# PRESSURE INTENSIFICATION ON THE FRONT OF A SHOCK WAVE PROPAGATING IN A HETEROGENEOUS SYSTEM

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An increase in pressure in the wave front as compared to the pulse initiating the wave has been observed experimentally in a study of shock-wave propagation in aqueous suspensions of bentonite [1]. In suspensions in which the solid phase is in the form of colloidal size particles  $\delta = 10^{-7} - 10^{-8}$  m of the mineral montmorillonite with mass content c = 6%, with multishock loading an intensification of this effect from experiment to experiment was observed [2]. In order to study the principles involved in this anomalous "intensification" of pressure on the shock wave front, as well as to clarify the effect of the nature of the material in the dispersed phase, experiments were performed with particles of another broad class of clay-like minerals — kaolinite.

## 1. Formulation of the Experiment

The experiments were performed in a vertical shock tube [2], in which waves were created by rupture of a diaphragm separating the medium to be studied from the high-pressure chamber.

One experiment consisted of a sequence of shock loadings of the same column formed by the dispersed system. The shocks followed each other at intervals of 5-6 min and were initiated by a continuously acting "piston" with identical working gas pressure in the highpressure chamber  $p_e = 2.4$  MPa in all experiments.

To obtain a general pattern of wave motion along the tube, longtime oscilloscope displays  $\approx 10$  msec were used. Results of pressure measurements in various sections of the tube A, B, C were used to construct wave diagrams in the plane x (mixture column height)-t (time) (the construction method is explained in [3]). At the beginning and end of each series of shock loadings, calibration experiments were performed with water. A typical test is shown in Fig. 1. The intensity of the incident wave  $p_1 \approx p_e = 2.4$  MPa, the initial mixture pressure  $p_0 = 0.1$  MPa. The wave diagram in the coordinates x, t clarifies the process of passage of the compression (solid curve) and rarefaction (dashed curve) waves. Analysis of the test experiments revealed that the results were in agreement with the acoustical approximation of shock wave theory.

#### 2. Waves in the Kaolinite Suspension

An experiment involving ten shock loadings of a water suspension of kaolinite clay powder is illustrated by the wave patterns and oscillograms of Fig. 2, where loadings 2, 4, and 9 are depicted. The mixture density  $\rho_0 = 1.18 \cdot 10^3 \text{ kg/m}^3$ , with 8.5 kg of clay powder being added to 25 kg of water, which corresponds to a mass concentration of particles in water of c = 25%.

Qualitative changes take place in the structure of the compression wave front from experiment to experiment. The amplitude of the pressure discontinuity increases. Thus, for the first shock the amplitude is in fact equal to the pressure  $p_e = 2.4$  MPa created by the high-pressure chamber "piston." By the fourth or fifth loading, the pressure has smoothly increased by a factor of 3-4 times, to 5-7 MPa more than the initiating pressure, which remained constant for each shock wave. The amplitude then stabilized, but an extended zone of excess pressure  $\Delta p = p_1 - p_{10}$  of duration  $\Delta \tau$  following the discontinuity (Fig. 3a) was formed. With time this zone relaxes to the equilibrium pressure  $p_{10}$  of a wave in water in the absence of particles, initiated by a pulse of the same intensity. The fall in  $\Delta p$ to zero can be interpreted in the x-t diagrams as a rarefaction wave, delayed from the compression discontinuity by a time  $\Delta \tau$ . Figure 3b shows experimental data on the increase in

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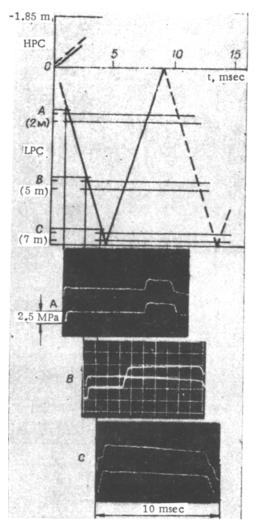


Fig. 1

in excess pressure  $\Delta p_{max}$  and the length of the relaxation layer  $\Delta \tau$  as functions of the shock number n, which characterize the effect under consideration for tenfold shock loading of a 25% kaolinite clay suspension. Also shown here are the values of  $\Delta p_{max}$  and  $\Delta \tau$  for n = 14, 15, 16, obtained in a similar experiment performed in a suspension with clay content c = 33%. With increase in particle concentration and shock number the effect intensifies.

