Numerical simulation for the propagation and action of shock waves during explosive synthesis

S A Zelepugin\textsuperscript{1,2}, O V Ivanova\textsuperscript{1}, A S Yunoshev\textsuperscript{3} and A S Zelepugin\textsuperscript{1,2}

\textsuperscript{1} National Research Tomsk State University, 36 Lenin Ave., 634050, Tomsk, Russia
\textsuperscript{2} Tomsk Scientific Center of the Siberian Branch of the Russian Academy of Sciences, 10/3 Akademichesky Ave., 634055, Tomsk, Russia
\textsuperscript{3} Lavrentyev Institute of Hydrodynamics of the Siberian Branch of the Russian Academy of Sciences, 15 Lavrentyev Ave., 630090, Novosibirsk, Russia

Abstract. The paper presents numerical and experimental studying the behavior of porous reactive mixtures (Al/S, Al/Tf and Al/Tf/C) under explosive loading using a multicomponent medium model. The initial dispersion of reactive mixtures is varied to evaluate its contribution to the development of chemical reactions. The results have shown that 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 and the sharp increase in pressure lead to the ampoule fracture. It is found that the higher the initial dispersion of the mixture components is, the more intensive the development of chemical reactions and the ampoule fracture are.

1. Introduction

The obtaining and use of new advanced materials is often connected with extreme conditions such as high-speed processes, high pressures and temperatures. At present, explosive technologies are used mainly in the metalworking production for molding, welding, cutting, hardening and compacting. Many of these technologies have been already introduced into production, while the potentials and perspectives of explosive loading have not been studied thoroughly yet, and so far this direction has not become a technology due to the lack of experimental data and numerical multicomponent medium models which can consider the cumulative effect of mechanical, chemical and physical processes and estimate the role of each factor. During explosive loading, 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 that the initiation and development of shock-induced reactions are determined not only by the temperatures reached but also by the dispersion of initial components [4]. In addition, the initial dispersion of components can contribute to the fracture of the cylindrical ampoule. For the analysis of these processes, it is necessary to develop mathematical models which in combination with available experimental data will provide reliable information on the behavior of materials, including the mechanisms and kinetics of physical and chemical transformations and ways to form the new states of substances.

In the work, the aluminum-sulfur, aluminum-fluoroplast and aluminum-fluoroplast-carbon mixtures placed in a cylindrical ampoule were studied under explosive loading on the basis of a mathematical multicomponent medium model considering the dispersion of the initial components.
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 [5]:

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

\[
\alpha_i \rho_i \frac{d}{dt} \mathbf{v}_i = \nabla \sigma_i + \frac{\partial}{\partial \mathbf{x}} \mathbf{R}_i, \quad (i = 1, 2, \ldots, N),
\]

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

where \( \frac{d}{dt} = \frac{\partial}{\partial t} + \mathbf{v}_i \cdot \nabla \).

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, \( \sigma_i = -P \mathbf{\hat{b}}_i + S_i \) is the stress tensor, \( P_i \) is pressure, \( S_i \) is the stress deviator, \( R_\mu \) 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 \( \alpha_1 + \alpha_2 + \ldots + \alpha_N = 1 \), \( \alpha_i \geq 0 \), \( \alpha_i = \rho_i^* / \rho_i \), where \( \rho_i^* \) is the reduced density (mass of the \( i \)-th component per unit volume).

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, 6]:

\[
J_{ji} = \frac{dn_i}{dt} = \begin{cases} 
0, & \text{if } \eta = 1 \text{ or } (T_i < T_\eta \text{ and } P < P_\eta), \\
 \{f(P_\eta), & \text{if } \eta < 1 \text{ and } (T_i \geq T_\eta \text{ or } P \geq P_\eta), \\
 \{K_0, & \text{if } P < P_\eta, \\
 \{K_\eta K_0, & \text{if } P \geq P_\eta, 
\end{cases}
\]

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

Evolution of porosity in the material (compression and growth of pores) is simulated using a kinetic model of the active type, which determines changes in the specific volume of pores influencing on the material properties and causing stress relaxation [7]:

\[
\frac{dV_{\text{p}}}{dt} = \begin{cases} 
0 \text{ for } |P_\text{p}| \leq P_\text{p}^* \text{ or } (P_\text{p} > P_\text{p}^* \text{ and } V_{\text{p}} = 0), \\
-\text{sign}(P_\text{p}) \cdot K_\eta \cdot |P_\text{p}| \cdot \left( P_\text{p} > P_\text{p}^* \right) \cdot (V_{\text{p}} - V_{\text{p}}^*), \text{ for } P_\text{p} < -P_\text{p}^* \text{ or } (P_\text{p} > P_\text{p}^* \text{ and } V_{\text{p}} > 0),
\end{cases}
\]

where \( P_\text{p}^* = P_\text{p}_i V_{\text{p}} (V_{\text{p}} + V_{\text{p}}^*_i) \), \( P_\text{p}_i \) is the pressure in the solid (undamaged) part of the \( i \)-th component in the mixture, \( V_{\text{p}}^*_i, V_{\text{p}}^*_2, P_\text{p}_2, K_\eta \) are the experimentally determined constants of the material.

