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Thermally Coupled SHS Processes: Numerical Modeling

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Abstract—Unsteady spatial modes of gasless combustion in parallelepiped-shaped sample containing two powder mixtures separated by low-melting inert layer were numerically modelled. Samples with square cross section were found to burn both in stationary and in unsteady periodic modes depending on the thermal conductivity of the inert inner layer. Combustion of sample with an active inner layer in quasi-stationary control modes when the average burning velocities of the donor and acceptor mixtures are the same were studied.

Keywords: gasless combustion, donor and acceptor mixture, thermally coupled SHS processes, spinning combustion

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1. INTRODUCTION

Thermal coupling of layered structure is used in SHS production of materials from weakly exothermic or even endothermic mixture [1]. The process occurs so that there is no mass transfer between the layers. The layered structure represents a highly exothermic (donor) mixture with a low-caloric (acceptor) one [1]. In [2, 3], high-caloric metallothermic or gas-free compositions with combustion temperature of 1500–4000 K were used as donor layers. Heat from these mixtures transfers in the main synthesis (acceptor) layer for additional heating of reagents. 2D and 3D modeling of such processes is directly related to instability of propagation of spatial waves, specifically spinning waves, of gasless combustion [4–6]. Spinning combustion waves are observed in case of unstable propagation of exothermic reaction front in gasless and low-gas systems [7–10].

In this report, I suggest a 3D mathematic model for high-temperature synthesis of layered metal composites—donor and acceptor layers with different kinetic and thermophysical characteristics—prepared from rectangular rods.

2. NUMERICAL MODEL

A model is defined as three-layered "donor-acceptor-donor" rectangular parallelepiped (Fig. 1). Parameters and composition of donor layers are assumed to be the same; the inner acceptor layer has homogeneous composition and macrostructure. The outer layers transfer thermal energy to the inner one and thereby accelerates the synthesis of reagents in it. At the boundaries between the layers, a conjugate heat transfer mode is implemented. The parallelepiped-shaped sample is ignited over the entire surface of the upper face and the combustion wave propagates downward (see Fig. 1).

In this case, governing equations in a Cartesian coordinate system can be written in the form:

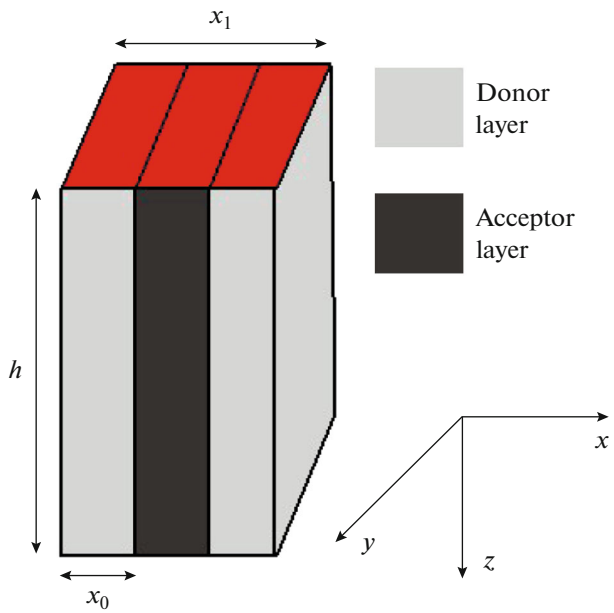

Fig. 1. Sample geometry.

$$[1 + \mathrm{Ph}\delta_D(\theta_1 - \theta_L)]\frac{\partial\theta_1}{\partial\tau} = \frac{\partial^2\theta_1}{\partial x^2} + \frac{\partial^2\theta_1}{\partial y^2} + \frac{\partial^2\theta_1}{\partial z^2} + \frac{1}{\gamma_1}\frac{\partial\eta_1}{\partial\tau}, \quad (1)$$

