MECHANISM OF LAMINATION OF PARTICULATE COMPOSITES IN SHOCK LOADING

N. A. Kostyukov

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It is known that the two-dimensional shock loading of particulate composite materials (PCM) – which are mechanical mixtures of dissimilar solid particles – often leads to an extremely nonuniform distribution (stratification) of the components after loading [1, 2]. For example, the following situation might occur: in a certain volume of a shock-compressed specimen, the concentration of one of the components becomes several times greater in one region than in adjacent regions. This property of PCM's – making their practical use more difficult – makes it very important to establish a stratification criterion. The question to be answered here is "what are the shock-wave parameters for which the material will remain uniform after loading?"

The first step in answering this question is the creation of a physical model that will accurately reflect the mechanism of the process that leads to lamination. The authors of [1, 2] examined possible approaches to the model description of results obtained for mixtures of copper–boron-nitride and copper–graphite. Below, we analyze these models, present new empirical data that lies outside the framework of existing representations, and propose a new physical model which qualitatively describes all of the results on PCM lamination known to date.

Analysis of Existing Lamination Models. In the explosive compaction of a mixture of powders of boron nitride and copper in cylindrical ampuls, the authors of [1] observed the formation of regions of high copper concentration in the central part of the pressed specimen. The specimen had the form of an annular layer which was symmetrical relative to the axis of the ampul. To explain this phenomenon, they proposed a model in which stratification occurred in those cases when an irregular (Mach) shock-wave regime of flow is realized in the loaded mixture. In this model, it is assumed that such layering is due to the presence of a subtantial radial gradient of mixture flow velocity near the surface of tangential discontinuity that divides the flow regions behind the fronts of the Mach and reflected shock waves (SW). Actually, all this reduces to the familiar effect of the migration of suspended particles toward the pipe axis in Poiseuille flow.

For approximate numerical calculations, it was assumed that the copper is in the molten state in the region where the maximum gradient of flow velocity exists. This was suggested by the results of study of the structure of preserved specimens. The estimates were made on the basis of experimental data from the determination of the force acting on a particle 1.55-32 mm in diameter in the boundary region of a Poiseuille flow [3]. The authors of [1] obtained satisfactory agreement between the theoretical and empirical values of the width of the annular layer in which the concentration of the solid component (boron nitride) is reduced. Such agreement was obtained with the assumptions that the viscosity of the liquid copper in the shock-compressed state was equal to its viscosity at atmospheric pressure, that the difference in the flow velocities behind the Mach and reflected waves was on the order of the velocity of the Mach wave front, and that the gradient flow lasted several microseconds.

However, these estimates are valid only for single smooth spheres in a steady laminar flow, i.e., for conditions which for certain do not conform to the compaction of a PCM in a cylindrical ampul. It was shown empirically in [4] that even small roughnesses of a spherical particle lead to a substantial increase in the force associated with its interaction with the surrounding flow. For PCM particles having complex morphology, the presumption of sphericity may deviate greatly from the actual flow conditions. Moreover, the viscosity coefficient depends on the state parameters of the substance and the rate of its loading [5, 6]. Thus, the use of values of viscosity at atmospheric pressure to describe processes taking place behind a shock front is questionable, and the estimates presented in [1] are unconvincing.

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The authors of [1] allow that other stratification mechanisms might exist, since they experimentally observed separation of particles within a broad range of concentrations of the mixture's components — including the case when one of the components cannot be regarded as being "suspended" in the flow of the other component.

The lamination of a copper–graphite mixture was observed in [2]. The setup of the experiments was similar to [1], but the outward manifestation of the phenomenon was different: after loading, graphite particles were concentrated near the pressing axis and formed a cylindrical core. In this case, copper was displaced toward the periphery. To explain this outcome, the author of [2] also used the hypothesis of the existence of an irregular shock-wave regime in the ampul and fusion of copper behind the Mach wave. However, he proposed a different lamination mechanism. He proposed that the mass flow behind the Mach wave had a complex spiral structure which combined displacement along the axis of the ampul with a powerful vortical flow around the axis. As a result, the copper melt was thrown toward the peripheral region of the flow, while the solid particles of graphite were concentrated near its axis.

