subscript denotes values at a reference point $x = x_0$.

In the gravity-dominated regime described by equation (2), $h(x)$ is seen to increase in the same direction as σ for small thicknesses, and in the same direction as ρ^{-1} for larger thicknesses. The origin of the driving force is mainly capillary : σ' (mainly barometric : $\rho g h^2 \beta$) in the first (second) case. Usually, both σ and ρ increase towards the cold end. So, both types of convection (thermocapillary and bulk-thermal) drive the upper part of the liquid from the hot to the cold site.

Our aim in this work is to provide a detailed discussion of free-surface deformation in both regimes where the restoring force is governed by gravity g (as in Ref. [3] and Eq. (2)), or surface tension σ. A well-defined criterion allows us to distinguish between the two. The latter regime corresponds to low-gravity conditions. Throughout, we neglect the weak, induced, transverse temperature gradient which decreases as h times Pr [2] (the Prandtl number Pr is much smaller than one in liquid metals).

In the next section, we describe a general formalism, based on non-dimensional analysis and covering the various regimes for driving and restoring forces, and various cell aspect-ratios. In section 3, coming back to the gravity-dominated regime, we stress that surface deformation basically reduces to a small, uniform tilting of the free surface — which opens up the principle of a novel method for measuring the surface-tension temperature coefficient σ' (Sect. 5). In section 4, we show that the capillarity-dominated regime also, is describable in simple physical terms, both for narrow cells (width ℓ much smaller than length L) and wide ones ($\ell \gg L$). In the latter case, the tilt-angle, in the middle of the cell, goes as $\dfrac{L^2}{h}\sigma'$.

2. Dimensionless-number formalism.

Free-surface shapes, in the weak-deformation limit, have been studied in the literature in terms of non-dimensional analysis (see for ex. Ref. [4]), e.g. in the zone-melting context. In this section, we

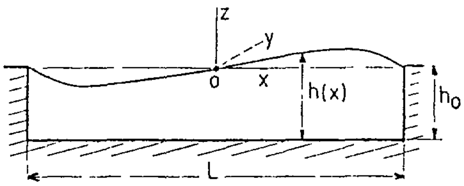

Fig. 1. — Geometrical characterization of free-surface deformation ; temperature gradient is directed along Ox. Cell width, along Oy, is ℓ.

adapt the corresponding formalism to our liquid-layer problem. In sections 3 and 4, we show that the physically interesting limiting cases where gravity or surface tension, respectively, dominate the restoring force, admit of simple direct solutions.

In terms of the geometry defined in figure 1, we introduce dimensionless lengths and velocities :

$$\begin{aligned} H &= \frac{h-h_0}{h_0},\ X = \frac{x}{L},\ Y = \frac{y}{\ell},\ Z = \frac{z}{h_0} \\ V_x &= \frac{v_x}{v_{0x}},\ V_z = \frac{v_z}{v_{0z}} \end{aligned} \tag{3}$$

where the subscript "zero" refers to the middle-point O. Defining three dimensionless numbers which refer velocity, gravity and surface tension, respectively, to viscosity :

$$\mathcal{R}\mathrm{e} = \frac{v_{0x}h_0}{\nu},\ \mathcal{G} = \frac{gh_0^3}{\nu^2},\ \mathcal{S} = \frac{\sigma h_0}{\rho\nu^2} \tag{4}$$

($\mathcal{R}\mathrm{e}$, for example, is the Reynolds number), and two aspect ratios :

$$A = \frac{h_0}{L},\ B = \frac{h_0}{\ell} \tag{5}$$

we get the dimensionless pressure field as :

$$P = \frac{h_0}{\eta v_{0x}}\left[p(x,y,z) - p_0 + \rho g z\right] = \frac{\mathcal{G}}{\mathcal{R}\mathrm{e}}Z + A\frac{\partial V_z}{\partial Z} - \frac{\mathcal{S}}{\mathcal{R}\mathrm{e}}\left[A^2\frac{\partial^2 H}{\partial X^2} + B^2\frac{\partial^2 H}{\partial Y^2}\right] \tag{6}$$

$p(x,y,z)$ obeying Navier-Stokes equation of course, with the associated boundary conditions (see, e.g., Eqs. (19) to (22), in Sect. 4) ; p_0 is the gas pressure at free surface.

