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Destruction of cylinder ampoules with solid phase reactive mixtures under explosive loading

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Peculiarities of solid-phase synthesis in aluminum-sulfur and aluminum-fluoroplast mixtures in cylinder ampoules under explosive loading have been studied in experiments and numerically. In experiments the chosen parameters of explosive loading were similar to those which are usually used for explosive compacting of the inert porous materials in cylinder ampoules. The results of experiments show that cylinder ampoules are destructed in explosive loading conditions when mixture used as a filler is capable of very fast exothermal reactions. To find out the reasons of ampoules destruction numeric calculations have been carried out by the method of finite elements on the base of multicomponent media. While studying the deformation of multicomponent media it is necessary to take into account the state and reaction of each component and components displacement inside a chosen released volume of the mixture. As a condition of compatible deformation of the components a pressure balance of mixture components was chosen. Synthesis reaction in the mixture was described by the phenomenological model of irreversible chemical transformations based on zero-order kinetics. Simulation of compaction of the porous mixture was carried out on the base of a kinetic model of active type, which describes the growth or collapse of the pores resulting in a permanent change of the material properties and stress relaxation. It was found that pressure is increased sharply at the bottom of the ampoule when shock wave is reflected from the bottom ampoule lid as a compressive wave which is followed by chemical transformation rate growth. High rate of heat release during reaction in a lower ampoule part resulted in gas phase formation which leads to a further pressure increase and becomes the reason of ampoules destruction.

Key words: solid-phase synthesis, explosive loading, multicomponent medium, numerical simulation.

1. Introduction

Nowadays the processes connected with mechanic and physicochemical transformations caused by shock waves passed through metals, minerals, polymers and other solids [1-4] attract an undoubted interest. It is caused by the development of new technological processes to get advanced materials and necessity to reduce the costs in aviation and space engineering, power and machine engineering, chemistry and mining. Besides, to initiate these processes explosive charge is used being a compact, light and cheap energy source. Explosive technologies are developed mainly in metalworking production for molding, welding, cutting, hardening and compacting. Research in the sphere of explosive compacting of ceramic powders presented mainly by experimental works are promising [5]. Experiments aimed to get new materials by a combination of different physical processes are carried out. For instance, in [6] a technology has been developed to get composites on the base of titanium and chromium boride by one-stage method combining self-propagating high temperature synthesis (SHS) and hot product compacting. The possibility to combine welding and explosive compacting with SHS to get layered metal-ceramic materials has been shown [7].

Many of these methods have been already introduced into production, while the potentials and perspectives of solid-phase synthesis of materials under explosive loading have not been studied thoroughly yet, so far this direction has not become a technology due to the lack of experimental data and numerical methods to describe this process correctly.

Actually, synthesis of new materials according to equilibrium phase diagrams is exhausted. The perspectives are connected with getting of metastable combinations in nonequilibrium conditions. Explosive loading gives big opportunities to create and control such conditions. High pressure and speed of substances make extreme conditions to get materials with unique properties. But substance reaction to such extreme effect can vary depending on substance characteristics. It is necessary to take into account short duration of explosive loading process (~10^{-6} s). As various processes and their stages require different time intervals for their development, not all the processes have enough time to develop within a short period of time which explosive loading is characterized by [8].

Currently the possibility of solid-phase chemical reactions in detonative regime remains underinvestigated [4, 8-10]. During shock compression, exothermic reactions with considerable heat production can take place in reaction mixtures that can result in self-sustaining detonation-like propagation of chemical reaction in the mixture. If reaction rate is high enough, the conditions for solid-phase (or gas free) detonation can be formed.

The development of mathematic models with introduction of additional parameters and equations to describe kinetics of chemical transformations taking into account initial parameters and characteristics of interactive media components as well as development of appropriate computing algorithm are required for analysis of these processes. In this connection, a mathematical model of multicomponent media was suggested and numerical code, which allows to simulate the behavior of reacting porous mixtures taking into account variations of initial components dispersion, explosion load duration and geometrical dimensions of materials under study was developed [11-14].

The following dependencies have been obtained: chemical transformation efficiency in reacting mixture; compacting level in inert mixture; relation between geometrical dimensions of recovery ampoules after loading on the layer thickness of explosive in both the axial and radial directions [15,16]. This approach, along with available experimental data, expands application area of the designed models and provides the possibility to get reliable information on reacting medium behavior including mechanisms and kinetics of physical-chemical transformations and ways of formation of new states of the substances.