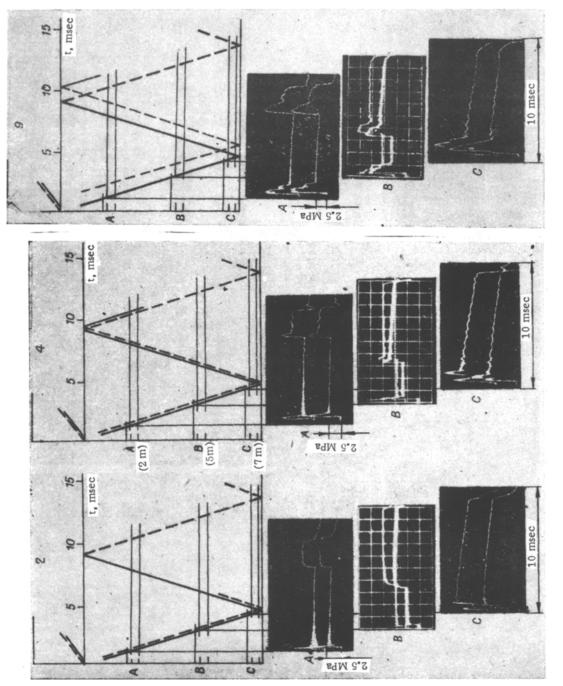
In all experiments the velocity of the incident wave front was equal to that of the reflected,  $D_1 = D_2 = 1450 \pm 50$  m/sec, which corresponds to the speed of sound in water. Pressure oscillograms indicate that the character of reflection of the compression and rarefaction waves from rigid wall and free surface is acoustical.

In order to study the structure of the wave front, the preceding experiment was repeated using "fast" oscilloscope sweeps of ~1 msec over the entire screen (Fig. 4). The vertical columns of three oscillograms correspond to single experiments. The tendencies observed previously were completely reproduced in this experiment. In the first experiment one sees

a decrease in  $\Delta p_{\max}$  and excess momentum  $\Delta I = \int_{0}^{\Delta v} \Delta p dt$  with distance x, while in further shock

actions Apmax is approximately the same as the wave evolves along the tube.

Multiple shock-wave action on the suspension leads to the appearance in the front of excess (relative to the initiating pulse) pressure, the mean value of which increases monotonically from experiment to experiment. The intensification of the excess pressure manifests itself in a smooth increase in amplitude and increase in the duration of the increased pressure zone. A similar effect was observed in a shock wave front in suspensions in [2], where the dispersed phase consisted of the mineral montmorillonite with c = 6% and  $\delta = 10^{-7}$ -





 $10^{-8}$  m. However, in those experiments increase in  $\Delta \tau$  and the pressure at the discontinuity itself increased up to the fourth shock, after which the amplitude and duration of the excess pressure zone stabilize. In the experiments with the kaolinite suspension "saturation" of  $\Delta \tau$  was noted only after the fifteenth or sixteenth shock, with practically constant amplitude  $\Delta p_{max}$ .

We will note one more difference. In the suspension with kaolinite particles at c = 20-30%, independent of the number of the shock in all experiments the propagation velocities of incident and reflected waves were practically equal to the speed of sound in pure water, 1450 m/sec. Reflection of waves from the contact surfaces was acoustic. In the suspension with montmorillonite even at c = 10-15% for initiating pressures of  $p_e = 2-3$  MPa there is a clearly expressed nonlinearity - a retardation of the incident wave motion to 450-250 m/sec, nonacoustic reflection from the rigid wall  $p_2/p_1 = 7-15$  with a velocity close to the speed of sound in water. Here  $p_2$  is the pressure in the reflected wave front. The reduction in the velocity of the incident wave and its nonacoustic reflection for the given montmorillonite particle concentration are apparently related to formation ahead of the front of a so-called thixotropic-coagulation structure and its complete destruction behind the discontinuity [3, 4].

### 3. Evaluation of Results

The excess pressure effect in the front and the monotonic increase in momentum  $\Delta I$  from experiment to experiment appear in the dispersed system without heat and mass exchange with the surroundings at constant volume of the dispersed phase. Nevertheless, preliminary shock processing of the suspension affects its initial state before experiment.

The pressure increase observed in experiment should be accompanied by a corresponding increase in energy per unit volume of the mixture. We will demonstrate this for a heterogeneous medium.