The equal pressures of the components are considered to be a condition for joint deformation of components during the interaction in the mixture, which determines the volume concentrations of the components:

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

3.1. Explosive loading of the aluminium-sulfur mixture

To study experimentally the explosive synthesis, the powder mixture of aluminum and sulfur (Al/S) 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 65/35 (Al/S), which was close to the stoichiometry of the aluminum sulfide (Al₂S₃) formation. The initial porosity of the mixture components to be loaded was 0.4 in the experiments. The pressed mixture was placed in a steel cylindrical ampoule. 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 and 20 mm in sickness. The ampoule was loaded by an explosive (ammonite 6GV) with addition of NaCl in mass proportion 1/1. The external diameter of the explosive was 50 mm. The density of the explosive was 1.0 g/cm³, and the detonation velocity was 2.8 km/s.

During loading the ampoule was broken (Figure 1). The ampoule was first broken at the bottom part, and then it was broken 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.

![Figure 1](image1.png)

**Figure 1.** The ampoule after the experiment.

3.2. Explosive loading of the aluminium-fluoroplastic mixture

In the following experiment (Figure 2), the powder mixture of aluminum and fluoroplastic (Al/Tf) was used as a loaded material. 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 (AlF₃) formation.

![Figure 2](image2.png)

**Figure 2.** Fragments of the ampoule after explosive loading.

The initial porosity of the loaded mixture was 0.5 (the ratio between the volume of pores and the total volume). The pressed mixture was placed in a steel cylindrical ampoule. The height of the cylindrical
3.3. Explosive loading of the aluminium-fluoroplastic-carbon mixture

In the following experiment, a cylindrical ampoule was loaded, which contained a three-component mixture of aluminum, fluoroplastic and carbon. Carbon was added to the mixture of aluminum and fluoroplastic in a proportion of 2/3, where graphite had two mass fractions and the mixture of Al/Tf had one mass fraction. The components were mixed in an AGO-2U planetary mill, and then the pressed mixture was placed in a steel cylindrical ampoule. The height of the cylindrical ampoule was 81 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 21 and 13 mm in sickness. The measured porosity was 0.25±0.005. 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/cm³, and the detonation velocity was 3.85 km/s. After experiment the ampoule was uniformly compressed from top to bottom. To determine the phase composition of the sample, the ampoule was cut into 5 parts (Figure 3). Detonation wave propagated from the left sample to the right one. The difference is clearly observed in the central and peripheral part of the sample. The reaction was complete only at the center of the sample. Carbon and AlF₃ are present among the products of the reaction.

![Figure 3. Ampoule cut into 5 parts after explosive loading.](image)

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 [7-9]. 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 (Al₂S₃) 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 Δh = 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, Tₜ = 933 K (aluminum melting temperature), and pressure, Pₜ = 1.6 GPa. Reaction rate K₀ was taken to be 301.6 GJ/(kg s), and...
K_0 = 2.0. For the aluminum and sulfur powders with high dispersion, we chose T_0 = 933 K and P_0 = 1.0 GPa.

Figure 4 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 4. 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) 15 μs, (b) 27 μs, (c) 28 μs, and (d) 30 μs.

A shock wave caused by the explosion products propagates along 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. At the same time, high pressures are observed in the sample for some time (Figure 4d), while the shock wave propagated along the ampoule already stopped acting.

Figure 5 demonstrates the explosive synthesis of aluminum sulfide in the Al/S high dispersion mixture placed in a cylindrical ampoule at different moments of time.

Figure 5. Explosive synthesis of aluminum sulfide in the high dispersion reactive mixture (Al/S) in an axial section of the ampoule at different moments of time: (a) 15 μs, (b) 27 μs, (c) 28 μs, and (d) 30 μs.

The results show (Figure 5) that the reaction in the high dispersion Al/S mixture was complete with the formation of aluminium sulfide (100%).
Figure 6 demonstrates the distribution of pressure profiles in the Al/S low dispersion mixture.

**Figure 6.** 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) 15 μs, (b) 27 μs, (c) 28 μs, and (d) 30 μs.

During the propagation of the shock wave, the higher pressure is maintained for a longer time in the high dispersion sample compared to the low dispersion one, which indicates the development of a faster chemical reaction and the contribution of porosity to the increase in pressure and 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 6b) and is greater than 18 GPa. After the encounter, the shock waves start propagating in opposite directions (Figure 6c). By 30 μs, high residual pressures (3.9 GPa) are observed in the high dispersion mixture (Figure 4d), while only negative residual pressures (-0.1 GPa) are observed in the low dispersion mixture (Figure 6d).

Figure 7 demonstrates the explosive synthesis of aluminum sulfide in the Al/S low dispersion mixture placed in a cylindrical ampoule at different moments of time.

**Figure 7.** Explosive synthesis of aluminum sulfide in the low dispersion reactive mixture (Al/S) in an axial section of the ampoule at different moments of time: (a) 15 μs, (b) 27 μs, (c) 28 μs, and (d) 30 μs.

The results show (Figure 7) that not all the low dispersion mixture components reacted to form aluminium sulfide.
5. Conclusion
The analysis of numerical and experimental results allows us to conclude about the contribution of the porosity and dispersion of mixtures to the dynamics of the shock wave propagation and the development of chemical reactions during the solid-phase synthesis in cylindrical ampoules subjected to explosive loading. During the propagation 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 in the low and middle part of the sample. It should be noted that the reaction in the high dispersion Al/S mixture is complete with the formation of aluminium sulfide (100%). Despite the sharp increase in pressure and temperature, not all the low dispersion mixture components react to form aluminium sulfide. The contribution of high pressures and temperatures is not sufficient for the complete reaction in the low dispersion mixture. The experimental and numerical results allowed us to find that the higher the initial dispersion and porosity of the mixture components, the more intensive the ampoule fracture. In some experiments, there was a complete fracture of the ampoule.

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