Expanding, for a thin layer ($A, B \ll 1$), the pressure and velocity fields to first order in deformation, we recognize the viscosity term $A\frac{\partial V_z}{\partial Z}$ to be of order θ the local slope of the deformed surface. Thus, $A\frac{\partial V_z}{Z}$ is much smaller than the pressure P. To order zero, the latter reduces to Birikh's pressure. So, we find the local thickness $H(X)$ as a solution of :

$$P_0(X) = \frac{3}{2A\mathcal{R}\mathrm{e}}\left[\frac{1}{4}\mathrm{Gr} - \mathrm{Re_M}\right]X = \frac{\mathcal{G}}{\mathcal{R}\mathrm{e}}H - \frac{\mathcal{S}}{\mathcal{R}\mathrm{e}}\left[A^2\frac{\partial^2 H}{\partial X^2} + B\frac{\partial^2 H}{\partial Y^2}\right] \tag{7}$$

where $P_0(X)$ is Birikh's solution [2] for the pressure field at the surface. Let us, for example, consider very wide cells : $\ell \gg L$, i.e. $B \ll A$. The last equation gives :

$$H(X) = \frac{3}{2}\frac{\mathrm{Re_M} - \mathrm{Gr}/4}{\mathcal{G}A}\left[\frac{\mathrm{ch}\left[\frac{1}{2A}\left(\frac{\mathcal{G}}{\mathcal{S}}\right)^{1/2}\right]}{\mathrm{sh}\left[\frac{1}{A}\left(\frac{\mathcal{G}}{\mathcal{S}}\right)^{1/2}\right]}\mathrm{sh}\left[\frac{X}{A}\left(\frac{\mathcal{G}}{\mathcal{S}}\right)^{1/2}\right] - X\right] \tag{8}$$

When gravity dominates $(\mathcal{G} \gg \mathcal{S}A^2)$, this equation reduces to :

$$H(X) = \frac{3}{2}\frac{\mathrm{Re_M} - \mathrm{Gr}/4}{\mathcal{G}A}\left[\frac{1}{2}\left\{\mathrm{e}^{-(\mathcal{G}/\mathcal{S})^{1/2}A^{-1}\left(\frac{1}{2} - X\right)} - \mathrm{e}^{-(\mathcal{G}/\mathcal{S})^{1/2}A^{-1}\left(\frac{1}{2} + X\right)}\right\} - X\right] \tag{9}$$

The surface is uniformly tilted over most of cell length, and edge effects occur only over a characteristic reduced distance $\left(\frac{\mathcal{S}A^2}{\mathcal{G}}\right)^{1/2}$, proportional to the capillary length $(\sigma/\rho g)^{1/2}$ H reaches its maximum value near the end wall :

$$H_{\mathrm{Max}} \simeq \frac{3}{4}\frac{\mathrm{Re_M} - \mathrm{Gr}/4}{\mathcal{G}A} \tag{10}$$

When capillarity dominates $(\mathcal{S}A^2 \gg \mathcal{G})$, the surface profile obtains as an expansion of the hyperbolic-line functions in equation (8) :

$$H(X) = \left(4\mathcal{S}A^3\right)^{-1}(\mathrm{Re_M} - \mathrm{Gr}/4)\left(X^3 - \frac{X}{4}\right) \tag{11}$$

(with our notations, $X = \pm\frac{1}{2}$ at the cell ends). The deformation is now a cubic, instead of linear, function of distance X ; and the deformation amplitude is given by :

$$H_{\mathrm{Max}} = \left(4\mathcal{S}A^3\right)^{-1}(\mathrm{Gr}/4 - \mathrm{Re_M})\left(12\sqrt{3}\right)^{-1} \tag{12}$$

and occurs at $X = \left(2\sqrt{3}\right)^{-1}$

3. Free-surface tilting under gravific restoring force.

From the above analysis, we recover the limit where gravity provides for the restoring force. Equation (9) is equivalent to an expansion of equation (2) to first order in deformation :

$$\frac{\mathrm{d}\zeta}{\mathrm{d}x} = \theta = \frac{3}{2\rho g h}\left[\frac{\mathrm{d}\sigma}{\mathrm{d}x} - \frac{gh^2}{4}\frac{\mathrm{d}\rho}{\mathrm{d}x}\right] = \frac{3G}{2\rho g h}\left[\sigma' + \frac{1}{4}\rho g h^2 \beta\right] \tag{13}$$

where $\zeta(x)$ is local vertical coordinate of free surface ($h(x)$ in Sect. 2). The surface deformation essentially reduces to a small, uniform tilt with slope θ : the curvature is negligible. The above solution, of course, is valid provided the cell dimensions L, and ℓ, are much larger than the capillary length $(\sigma/\rho g)^{1/2}$ — a few millimeters usually.