This work is aimed to study the processes of materials synthesis in a solid-phase regime and to discover the reasons of destruction of cylinder recovery ampoules after explosive loading. Investigations are characterized by complex approach using experiments and numerical simulation.

2. Experimental results

2.1. Explosive loading of aluminum and sulfur mixture (Al/S)

Mixture of aluminum powder ASD4 grade PAP2 (flakes with sizes 20 μm, thickness several microns) and sulfur powder [11, 13] was used as a loading material for experiments. The powders were mixed in a planetary mill AGO-2U in mass proportion 35/65 (Al/S), which corresponds to stoichiometry of aluminum sulfide formation Al_2S_3. After that the powders were compressed into eight tablets with diameter 8 mm each and porosity 0,393±0,005. The tablets were placed in a steel cylinder ampoule with external diameter 20 mm, internal diameter 14 mm, length 95 mm. Both ampoule ends were capped.

Explosive load parameters were chosen close to the ones usually used for explosive compaction of inert porous fillers in cylinder ampoules [17, 18]. During dynamic compacting pressure behind the shock wave front should exceed 2 HV, where HV – Vickers hardness of the material pressed [17]. In the present case ampoule was loaded by ammonite 6LH with addition of NaCl in mass proportion 1/1. Explosive density is 1.2 g/cm^3. External diameter of explosive was equal to 50 mm that slightly exceeds the critical one to avoid misfire. Measured detonation speed was 2.8 km/s.

During the loading the ampoule was broken (fig.1).

The ampoule first broke at the bottom part, then it broke along the full length. After the experiment many crystallized drops with sizes up to 4 mm were found on the ampoule internal surface. X-ray phase analysis of the material collected from the ampoule showed that this was aluminum sulfide (the phases alpha and omega).

2.2. Explosive loading of aluminum and fluoroplastic mixture (Al/Tf)

In this series of experiments the mixture of aluminum and fluoroplast (Al/Tf) put in cylinder ampoule was used as loading material. The powders were mixed in a planetary mill AGO-2U in mass proportion 30/70 (Al/Tf), which corresponds to stoichiometry of aluminum fluoride formation AlF_3. Besides, the initial porosity of loaded mixture components was varied in experiments. The initial porosity was 0.5 in the first case and 0.045 in the second case. Ampoules identical to above mentioned ones were used. The ampoule was loaded by ammonite 6LH. The external charge diameter of explosive was equal to 45 mm. The explosive density amounted to 1.0 g/cm^3, detonation speed to 3.85 km/s.

The first ampoule was broken into coarse fragments (fig. 2).

The second ampoule cracked throughout the whole length (fig. 3).

Fig. 1. Ampoule after explosive loading.

Fig. 2. Fragments of the ampoule after explosive loading.

Fig. 3. The ampoule and remained layer of the sample from the top of the ampoule after explosive loading.

Only the upper sample layer of 5 mm has been preserved, which is presented on the right of the figure 3 including a scaled-up image. According to X-ray diffraction analysis, there are no traces of reaction in this part of the sample. Numerical computations based on multicomponent media model have been carried out to find out the reasons of the ampoule break.

3. Numerical results

3.1 Mathematical description of multicomponent media behaviour and physico-chemical transformations in them in the conditions of explosive loading

Equation system to describe unstable adiabatic motion of each component in some fixed in space volume of a solid compressible mixture V, limited by surface S, which takes into account appropriate exchange of mass, momentum and energy between the components inside the volume V, as well as generation and evolution of microdamages, consists of equations of continuity, motion, energy, the rate of change in the specific volume of microdamages [11-16].

In a heterogeneous mixture each component takes only a part of mixture volume ($V_1 + V_2 + ... + V_N = V$). Hence, in the theory of multicomponent mixtures a value α_i ($i = 1,2,\ldots,N$) is used to characterize the part of each element in mixture volume: $\alpha_1 + \alpha_2 + \ldots + \alpha_N = 1$, ($\alpha_i \geq 0$), $\alpha_i = \rho_i^*/\rho_i$, where ρ_i^* is reduced density (the mass of the i-th component in one unit of medium volume) [19].

Also the theory of multicomponent mixtures uses the quantities β_i ($i = 1,2,\ldots,N$) to characterize the mass concentration of each mixture component: $\beta_1 + \beta_2 + \ldots + \beta_N = 1$, ($\beta_i \geq 0$), $\beta_i = m_i/m$, where m_i is the mass of i-th component, m is mixture mass [19].