$$\frac{\partial\theta_2}{\partial\tau} = \delta\left(\frac{\partial^2\theta_2}{\partial x^2} + \frac{\partial^2\theta_2}{\partial y^2} + \frac{\partial^2\theta_2}{\partial z^2}\right) + \frac{\Omega}{\gamma_2}\frac{\partial\eta_2}{\partial\tau}, \quad (2)$$

$$\frac{\partial\eta_1}{\partial\tau} = \gamma_1(1 - \eta_1)\exp\left(\frac{\theta_1}{1+\mathrm{Ar}\theta_1}\right), \quad (3)$$

$$\frac{\partial\eta_2}{\partial\tau} = \gamma_2(1 - \eta_2)\exp\left(\frac{\sigma\theta_2}{1+\mathrm{Ar}\theta_2}\right). \quad (4)$$

The boundary conditions are the following:

$$\tau = 0:\ \theta_i(x,y,z,0) = \theta_0,\ \ \eta_i(x,y,z,0) = 0, \quad (5)$$

$$z = 0:\ \theta_i(x,y,0,\tau) = \theta_w(\tau < \tau_w),\ \ \partial\theta_i(x,y,0,\tau)/\partial z = 0(\tau < \tau_w), \quad (6)$$

$$x = 0,\ \ x = x_1:\ \partial\theta_i(0,y,z,\tau)/\partial x = 0,\ \ \partial\theta_i(x_1,y,z,\tau)/\partial x = 0, \quad (7)$$

$$y = 0,\ \ y = y_1:\ \partial\theta_i(x,0,z,\tau)/\partial y = 0,\ \ \partial\theta_i(x,y_1,z,\tau)/\partial y = 0, \quad (8)$$

$$z = h:\ \partial\theta(x,y,h,\tau)/\partial z = 0, \quad (9)$$

$$x = x_0,\ \ x = x_1 - x_0:\ \theta_1 = \theta_2,\ \ \frac{\partial\theta_1}{\partial x} = \Lambda\frac{\partial\theta_2}{\partial x}. \quad (10)$$

Dimensionless variables and parameters:

$$\gamma_1 = \frac{c_1RT_*^2}{Q_1E_1},\ \ \gamma_2 = \frac{K_2(T_*)}{K_1(T_*)}\gamma_1,\ \ \theta_i = \frac{(T_i - T_*)E_1}{RT_*^2},$$

$$\theta_L = \frac{(T_L - T_*)E_1}{RT_*^2},\ \ \theta_0 = -\frac{1}{\gamma_1},\ \ \mathrm{Ar} = \frac{RT_*}{E_1},$$

$$x = \frac{r_1}{r_*},\ \ y = \frac{r_2}{r_*},\ \ z = \frac{r_3}{r_*},\ \ r_* = \sqrt{\frac{\lambda_1 t_*}{c_1\rho_1}},\ \ t_* = \frac{c_1RT_*^2}{Q_1E_1K_1(T_*)},$$

$$K_i(T_*) = k_{0i}\exp(-E_i/RT_*),\ \ \tau = \frac{t}{t_*},\ \ \tau_w = \frac{t_w}{t_*},$$

$$x_1 = \frac{R_1}{r_*},\ \ x_0 = \frac{R_0}{r_*},\ \ y_1 = \frac{R_2}{r_*},\ \ h = \frac{R_3}{r_*},\ \ \mathrm{Ph} = \frac{Q_L}{Q_L\gamma_1},$$

$$\Omega = \frac{Q_2K_2(T_*)c_1}{Q_1K_1(T_*)c_2},\ \ \delta = \frac{\lambda_2c_1\rho_1}{\lambda_1c_2\rho_2},\ \ \sigma = \frac{E_2}{E_1},\ \ \Lambda = \frac{\lambda_2}{\lambda_1},$$

where $T_* = T_0 + Q_1/c_1$ is the adiabatic combustion temperature of donor mixture; r_1, r_2, r_3 are the Cartesian coordinates; t is the time; R_1, R_2, R_3 are sample dimensions; R_0 is the donor layer width; Q_L stands for heat of phase transformation in the donor mixture; T_L is the melting point; $\delta_D(\theta_1 - \theta_L)$ is the Dirac function; ρ_i, c_i, λ_i are density, heat capacity, and thermal conductivity, respectively; Q_i, E_i are reaction heat and activation energy; k_{0i} and $K_i(T_*)$ are pre-exponential factor and reaction rate at T_*; η_i is the extent of conversion. Note that $i = 1$ for donor mixture, $i = 2$ for acceptor inner layer.