In a dispersed system with surface area of all particles totaling A and volume V, the total free energy can be represented in the form (see, for example, [5])

$$F = F' + F^s + \sigma A, \tag{3.1}$$

where  $F^s$  and  $F^{\ell}$  are the volume components of the energies of the solid and liquid phases;  $\sigma A$  is the boundary-layer surface energy;  $\sigma$  is the surface tension for the boundaries considered. It follows from the expression for energy density

$$F_0 = F_0^l + F_0^s + \sigma A/V$$

that increase in free energy per unit volume of the mixture may occur because of increase in the system surface area; for example, because of fractionation of aggregates of the dispersed phase in the shock wave front.

The physical phase boundary surface has a finite thickness, dependent on the radius of action of intermolecular and electrostatic forces in the layer. In this transition region the free energy density differs from its value within the phases. For individual materials the loss in potential surface energy is proportional to the area of the boundary. In those cases where overlapping of the boundary layers occurs, the free energy begins to depend on the area in a more complex manner, the surface energy contributions of the form of Eq. (3.1) becoming nonadditive.

The given situation develops in deformation of so-called coagulation contacts [6], formed at points of closest approach of particles when their concentration is sufficiently high in the dispersed phase. The surface tension of the interlayer does not coincide with the free energy per unit surface f = F/A. The relationship between f and  $\sigma$  has the form  $\sigma = f + A\partial f/\partial A$ . For a thin film of thickness h, the quantity  $\sigma$  is some function of h [7]:  $\sigma = f - h\partial f/\partial h$ . Change in the free energy of the system also depends on h:

$$dF = dF^{l} + dF^{s} + \sigma_{0}dA - A^{*}\frac{\partial f}{\partial h}dh.$$

Here  $\sigma_0 = f$  is the surface tension of a thick layer;  $A^* = \chi A_1$  is the total surface of the coagulation contacts between particles;  $\chi$  is the number of particles;  $A_1$  is the area of a single contact. The corresponding change in free energy, equal to  $dF_1 = A_1(\partial f/\partial h)dh$  shows that for a change in contact thickness in an equilibrium isothermal process an additional

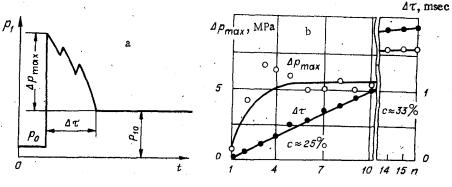


Fig. 3

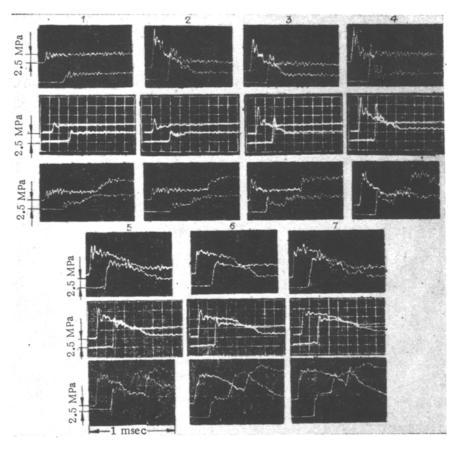


Fig. 4

expenditure of work is required, defined by the expression  $dF_1 = A_1\Pi(h)dh$  ( $\Pi(h) = -\partial f/\partial h$  is the cleavage pressure [8]). When  $\Pi(h) > 0$ , the film tends to thicken under the action of the excess pressure within the liquid volume of which it is a part:

$$p_1 = p_{10} + \Pi(h), \tag{3.2}$$

where  $p_{10}$  is the pressure in the thin film;  $p_1$  is the pressure in the liquid volume.

The contribution of the cleavage pressure forces N(h) at the contacts  $\chi_s$  per unit surface area of the dispersed phase can be written in the form

$$p_1 = p_{10} + N(h)\chi_s. \tag{3.3}$$

According to [9] the quantity  $\chi_{\mathbf{S}}$  is defined by the expression

$$\chi_{s} = (3/2)(k\alpha)/(\pi\delta^{2}).$$
(3.4)

Here k is the coordination number,  $\alpha$  is the volume content of particles. It has been shown [6] that independent of its nature the interaction force between particles can be expressed in terms of the cleavage pressure

$$N(h) = \pi \overline{R} f(h) \tag{3.5}$$

(where  $\bar{R}$  is the radius of curvature of the surface at the contact point). For spherical particles of radius R, from Eqs. (3.2)-(3.5) we have

$$p_1 = p_{10} + k_1 f(h) \alpha / R. \tag{3.6}$$

It is evident that the pressure component related to change in h is proportional to the dispersion of the system and the volume content  $\alpha$  (where  $k_1$  is a coefficient of the order of unity).