Chemical reaction in a compressed porous multicomponent mixture is described by a phenomenological model of irreversible chemical transformations P based on zero-order kinetics [19-23]:

$$J_{ji} = \frac{d\eta}{dt} = \begin{cases} 0, & \text{if } \eta = 1 \text{ or } (T_i < T_\eta \text{ and } P < P_\eta) \\ f(P_\eta), & \text{if } \eta < 1 \text{ and } (T_i \geq T_\eta \text{ or } P \geq P_\eta) \end{cases}$$

$$f(P_\eta) = \begin{cases} K_0, & \text{if } P < P_\eta \\ K_p K_0, & \text{if } P \geq P_\eta, \end{cases}$$

where T_i is the temperature, P is the consistent components pressure, T_η, P_η, K_p, K_0 are the constants, η is the degree of transformation of the substance.

To account destructions in multicomponent media the model of damaged medium characterized by microcavities (pores, cracks) is applied.

The total volume of each component W consists of a non-damaged part with volume W_s and density ρ_s and microcavities (pores) with volume W_f, where the material density is supposed to be zero. Average density of a damaged component relates to the introduced parameters as $\rho = \rho_s (W_s/W)$. Component damage level is characterized by a microdamage specific volume $V_f = W_f (W^*\rho)$. All the microcavities are supposed to be located inside the components and not to be connected with the outside surface, and damaged medium model is used independently for each component.

Material destruction simulation is carried out with a help of kinetic destruction model of active type which determines pores growth or collapse resulting in permanent changes of material properties and stress relaxation.

Pressure in undamaged mixture component is a function of specific volume, specific internal energy, it is calculated by Mie-Gruneisen equation of state within the whole range of loading conditions.

While studying the medium deformation within the frames of multicomponent model it is necessary to take into account the state and reaction of each component and, unlike one-component mixture [21-23], not only displacement of external boundaries of the assigned volume but displacement of components inside the assigned mixture volume. That is why the theory of multicomponent medium motion should take into account the conditions of compatible deformation of components, which take into account not only physical properties of the components but generally their structure as well [19]. The present work studies the propagation of shock waves resulting in phase transition in solid mixture components, which in its turn leads to an increased with pressure but limited strength of material, and at high pressures the properties of the solid to some extent become similar to the properties of liquids. Apart from this, the compressibilities, densities and specific heats of the solids are fairly similar that reduces the effects of pressure differences in the components. In this case, the equilibrium state which the mixture every time comes to, is determined first of all by a balance of the pressures of the components, which in their turn are determined by the specific volumes and specific internal energies of the mixture components. The above mentioned suggestions allow the equal pressures to be used during the interaction of components as a condition for joint deformation of components in the mixture:

$$P = P_1(V_1, E_1) = P_2(V_2, E_2) = \ldots = P_N(V_N, E_N).$$

3.2 Cylinder ampoule explosive loading simulation

The task of explosive loading of cylinder ampoule with porous mixture Al/S was studied numerically in an axisymmetrical formulation. The temperature $T_\eta = 933$ K (aluminum melting point) and pressure $P_\eta = 0.9$ GPa [16] were chosen as criteria for the initiation of chemical reaction. The rate of chemical transformation K_0 was set equal to 260.8 GJ/(kg*s), and $K_p = 20.0$. Numeric calculations have been carried out by research software based on finite elements method modified for the purposes of high-speed loading simulation [24,25].

Figure 4 shows the distribution of pressure profiles in the mixture Al/S placed into a cylinder ampoule at different moments of time characterizing dynamics of shock wave process during the explosive synthesis of aluminum sulfide.

Shock wave propagates along the sample under the action of explosion products. In this process, the wave front in the filler remains behind the front in the ampoule body due to time required for pores collapse in the filler. Pressure at the sample bottom is considerably increased (fig. 4c) and exceeds 6 GPa when the shock wave is reflected from the ampoule bottom as a compressive wave.

Taking into account the analysis of experimental and numerical results it is possible to suppose the following dynamics of chemical transformations process development in exothermal mixtures at explosive loading. In the upper and central parts of the ampoule the reaction is initiated in the transmitting shock wave. When the shock wave is reflected from the bottom ampoule lid as a compressive wave, the pressure in the bottom is increased sharply that results in a rise of the chemical transformations rate in the lower part of mixture. High rate of heat release in the course of chemical reaction in the lower ampoule part causes gas phase formation, which in its turn brings to pressure increase in this part and ampoule destruction, and it is the lower ampoule part where the destruction process is initiated.

