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OCCURRENCE OF FREE CONVECTION IN A PLANE LAYER IN THE PRESENCE OF A CHEMICAL TRANSPORT REACTION

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We consider the conditions for the occurrence of natural convection when a volatile compound is transported through a gaseous phase as a result of a chemical reaction with a solid substance. We determine the variation of the critical Rayleigh number for the principal level of instability as a function of the parameters of the process.

In an infinite plane horizontal gaseous layer with solid boundaries we are given thermal boundary conditions of the first kind. On both of the boundaries there takes place a heterogeneous reversible exothermic reaction of the type $\nu_{AA} + \nu_{SS} = \nu_{BB}$; A is the initial gas; S, solid material of the wall; B, gaseous reaction product; ν_{A} , ν_{S} , ν_{B} , stoichiometric coefficients. The mixture of gases in the layer may be considered binary, since the reaction takes place in a heterogeneous manner and the vapor pressure of the solid material is negligibly small. For $T_1 > T_2$ (Fig. 1), since the reaction is exothermic, the thermodynamic equilibrium is shifted in such a way that the rate of the direct reaction, and consequently the gas flow rate A, on surface I will be less than on surface II. For the reaction product B the situation is reversed. The difference in concentrations gives rise to flows of the components caused by diffusion and convection. If the solid material of the surface takes part in the reaction, there will be mass Stefan flow in the system. In stationary conditions, the presence of Stefan flow is analogous to blowing into the system at a constant velocity. For the given conditions, there may be thermal and concentration nonuniformities in density in the mixture of gases. We write the concentration of the light component as $P_A/P = c$, and the density of the

mixture as $\rho = \frac{P}{RT} [\mu_A + (\mu_A - \mu_B)c], \mu_A, \mu_B$ being the molecular masses of the components. Following [1], we

shall assume that the density of the mixture admits of a linear expansion with respect to the average values of T and c, i.e., $\rho = \rho_0 (1 - \beta_1 T' - \beta_2 c')$, where T' and c' are the deviations from the average values;

$$\beta_{1} = -\frac{1}{\rho_{0}} \left(\frac{\partial \rho}{\partial T} \right)_{P,c}; \quad \beta_{2} = -\frac{1}{\rho_{0}} \left(\frac{\partial \rho}{\partial c} \right)_{P,T}.$$

We write the equations of free convection of the mixture, considering it incompressible [1]. It should be noted that $\mathbf{v} = \mathbf{v}_{c} + \mathbf{v}_{0}$ is the total hydrodynamic velocity; \mathbf{v}_{c} is the convective velocity; \mathbf{v}_{0} is the Stefan velocity corresponding to the average density ρ_{0} .

Disregarding thermal diffusion and diffusive heat conduction in the heat and mass flows and assuming that the nonuniformity in density is essential only in the expression for the lifting force (the Boussinesq approximation), we obtain a system of equations,

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Fig. Scheme of the process.

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\nabla) \mathbf{v} = -\frac{1}{\rho_0} \nabla^P + \mathbf{v}\Delta \mathbf{v} + g(\beta_1 T' + \beta_2 c') \cdot \gamma,$$

$$\frac{\partial \mathbf{v}}{\partial t} = 0,$$

$$\frac{\partial T}{\partial t} + \mathbf{v}\nabla T = a\nabla T,$$

$$\frac{\partial c}{\partial t} + \mathbf{v}\nabla c = D\nabla c,$$
(1)

where γ is the unit gravitational vector; a, thermal conductivity; D, diffusion coefficient.

Let us consider the boundary conditions for system (1). For the velocity and temperature z = 0, $v = v_0$, $T = T_1$; z = h, $v = v_0$, $T = T_2$. The boundary conditions for the concentrations can be obtained in the quasistationary approximation, assuming that the rate of the heterogeneous reaction with respect to a given component is equal to the flow of this component to the surface [2].

