Propagation of cellular detonation in a gas suspension in the presence of a concentration gradient

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Abstract. Models for the description of detonation flows in inhomogeneous gas particle mixtures are presented. The physical and mathematical model of heterogeneous detonation of aluminum suspensions is based on approaches to the mechanics of heterogeneous media and semi-empirical laws of ignition and combustion of particles. The model is extended for suspensions that are inhomogeneous in particle concentrations. This suggests the solution of an additional equation for the spatial distribution of initial concentrations. This determines the concentration distribution from the solution of the stationary problem of the structure of the detonation wave and its agreement with empirical data on the rate of normal detonation. The model for describing detonation in mixtures of reacting gases with inert particles is based on the analysis of the state of stoichiometric compositions. The problem of the initiation and development of cellular detonation in a flat channel in the presence of a longitudinal or transverse gradient of the concentration of aluminium particles is considered. The limits of changes in concentrations from poor to super stoichiometric compositions are considered for 1 μm particles. When detonation propagates along a channel with a longitudinal positive (negative) concentration gradient, the cellular structures do not change, but the average and peak values of pressure and phase density increase (decrease). With a transverse concentration gradient, cell-like structures with a curved front, asymmetric and strongly elongated cells form in the channel. The results are consistent with studies of similar gas detonation flows.

1. Introduction
Detonation studies are related both to problems of explosion and fire safety, and to the development of detonation technologies. In detonation engines, a gaseous medium or dispersed suspension can be used as a working mixture. One way to control processes is to organize and use various kinds of heterogeneities. The study of gas and heterogeneous detonation, both theoretically and experimentally, was carried out, as a rule, for mixtures with a uniform concentration. The problems of suppression of detonation by dispersed clouds were considered in [1, 2], in [3] the features of cellular detonation in a two-fuel mixture were described. The homogeneity of the composition of gas mixtures is achieved quite quickly; nevertheless, the question of the detonation characteristics in inhomogeneous mixtures was considered in [4–7]. For two-phase media, these questions were considered in the problems of lifting, ignition, and detonation combustion of dust layers [8–10].
One of the problems is the description of the combustion of powders in the absence of an oxidizing agent. Combustion of aluminum can proceed both with the formation of oxide $\text{Al}_2\text{O}_3$ and with the formation of suboxides $\text{AlO}$, $\text{AlO}_2$, $\text{Al}_2\text{O}$. The review [11] discusses models of detonation combustion, in which the formation of suboxides is taken into account in various ways. The issues of modeling the deflagration combustion of aluminum with an oxidizer deficiency are discussed in [12]. In detonation combustion, the total heat release and detonation velocity are determined by the equilibrium composition of the detonation products. Some data of thermodynamic calculations of the equilibrium composition of aluminum detonation products are given in [13]. In [14], the dependence of the detonation velocity of aluminum suspensions in oxygen on the initial concentration of particles is presented, which will be used to construct a model of detonation combustion of aluminum in a gas suspension inhomogeneous in concentration.

2. Physical and mathematical model of detonation in non-uniform suspensions of reactive particles in gas oxidiser

Two-dimensional detonation flows are described within the framework of the mechanics of interpenetrating continua by the Euler equations for the gas phase and particles. The system is closed by the equations of state and relations describing the interphase exchange of masses, momentum and energy.

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} + \frac{\partial \rho v}{\partial y} = -(1) J$$

$$\frac{\partial \rho u}{\partial t} + \frac{\partial \left[ \rho u^2 + (2-i) p \right]}{\partial x} + \frac{\partial \rho u v}{\partial y} = -(1) (f_y - Ju_2) $$

$$\frac{\partial \rho v}{\partial t} + \frac{\partial \left[ \rho u v + (2-i) p \right]}{\partial x} + \frac{\partial \rho v^2}{\partial y} = -(1) (f_x - Ju_2) ,$$