The author restricted himself to a qualitative explanation, not involving himself with numerical estimates. However, a simple analysis immediately shows the basic flaw in the proposed mechanism. It follows from considerations discussed in [7] that to ensure a finite rate of radial displacement of the solid particles in the given model, the angular velocity of the flow behind the Mach wave should increase with a decrease in the radius of rotation as $r^{-1/2}$. This requirement conflicts with the result of analysis of the reasons for the swirling of the flow [2]. This result shows that the radius of rotation approaches zero, the rate of rotation of the flow decreases until it stops completely. Thus, the vortex model of stratification is internally inconsistent.

New Experimental Data. We will present results from experimental studies of the lamination effect for a considerably broader range of PCM's than was studied in [1, 2]. We found that it is possible to have cases of lamination that cannot be explained by the above mechanisms, since they take place under conditions that are inconsistent with the assumptions made in [1, 2].

The new experiments were conducted on binary mixtures which included powdered metals, carbides, nitrides, oxides, and other materials having a broad range of thermophysical and mechanical characteristics. The dimensions of the particles of both components were roughly the same and did not exceed 100 μm. The initial density of the composite was 40-80% of the density of the monolithic state. The setup of the experiments was similar to that described in [1, 2]. The inside diameters of the ampuls ranged from 3.7 to 15 mm, while their lengths ranged within 10-15 diameters. Charges of explosive (EX) were prepared from ammonite 6ZhV, hexogen, and mixtures of these two substances. After shock loading, we performed metallographic studies of the structure of the intact specimens. To prepare the metallographic sections, the ampuls were cut along planes perpendicular and parallel to the axis.

The results are shown in Table 1. They permit the following conclusions.

1. The above representations notwithstanding, lamination of the PCM is possible without the transformation of one of the components to the liquid state. For example, in experiments 4, 9, 15, and 16, we saw an increase in the concentration of the heavy component near the specimen axis even though the energy of the SW was considerably lower than the energy necessary for complete melting of the component with the lowest melting point.

In principle, even a weak shock wave can fuse a particle from the surface [8, 9]. However, the presence of the "cold" cores and the relatively small thicknesses of the fluid layer mean that the lamination models set forth in [1, 2] are invalid. To estimate the maximum possible thickness of the melt, we used an idealization of the shock compression of a PCM which is based on the following assumptions: a) the fraction of energy expended on the creation of defects in the crystalline structure of the particles and their brittle fracture is much smaller than the total energy of the shock compression; b) elastic energy is not stored, and all of the energy of the shock compression is expanded on increasing the thermal energy of the substance; c) heat energy is released instantaneously and is concentrated on the surface of the particles; d) all of the energy of the shock compression is stored in one (the i-th) component of the mixture.

We then arrive at the following relation to determine the mass fraction of the melt of the i-th component L_i

TABLE 1

No. of experiment	Composition of PCM	Mass ratio of the components	Relative density, %	Diameter of ampul, mm	Explosive	Character of lamination	Source
1	Boron-nitride–copper	1/37	50	—	Hexogen + sodium bicarbonate	HA	[1]
2	Iron–copper	1/1	50	—	» » »	None	[1]
3	Graphite–copper	—	—	6	Hexogen	LC	[2]
4	» »	—	—	6	Ammonite 6ZhV	LC	[2]
5	Graphite–titanium	1/4	62	15	» 6ZhV	HA	Present study
6	» »	1/4	45	15	» 6ZhV	HC	»
7	» »	1/4	62	3,7	» 6ZhV	None	»
8	Titanium-carbide–steel-G13	1/1	80	10	» 6ZhV	»	»
9	» » » G13	1/1	80	15	Hexogen	HC	»
10	Titanium-carbide–titanium nickelide	1/1	59	15	Ammonite 6ZhV	None	»
11	» » » »	1/1	80	15	Hexogen	»	»
12	Magnesium–lead	1/1	59	15	Ammonite 6ZhV	HC	»
13	Magnesium–tungsten	1/2	64	15	» 6ZhV	HC	»
14	» »	1/2	64	6	» 6ZhV	None	»
15	Boron-nitride–tungsten	2/1	53	15	» 6ZhV	HA	»
16	Boron-nitride–nichrome	3/17	41	15	» 6ZhV	HC	»
17	Titanium–tungsten	1/1	67	15	Hexogen	None	»
18	Aluminum-oxide–aluminum-nitride	1/1	53	15	»	»	»
19	Magnesium-oxide–copper	1/2	57	15	Hexogen + ammonite 6ZhV (1/2)	»	»
20	Sand–tungsten	1/2	42	15	» » »	»	»

