Fracture of cylindrical ampoules during solid phase synthesis under explosive loading

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Abstract. The paper presents the numerical and experimental study of the behavior of porous reactive mixtures (Al-S and Al-Tf) under explosive loading using a multicomponent medium model. The initial dispersion of the reactive mixture is varied to evaluate its contribution to the fracture of the cylindrical ampoule. The results have shown that the high rate of heat release during the chemical reaction in the bottom part of the ampoule and a sharp increase in pressure lead to the formation of the gas phase, which causes the ampoule fracture. It is found that the higher the initial dispersion of the mixture components is, the more intensive the ampoule failure is.

1. Introduction

By now, the solid-phase synthesis of materials placed in cylindrical ampoules under explosive loading has been insufficiently studied and, therefore, it is of great interest for further research. This direction has not yet reached the level of technology due to the lack of experimental data and numerical techniques correctly describing the process. During shock-wave compression, there is a significant energy release due to exothermic reactions in reacting mixtures, which, on the one hand, can lead to a self-sustaining propagation of chemical reactions in mixtures and, on the other hand, to the failure of ampoules, as well as the complete fracture [1-3]. It is necessary to mention the investigations directed to the study of solid-phase chemical reactions in the detonation regime [4-6]. If the reaction rate is high enough, then it can lead to the conditions which will allow the solid phase (or gasless) detonation to occur.

For the analysis of these processes, it is necessary to develop mathematical models with the introduction of additional parameters and equations describing the kinetics of chemical reactions and considering the initial parameters and characteristics of reactive components, as well as the appropriate computational algorithms. The developed numerical models in combination with the available experimental data will provide the possibility to receive reliable information on the behavior of reactive media, including the mechanisms and kinetics of physical and chemical transformations and ways to form the new states of substances.

The goal of this work is to study the problems and peculiarities which occur during the solid-phase synthesis in the cylindrical ampoules subjected to explosive loading.
2. Formulation of the problem

The governing equations of continuity, momentum, and energy for the nonstationary adiabatic motion of each component in some fixed volume V of a compressible mixture bound by surface S can be written in the following form [7]:

\[
\frac{\partial}{\partial t}(\alpha_i \rho_i) + \nabla \alpha_i \rho_i \mathbf{v}_i = 0, \quad (i = 1, 2, \ldots, N),
\]

\[
\alpha_i \rho_i \frac{d \mathbf{v}_i}{dt} = \nabla \sigma_i + \alpha_i \sum_{j=1}^{N} \alpha_j \mathbf{R}_{ij}, \quad (i = 1, 2, \ldots, N),
\]

\[
\alpha_i \rho_i \frac{d E_i}{dt} = \sigma_i \varepsilon_i + \alpha_i \sum_{j=1}^{N} \alpha_j F_{ij}, \quad (i = 1, 2, \ldots, N),
\]

where

\[
\frac{d \varepsilon_i}{dt} = \frac{\partial \sigma_i}{\partial t} + \mathbf{v}_i \cdot \nabla \mathbf{v}_i.
\]

Here, \( t \) is the time, \( \rho_i \) is the density of the \( i \)-th component equal to the mass of the \( i \)-th component per unit volume of the \( i \)-th component, \( \mathbf{v}_i \) is the velocity vector, \( E_i \) is the internal specific energy, \( \varepsilon_i \) is the strain rate tensor, \( \sigma_i = -P_i \delta_i + S_i \) is the stress tensor, \( P_i \) is pressure, \( S_i \) is the stress deviator, \( R_{ij} \) is the intensity of the momentum exchange between the \( j \)-th and \( i \)-th components, \( F_{ij} \) is the intensity of the energy exchange between the \( j \)-th and \( i \)-th components, \( N \) is the number of components.

Volume fractions of the mixture occupied by each component are given by:

\[
\alpha_1 + \alpha_2 + \ldots + \alpha_N = 1, \quad (\alpha_i \geq 0), \quad \alpha_i = \rho_i^{*}/\rho_i,
\]

where \( \rho_i^{*} \) is the reduced density (mass of the \( i \)-th component per unit volume).

The mixture components interact with each other by exchanging momentum \( R_{ij} \), energy \( F_{ij} \), and mass \( J_{ij} \) in the presence of chemical reactions within the framework of the multicomponent medium model.

In modeling chemical reactions under shock wave loading, we use the zeroth-order kinetic relation characterized by a constant rate of chemical transformations [1, 2, 8]:

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

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

where \( T_i \) is the temperature, \( P \) is the matched pressure of components, and \( T_\eta, P_\eta, K_P, K_0 \) are the constants, \( \eta \) is the conversion degree.

Studying the deformation of multicomponent media, it is necessary to take into account the state and response of each component, and, in contrast to a homogeneous mixture, not only the displacement of the external boundaries of the selected volume, but also the displacement of components in the selected volume of the mixture. Here, we consider the equal pressures of the components to be a condition for joint deformation of components during the interaction in the mixture, which determines the volume concentrations of the components:

\[
P = P_i(V_i, E_i) = P_j(V_j, E_j) = \ldots = P_N(V_N, E_N).
\]
3. Experimental results

To study experimentally the explosive synthesis, the powder mixture of aluminum and fluoroplastic (Al/Tf) was used as a loaded material that was placed in a cylindrical ampoule. The powders were mixed in an AGO-2U planetary mill. The mass fractions of components were 30/70 (Al/Tf), which was close to the stoichiometry of the aluminum fluoride (AlF$_3$) formation. The initial porosity of the loaded mixture components was varied in the experiments. In the first case, the initial porosity of the mixture components was 0.5 (the ratio between the volume of pores and the total volume) and in the second case, the initial porosity was 0.045. The pressed mixture was placed in a steel cylindrical ampoule (Figure 1). The height of the cylindrical ampoule was 95 mm, the external diameter was 20 mm. The thickness of the lateral wall of the ampoule was 3 mm. The ampoule was closed with lids 10 mm in thickness.