3. MATHEMATICAL SOLUTION

Equations (1)–(10) were solved numerically by the finite difference scheme using coordinate-wise splitting and sweep method. I applied adiabatic boundary conditions to the side faces of the parallelepiped, which were second-order approximated, and then checked the approximate convergence of the difference scheme. To solve the task with phase transition, an economic scheme of through counting with smoothing coefficients and no explicit separation of the interface [11] was used. Thus, the delta function $\delta_D(\theta_1 - \theta_L)$ in Eq. (1) was replaced by a function:

$$\Phi(\theta) = \frac{\varphi(\theta)\,\mathrm{erf}^{-1}(\sqrt{2})}{\Delta\theta\sqrt{2\pi}}\exp\left(\frac{(\theta_L - \theta)^2}{2\Delta\theta^2}\right),$$
$$\varphi(\theta) = \begin{cases} 0, \theta > \theta_L + \Delta\theta \\ 1, \theta_L - \Delta\theta < \theta < \theta_L + \Delta\theta, \\ 0, \theta < \theta_L - \Delta\theta \end{cases} \quad (11)$$

where at least five calculation points are in the temperature range of smoothing $\Delta\theta$ for each direction.

As the reaction front propagates along the sample, there is a decrease in the calculated area as viewed from burned mixture that keeps pace with its increase on the source side of green mixture. This axial "shift" made it possible to reduce the time of calculations until reaching the steady state combustion regime. For all calculations, I assumed that the time of contact with the heated surface $\tau_{ign} = 500$, $\mathrm{Ph} = 0.6$, $\theta_L = -2.5$, and $\delta = 1$. To reduce the time to reach a stable nonstationary combustion regime, a small initial perturbation of the temperature field, which determined the direction of rotation of the hot spots, was set.

The suggested 3D mathematical model (Eqs. (1)–(5)) allows to calculate the spatial modes of combustion of the three-layer parallelepiped-shaped sample, as well as homogeneous sample. The sample has symmetry planes passing through the center of inner layer, but the symmetry condition for solving Eqs. (1) and (2) is not given. It was shown in [6] that an increase in the area of the parallelepiped base leads to complex spatial combustion regimes with a large number of hot spots. In this case, the combustion spots can follow both spiral trajectories along the side surface and inner trajectories normal to the side faces.

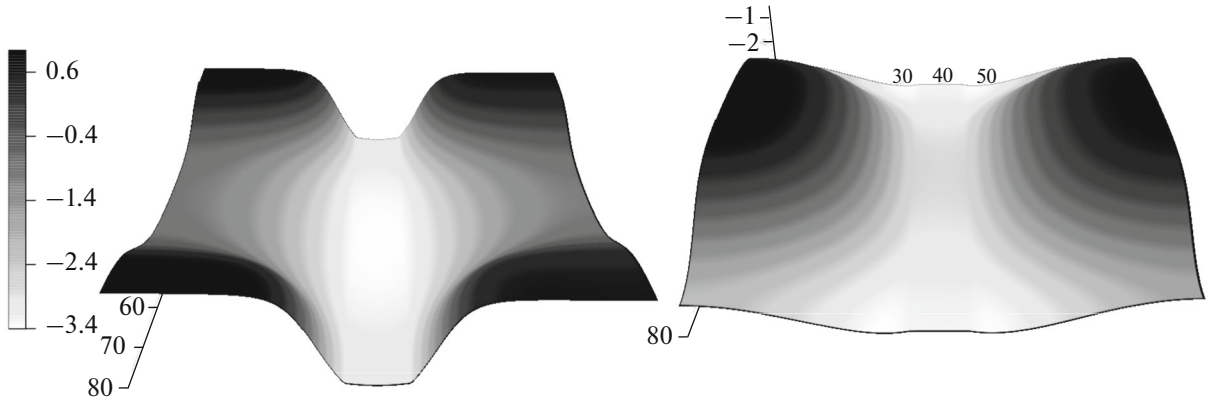

Fig. 2. Temperature fields in case of unsteady combustion mode in sample with inert inner layer: $x_1 = 80$, $y_1 = 80$, $x_0 = 35$, $\theta_0 = -7$, Ar = 0.13, $\Omega = 0$, $\Lambda = 7.5$.