The rate of the reverse reaction has the form

$$W_{A} = -\frac{d[A]}{dt} = -\frac{P}{RT} \frac{dc}{dt} = v_{A} \frac{P}{RT} [k^{+}c^{v_{A}} - k^{-}(1-c)^{v_{B}}], \qquad (2)$$

where

$$k^{+} = A_{0}^{+} \left(\frac{P}{RT}\right)^{\mathbf{v}_{A}-1} \exp\left(-\frac{E_{+}}{RT}\right);$$

$$k^{-} = A_{0}^{-} \left(\frac{P}{RT}\right)^{\mathbf{v}_{B}-1} \exp\left(-\frac{E_{-}}{RT}\right);$$

 A_0^+ , A_0^- are preexponents; E_+ , E_- are the activation energies of the forward and reverse reactions, respectively.

The total flow of component A is

$$j_A = \frac{P}{RT} \frac{D}{\alpha} \frac{dc}{dz} + v_0 \frac{P}{RT} c, \qquad (3)$$

where $\alpha = 1 + \left(\frac{\mu_A - \mu_B}{\mu_A}\right) \vec{c}$; \vec{c} is the average value of the concentration.

From (2) and (3) we obtain the boundary conditions:

$$z = 0 \quad \frac{dc_1}{dz} = \frac{\mathbf{v}_A \alpha h}{D} \quad k_1^- [K_p^{(2)} \, c_1^{\nu_A} - (1 - c_1)^{\nu_B}] - \frac{v_0}{D} \, c_1,$$

$$z = h \quad \frac{dc_2}{dz} = \frac{\mathbf{v}_A \alpha h}{D} \quad k_2^- [K_p^{(2)} \, c_2^{\nu_A} - (1 - c_2)^{\nu_B}] + \frac{v_0}{D} \, c_2.$$
(4)

We reduce Eqs. (1) and conditions (2)-(4) to dimensionless form, making use of the variables

$$\tau = \frac{D}{h^2} t, \quad \zeta = \frac{z}{h}, \quad u = \frac{vh}{D}, \quad p = \frac{h^2}{\rho_0 v D} P, \quad \theta = \frac{T - T_2}{T_1 - T_2}, \quad \eta = \frac{c - c_2}{c_1 - c_2}$$



Fig. 2. Variation of Ra₁^{*} as a function of log s: 1) corresponds to the case γ_1 , $\gamma_2 \gg 1$; 2) γ_1 , $\gamma_2 \ll 1$; 3) calculated for the transport reaction Ni + 4CO = Ni(CO)₄ when $\gamma_0 = 10^2$.

We obtain the system

$$\frac{\partial \mathbf{u}}{\partial \tau} + (\mathbf{u}\nabla) \mathbf{u} = -\Pr \nabla p + \Pr \Delta \mathbf{u} + (\operatorname{Ra}_{1}\theta + \operatorname{Ra}_{2}\eta) \cdot \gamma,$$

$$\operatorname{div} \mathbf{u} = 0.$$

$$\frac{\partial \theta}{\partial \tau} + \mathbf{u}\nabla \theta = \mathrm{L}\Delta\theta,$$

$$\frac{\partial \eta}{\partial \tau} + \mathbf{u}\nabla \eta = \Delta\eta$$
(5)

with the boundary conditions

$$\frac{d\eta}{d\zeta} = \frac{A_{1}}{c_{1} - c_{2}} \left\{ K_{p}^{(1)} \left[\eta \left(c_{1} - c_{2} \right) + c_{2} \right]^{\nu_{A}} - \left[1 - \eta \left(c_{1} - c_{2} \right) - c_{2} \right]^{\nu_{B}} + \frac{Pe}{c_{1} - c_{2}} \left[\eta \left(c_{1} - c_{2} \right) + c_{2} \right] \right\},$$

$$\zeta = 1, \quad u = Pe, \quad \theta = 0,$$

$$\frac{d\eta}{d\zeta} = \frac{-A_{2}}{c_{1} - c_{2}} \left\{ K_{p}^{(2)} \left[\eta \left(c_{1} - c_{2} \right) + c_{2} \right]^{\nu_{A}} - \left[1 - \eta \left(c_{1} - c_{2} \right) - c_{2} \right]^{\nu_{B}} + \frac{Pe}{c_{1} - c_{2}} \left[\eta \left(c_{1} - c_{2} \right) + c_{2} \right] \right\}.$$

$$(6)$$

Here $\Pr = v/D$, L = a/D. From this point on, we shall assume that for the gases $\Pr = L = 1$, $\operatorname{Ra}_{i} = g\beta_{1}(T_{1} - T_{2}) \times h^{3}/D^{2}$, $\operatorname{Ra}_{2} = g\beta_{2}(c_{1} - c_{2})h^{3}/D^{2}$, thermal Rayleigh number and its concentration analog; $A = v_{A}\alpha h^{2}k^{-}/D$, ratio of the scales of the rate of the reverse reaction and the diffusion rate; $\operatorname{Pe} = v_{0}h/D$, Péclet number.