The sign of tilt angle θ depends (for a given sign of $G = \nabla T$) on average thickness h (denoted h_0 in the previous section). In the usual case where thermal expansion β is positive, while surface-tension temperature coefficient σ' is negative, we can define a characteristic thickness h_c given by :

$$h_c^2 = -\frac{4\sigma'}{\rho g \beta} = 4\left(\frac{h^2 \mathrm{Re_M}}{\mathrm{Gr}}\right) \tag{14}$$

With ordinary values of parameters for liquid metals, h_c is about half a centimeter typically. The surface slope θ is positive (negative) for h smaller (larger) than h_c. h_c is the thickness for which bulk and surface convections conspire to restore horizontality of free surface. (Of course, for h_c to be properly defined by equation (14), our initial criterion : $\mathrm{Gr} < (\mathrm{Gr})_c$ must be fulfilled, that is : with $\beta = 10^{-4}\,\mathrm{K}^{-1}$ and $G = 10\,\mathrm{Kcm}^{-1}$, kinematic viscosity $\nu = \eta/\rho$, in $\mathrm{cm^2s^{-1}}$, must be larger than $\simeq \left(\frac{h(\mathrm{cm})}{10}\right)^2$). This suggests an original way of measuring σ', as we shall see in section 6.

On the other hand, the flow velocity on the free surface itself may be shown [2] to vanish for equal and opposite Grashof and Reynolds-Marangoni numbers. For a given imposed thermal gradient, the overall intensity of flow is then considerably reduced — which may have interesting applications, in directional solidification for instance. (It should be stressed, though, that the velocity profile $v_x(z)$ now displays an inflection point in the middle plane of the layer, which in the limit of low viscosity may result in non stationary instabilities (see e.g. Ref. [5]).) The corresponding characteristic thickness h_* is given by [2] :

$$h_*^2 = \frac{12\sigma'}{\rho g \beta} \tag{15}$$

A short development is in order here. The case $\beta > 0$ and $\sigma' > 0$ is more interesting in practice than the one considered in reference [2] (both β and σ' negative). It could be arrived at by doping with a surface-active agent under appropriate conditions of temperature and chemical potentials (e.g. bismuth in tin, see Refs. [6, 7]). Then $\sigma' \equiv \mathrm{d}\sigma/\mathrm{d}T$ has a maximum as a function of temperature. The point is that surface tension is density of surface free energy :

$$\sigma = f_s = e_s - T s_s \tag{16}$$

The high-temperature side is dominated by increase in entropy, so that σ' is negative (as in a pure liquid) — while the low-temperature side is dominated by increase in energy e_s (since more and more Bi atoms leave the surface as $T\nearrow$).

With ordinary values of parameters, h_* is of order 1 cm.

4. Free-surface tilt under Marangoni-induced pressure field.

This case corresponds to low-gravity conditions. The "barometric" pressure is now negligible compared to flow-induced pressure. The corresponding criterion writes simply :

$$\rho g a \ll \sigma / r \tag{17}$$

(or $\mathcal{G} \ll SA^2$, see Sect. 2), where a is deformation amplitude and $1/r$ is maximum characteristic surface curvature. The latter is taken to be modest in the sense that $r \gg L, \ell$, the cell length L and width ℓ themselves being on the scale of centimeters for example. Only in a low-gravity environment ($g \lesssim 10^{-2} g_0$, typically), will condition (17) be fulfilled.

Let us consider a shallow container which is wetted by the liquid, with an amount of liquid filling exactly the container : the free surface is pinned at the wall edges and tends to stay parallel to the bottom wall. The temperature gradient is directed along Ox (e.g. T decreasing from left to right, Fig. 2).

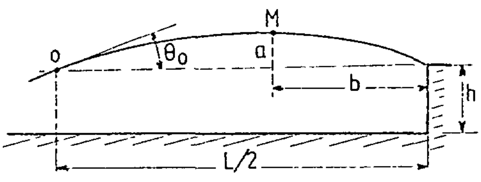

Fig. 2. — Schematics of free-surface deformation in a wide cell : $\ell \gg L$, when capillarity provides for the restoring force ; axis of symmetry : perpendicular to plane of drawing, through point O.