4. Conclusions

Peculiarities of solid-phase synthesis in aluminum-sulfur and aluminum-fluoroplast mixtures under explosive loading at variations of initial porosity of initial components have been studied in experiments and numerically. The results of experiments showed that cylinder ampoules are destructed in explosive loading conditions when mixture used as a filler is capable of very fast exothermal reactions. As a result of numeric studies it was found that pressure is increased sharply at the ampoule lower part when shock wave is reflected from the bottom ampoule lid as a compressive wave that is followed by a growth of the chemical transformation rate. High rate of heat release in the course of reaction in the lower ampoule part resulted in gas phase formation that brings to a further pressure increase and becomes the reason of ampoules destruction.

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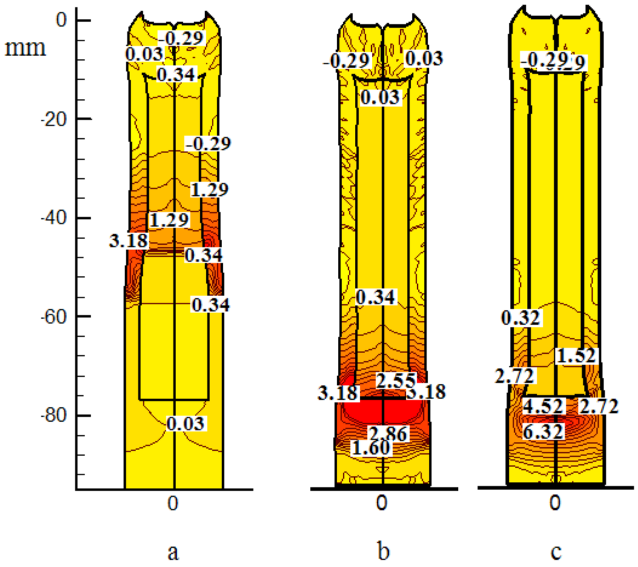

Fig. 4. Distribution of pressure profiles (GPa) in a mixture of reacting porous components Al/S in an axial section of the ampoule at different moments of time: (a) 20 μs, (b) 31 μs, (c) 32 μs.

References

1. Physics of an explosion, Ed. by L.P. Orlenko. Moscow, Fizmatlit. (2002) 1488 p. (in Russian) [Физика взрыва / Под ред. Л.П. Орленко. М., Физматлит. 2002. 1488 с]
2. V.V. Selivanov, S.A. Novikov, I.F. Kobylkin. Explosive technologies. Moscow, BMSTU. (2008) 648 p. (in Russian) [В.В. Селиванов, С.А. Новиков, И.Ф. Кобылкин. Взрывные технологии. М., МГТУ им. Баумана. 2008. 648 с.]
3. V.V. Danilenko. Explosion: physics, engineering, technology. Moscow, Energoatomizdat. (2010). 784 p. (in Russian) [В.В. Даниленко. Взрыв: физика, техника, технология. М., Энергоатомиздат. 2010. 784 с.]
4. S.A. Zelepugin, A.Yu. Dolgoborodov, O.V. Ivanova, A.S. Zelepugin. Shock wave synthesis in solid mixtures: Monograph. Tomsk, Institute of Atmospheric Optics SB RAS. (2012) 230 p. (in Russian) [С.А. Зелепугин, А.Ю. Долгобородов, О.В. Иванова, А.С. Зелепугин. Ударно-волновой синтез в твердых смесях. Монография. Томск, ИОА СО РАН, 2012. 230 с.]
5. L.B. Pervukhin, M.I. Alymov, I.V. Saikov, R.D. Kapustin, O.L. Pervukhina, E.V. Petrov. Letters on materials. **5**(1), 57 (2015). (in Russian) [Л.Б. Первухин, М.И. Алымов, И.В. Сайков, Р.Д. Капустин, О.Л. Первухина, Е.В. Петров. Письма о материалах. **5**(1), 57 (2015)]
6. V.A. Shcherbakov, A.N. Gryadunov, N.V. Sachkova, A.V. Samokhin. Letters on materials. **5**(1), 20 (2015). (in Russian) [В.А. Щербаков, А.Н. Грядунов, Н.В. Сачкова, А.В. Самохин. Письма о материалах. **5**(1), 20 (2015)]
7. M.I. Alymov, L.B. Pervukhin, A.S. Rogachev, O.L. Pervukhina, I.V. Saikov. Letters on materials. **4**(3), 153 (2014). (in Russian) [М.И. Алымов, Л.Б. Первухин, А.С. Рогачев, О.Л. Первухина, И.В. Сайков. Письма о материалах. **4**(3), 153 (2014).]
8. A.Yu. Dolgoborodov, M.N. Makhov, A.N. Streletskij, I.V. Kolbanev, M.F. Gogulya, V.E. Fortov. Khimicheskaya Fizika. **23**(9) 85 (2004). [А.Ю. Долгобородов, М.Н. Махов, А.Н. Стрелецкий, И.В. Колбанёв, М.Ф. Гогуля, В.Е. Фортов. Химическая физика. **23**(9) 85 (2004)].
9. S.A. Rashkovskii, A.Y. Dolgoborodov. Technical Physics Letters. 41(6) 575 (2015). Doi: 10.1134/S1063785015060279 [С.А. Рашковский, А.Ю. Долгобородов. Письма в ЖТФ. 41(12) 24 (2015)].
10. A.Yu. Dolgoborodov. Combustion, Explosion and Shock Waves. **51**(1) 86 (2015). Doi: 10.1134/S0010508215010098 [А.Ю. Долгобородов. Физика горения и взрыва. **51**(1), 102 (2015).]
11. O. Ivanova, S. Zelepugin, A. Yunoshev, V. Silvestrov. J. Energ. Mater. **28**(1) 303 (2010). Doi: 10.1080/07370652.2010.505940
12. O.V. Ivanova, S.A. Zelepugin, A.S. Yunoshev, V.V. Sil'vestrov. Eurasian Chemico-Technological Journal. **16**(1) 3 (2014).
13. S.A. Zelepugin, O.V. Ivanova, A.S. Yunoshev, V.V. Sil'vestrov. Doklady Physical Chemistry **434**(2) 172 (2010). Doi: 10.1134/S0012501610100052 [С.А. Зелепугин, О.В. Иванова, А.С. Юношев, В.В. Сильвестров. Доклады Академии наук. **434**(5) 643 (2010)].

