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VOLUME-VISCOUS PROPERTIES OF MERCURY CONTAINING A GASEOUS PHASE

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The rheological model of a liquid with heredity is used to analyze experimental results on normal stress relaxation in mercury. A low end estimate of viscosity at low frequencies is obtained.

A model of a viscous compressible liquid with heredity, generalizing the linear-viscous Navier—Stokes model, was proposed in [1]. This model has been employed to describe a liquid with large volume relaxation time. In the present study the theory of [1] will be used to describe behavior of liquid mercury containing fine air bubbles. For rapid volume deformations of the mixture, system equilibrium is disrupted as regards the processes of solution and heat exchange between the components, which manifests itself macroscopically by the appearance of relaxation processes in the liquid, i.e., heredity.

The fundamental relationships of [1] will be presented below in slightly changed notation.

We assume that the stress tensor in a liquid particle in a fixed Cartesian coordinate system at the time t_0 can be represented in the form

$$p^{ij}(t_0) = -p(\rho_0, T_0)\delta^{ij} + \tau^{ij}(t_0), \tag{1}$$

where $\rho_0 = \rho(t_0)$, $T_0 = T(t_0)$ are the density and temperature in the particle at the time t_0; $p = p(\rho, T)$ is the pressure; the viscous stress tensor τ^{ij} depends on ρ_0 and the values of the temperature T and the deformation rate tensor $e_{ij} = 1/2(v_{i,j} + v_{j,i})$ in the particle at all times preceding to:

$$\tau^{ij}(t_0) = \tau^{ij}[\rho_0, T(t \leqslant t_0), e_{kl}(t \leqslant t_0)]. \tag{2}$$

It will be assumed that if at $t \leq t_0$ $T = T(t) = T_0$ and $e_{kl} = 0$, then $\tau^{ij}(t_0) = 0$. We introduce the notation $\tilde{e} = e^i_i$, $S_{ij} = e_{ij} - (1/3)\tilde{e}\delta_{ij}$. Let the liquid be isotropic, while functional (2) is a linear integral operator of $\theta = dT/dt$ and e_{ij}. Then because of isotropicity, to specify Eq. (2) three kernels $K_i = K_i(\rho_0, T_0, t)$, $i = 1, 2, 3$ are sufficient:

$$\tau^{ij}(t_0) = \delta^{ij}\int_{-\infty}^{t_0} K_1(\rho_0, T_0, t_0 - t)\tilde{e}(t)\,dt + 2\int_{-\infty}^{t_0} K_2(\rho_0, T_0, t_0 - t)s^{ij}(t)\,dt + \delta^{ij}\int_{-\infty}^{t_0} K_3(\rho_0, T_0, t_0 - t)\theta(t)\,dt. \tag{3}$$

It should be noted that Eq. (3) is of limited applicability. In the general case of nonlinear viscoelasticity in place of the tensor e_{ij} in Eqs. (2), (3), we must use the tensor ε_{ij} (where ε_{ij} is the finite deformation tensor [3, 4]). Moreover in the expression for τ^{ij} terms nonlinear in ε_{ij} may appear. It can be shown that the general case leads to Eq. (3) for small deformations or for media with sufficiently short relaxation times.

Integration over time in Eq. (3) is performed for a fixed liquid particle. The kernels $K_i = K_i(\rho, T, t)$, $i = 1, 2, 3$ are in fact defined for $t \geq 0$, however it is convenient to predefine them, taking $K_i(\rho, T, t) = 0$ at $t < 0$. We may then take the upper integration limit in Eq. (3) equal to infinity. For brevity, we will omit the dependence of K_i on ρ and T below.

Following [2], we require that for cyclical processes, where $\rho \to \rho_1$, $T \to T_1$, $e_{ij} \to 0$ as at $t \to \pm\infty$.