 $\xi = 0, \quad \mu = \text{Pe}, \quad \theta = 1,$

We consider the conditions for stationary equilibrium of the system. We obtain the equations

$$\operatorname{Pe} \nabla \theta_0 = \Delta \theta_0, \quad \operatorname{Pe} \nabla \eta_0 = \Delta \eta_0.$$

The effect of Pe on the initial temperature and concentration profiles was investigated in [3], in which it was shown that for $Pe \leq 1$ the initial profile practically coincides with the linear.

In [4] an expression was obtained for v_0 in the presence of a heterogeneous reaction in stationary conditions:

$$v_0 = \frac{\delta D}{\gamma h \alpha} \frac{\mu_A}{\mu_B} \ln \frac{1 - \gamma c_0^2}{1 - \gamma c_1^0}, \quad \text{where} \quad \delta = 1 - \frac{\mu_B v_B}{\mu_A v_A}, \quad \gamma = 1 - \frac{v_B}{v_A}.$$

Estimates indicate that for typical transport reactions the value of Pe varies between 0.01 and 0.5. Therefore, the effect of the initial Stefan flow on the limit at which convection occurs may be disregarded. In this case the conditions for stationary equilibrium have the form

$$\Delta \theta_0 = 0, \quad \Delta \eta_0 = 0, \quad \eta_0 = \theta_0 = 1 - \zeta, \quad \frac{d\eta_0}{d\zeta} \Big|_{0,1} = -1.$$

The conditions for the initial concentrations reduce to the form

$$c_{1}^{0} - c_{2}^{0} = A_{1} [K_{p}^{(1)} c_{1}^{0^{\nu} A} - (1 - c_{1}^{0})^{\nu B}],$$

$$c_{1}^{0} - c_{2}^{0} = -A_{2} [K_{p}^{(2)} c_{2}^{0^{\nu} A} - (1 - c_{2}^{0})^{\nu B}].$$
(7)

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tion drop $\Delta \overline{c^0}$ as a function of Δt .

Hereafter we shall assume that $\nu_A = 2$, $\nu_B = 1$. We introduce small perturbations in equilibrium and substitute them into the initial equations (5) and the conditions (6), linearizing with respect to the perturbations; we then obtain the equations for linear perturbations:

$$\frac{\partial \mathbf{u}}{\partial \mathbf{r}} = -\nabla p + \Delta \mathbf{u} + (\operatorname{Ra}_{1} \theta + \operatorname{Ra}_{2} \eta) \gamma,$$

$$\operatorname{div} \mathbf{u} = 0,$$

$$\frac{\partial \theta}{\partial \tau} + \mathbf{u} \nabla \theta_{0} = \Delta \theta,$$

$$\frac{\partial \eta}{\partial \tau} + \mathbf{u} \nabla \eta_{0} = \Delta \eta$$
(8)

and the boundary conditions:

$$\zeta = 0, \quad u = 0, \quad \theta = 0, \quad \frac{d\eta}{d\zeta} = B_1 \eta,$$

$$\zeta = 1, \quad u = 0, \quad \theta = 0, \quad \frac{d\eta}{d\zeta} = -B_2 \eta,$$
(9)

where $B_1 = A_1 [2K_{\rho}^{(1)} c_1^0 + 1], \quad B_2 = A_2 [2K_{\rho}^{(2)} c_2^0 + 1].$

Eliminating the pressure in (8) and setting the perturbations proportional to $\exp[-\lambda t + i(k_1x + k_2y)]$ for $\lambda = 0$, we obtain a boundary-value problem for neutral perturbations:

$$u^{1V} - 2k^{2}u'' + k''u = (Ra_{1}\theta + Ra_{2}\eta)k^{2},$$

$$\theta^{11} - k^{2}\theta = -u, \quad \eta^{11} - k_{2}\eta = -u_{\bullet}$$
(10)