$$\frac{\partial \rho E_i}{\partial t} + \frac{\partial \rho u E_i}{\partial x} + \frac{\partial \rho v E_i}{\partial y} = -(1) \left( q + f_y u_2 + f_x v_2 - JE_2 \right) ,$$

$$p = \rho_1 RT_i, \quad E_i = \frac{u_i^2}{2} + c_i T_i + (i-1)Q .$$

Here $p$ is pressure, $\rho_i, u_i, v_i, T_i, E_i, c_i$ are mean density, longitudinal and transversal velocity components, temperature, total energy per mass unit and heat capacity of the $i$-th phase (1, 2 indicate gas and particles, respectively). Mass concentrations of gas and particles are defined as $\xi_i = \rho_i / \rho$, $\rho = \sum_i \rho_i , \rho_i = \rho m_i, m_i$ is the volume concentration of the $i$-th phase.

Closure relations for the exchange of mass, momentum, and energy of micro- and nano-sized particles are presented in [15]. Particle combustion is described in the framework of the simplest model of one-stage kinetics, taking into account incomplete particle burning (the presence of residual concentration of unburned particles) and the transitional regime from diffusion-limited to kinetically-controlled combustion for particles with a diameter of less than 1 micron [16]. The dependence of the characteristic burning time of aluminum particles in the submicron range from 135 nm to 1 μm is taken as:

$$J = \frac{\rho}{\tau_{\xi}} (\xi - \xi_k) \exp(-E_a / RT_i) \quad \text{at} \quad T > T_{ign}, \quad \xi > \xi_k, \quad J = 0, \quad \text{at} \quad T < T_{ign} \quad \text{or} \quad \xi \leq \xi_k .$$

$$\tau_{\xi} = \tau_{0}(d / d_0)^{0.3} \exp(E_a / RT_i)(p / p_i)^{-m}$$

(2)
where \( m = -0.25 \ln d [\mu m] \), \( E_c = 0.5[E_{\text{micro}}(2 + \ln d [\mu m] - E_{\text{nano}} \ln d [\mu m]) \), and \( E_{\text{micro}} \) is the activation energy for micron particles (32 kJ/mol [11]), \( E_{\text{nano}} \) is the activation energy for nanoparticles (60 kJ/mol [17]), \( p \) is pressure, and \( d \) is the particle diameter.

The heat release of chemical reactions \( Q \) in a homogeneous suspension can be determined from empirical data or thermodynamic calculations of the normal detonation velocity, which for the stoichiometric composition of a mixture of aluminum and oxygen is about 1.6 km/s. The relationship between the detonation velocity and integral heat release is determined from the problem of the structure of a stationary wave. The solution corresponding to the Chapman-Jouguet regime has either an internal sound point (in terms of the frozen speed of sound) or a final equilibrium state, sound in terms of the equilibrium speed of sound [17]. In the pre-stoichiometric interval, according to experimental data [18], it is assumed that \( \xi_0 = 0.1 \xi_0 \). Approximating dependence of the parameter \( Q \) on the initial concentration in the form \( Q(\xi_0) = 20.3 \exp(-3.17 \xi_0) \) provides agreement with the data of [14] at \( 0.2 < \xi_0 < 0.55 \). For high load conditions (super stoichiometry), an additional determining parameter is \( \xi_k \). The values of the residual concentration at a value of \( Q=3.52 \) MJ/kg, corresponding to stoichiometry, are approximated by a function \( \xi_k = 0.1 \xi_0 + 3.3(\xi_0 - 0.55)(\xi_0 - 0.4) \), approaching 1 at \( \xi_0 \to 1 \). In an inhomogeneous suspension, the initial concentration distribution is determined by a function \( \xi_0(x, y) \). Accordingly, the thermal effect of the reaction and the fraction of unburnt particles at each point in space vary and depend on what was the initial concentration of the volume of particles that arrived at this point in space. To do this, the function \( \xi(x, y) \) is introduced as follows. The initial distribution corresponds to the concentration distribution \( \xi_0(x, y) = \xi_0(x, y) \), and as the mixture moves, it is determined from the equation \( D \xi / Dt = 0 \). With (1), it is equivalent to

\[
\frac{\partial \rho_x \xi}{\partial t} + \frac{\partial \rho_x u_x \xi}{\partial x} + \frac{\partial \rho_y v_y \xi}{\partial y} = -J \xi .
\]  

Parameters \( Q(\xi) \) and \( \xi_k(\xi) \) are defined by the above functions.