4. COMBUSTION OF DONOR-INERT ACCEPTOR-DONOR SAMPLE

The tapping of highly heat-conducting metal elements permits to increase the burning velocity of condensed system. In this case, addition of heat-conducting metal plate or wire with relatively small thickness increases the local burning velocity by 2–3 times. Combustion of sample consisting of inert highly thermally conductive inner layer from a gas-free mixture is made possible both in stationary and in unsteady regime depending on the thermal conductivity and thickness of inner layer. The average burning velocity of such system can be significantly higher than that of homogeneous donor sample. Now let us consider gasless combustion in the sample with inert inner layer (foil, plate) possessing high thermal conductive properties. For the given parameters: $\Omega = 0$, $\Lambda = 15$, $x_1 = y_1 = 80$, $x_0 = 35$, $\theta_0 = -7$, Ar = 0.13, the average burning velocity was found to be higher by 17% than that of homogeneous sample. This is associated with heat recovery: heat transfer from combustion products to the region of heating due to higher thermal conductivity of the inner layer. Furthermore, the temperature of the inner layer is approximately 3 characteristic intervals lower than the combustion temperature of the active layer in the sample section passing through the combustion front. The ratio of thermal conductivity coefficients $\Lambda = 15$ corresponds to nickel–aluminum compact and steel. A decrease in the thermal conductivity of the inner layer, for example, due to a change in the composition or an increase in the porosity of the acceptor mixture, transforms combustion in unsteady regime with symmetrical motion of hot spots (Fig. 2).

The unsteady combustion proceeds in a periodic two-spot mode, which is symmetrical with respect to the plane passing through the point $x = 40$ parallel to the axis z. The cycle is born conventionally in two hot spots: $x = 0$, $y = 40$ and $x = 80$, $y = 40$. Then each spot splits into two ones propagating in opposite directions to corners (parallelepiped edges). A short time later, these spots move back and merge into a new spot at $x = 40$.

Thus, the use of inert highly thermally conductive plate with small size $x_1 - 2x_0 < x_0$ in gas-free system is beneficial for stable propagation of combustion wave at a constant τ_{ign}. Heat-conducting elements initiate combustion of gas-free powder mixtures when independent combustion of homogeneous mixture in the frontal mode becomes impossible [12].

5. COMBUSTION OF DONOR-ACTIV ACCEPTOR-DONOR SAMPLE

Spatial periodic combustion regimes in a sample with an active inner layer are more diverse than those in a sample with an inert inner layer and share a number of traits with combustion regimes in a homogeneous sample [6]. Unlike above-described case, Ω, γ_2, and σ parameters responsible for reaction kinetics in the acceptor mixture were added. Note that merely certain of these modes can be determined and described within the framework of the numerical solution method. If there is a strong difference between the reaction times in the donor and acceptor mixtures, the combustion wave propagates through the donor mixture much faster than that in the acceptor mixture ("chemical furnace" mode [1]). In this case, attempts to study numerically the process with reaching steady state mode fared poorly because the distance between the reaction fronts in the donor and acceptor mixtures grows constantly. For a fixed sample height (z axis), there comes a moment when the interfrontal distance becomes comparable to or greater than the sample height h. Therefore, the choice of the parameters for donor and acceptor layers is limited by the fusion and control regimes [13], in which the burning velocity values for donor and acceptor mixtures are close.