Here $k^2 = k_1^2 + k_2^2$, where k is the wave number. The boundary conditions correspond to the conditions (9). System (10) can be solved by the Bubnov-Galerkin method [5]. The perturbation in the rate is given in the form $u = \zeta^2(1-\zeta)^2$. As a result, we obtain an expression for the Rayleigh number of the principal level of instability in the form

$$Ra_{i} = Ra_{1}^{0} / \left(1 + s \frac{I_{i}}{I_{2}}\right), \qquad (11)$$

where $\operatorname{Ra}_{1}^{0} = \operatorname{Lk}^{4}/I_{1}$ is the solution of the problem for purely thermal convection with solid isothermal boundaries, in which

$$L = k^{4} + 24k^{2} + 504; \quad I_{1} = A + 1260k (c_{1}f_{1} + c_{2}f_{2});$$

$$I_{2} = A + 1260k (c_{3}f_{1} + c_{4}f_{2}); \quad c_{1} = -a \frac{(1 - ch k)}{sh k}; \quad c_{2} = -a;$$

$$a = 2k^{-4} + 24k^{-6}; \quad b = 12k^{-4}; \quad f_{1} = (12 + k^{2}) (ch k - 1) - 6k sh k;$$

$$f_{2} = (12 + k^{2}) sh k - 6k (ch k - 1);$$

$$c_{3} = [(aB_{1} + b) (k sh k + B_{2} ch k) - B_{1} (aB_{2} + b)] z^{-1};$$



Fig. 4. Limits of the variation of s as a function of Δt : 1) $\gamma_0 =$ 10^2 ; 2) $\gamma_0 = 1$; 3) $\gamma_0 = 10^{-2}$; 4) $\gamma_0 \rightarrow \infty \ (\Delta c^0 = \Delta c^0)$.

$$c_4 = - [(aB_1 + b)(k \operatorname{ch} k + B_2 \operatorname{sh} k) + k(aB_2 + b)]z^{-1};$$

$$z = k(B_1 + B_2)\operatorname{ch} k + (k^2 + B_1B_2)\operatorname{sh} k.$$

The dimensionless complex s represents the ratio of the concentration Rayleigh number to the thermal

Rayleigh number: $s = \frac{\beta_2}{\beta_1} \frac{\Delta c^0}{\Delta T}$. The minimum of the expression (11) with respect to the wave number k

yields the minimum critical number Ra^{*}₁, which depends on the dimensionless parameters s, B₁, and B₂. To analyze this relation, we determine the initial concentration drop Δc^0 from conditions (7). The solution of system (7) is carried out by means of a linear expansion in a Taylor's series with respect to the thermodynamic equilibrium value of the concentrations c_1^0 and c_2^0 .

As is known,

$$K_p = \frac{1-\overline{c}}{\overline{c^2}} = \exp\left[-\frac{\Delta H^0}{RT} + \frac{\Delta S^0}{R}\right],$$

where ΔH^0 is the variation in enthalpy or the thermal effect of the reaction; ΔS^0 is the variation in entropy as a result of the reaction. Hence

$$\overline{c} = \frac{\sqrt{1+4K_p}-1}{2K_p} \, .$$

The concentration drop has the form

$$\Delta c^{0} = \varphi \Delta \overline{c^{0}}, \tag{12}$$

where $\Delta c_0 = c_1^0 - c_2^0$ is the equilibrium concentration drop; $\varphi = \left(1 + \frac{1}{2\gamma_1} + \frac{1}{2\gamma_2}\right)^{-1}$ is a factor determining the

macrokinetic regions in which the process takes place; $\gamma = A\sqrt{1 + 4K_p}$ is a dimensionless parameter characterizing the ratios of the scales of the rate of a reversible reaction to the diffusion rate. It should be noted that the parameters γ and B, from the boundary conditions (9), are practically equal for all values of K_p, and therefore the boundary conditions for the perturbations in concentration can be written in the form

$$\frac{d\eta}{d\zeta}\Big|_{0} = \gamma_{1}\eta, \quad \frac{d\eta}{d\zeta}\Big|_{1} = -\gamma_{2}\eta.$$
(13)