14. O.V. Ivanova, S.A. Zelepugin. Russian Physics Journal **55**(9-3) 46 (2012). (in Russian) [О.В. Иванова, С.А. Зелепугин. Известия вузов. Физика. **55**(9-3) 46 (2012)].
15. O.V. Ivanova, S.A. Zelepugin. Applied Mechanics and Materials. 770. 174 (2015). Doi: 10.4028/www.scientific.net/AMM.770.174.
16. O.V. Ivanova, S.A. Zelepugin. International Journal of Self-Propagating High-Temperature Synthesis. **23**(4) 192 (2014). Doi: 10.3103/S1061386214040025.
17. O.L. Pervukhina. Izvestiya VolgGTU. (9) 60 (2006). (in Russian) [О.Л. Первухина. Известия ВолгГТУ. (9) 60 (2006)].
18. V.A. Corelski, S.A. Zelepugin. International Journal of Self-Propagating High-Temperature Synthesis. **8**(1) 43 (1999).
19. R.I. Nigmatulin. Dynamics of multiphase media. 1. New York, Hemisphere. 507 p. (1991). [Р.И. Нигматулин. Динамика многофазных сред. Ч.1. М., Наука. 1987. 464 с.]
20. A.S. Shteinberg, A.A. Berlin, A.A. Denisaev, A.S. Mukasyan. International Journal of Self-Propagating High-Temperature Synthesis. **20**(4) 259 (2011). Doi: 10.3103/S1061386211040030.
21. S.A. Zelepugin, V.B. Nikulichev. Combustion, Explosion and Shock Waves. **36**(6) 845 (2000). [С.А. Зелепугин, В.Б. Никуличев. Физика горения и взрыва. **36**(6) 186 (2000)].
22. S.A. Zelepugin, V.B. Nikulichev, O.V. Ivanova, A.S. Zelepugin. Khimicheskaya Fizika. **24**(10) 76 (2005). (in Russian). [С.А. Зелепугин, В.Б. Никуличев, О.В. Иванова, А.С. Зелепугин. Химическая физика. **24**(10) 76 (2005)].
23. S.A. Zelepugin, V.B. Nikulichev, O.V. Ivanova. AIP Conf. Proc. **845** 1177 (2006). Doi: 10.1063/1.2263534.
24. G.R. Johnson. International Journal of Impact Engineering. **38**(6) 456 (2011). Doi:10.1016/j.ijimpeng.2010.10.017.
25. V.A. Gorelski, S.A. Zelepugin, A.Yu. Smolin. Computational Mathematics and Mathematical Physics. **37**(6) 722 (1997). [В.А. Горельский, С.А. Зелепугин, А.Ю. Смолин. Журнал вычислительной математики и математической физики. **37**(6) 742 (1997)].