ALIGNMENT OF HOLLOW MICROSPHERES

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Currently there is considerable interest in the problem of creating hollow microspheres of uniform thickness made of amorphous materials of the glass, plastic, and polymer types. In the first instance this is connected with question of microencapsulation [1], and also development of new areas for the application of microspheres: laser thermonuclear targets; microballoons for storing hydrogen, oxygen, and rocket fuels under high pressure; a filler for structural panels of flying and underwater equipment (see [2] and the bibliography in it).

In this work the process of nonisothermal expansion of an almost spherical thin liquid shell is considered under the action of heated gas pressure in the cavity. A phenomenological model is suggested based on the laws of mass, pulse, and energy conservation in an approximation of a thin layer which are closed by thermodynamic fundamental relationships for a thin film [3]. A numerical study is made of the mechanism of microsphere alignment imposed on the model connected with the dependence of material film viscosity on temperature.

In the approximation of a thin film the position of a liquid shell at instant of time t is prescribed by a two-dimensional surface $\Gamma = \Gamma(t)$ at each point $\mathbf{x}$ of which velocity $\mathbf{v}$, thickness h, absolute temperature θ, tension τ, and internal surface energy ε are determined. We assume that a polytropic gas within the cavity forms a closed thermodynamic system, i.e., its mass M is constant, and volume V, internal energy E, absolute temperature θ_0, and pressure p_0 only depend on time and they are connected by the equations

$$p_0V = MR_0\theta_0/m,\quad E = Mc_0\theta_0 \tag{1}$$

(R_0 is gas constant, m is molecular weight, c_0 is gas heat capacity). Correspondingly an external gas will play the role of a heat and pressure reservoir with characteristics θ_∞, p_∞.

Let ρ, c, σ, $\varkappa$, and $\mu(\theta)$ denote density, coefficients of specific heat capacity, surface tension, thermal conductivity, and film material viscosity, and

$$\mu(\theta) = \rho\nu\exp(\theta_*/\theta) \tag{2}$$

(ν, θ_* are constant values). Equation (2) is a good approximation of the dependence of dynamic viscosity coefficient for amorphous material on temperature. For simplicity ρ, c, σ, $\varkappa$ are assumed to be constant.

We take as independent thermodynamic parameters of the film thickness h and temperature θ and we assume the equalities $\tau = 2\sigma$, $\varepsilon = 2\sigma + \rho ch\theta$. In other words, film tension is made up of surface tensions of facial interphase boundaries which here are assumed to be noninteracting ($\tau_h = 0$), and the expression for energy emerges from the condition for existence of entropy: $h^2(\varepsilon/h)_h = \theta^2(\tau/\theta)_\theta$. We fix fundamental relationships for stress tensor T and heat flow vector $\mathbf{q}$ of film Γ [3]:

$$T = [2\sigma + 3\mu(\theta)h\,\mathrm{div}_\Gamma\mathbf{v}]\nabla_\Gamma\mathbf{x} + 2\mu(\theta)hD_\Gamma,\quad q = -\varkappa h\nabla_\Gamma\theta. \tag{3}$$

Here ∇_Γ is surface gradient; D_Γ is strain rate tensor deviator of Γ. It is noted that coincides with the metric tensor of Γ and $\Delta_\Gamma\mathbf{x} = k\mathbf{n}$ ($k = -\mathrm{div}_\Gamma\mathbf{n}$ is the sum of principal curvatures of Γ, $\mathbf{n}$ is vector of the unit normal to Γ).

In addition, we assume the following equality for the surface density of heat sources:

$$Q = \beta(\theta_\infty - \theta) - (E_t + p_0V_t)/A \tag{4}$$

(β is interphase heat exchange coefficient for the film material with the external gas, A is area of Γ). For an almost spherical microballoon, with an average radius of $r_0 = (3V/4\pi)^{1/3}$ it is reasonable to use the equation $\beta = \varkappa_\infty/r_0$ ($\varkappa_\infty$ is thermal conductivity coefficient for the external gas). In (4) no consideration is given to radiation heat exchange according to the

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Stefan–Boltzman rule since in calculations for moderate temperature its contribution is insignificant. Finally, the following heuristic relationship

$$\theta_0 = \frac{1}{A}\int_\Gamma \theta d\Gamma \tag{5}$$

establishes the connection of the temperatures of the film and gas in the cavity.

We write asymptotically differential rules for mass, pulse, and energy conservation accurate with hk → 0 [3]:

$$\dot{h} + h\,\mathrm{div}_\Gamma \mathbf{v} = 0; \tag{6}$$

$$\rho h\dot{\mathbf{v}} = \mathrm{div}_\Gamma T + (p_0 - p_\infty)\mathbf{n}; \tag{7}$$

$$\rho ch\dot{\theta} = \mu(\theta)h[3(\mathrm{div}_\Gamma \mathbf{v})^2 + 2D_\Gamma : D_\Gamma] + Q - \mathrm{div}_\Gamma \mathbf{q}. \tag{8}$$

Here the action of mass forces is ignored; normal $\mathbf{n}$ is directed into the external gas; the period above indicates the total derivative with respect to time.