The Birikh solution [2] provides a valid approximation to the velocity profile $v_x(z)$ for both thermal (bulk) and thermocapillary convections. This is a one-dimensional solution however ($\theta = 0$) and, as such, it fails to describe properly the "internal" pressure field associated with the flow. To do so, the full 2d problem ($\theta \neq 0$ and $v_z \neq 0$) must be solved self-consistently, that is, with the constraint : $v_z = \theta v_x$ at free surface. This was not necessary, in section 1, with the "external" barometric pressure field $\rho g z$.

The Navier-Stokes equations now read :

$$\begin{aligned} \frac{\partial p}{\partial x} &= \eta \left[\frac{\partial^2 v_x}{\partial z^2} + \frac{\partial^2 v_x}{\partial x^2} \right] \\ \frac{\partial p}{\partial z} &= \eta \left[\frac{\partial^2 v_z}{\partial z^2} + \frac{\partial^2 v_z}{\partial x^2} \right] \end{aligned} \tag{18}$$

the velocity components being linked by the condition of incompressibility : $\frac{\partial v_x}{\partial x} + \frac{\partial v_z}{\partial z} = 0$.

The boundary conditions are : (i) no slip at wall, (ii) vanishing normal and tangential stress components at free surface, (iii) geometrical constraint on velocity at free surface :

$$z = 0 \ : \ v_x = v_z = 0 \tag{19}$$

$$z = \xi \ : \ p - p_0 = 2\eta \frac{\partial v_z}{\partial z} + \frac{\sigma}{r} \ ; \ \frac{\mathrm{d}\sigma}{\mathrm{d}x} = \eta \left(\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x} \right) \tag{20}$$

$$z = \xi \ : \ v_z = \theta v_x \tag{21}$$

($\zeta(x)$: z - coordinate of free surface at abscissa x ; $z = 0$ at bottom wall).

The (small) tilt angle $\theta = \frac{\partial \zeta}{\partial x}$ results from subjecting the velocity field v_x, at every abscissa x, to the return-flow condition :

$$\int_0^{\zeta} v_x(z) \mathrm{d}z = 0 \tag{22}$$

The general situation is relatively complex because the solution must involve an optimized trade-off, at every point on the free surface, between the *two* radii of curvature r_L and r_ℓ respectively along length $L(/\!/ \ \mathrm{O}x)$ and width $\ell(/\!/ \ \mathrm{O}y)$ of cell.

Let us first assume the cell to be very wide : $\ell \gg L$. Then the curvature is essentially longitudinal (i.e. along L) $r_\ell \gg r_L$. At every point x, $r^{-1} \equiv r_L^{-1} = -\dfrac{\partial^2\zeta}{\partial x^2}$ is given by Laplace formula (the small viscosity term $2\eta\,\dfrac{\partial v_z}{\partial z}$ being neglected) :

$$\left(\frac{\partial p}{\partial x}\right) x = \frac{\sigma}{r} \tag{23}$$

($x = 0$ at mid-point O, Fig. 2). The "Poiseuille" overpressure gradient being given by [2] :

$$\frac{\partial p}{\partial x} = \frac{3G\sigma'}{2h} \tag{24}$$

Direct integration gives :

$$\frac{\partial\zeta}{\partial x} = -\frac{(\partial p/\partial x)}{\sigma}\frac{x^2}{2} + \theta_0\ ;\ \zeta = -\frac{(\partial p/\partial x)}{\sigma}\frac{x^3}{6} + \theta_0 x \tag{25}$$

$\left(\zeta(x=0) = \zeta\left(x = \dfrac{L}{2}\right) = h\right)$, so that :

$$\theta_0 = \theta(x=0) = \frac{G}{16}\frac{L^2}{h}\frac{\sigma'}{\sigma} \tag{26}$$

in agreement with equation (11) of section 2.