![Figure 1. The ampoule before the experiment.](image1)

Figure 2 shows the structure of the Al/Tf sample surface before the experiment.

![Figure 2. The structure of the Al/Tf sample surface.](image2)

The ampoule was loaded by an explosive (ammonite 6GV). The external diameter of the explosive was 45 mm. The density of the explosive was 1.0 g/cm$^3$, and the detonation velocity was 3.85 km/s.

The first ampoule was broken into large fragments (Figure 3).
The second ampoule was damaged along the whole length of the material. Only the upper layer of the sample, which is 5 mm in thickness, was saved after explosive loading. There are no traces of the reaction in this part of the sample according to the X-ray diffraction analysis.

4. Numerical results

To study the explosive synthesis, the numerical computations were carried out by using the model of a multicomponent medium and the finite element method [9-10]. We numerically solved an axisymmetric problem of a cylindrical steel ampoule containing a porous Al–S mixture under explosive loading. The mass fractions of the components were as follows: Al – 0.35; S – 0.65, which corresponded to the stoichiometry of the aluminum sulfide (Al$_2$S$_3$) formation after initiation of a chemical reaction.

The height and the diameter of the cylindrical mixture sample were 65 mm and 14 mm, respectively. The thickness of the lateral wall of the ampoule was $\Delta h = 3$ mm; the thickness of the lids was 20 mm. The external diameter of the ampoule was 20 mm. In the computations, the action of the explosive surrounding of the ampoule was modeled as the pressure of the explosion products acting on the horizontal and vertical layers of the ampoule during propagation of the detonation front. The detonation velocity was 3.3 km/s and corresponded to the experimental evaluations.

In the work, we assume that a chemical reaction initiation criterion (pressure criterion) and a reaction rate depend on the initial value of the mixture dispersion. For the aluminum and sulfur powders with low dispersion, we chose the reaction initiation criteria in terms of temperature, $T_\eta = 717$ K (sulfur evaporation temperature), and pressure, $P_\eta = 2.0$ GPa. Reaction rate $K_0$ was taken to be 301.6 GJ/(kg s), and $K_p = 2.0$. For the aluminum and sulfur powders with high dispersion, we chose $T_\eta = 717$ K and $P_\eta = 1.0$ GPa.

Figure 4 demonstrates the distribution of pressure profiles in the Al–S low dispersion mixture placed in a cylindrical ampoule at different moments of time, describing the dynamics of the shock wave process during the explosive synthesis of aluminum sulfide.
Figure 4. Distribution of pressure profiles (GPa) for the low dispersion reactive mixture (Al-S) in an axial section of the ampoule at different moments of time: (a) 10 μs, (b) 26 μs, (c) 27 μs, and (d) 28 μs.

A shock wave caused by the explosion products propagates along the sample. It should be noted that the wave front propagating in the sample is behind the front propagating along the ampoule due to collapsing the pores in the sample. When the shock wave is reflected from the bottom of the ampoule in the form of a compression wave, the pressure at the bottom of the sample increases by several times (Figure 4b) and is greater than 18 GPa.

Figure 5 demonstrates the distribution of pressure profiles in the Al–S high dispersion mixture placed in a cylindrical ampoule at different moments of time, describing the dynamics of the shock wave process during the explosive synthesis of aluminum sulfide.

Figure 5. Distribution of pressure profiles (GPa) for the high dispersion reactive mixture (Al-S) in an axial section of the ampoule at different moments of time: (a) 10 μs, (b) 26 μs, (c) 27 μs, and (d) 28 μs.

During the propagation of the shock wave, the higher pressure is observed in the high dispersion sample compared to the low dispersion one, which indicates a faster chemical reaction and its contribution to the increase in the pressure and the temperature. The shock wave reflected from the bottom of the ampoule encounters the shock compression wave propagating in the sample, which leads to a sharp increase in the pressure at the bottom of the sample (Figure 5b) and is greater than 18 GPa. After the encounter, the shock waves start propagating in opposite directions (Figure 5c). The
high residual pressures are observed in the sample and reach more than 4 GPa for the high dispersion mixture (Figure 5d) and 2 GPa — for the low dispersion one (Figure 4d).

5. Conclusion

Based on the analysis of numerical and experimental results, we can assume the following dynamics of the shock wave propagation during the solid-phase synthesis in cylindrical ampoules subjected to explosive loading. During the propagation of the shock wave, an exothermic reaction is initiated in the sample and the higher pressures are observed for the high dispersion sample compared to the low dispersion one. The shock wave, reflected from the bottom of the ampoule in the form of a compression wave, encounters the shock wave propagating in the sample, which leads to a sharp increase in the pressure and the rate of chemical reactions at the bottom of the sample. The high rate of heat release during the chemical reaction in the bottom part of the ampoule and a sharp increase in pressure lead to the formation of the gas phase, which causes the ampoule fracture.

The initial dispersion of the reactive mixture was varied to evaluate its contribution to the fracture of the cylindrical ampoule. The experimental and numerical results allowed us to find that the higher the initial dispersion of the mixture components was, the more intensive the ampoule failure was. In some experiments, there was a complete fracture of the ampoule.

Acknowledgments

The reported study was funded by the Russian Foundation for Basic Research, according to research project No. 14-03-00666.

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