The statement of the problem of shock-wave initiation, formation and propagation of cellular detonation in a flat channel is similar to [15]. The numerical technique is based on the conservative flux-splitting schemes: the TVD scheme by Harten for gas and the Gentry-Martin-Daly scheme for particles. The numerical method has been tested earlier and applied for 2-D numerical simulations of the shock wave and detonation flows. The step of the finite-difference grid was determined according to the scale of the relaxation zones, varied in the test calculations.

3. Description of detonation flows of inhomogeneous gas suspensions of inert particles based on an analysis of the equilibrium of the gas chemical composition

It is assumed that incompressible particles are in mechanical and thermal equilibrium with gas, and their volume fraction is small. The influence of partial pressure and pressure of saturated vapors of the condensed phase is also neglected. The density and internal energy of a two-phase mixture are calculated by the relations:

\[
\rho = \rho + \rho_c, \quad \rho_c = (1-\alpha)\rho, \quad \rho_c = \alpha \rho_c, \quad U^* = (1-\alpha)U + \alpha U^c ,
\]

where \( U^*, U, U^c \) are internal energy of two-phase mixture, gas and particles, \( \alpha \) is mass fraction of particles in two-phase mixture, \( \rho, \rho_c, \rho \) are density of a gas, volume density of particles and gas-particles mixture.

According to the model [19-21], molar mass \( \mu \) and internal energy of a gas \( U \) can be calculated via the formulas:
\[
\mu(\rho, T) = \frac{B\mu_{\text{min}} - 2\mu_{\text{max}} + \sqrt{D}}{2(A^* - 1)},
\]
(5)

\[
B = \frac{AT^{3/4}}{4K_\gamma} \left(1 - e^{-\theta/T}\right)^{2/3} e^{-E/RT} \frac{\mu_{\text{max}}^2}{\mu_{\text{min}}},
\]
\[
D = (B\mu_{\text{min}} - 2\mu_{\text{max}})^2 + 4(B - 1)\mu_{\text{max}}^2,
\]

\[
U(T, \mu) = \left[\frac{3}{4} \left(\frac{\mu}{\mu_a} + 1\right) + \frac{3}{2} \left(\frac{\mu}{\mu_a} - 1\right) e^{\theta/T} - 1\right] \frac{RT}{\mu} + E\left(\frac{1}{\mu} - \frac{1}{\mu_{\text{min}}}\right).
\]

Here \(T\) is the temperature, \(R\) is the universal gas constant, \(\mu_a, \mu_{\text{min}}, \mu_{\text{max}}, K_\gamma, A, \theta, E\) are constants. From the first law of thermodynamics, the equation of state of an ideal gas, the adiabatic condition, and (4) - (6) the following formula follows for calculating the speed of sound \(c^*\) and the adiabatic index \(\gamma^*\) of a two-phase mixture:

\[
c^* = \frac{dP}{d\rho^*} = \gamma^* \frac{P}{\rho^*}, \quad \gamma^* = \frac{d\ln P}{d\ln \rho^*} = \frac{d\ln P}{d\ln \rho} = 1 - \frac{P}{\rho} \frac{\mu}{\mu_a} + \frac{P}{T} \left(1 - \frac{1}{\mu} \frac{d\rho}{dT}\right),
\]

\[
dT = \frac{1}{1 - \alpha} \frac{d\rho}{d\rho_a} = -(1 - \alpha)\left(U_p\mu_a - RT / \rho\mu\right),
\]

\[
(1 - \alpha)(U_T + U_p\mu_T) + \alpha U_T C.
\]

Algebraic formulas for calculating \(U_p, U_T, \mu_p, \mu_T\) can be obtained from the equation of state of an ideal gas and (4), (5).