It is evident that for the position Γ and distribution of θ Eqs. (1) and (5) entirely determine the thermodynamic gas constant in the cavity, and therefore the functions sought will be h, v, θ at moving surface Γ whose points satisfy the equation $\dot{\mathbf{x}} = \mathbf{v}(\mathbf{x}, t)$. It is necessary to set initial conditions for them.

We assume that surface Γ is star-shaped with respect to the center of mass

$$\mathbf{a} = \int_\Gamma h\mathbf{x}d\Gamma \Big/ \int_\Gamma hd\Gamma, \tag{9}$$

then it is possible to parametrize it by a unit sphere $S = \{|\mathbf{s}| = 1\}$ by means of stereographic projection $\mathbf{x} = \mathbf{a}(t) + r(\mathbf{s}, t)\mathbf{s}$ with the center at point $\mathbf{a}$. If in the equality $\dot{\mathbf{x}} = \mathbf{a}_t + (r_t + \dot{\mathbf{s}}\cdot\nabla_S r)\mathbf{s} + r\dot{\mathbf{s}} = \mathbf{v}(\mathbf{x}, t)$ set $r\dot{\mathbf{s}} = \mathbf{u}(\mathbf{s}, t)$, then we obtain

$$\mathbf{v} = \mathbf{a}_t + (r_t + r^{-1}\mathbf{u}\cdot\nabla_S r)\,\mathbf{s} + \mathbf{u},\ \dot{f} = f_t + r^{-1}\mathbf{u}\cdot\nabla_S f \tag{10}$$

[$f(\mathbf{s}, t)$ is an arbitrary function]. These equations make it possible to rewrite problem (1)-(8) in terms $\mathbf{a}(t)$, $r(\mathbf{s}, t)$, $h(\mathbf{s}, t)$, $\theta(\mathbf{s}, t)$, $\mathbf{u}(\mathbf{s}, t)$ obtained as a result of substituting variables. It is evident that vector field $\mathbf{u}$ is tangential to sphere S.

From (6), (7), (9) it follows that $\mathbf{a}_{tt} = 0$, i.e., it is possible to assume that $\mathbf{a} = 0$, since the original equations permit Galilean transfer. We consider the main spherically symmetrical solution $h(t)$, $r(t)$, $\theta(t)$, $\mathbf{u} = \mathbf{0}$ and its small perturbations $H(\mathbf{s}, t)$, $R(\mathbf{s}, t)$, $\Theta(\mathbf{s}, t)$, $\mathbf{u}(\mathbf{s}, t)$ in terms of which the following linearized equalities are valid

$$\begin{gathered}\mathbf{n} = \mathbf{s} - r^{-1}\nabla_S R,\ \nabla_\Gamma = r^{-2}[(r - R)\nabla_S + \mathbf{s}\nabla_S R\cdot\nabla_S],\\ \nabla_\Gamma \mathbf{x} = \nabla_S \mathbf{s} + r^{-1}(\mathbf{s}\otimes\nabla_S R + \nabla_S R\otimes\mathbf{s}),\ \mathrm{div}_\Gamma \mathbf{v} = 2r_t/r + 2(R/r)_t + r^{-1}\,\mathrm{div}_S \mathbf{u},\ D_\Gamma = r^{-1}D_S\end{gathered} \tag{11}$$

(⊗ is a symbol of a tensor product, D_S is deviator of the symmetrical part of tensor $\nabla_S \mathbf{u}$). Taking account of (10) and (11) we obtain equations for determining the main solution

$$\begin{gathered}(hr^2)_t = 0,\ \rho hr_{tt} = p_0 - p_\infty - 4r^{-1}[\sigma + 3\mu(\theta)hr_t/r],\\ \rho ch\theta_t = 12\mu(\theta)h(r_t/r)^2 + Q;\end{gathered} \tag{12}$$

$$p_0 = 3MR_0\theta/(4\pi r^3 m),\ Q = \beta(\theta_\infty - \theta) - p_0 r_t - Mc_0\theta_t/(4\pi r^2) \tag{13}$$

and its perturbation

$$(H/h + 2R/r)_t + r^{-1}\,\mathrm{div}_S \mathbf{u} = 0; \tag{14}$$

$$\rho(hR_{tt} + r_{tt}H) = 2r^{-2}[\sigma + 3\mu(\theta)hr_t/r](\Delta_S R + 2R) + 6r^{-1}[\mu(\theta)H_t - 2\mu_\theta(\theta)\Theta hr_t/r]; \tag{15}$$