Note. Increased in concentration: HC, HA, heavy component in the cylindrical core and the annular layer; LC, LA, light component in the cylindrical core and annular layer.

$$L_i = (D\Delta V)^2/\{2V_0^2[c_{pi}(T_{*i} - T_0) + H_i]\alpha_i\rho_i\},$$

where D is the velocity of propagation of the SW; V_0 is the specific volume of the mixture in the undisturbed state; ΔV is the change in the specific volume of the mixture during shock compression; T_0 is the initial temperature; ρ_i, c_{pi}, T_{*i}, H_i, and α_i are the density, heat capacity, melting point, latent heat of fusion, and mass fraction of the i-th component. The thickness of the molten layer $h \approx L_i d/6$ (d is the diameter of the particles). Numerical estimates show that $h/d \ll 1$ in our experiments. Considering that each of the above assumptions leads to overestimation of the mass fraction of melt that is present, it can be expected that the actual thickness of the fluid layer on the particle surface will be considerably less than the predicted thickness.

2. The presence of a high-speed flow behind the Mach wave is not a necessary condition for the origination of lamination. In a special series of experiments conducted using the above-described powder mixture, we placed copper plates 0.5-1.0 mm thick in the mixture with an orientation normal to the axis of the ampul. The plates were inserted to intercept the high-speed flow. Study of the specimens that were still intact after loading showed that lamination of the mixture occurs on both sides of the plates (Fig. 1). This finding is inconsistent with the model presented in [1]. Figure 1 presents a photograph of a metallographic section from a specimen after shock loading (experiment 13): 1) is the wall of the ampul; 2) is the copper plate; 3) is the region in which there is a high concentration of the heavy component. The arrow indicates the direction of propagation of the detonation wave.

3. Along with the already-observed [2] increase in the concentration of the light component near the specimen axis, the reverse effect is also possible in certain cases, i.e., there may be an increase in the concentration of the heavy component near the axis (see Table 1, experiments 6, 9, 12, 13, and 16). This effect is seen not only when the energy of the Mach wave is greater than the energy required for melting one of the components, but also when $h/d \ll 1$.

Thus, the arguments made in [1, 2] concerning the mechanisms responsible for stratification of a PCM during shock compression either are incorrect or are not the only possible explosions. It should be noted that they were obtained with the use of several assumptions and are based on indirect data – the results of studies of the structure of specimens that were intact after loading. Such information often leads to erroneous conclusions, since the most valuable information – information on the behavior of the material at the moment of loading – is lost.

Inertial Model of Lamination. One source of objective information in the search for other lamination mechanisms is direct observation of the flow pattern under dynamic conditions. Of considerable interest here is experimental study of the flow of binary mixtures of solid particles when they are subjected to shock loading [10]. The results obtained here are distinguished by their clarity and unambiguity, thanks to the simplicity and clarity of the analytical method employed – impulsive x-ray diffraction study.

Proceeding on the basis of these results, it can be stated that all of the data obtained to date on the stratification of PCM's can be fully explained by features of the structure of the mass flow at the moment of shock loading. Let us explain this conclusion with specific examples. Figure 2 shows some of the flow variants recorded in [10]. The dark and light points represent particles of the heavy and light components of the mixture. The solid and dashed lines represent their trajectories in a movable coordinate system connected with the detonation front from the explosive. In the figure, AC is the front of the detonation wave, AO is the front of the incident SW, OB is the front of the reflected SW, OO_1 is the front of the Mach SW, OT is the surface of tangential discontinuity, and MD is the reflecting surface (in a two-dimensional formulation) or the axis of symmetry (in an axisymmetric formulation). Having a greater kinetic energy than the lighter particles, the heavier particles are slowed less than the former when they pass through the shock wave. They continue to move for a certain period of time, smoothly changing direction under the influence of the particle interaction forces. In the general case, the velocities of the heavy and light particles eventually become equal a certain distance from the front. The situation depicted in Fig. 2a corresponds to weak mechanical interaction between the particles of the heavy and light components. This accounts for the existence of a significant velocity disequilibrium during the entire time of loading. The particles of the heavy component ultimately accumulate near the interface with the explosive. In our opinion, it is this phenomenon that accounts for the specific features of the lamination of the copper–graphite mixture observed in [2].