We introduce the dimensionless temperature $t = \frac{|\Delta H^0|}{RT_0^2} (T - T_0)$ [2], where $T_0 = \Delta H^0 / \Delta S^0$ is the value of

the temperature at which $K_p = 1$. Expressing T in terms of t, we obtain

$$K_p(t) = \exp\left[-t\left(1+\left|\beta\right|t\right)^{-1}\right], \quad \gamma = \gamma_0 f(t),$$

where

$$\gamma_{0} = \frac{2\sqrt{5}A_{0}^{-}\alpha h^{2}}{D} \exp\left(-\frac{E_{-}}{RT_{0}}\right);$$

$$f(t) = \sqrt{1+4K_{p}(t)} \exp\left(\frac{E_{-}}{\Delta H^{0}}\frac{t}{1+|\beta|t}\right); \quad \beta = \frac{R}{\Delta S^{0}}.$$

The parameter $s = \frac{d}{|\beta|} \frac{\Delta c^0}{\Delta t}$, where $d = \left(\frac{\mu_B}{\mu_B - \mu_A} - \tilde{c}_0\right)^{-1}$. We consider the limits of variation of s.

In the limiting case γ_1 , $\gamma_2 \gg 1$ the boundary conditions (13) become conditions of the first kind. The concentration drop is equal to the thermodynamic equilibrium concentration drop, and the process takes place in the diffusion region. The maximum value of s is found from the conditions $\Delta c^0 = \Delta c^0 \simeq 1$, $\Delta t = \Delta t_*$. The quantity Δt_* means the temperature width of the zone of reversibility of the reaction and is determined by the temperature dependence of the equilibrium constant. This quantity may be calculated from the condition $\Delta t_* = \alpha^{-1}$,

where
$$\alpha = \frac{dc^{0}}{dt}\Big|_{t=0}$$
. From the estimates for typical transport reactions, $s_{max} \simeq 20$. For γ_1 , $\gamma_2 \ll 1$ the

boundary conditions (13) correspond to the absence of a concentration flow (the analog of thermal adiabatic conditions). The process takes place in the kinetic region, $\Delta c^0 \ll \Delta \overline{c^0}$. For any Δt , d, $|\beta|$, the value of $s \ll 1$. It should be noted that for small values of Δt , for all reactions, $s \ll 1$, and in the limit as $\Delta t \rightarrow 0$, $\Delta s \rightarrow 0$.

Figure 2 shows the variation of Ra_1^* as a function of the parameters s, γ_1 , and γ_2 . The number Ra_1^* decreases sharply as s increases, and this reflects the relative contribution of the concentration convection. With decreasing γ_1 , γ_2 , the values of which determine the character of the boundary conditions, Ra_1^* also decreases, a fact which was noted for purely thermal convection and boundary conditions of the third kind [5].

Thus, taking account of the heterogeneous reversible exothermic reaction leads to a considerable decrease (as much as 95%) in the threshold of convective instability and a widening of the boundaries of the influence of the concentration convection.

As an example, let us consider the specific transport reaction Ni + 4CO = Ni(CO)₄. Corresponding to this reaction we have the kinetic equation (2) when $\nu_A = 2$, $\nu_B = 1$, $|\Delta H^0| = 40 \text{ kcal} \cdot \text{mole}^{-1}$; $|\Delta S^0| = 100 \text{ cal} \cdot \text{mole}^{-1} \cdot \text{deg}^{-1}$; $E_- = 52 \text{ kcal} \cdot \text{mole}^{-1}$ [6, 7]. The quantity γ_0 can vary within wide limits essentially as a result of the quantity A_0^- , which characterizes reactivity of the surface, and the system dimension h. The interval of variation is $\gamma_0 = 10^{-2} - 10^2$.

Figure 3 shows the variation of φ and $\Delta \overline{c}^0$ (4) as functions of the temperature drop $\Delta t = t_1 - t_2$. The value of t_1 was fixed at $t_1 = +5$, and t_2 varied from -10 to +5.