In order to synthesize materials, a necessary condition is the combination of high-and low-exothermic layers. The thermal conductivity and calorific value of

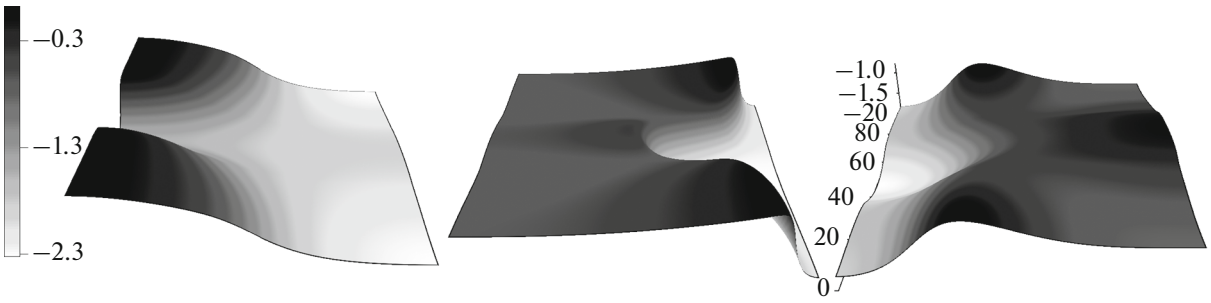

Fig. 3. Temperature fields in case of unsteady combustion mode in sample with active inner layer: $x_1 = 93$, $y_1 = 93$, $x_0 = 31$, $\theta_0 = -7$, Ar = 0.13, $\Omega = 0.65$, $\Lambda = 0.6$.

the donor layer are much higher than those of the acceptor layer, which ensures high heating rate for inner layer. The main results of numerical calculations were obtained for the following parameters for inner layer: $\gamma_2 = \gamma_1$, $\sigma = 1.15$, $\Lambda = 0.6$, and $\Omega = 0.65$ (typical of "chemical furnace" method). At these values, a periodic control mode is implemented. In this case, the linear burning velocities in the sample plane $y = 0.5y_1$ for inner and outer layers are equal, but the combustion front in the acceptor layer can lag behind the combustion fronts in the donor layers by approximately the thickness of the heated donor layer. Figure 3 shows the temperature distribution in the sample cross section (z = const) passing through the combustion zone of donor layers. Large combustion spots move along the side faces of the sample in the outer donor layers, thus creating favorable conditions for the propagation of combustion spot in the inner layer. The mode with alternating combustion spots (from outer layers to donor one and vice versa) can be called the control mode as for a 2D theoretical model [13]. The maximum temperature in the hot spots does not exceed the adiabatic combustion temperature of the donor mixture and is 2–3 characteristic intervals lower than the maximum temperatures in unsteady combustion modes in a homogeneous sample [6]. With a decrease in the calorific value of the acceptor mixture by 23% ($\Omega = 0.5$) at fixed sizes and ignition conditions, it was not possible to initiate the synthesis of a three-layer sample in the frontal mode. A similar result exists with increasing acceptor layer volume.

An increase in the sample size gives rise to complex spatial combustion modes with a large number of hot spots. In this case, the numerical calculation is more time consuming.

CONCLUSIONS

A 3D theoretical model of high-temperature synthesis of parallelepiped-shaped sample consisting of donor-acceptor-donor layers with phase transformation of components is proposed and a numerical solution technique is developed.

Combustion of a three-layer sample with an inert highly thermally conductive inner layer is found to be possible both in stationary and in unsteady mode depending on the thermal conductivity and thickness of inner layer. The average burning velocity of such sample is higher than that of a homogeneous sample.

Unsteady periodic combustion modes in a sample with an active inner layer are revealed. The hot spots can move either along the trajectories over the side surfaces or along the inner trajectories with the leading reaction zone in donor layers. The combustion of samples with thermally thin inner layer proceeds in the control regime [13]: the reaction front propagating through the acceptor mixture lags behind the reaction fronts in the donor layers by a certain distance.

The suggested mathematical model for combustion of layered system can be useful for estimating the optimal conditions for synthesis of metal composites in the frontal combustion mode.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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