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$$\int_{-\infty}^{+\infty} \tau^{ij}(t)\, e_{ij}(t)\, dt \geqslant 0, \tag{4}$$

which implies compatibility of Eq. (3) with thermodynamics. Since the functions $\theta = \theta(t)$ and $e_{ij} = e_{ij}(t)$ can vary over wide limits, inequality (4) imposes definite limitations on the possible for of the kernels $K_i = K_i(t)$. If we transfer from the kernels $K_i = K_i(t)$ to their Fourier transforms $\tilde{K}_i = \tilde{K}_i(\omega)$, then Eq. (4) reduces to a system of inequalities

$$\int_0^{+\infty} |f(\omega)|^2 \operatorname{Re} \tilde{K}_1(\omega)\, d\omega \geqslant 0, \qquad \int_0^{+\infty} |g(\omega)|^2 \operatorname{Re} \tilde{K}_2(\omega)\, d\omega \geqslant 0, \tag{5}$$

$$\int_0^{+\infty} \operatorname{Re}[f^*(\omega)\, h(\omega)\, K_3(\omega)]\, d\omega \geqslant 0. \tag{6}$$

Here $f(0) = g(0) = 0$, and in the remaining expressions the complex functions f, g, h are arbitrary. It is clear that Eq. (6) can be satisfied only if $K_3 = 0$. Inequality (5) can be satisfied, for example, by taking

$$K_j(\omega) = \sum_{n=0}^{+\infty} \frac{A_n^j}{1 + i\omega\tau_n}, \quad \tau_n > \tau_{n+1} > 0,$$
$$A_n^j \geqslant 0, \quad K_j(t) = \sum_{n=0}^{+\infty} \frac{A_n^j}{\tau_n} e^{-t/\tau_n}, \quad j = 1, 2, \tag{7}$$

which implies the presence of a countable relaxation time spectrum. The quantities A_n^j, τ_n depend on ρ and T.

As can easily be seen, the model of Eqs. (1), (3), (7) is a generalization of the Navier–Stokes model of a linear-viscous liquid. In fact, for slow processes when all parameters within the particle change with characteristic times much longer than τ_0, Eq. (1) transforms to

$$p^{ij} = -p(\rho, T)\delta^{ij} + \eta_v(\rho, T)\tilde{e}\delta^{ij} + 2\eta_s(\rho, T)s^{ij},$$
$$\eta_v = \sum_{n=0}^{+\infty} A_n^1, \quad \eta_s = \sum_{n=0}^{+\infty} A_n^2,$$

where η_v, η_s are coefficients of liquid volume and shear viscosity.

Let the layer of liquid described by the model of Eqs. (1), (3), (7) be at rest at $t \leq 0$, while at $t > 0$ its thickness $l = l(t)$ changes from $l_0 = l(0)$ to $l(\Delta t) = l_0 + \Delta l$, with the liquid temperature remaining constant and equal to T_0, while the density changes from $\rho_0 = \rho(0)$ to $\rho_1 = \rho(\Delta t)$. At $t \geq \Delta t$ the normal stress on the layer surface has the form

$$\sigma(t) = -p_1 + \int_0^{\Delta t} \left(K_1(t-\tau) + \frac{4}{3}K_2(t-\tau)\right) \frac{\dot{l}(\tau)}{l(\tau)}\, d\tau,$$
$$p_1 = p(\rho_1, T_0), \quad \rho_1 = \rho_0 l_0/(l_0 + \Delta l). \tag{8}$$

For $\Delta l/l_0 \ll 1$ and $t, \tau_0 \gg \Delta t$ Eq. (8) reduces to

$$\sigma(t) = -p_0 + c_0^2\rho_0 \frac{\Delta l}{l_0} + \frac{1}{\tau_0}\left(A_0^1 + \frac{4}{3}A_0^2\right) e^{-t/\tau_0} \frac{\Delta l}{l_0}, \tag{9}$$

where $p_0 = p(\rho_0, T_0)$, $c_0^2 = \dfrac{\partial p}{\partial \rho}(\rho_0, T_0)$. In Eq. (9) only the dominant exponential remains.

Thus observation of the normal stress relaxation process permits determination of τ_0 and the quantity $\eta = A_0^1 + (4/3)A_0^2$, thus allowing estimation of a lower limit for the volume viscosity η_v.