As can be seen from Fig. 2, when $\gamma_0 \ll 1$, the process takes place only in the kinetic region. As γ_0 increases, there is a shift in the diffusion region with respect to Δt , and the larger the value of γ_0 , the sooner this shift occurs. The temperature width of the zone of reversibility is $\Delta t_* \simeq 10$ ($\Delta \overline{c}^0 \approx 1$). The limits of variation of s with Δt for the indicated cases with respect to γ_0 are shown in Fig. 4.

If the process takes place in the kinetic region, i.e., $\gamma_0 \ll 1$, and even for small values of Δt , the quantity $s \ll 1$, which corresponds to the upper segment of curve 2 in Fig. 2. If the process can take place both in the kinetic region and in the diffusion region ($\gamma_0 \gg 1$), then the variation of Ra₁^{*} as a function of s takes the form of curve 3 in Fig. 2.

Thus, we have obtained an expression for the critical Rayleigh number of the principal level of instability as a function of the dimensionless parameters of the process s, γ_1 , γ_2 . The parameter s describes the relative contribution of the concentration convection to the picture of the general instability of the mixture, while γ_1 and γ_2 reflect the nature of the boundary conditions with respect to concentration. As s increases, the conditions for the occurrence of convection become much less severe. The critical value of Ra^{*} can be reduced by 90-95% if the process takes place in the diffusion region and the temperature drop is equal to the thermodynamically optimal drop Δt_* at which the concentration drop is maximum ($\Delta c^0 = \Delta \overline{c}^0 = 1$).

A decrease in γ_1 and γ_2 also brings a decrease of 10-20% in Ra_1^* . However, for small values of γ_1 and γ_2 the process can take place only in the kinetic region, the value of $s \ll 1$, and therefore the total decrease in Ra_1^* is small. For a specific transport reaction we have shown the regions in which the process takes place,

the limits of variation of the parameter s for different values of γ_1 and γ_2 , and also the variation of Ra^{*} as a function of these parameters.

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MOTION OF GAS BUBBLES IN AN INFINITE VOLUME OF STATIONARY LIQUID IN A

GRAVITATIONAL FIELD

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The resisting forces, velocities, and shape parameters of gas bubbles rising in an infinite volume of liquid are found analytically.

The laws of motion of gas bubbles relative to a liquid are fundamental for the construction of a theory of two-phase media [1]. Ordinarily in the theoretical description of the laws of motion of bubbles in a liquid it is assumed that the bubbles are spheres of radius a and that their motion in the liquid is potential and satisfies the boundary-value problem [2]

$$\Delta \varphi = 0; \quad \mathbf{u} = \operatorname{grad} \varphi; \quad u_r = 0 \quad \operatorname{at} \quad r = a; \quad \mathbf{u} \to \mathbf{U} \quad \operatorname{as} \quad r \to \infty.$$
(1)

The solution of problem (1) determines the behavior of the normal u_r and tangential u_η velocity components in the neighborhood of a bubble:

$$u_r = U \left[1 - \left(\frac{a}{r}\right)^3 \right] \sin \eta; \quad u_\eta = -U \left[1 + \frac{1}{2} \left(\frac{a}{r}\right)^3 \right] \cos \eta, \tag{2}$$

and the pressure distribution on the surface of a bubble is described by Bernoulli's equation

$$\frac{\rho}{2} \left[u_{\eta}^2 \right]_{r=a} + \rho_{\theta} = \text{const.}$$
(3)

Equations (2) and (3) are solved for gas bubbles satisfying the pressure balance condition

$$\rho_0 = \rho_b - \frac{2\sigma}{R} \,. \tag{4}$$

It follows from the condition of static equilibrium (4) that the bubbles can be spherical either if they are very small, when the second term is large, or are stationary with respect to the liquid. In other cases the nonuniformity of the pressure distribution over the surface of a bubble described by Eqs. (2) and (3) must lead to its deformation into an ellipsoid flattened in the direction of motion [2], and to an increase in the area of the interface and consequently to an increase in the dissipative forces as the bubble moves through a viscous liquid.

Thus, to find the laws of motion of a gas bubble it is necessary to solve the problem of the flow of an ellipsoidal bubble and the effect of its velocity with respect to the liquid on its shape.

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