In the presence of a spatial gradient \(\alpha\) the chemical equilibrium model under consideration remains unchanged. In the presence of a spatial gradient of the chemical composition of the gas phase, two options for using the model are possible. In the first version, distribution of the space of constants \(\mu_a, \mu_{\text{min}}, \mu_{\text{max}}\) is calculated, while the remaining model constants do not change. In the second version, the spatial distribution of all model constants is calculated. The first method for calculating thermodynamic parameters is easier to implement, while the second method is more accurate.

4. Results of calculations

4.1. The propagation of detonation waves in aluminium - oxygen mixtures with longitudinal concentration gradients

The concentration distribution along the channel was set by linear functions: decreasing \(\xi_0(x) = 0.55 - 0.3125x\) and increasing \(\xi_0(x) = 0.3 + 0.3125x\).

Figure 1. Maximal pressure fields at cellular detonation propagation in mixtures with longitudinal concentration gradients: negative (top), positive (bottom), \(\Delta t = 0.04\) ms.

Figure 1 shows the patterns of maximum cellular detonation pressures. It can be seen that the structure of the cells at the positive and negative concentration gradients is the same, their size remains
practically unchanged. In this case, the average and peak pressures along the channel vary in accordance with the initial loading of particles. Their values in areas of higher initial concentrations are higher than in areas of reduced initial particle concentration.

Figure 2 shows the instantaneous pressure profiles on the channel wall (\(y = 0\)) with a step of 0.04 ms that characterize cellular detonation flows. Here, as in Fig. 1, the maximum peak pressure values (due to the reflection of the transverse wave from the channel wall) also correspond to regions of higher values of the initial particle concentrations.

**Figure 2.** Pressure distributions on the wall \((y=0)\) at cellular detonation propagation in mixtures with longitudinal concentration gradients: negative (a), positive (b), \(\Delta t = 0.04\) ms.

**Figure 3.** Pressure gradient fields at cellular detonation propagation in mixture with transversal concentration gradient, \(\Delta t = 0.08\) ms.
4.2. The propagation of a detonation wave in a mixture with transversal concentration gradient

The concentration distribution across the channel was specified by a linear function $\xi(y) = 0.3 + 12.5y$. Figure 3 presents pictures of the field of pressure gradients (some analog of Schlieren images) with a time step of 0.08 ms, which reveal a strong curvature of the shape of the leading edge (thick lines) and the presence of many transverse waves. From Fig. 3, it can also be seen that the average propagation velocity of the curved front is constant. Figure 4 shows the field of maximal pressure history of cellular detonation. One can see that the system of transverse waves is irregular, and the distance between them in the lower part of the channel is greater than in the upper part. The result is a strongly elongated and asymmetric form of detonation cells. Moreover, the average and peak pressures in the upper part of the channel are much higher than in the lower part, which corresponds to the patterns revealed with longitudinal concentration gradients (Fig. 1). Asymmetric cells were previously modeled in inhomogeneous gas media in [22].

![Figure 4. Cellular detonation in the mixture with transversal concentration gradient: maximal pressure histories.](image)

5. Conclusions

The paper presents the models of reduced kinetics for the analysis of detonation flows in inhomogeneous gas particle suspensions. The models are based on an analysis of the equilibrium state or the use of the known dependence of the detonation velocity on the initial loading of reacting particles. Examples of calculating the propagation of cellular detonation in a gas suspension of aluminum particles in a channel with longitudinal and transverse particle concentration gradients are presented. Regular cellular structures are formed in channels with longitudinal concentration gradients; the cell size along the channel remains almost unchanged. In a channel with a transverse concentration gradient, irregular cells of an asymmetric and elongated shape with an inclined detonation front are formed. Peak and average pressure values are higher in areas of higher initial concentrations. The results are consistent with studies of similar gas detonation flows in [22].

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