$$\rho h(r\mathbf{u}_{tt} + r_{tt}\nabla_S R) = 2\mu(\theta)hr^{-1}\,\mathrm{div}_S D_S - 3\nabla_S\,[\mu(\theta)H_t - 2\mu_\theta(\theta)\Theta hr_t/r]; \tag{16}$$

$$\rho c(h\Theta_t + \theta_t H) = \varkappa hr^{-2}\Delta_S\Theta - 12\mu(\theta)h(r_t/r)(H/h)_t + 12(r_t/r)^2[h\mu_\theta(\theta)\Theta + \mu(\theta)H] - \beta\Theta. \tag{17}$$

It is noted that vector $\mathrm{div}_S D_S$ is tangential to sphere S, and in a potential perturbation of velocity $\mathbf{u} = r^{-1}\nabla_S U$ the equality $2\mathrm{div}_S D_S = r^{-1}\nabla_S(\Delta_S U + 2U)$ is valid. In this case (16) is transformed into a scalar equation for potential

$$\rho h(U_t + r_{tt}R) = \mu(\theta)hr^{-2}(\Delta_S U + 2U) + 6\mu_\theta(\theta)\Theta hr_t/r - 3\mu(\theta)H_t. \tag{18}$$

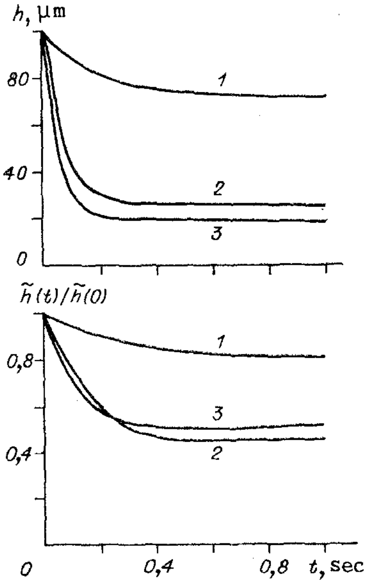

Fig. 1

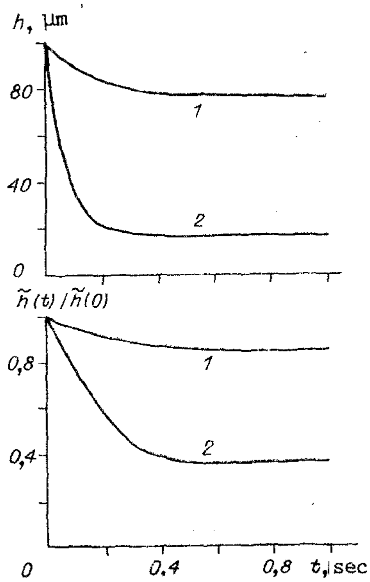

Fig. 2

In deriving equations in the versions (14)-(18) it is assumed that H, R, Θ, and U have a zero mean with respect to sphere S, and therefore A, V, θ_0, p_0, and β are not perturbed on account of the equations

$$A = 4\pi r^2 + 2r\int_S R dS,\quad V = \frac{4}{3}\pi r^3 + r^2\int_S R dS,\quad \theta_0 = \theta + \frac{1}{4\pi}\int_S \Theta dS.$$

It is evident that problem (14), (15), (17), (18) assumes separation of variables, and the first spherical harmonic which also determines the degree of microsphere alignment will attenuate weakest of all. If we limit ourselves to perturbations of this form, by retaining the center of mass $\mathbf{a} = \frac{r}{4\pi}\int_S\left(\frac{H}{h} + \frac{3R}{r}\right)\mathbf{s}dS$, and in fact ($\mathbf{e}$ = const) $H = \tilde h(t)\mathbf{e}\cdot\mathbf{s}$, $R = -(r/3h)\tilde h(t)\,\mathbf{e}\cdot\mathbf{s}$, $\Theta = \tilde\theta(t)\mathbf{e}\cdot\mathbf{s}$, then for $\tilde h(t)$, $\tilde\theta(t)$ we obtain normal differential equations

$$\rho[r_{tt}\tilde h - h(r\tilde h/3h)_{tt}] = 6r^{-1}[\mu(\theta)\tilde h_t - 2\mu_\theta(\theta)\tilde\theta h r_t/r]; \tag{19}$$

$$\rho c(h\tilde\theta_t + \theta_t\tilde h) = 12(r_t/r)^2[h\mu_\theta(\theta)\tilde\theta + \mu(\theta)\tilde h] - 12\mu(\theta)h(r_t/r)(\tilde h/h)_t - 2\varkappa h r^{-2}\tilde\theta - \beta\tilde\theta. \tag{20}$$

Thus, the set of Eqs. (12), (13), (19), (20) with respect to $h(t)$, $r(t)$, $\theta(t)$, $\tilde h(t)$, $\tilde\theta(t)$ with prescribed dependences $\mu(\theta)$ [Eq. (2)] and $\beta = \varkappa_\infty/r$ is the simplest model of microsphere alignment.