The following facts are assumed by the given physical model. Upon shock compression of a suspension with amplitude up to 100 MPa, thermal and inertial effects in the condensed phase with colloidal-sized particles are insignificant. The principal role is played by viscosity. In a coordinate system fixed to the front, the mixture flowing into the discontinuity with equilibrium  $h = h_0$  [such that  $\Pi(h_0) = 0$ ] brakes, and thus is compressed. In the viscous discontinuity a decrease in all interparticle distances occurs, i.e., dh < 0. Upon abrupt approach of particles due to overlapping of boundary layers the energy stored in coagulation contacts is released. This unavoidably leads to the appearance of an excess pressure  $\Pi(h) > 0$  and increases the curvature of the wave.

Behind the discontinuity there is a boundary layer relatively extensive in time ( $\approx \Delta \tau$ ), characterized by a drop in excess pressure to its equilibrium value. Scattering of the excess momentum of the incident flow takes place in this layer, and the energy liberated is transformed into uncompensated heat. The time required for conversion of the suspension from its initial to its final state is determined by  $\Delta \tau$ , and is much greater than the time required for establishment of pressure in the discontinuity, ~10 µsec, but is less than the characteristic time over the course of which the parameters change behind the boundary layer in the steady-state flow region. The pressure in this region depends solely on the velocity of the continuously acting high-pressure chamber "piston." The width of and pressure distribution within the wave front are strongly affected by the kinetics of nonequilibrium processes.

In each experiment any elementary volume of the medium entering the discontinuity undergoes the same sequence of changes of state as did the previous one. Therefore,  $\Delta p_{max}$  and, correspondingly, the excess momentum  $\Delta I$ , are approximately conserved during evolution of the wave. The duration of the boundary layer is obviously determined by the mutual effect of the scalar flows related to volume viscosity, the degree of dispersion of the system, and the rate of conversion of excess pressure energy into uncompensated heat.

We will perform an approximate, order-of-magnitude, calculation of the pressure defined by the second term of Eq. (3.6). The work of wetting  $\beta$  in a symmetric gap is defined as the change in surface energy (with opposite sign) upon displacement of gas (vapor) 1 from the gap by the liquid 2, previously bounded by the solids 3 which form the gap:

$$\beta = -2(\sigma_{23} - \sigma_{13}) > 0,$$

where  $\sigma_{23}$ ,  $\sigma_{13}$ , and  $\sigma_{12}$  are the corresponding surface tensions on the intermedium boundaries 2-3, 1-3, and 1-2.

Upon wetting of the free external surface, energy is liberated in the form of heat, which can be measured in a calorimeter; surface 3 is coated by a layer of liquid 2 of sufficiently great thickness, and when displacement of medium 1 by liquid 2 occurs in a narrow gap, the wetting energy is partially stored in the form of excess thin film energy. In the adiabatic approximation  $f(h) = -2(\sigma_{23} - \sigma_{13})$ . From the condition for wetting of surface 3 by liquid 2,  $\sigma_{23} + \sigma_{12} - \sigma_{13} < 0$  it follows that  $-(\sigma_{23} - \sigma_{13}) > \sigma_{12}$ .

The lower limit of the maximum possible cleavage energy pressure is determined by the surface tension of the dispersed phase. For water,  $\sigma_{12} = 0.073 \text{ J/m}^2 \approx 0.1 \text{ J/m}^2$ . For the characteristic dimension of the elementary plates of the solid phase which scale off the initial aggregate, we take [10]:  $\delta = 10^{-8}$  m, then for a volume content  $\alpha = 10\%$  for evaluation of Eq. (3.6) we have

$$\Delta p = p_1 - p_{10} = 2 \cdot 10^6 \text{ Pa}$$
 .

This estimate of the energy f(h), used in [11], agrees qualitatively with the experimental results.

In an individual contact the maximum excess pressure can be estimated as [7]

$$11(h_{\min}) \approx 2\sigma_{12}/h_{\min} \approx 300$$
 MPa.

where  $h_{\min} \approx 6 \cdot 10^{-10}$  m is the thickness of a monomolecular water layer.

According to the concepts presented above concerning the development of excess pressure in a wave, the effect should manifest itself when the mean distance between particles of the dispersed phase  $h_0$  is close to twice the radius of action of surface forces. To this distance there corresponds some critical concentration of the dispersed phase, which when attained assures collective overlapping of boundary layers in the front of a relatively weak compression wave. Thus, the nature, concentration, and aggregate states of the interacting phases affect the structure of the shock wave front.

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