Such an experiment was performed for mercury containing air in the form of fine bubbles using Leutert (Federal Republic of Germany) equipment containing a piston pump and reference manometer at 10^2 MPa. The mercury mass was 2.06 kg, with the ratio of air mass to mercury mass not exceeding $4.5 \cdot 10^{-7}$. The specimen was subjected to rapid loading and unloading over the pressure range 0.1-70 MPa at a temperature T = 289 K. After a rapid change in mixture volume normal stress relaxation was observed and values of σ_i, t_i, i = 1, ..., N were recorded. For an arbitrary set of three pairs of σ_i, t_i the relaxation time τ_0 was calculated, which was then averaged over all possible sets. Then the parameter η and the equilibrium pressure p were found by minimization of the function

$$F = F(\eta,\ p) = \sum_{i=1}^{N} e^{t_i/\tau_0}\left(\sigma_i + p - \frac{\eta \Delta l}{\tau_0 l_0} e^{-t_i/\tau_0}\right)^2.$$

As a result it was found that $\tau_0 = 12 \cdot (1 \pm 0.08)$ sec, $\eta = 2 \cdot 10^8 (1 \pm 0.5)$ Pa•sec. Within the limits of experimental accuracy τ_0 and η did not depend on pressure. Since η_s for mercury undoubtedly exceeds $1.6 \cdot 10^{-3}$ Pa•sec [5], we obtain the estimate $\eta_v \geq 10^8$ Pa•sec. This severely contradicts the value $\eta_v = 9.2 \cdot 10^{-3}$ Pa•sec, calculated from the known ultrasound absorption coefficient α in mercury [5]:

$$\alpha/\omega^2 = \frac{1}{2c_0^3\rho_0}\left(\eta_v + \frac{4}{3}\eta_s\right) = 2{,}7 \cdot 10^{-16}\ \text{sec}^2/\text{m}.$$

A simple explanation of this apparent contradiction is that in the model considered in the present study

$$\alpha/\omega^2 = \frac{1}{2c_0^3\rho_0}\operatorname{Re}\left[\tilde{K}_1(\omega) + \frac{4}{3}\tilde{K}_2(\omega)\right] = \frac{1}{2c_0^3\rho_0}\sum_{n=0}^{+\infty}\frac{A_n^1 + \frac{4}{3}A_n^2}{1 + (\omega\tau_n)^2}.$$

In the latter sum, at frequencies $\omega \gtrsim 10^6$ Hz only terms with $\tau_i \lesssim 10^{-6}$ sec produce a contribution, while the contribution of the term $\eta/[1 + (\omega\tau_0)^2]$ is strongly suppressed.

From the physical viewpoint the great difference in results of volume viscosity measurements at low and high frequencies is related to the fact that at low frequencies the dominant contribution to the measured quality is produced by processes of transport through bubble boundaries, while at high frequencies a significant role is played only by hydrodynamic processes near the bubbles.

We will note that apparently the first theoretical study which indicated that air bubbles may increase the effective volume viscosity of a liquid was that of Taylor [6], in which, however, relaxation processes were not considered.

NOTATION

t, t_0, t_i, time; ρ, ρ_0, ρ_1, density; T, T_0, T_1, temperature; p^{ij}, stress tensor; τ^{ij}, viscous stress tensor; δ^{ij}, Kronecker symbol; v_i, flow velocity components; e_{ij}, deformation rate tensor; s_{ij}, deviator portion of deformation rate tensor; η_v, η_s, volume and shear viscosities; ω, frequency; τ_n, relaxation times; α, ultrasound absorption coefficient; K_i, kernals characterizing liquid relaxation; A_n^i, weight coefficients characterizing contribution of n-th relaxation process; θ, rate of change of temperature; f, g, h, arbitrary functions of frequency; c_0, isothermal speed of sound at low frequency; σ, σ_i, normal stress; l, l_0, liquid layer thickness; Δl, change in layer thickness; Δt, time increment; η, phenomenological coefficent with dimensions of viscosity; p, p_0, p_1, pressure.

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