It is noted that from (19) in a quasistationary approximation (i.e., when forces of inertia are omitted) there follows the equation $\tilde h_t = 2\mu_\theta(\theta)\tilde\theta h r_t/[r\mu(\theta)]$ from which it is possible to estimate qualitatively the behavior of $\tilde h(t)$. In fact, if $\tilde\theta > 0$ with $\tilde h > 0$ ("thermal inertia" of the film predominates over the dissipative effect), then the alignment process is realized for a microballoon ($\tilde h_t < 0$) for $r_t > 0$, $\mu_\theta(\theta) < 0$.

The set of differential equations (12), (13), (19), (20) was integrated numerically with the initial conditions $r(0) = 500$ μm, $h(0) = 100$ μm, $\theta(0) = 2000$ K $\tilde\theta(0) = 0$, $\tilde h(0) \neq 0$ (original misalignment). In this case consideration of forces of inertia cause in calculations development of a sharp boundary layer with t = 0, and therefore a quasistationary approximation is used where velocity $r_t(0)$ is not prescribed. Glass was selected as a microsphere material with the following physical characteristics [4]: $\rho = 2200$ kg/m³, $c = 1437$ J/(kg·K), $\varkappa = 2.35$ W/(m·K), $\sigma = 0.3$ N/m. The internal gas was air whose mass was 0.02% of the mass of the shell, and the external gas had the characteristics: $\varkappa_\infty = 3\cdot10^{-2}$ W/(m·K), $\theta_\infty = 300$ K, $p_\infty = 1.01\cdot10^4$ Pa. The dynamic viscosity coefficient was determined by (2).

Given in Fig. 1 are the results of calculations for the following values: 1-3) $\nu = 4.55\cdot10^{-10}$, $4.55\cdot10^{-12}$, $4.55\cdot10^{-10}$ m²/sec, $\theta_* = 50{,}000$, 55,000, 45,000 K, respectively. Presented in Fig. 2 is the dependence of the solution on the change in internal pressure p_0 by an increase in mass of the gas: 0.2 and 2% (curves 1 and 2) of the shell mass with $\nu = 4.55\cdot10^{-10}$ m²/sec, $\theta_* = 55{,}000$ K. Noted in Fig. 3 is the effect of external pressure

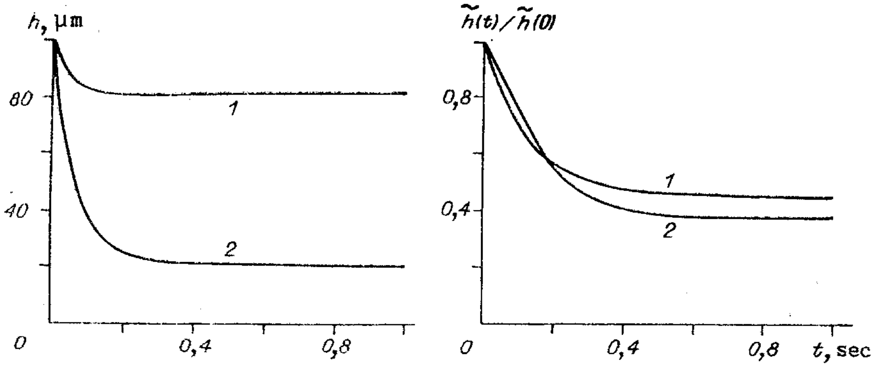

Fig. 3

$p_\infty \cdot 10^5$, $3.04 \cdot 10^3$ Pa (curves 1 and 2) with $\nu = 4.55 \cdot 10^{-12}$ m²/sec, $\theta_* = 55{,}000$ K.

The alignment mechanism described connected with the dependence of viscosity on temperature only operates with expansion of a shell ($\tilde{h}_t = 0$ with $h_t = 0$), and consequently it is possible to call it dynamic. Here it is difficult to work out an ideal result ($\tilde{h} \to 0$ with $t \to \infty$) in view of the short duration of the dynamic process. We also considered a thermodynamic alignment mechanism based on the dependence of surface tension coefficient σ on temperature θ which it is simple to consider in the model. However, the calculations performed showed the insignificance of this effect for the given size of glass microballoons (it is noted that alignment sets in with the natural condition $\sigma_\theta(0) < 0$). Apparently small microspheres warm up rapidly, they become almost isothermal, and therefore it is necessary to look for and to analyze other alignment mechanisms, for example connected with the dependence of tension τ on thickness h.

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