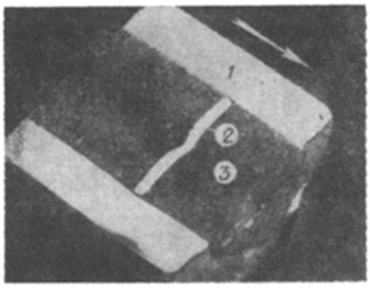

Fig. 1

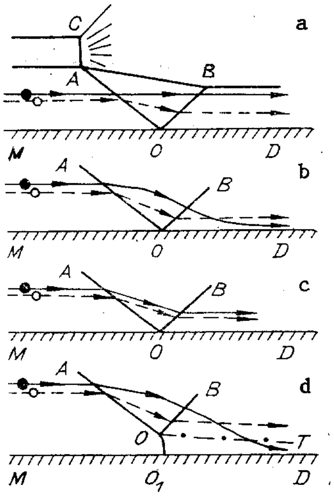

Fig. 2

The sizes of the regions of velocity disequilibrium behind the incident and reflected shocks may differ appreciably. Figure 2b shows the flow pattern for which the ratio of the dimensions of the regions is such that the particles of the heavy component begin to lose velocity only after having reached the reflecting surface MD. It is obvious that in such cases there is an increase in the concentration of heavy particles near the reflecting surface, i.e., the character of the stratification is directly opposite that examined above. Such a flow pattern could have been realized in experiments 6, 9, 12, 13, and 16 (see Table 1).

It should be noted that the preferential concentration of one of the components near the reflecting surface is not always characteristic of the given PCM. The lamination phenomenon may be absent when the test assembly has a different geometry (experiments 5, 7, and 13, 14).

Figure 2c shows the flow pattern for which the sizes of the regions of velocity disequilibrium behind the incident and reflected shock waves are smaller than the characteristic dimension of the particles of the mixture. Such flows arise when there is a small difference between the masses of the particles of both components or when the interparticle forces are large. Lamination will obviously be absent in these cases, and one-velocity models of the PCM can be used to describe the shock-wave processes. The data shown in the table indicates that the absence of lamination is seen mainly in mixtures consisting of high-strength components or components which are similar in strength.

We should especially note yet one more stabilizing factor which impedes the onset of lamination: subdivision of the particle surface. Comparison of experiments 9 and 10 shows that, given identical loading conditions and approximately the same ratios of component densities, the results obtained from the loading may differ appreciably. We connect this fact with differences in the morphology of the particles of titanium nickelide and steel G13: the first were irregular in form and had a subdivided surface, while the second were close to spherical. This resulted in a considerably greater interparticle force in experiment 11 compared to experiment 9.

In the case of irregular reflections regimes, the flows behind the incident and reflected shock waves are similar to those examined above. The lamination phenomena observed behind oblique shock waves are also possible behind the Mach wave in those regions where the incoming flow is not perpendicular to the surface of the front (such as near the branch point of the shock wave).

Deserving of special attention is the flow structure shown in Fig. 2d, which corresponds to the occurrence of filtration of heavy particles into the region compressed by the Mach wave. In this case, the geometry of the region in the intact specimens where there is a high concentration of heavy particles is determined by the interparticle forces. If these forces are great enough to prevent the particles from reaching the reflecting surface, then the high-concentration region will be a strip in the case of plane loading or an annular zone in the case of axisymmetric loading. This is seen in experiments 1, 5, and 15. Otherwise, the heavy particles accumulate near the reflecting surface or the specimen axis. It should be noted that the form of the high-concentration zone in the case of weak particle interaction will be the same as with a regular regime of SW reflection (Fig. 2b). Thus, special experiments must be performed to determine which of the two possible regimes (regular or irregular) will occur in experiments 5, 9, 12, 13, and 16.

Discussion of Results. In light of the inertial model of PCM lamination proposed in the present study, there are three main factors which determine the character of the lamination: the ratio of the masses of the particles of dissimilar components; the magnitude of the interparticle force; the duration of the shock wave, i.e., the time from the moment the flow crosses the shock wave to the moment it interacts with another shock front (such as the reflected shock wave) or the rarefaction wave.

Given fixed parameters characterizing a shock loading, the force associated with the particle interaction is determined by the thermophysical and strength characteristics of the materials of the particles and their morphologies. The time parameter depends on the geometric dimensions of the specimen. It follows from this that a change in the character of lamination can be expected to occur with a change in the diameter of the ampul. This is confirmed by the results of experiments 5, 7, and 13, 14.