Now, point M of figure 2, with maximum elevation a and abscissa $x_{\mathrm{M}} = \dfrac{L}{2} - b$, is determined by $\left.\dfrac{\partial\zeta}{\partial x}\right|_{\mathrm{M}} = 0$, that is :

$$b = \frac{L}{2}\left(1 - 1/\sqrt{3}\right) \simeq 0.4\,\frac{L}{2} \tag{27}$$

Then, a slightly overstimated evaluation of deformation a is :

$$a \lesssim \theta_0\left(\frac{L}{2} - b\right) = \theta_0\frac{L}{2\sqrt{3}} \simeq 0.3\,\theta_0 L \simeq 2\times 10^{-2} G\frac{\sigma'}{\sigma}\frac{L^3}{h} \tag{28}$$

Note the cubic dependence in cell length, as opposed to direct proportionality in the barometric case, section 3. Typically, with $\dfrac{\sigma'}{\sigma} = 10^{-4}\mathrm{K}^{-1}$, $G = 20\,\mathrm{Kcm}^{-1}$, $h = 0.3$ cm and $L = 2$ cm, we get : $a \lesssim 8\ \mu$m. But a increases rapidly with L, and our analysis breaks down of course as soon as deformation a ceases to be much smaller than thickness h.

Consider now the opposite case of a long cell : $L \gg \ell$, $r_L \gg r_\ell$. The main curvature is transverse and uniform along ℓ, given at every abscissa x by :

$$\frac{1}{r_\ell} = \frac{1}{\sigma}\left(\frac{\partial p}{\partial x}\right) x = \frac{3}{2}\frac{G}{h}\frac{\sigma'}{\sigma} x \tag{29}$$

i.e. a depression of free surface in the left-hand side of the cell, a swelling in the right-hand side, and an inflection point in the middle. The deformation amplitude $a(x)$ is now given by :

$$a(x) = \frac{(\ell/2)^2}{2r_\ell(x)} \tag{30}$$

With $\ell = 1$ cm, $L = 10$ cm, and the same values as above for σ'/σ, G and h, the maximum deformation $\left|a\left(x \simeq \pm\frac{L}{4}\right)\right|$ is about 30 μm.

This case is perhaps less interesting, experimentally, than the first one, although σ' shows up in formula (30) as it does in equations (26) and (28). A mixed cross-over $\left(r^{-1} = r_L^{-1} + r_\ell^{-1},\ r_L \approx r_\ell\right)$ is expected for cells with more or less square shape.

The treatment in this paper could be extended to a system of two fluid layers separated by an interface. One might even consider a phase-separated system approaching its critical point T_c. The equivalent of equation (26) would then lead to an evaluation of the quantity $-(T_c - T)\frac{\sigma'}{\sigma} = \mu$, that is, the critical exponent of surface tension $\sigma = \sigma_0 (T_c - T)^\mu$. (The experiment would be delicate though, due to the necessity of working with the smallest possible thermal gradient, and of maintaining the interface pinned against the walls.)

5. Conclusions. A method for measuring σ'.

We have discussed, as a function of layer thickness, the free-surface deformation under an applied horizontal thermal gradient. This was done with an analysis in terms of dimensionless quantities and with emphasis on both cases where gravity, or capillarity, provide for the restoring force.

The latter case is relevant to low-gravity conditions. It would be interesting, under such conditions, to try and check (e.g. photographically) the square L-dependence of angle θ_0 (formula (26)), or the cubic dependence of amplitude a (Eq. (28)), and the overall shape of the deformed surface as sketched in figure 2.

In the former case, under ordinary gravity, and temperature gradients G a few tens degrees per cm, the characteristic deviation θ is of order a fraction of 1 % (and the characteristic thickness h_c, Eq. (14), is a fraction of a centimeter for a liquid metal).

We stress again that we have neglected throughout any temperature variation of σ' and β themselves, as well as of viscosity. Should this assumption break down (under larger thermal gradients for instance), the various dimensionless numbers we have introduced in sections 1 and 2 would vary along ∇T, and the treatment would be invalidated. The Poiseuille gradient in equation (24) for example, would now have to be included in any integration along Ox.

These considerations yield the principle of a new, simple but accurate method for measuring the surface-tension temperature coefficient $\sigma' = \frac{\mathrm{d}\sigma}{\mathrm{d}T}$. Working with $h < h_c$, an optical measurement of the tilt-angle θ leads us through equation (13) to σ', if thermal expansion β is known (avoiding, at the same time, rotational-flow problems if $\mathrm{Gr}\,(h = h_c)$ happens to be $\gtrsim (\mathrm{Gr})_c$). Of course, expression (26) for the σ-restored (i.e. low-gravity) situation, also enables us to measure σ'. The relative precision of the two choices (normal or low gravity) depends, among other factors, upon L, the cell length.

This method would be ideally suited for liquid metals where the temperature dependence of σ' itself (and of β and η) is relatively weak, and vaporization effects can be ignored.

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