The physical model itself does not yet offer a quantitative connection between the quantities characterizing the process it describes. To solve this problem, it will be necessary to take the next step — construct a mathematical model. However, serious difficulties are encountered in any attempt to mathematically describe the PCM lamination phenomenon on the basis of an inertial model. These difficulties are connected mainly with the lack of any suitable method to determine the interparticle force behind the shock front and the lack of suitable approaches to solving a two-dimensional boundary-value problem concerning the loading of a layer of a PCM by a pressure charge with allowance for the effect of the ampul and the velocity disequilibrium of the components.

Mathematical modeling is possible in certain special cases. For example, if the volume fraction of the heavy component is small and the dynamic strength of this component is greater than that of the light component, then the pore space at the shock front will be filled mainly by the light component [11]. Here, the form of the heavy particles remains essentially unchanged. As in any adiabatic process, the deformation of the particles of the main component during shock loading inevitably leads to an increase in temperature. Given a shock loading of sufficiently high energy and assuming satisfaction of the inequality $d/u > d^2/\varkappa$ ($\varkappa$ is the diffusivity and u is the mass velocity), the material of the particles undergoes a thermally-induced loss of strength behind the shock front. This means that the interparticle force no longer depends on the initial strength characteristics of the material of the light component and is determined mainly by forces associated with surface friction.

If the heavy particles are close to spherical (as in the case of grains of a rapidly quenched material), then it becomes possible to describe the structure of the shock wave within the framework of the problem of the flow of a viscous liquid about nondeformable smooth spheres. This in turn makes it possible to predict the range of loading parameters in which the degree of lamination of the components in the front will not exceed a prescribed value. In this case, the effective value of the viscosity coefficient is assumed to be known.

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EFFECTIVE ELASTIC MODULI OF GRANULAR MEDIA

A. D. Zaikin

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The solution of problems concerning the deformation of heterogeneous media is often based on the hypothesis of effective homogeneity [1], which amounts to assuming that the heterogeneous medium can be replaced by a homogeneous continuum having certain effective parameters. The problem consists of determining the effective properties of the heterogeneous medium through the properties of the phases and some of their geometric characteristics. Finding the effective elastic moduli of sandstone oil and gas resevoirs in general and the velocities of longitudinal and transverse waves in particular, determining the relationship between the velocities and the structure of the pore space on the one hand and the properties of the fluid on the other hand – these are very important problems for seismic prospecting.

We will concern ourselves with the simpler situation of an empty (not containing fluid) consolidated granular skeleton. There are several approaches to solving this problem, but until recently the granular character of the skeleton has been accounted for only in solutions based on the Hertz problem concerning the deformation of two spheres at the point of contact under the influence of applied forces [2]. However, the presumption of point contact at the initial moment of loading does not conform to the condition of consolidation of rock and leads to a situation whereby elastic waves in such a model propagate only in the presence of external pressure. A number of other solutions [1] account only for the fraction of the volume corresponding to the pore space. In practice, an equation obtained from statistical analysis of a large number of laboratory measurements is widely used to relate the velocities of elastic waves with a certain characteristic of the structure (mainly porosity), as well as with mean grain size, permeability, etc. Thus, there is a need for new approaches to the calculation of the effective elastic moduli of granular media.

The author of [3] proposed the use of the variational approach to calculate the stress state of an individual main and the effective elastic moduli of an empty granular skeleton. He investigated a longwave approximation, i.e., a situation in which the length of the elastic wave is much greater than the sizes of the grains. This makes it possible to change over to the static equations for an individual grain. A granular body is subjected to a hypothetical unilateral compression

$$e_{11} = e_{22} = e_{21} = e_{13} = e_{23} = 0,\ e_{33} = 1. \tag{1}$$

It is assumed that the energy associated with the deformation of one grain is minimal with certain restrictions on the character of this deformation, i.e.: the grain as a whole does not undergo displacement or rotation, and the strain tensor at the center of the grain has the form (1). These requirements make it possible to completely determine the unknowns at the points of contact with the loading grain. However, the hypothesis on the character of the elastic strains of an individual grain needs to be more carefully substantiated. In any case, it should be consistent with the asymptotic solution for a continuum.

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