

A MODEL OF STRUCTURAL TRANSFORMATIONS IN A REACTIVE DISPERSED MEDIUM UNDER CONDITIONS OF NONGASIFYING COMBUSTION

O. B. Kovalev and V. M. Fomin

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Self-propagating high-temperature synthesis (SHS) [1], which is a highly exothermic physical and chemical interaction of reagents in the condensed phase, is an effective method for producing a wide class of high-melting compounds.

The methods of description of the processes in an SHS wave were initially based on the thermal theory of [1], which was built on the assumption of the temperature homogenization of the medium with constant thermal and physical characteristics and of the absence of the mass transfer of reagents. Mathematically, this theory was reduced to the equations of heat transfer and to the equations of the kinetics of chemical reactions:

$$c\rho \frac{\partial T}{\partial t} = \lambda \frac{\partial^2 T}{\partial x^2} + Q \frac{\partial \beta}{\partial t}; \quad (1)$$

$$\frac{\partial \beta}{\partial t} = k_0 \exp\left(-\frac{E}{RT}\right) \Phi(T). \quad (2)$$

Here T is the temperature, β is the degree of conversion, c and ρ are the specific heat and density, λ is the effective thermal conductivity, Q is the reaction heat, E is the activation energy, k_0 is the preexponent, and $\Phi(T)$ is a kinetic function. The thermal theory of [1] describes well the case of narrow reaction zones with complete transformation of substances in them. Wide zones of transformations with chemical stages [2], phase transitions [3, 4], capillary spreading [5], structural transformations [6], etc., were described by representing the kinetic function $\Phi(T)$ obtained empirically [2] or theoretically [3–6]. The thermal theory of [1] and various quasi-homogeneous modifications of it [2–6] failed to give a satisfactory description of the synthesis-wave structure. The concept of a narrow zone of chemical reactions allows one to obtain only approximate relations for the wave propagation velocity as a function of the initial temperature and other parameters of the system. In most cases, experimental temperature profiles are inconsistent with the concept of narrow reaction zones. The wave structure in actual processes have inflections and isothermal sections and cannot be described within the framework of the thermal theory.

The model of nongasifying combustion with phase transformations [7] is also based on the thermal theory. In this model, to describe phase transitions, the heat capacity of a substance is represented by the Dirac δ -function. Such a representation implies that the phase transition occurs instantaneously. In fact, the phase transformation (melting or crystallization) proceeds in time and is determined by the intrinsic kinetics of the process, which also affects the SHS-wave structure.

Aldushin and Nekrasov et al. [8–10] proposed a model of nongasifying combustion in which the reactive diffusion processes are considered at the level of a mixture cell with allowance for the scale of heterogeneous and multistage transformations, which enabled them to make the first quantitative estimates of SHS. However, the chemical composition and structure of the final product remain an open question.

Attempts to build a heterogeneous SHS model were undertaken in [11–13] and were a failure. Based, on the whole, on the equations of mechanics of a heterogeneous medium [14], Smolyakov introduced the kinetic equation for the degree of conversion, and this model is slightly different from the thermal theory. In

addition, the Dirac δ -function is used to describe phase transitions, as in [7]. Thus, all the advantages of the heterogeneous approach were nullified.

Macrostructural transformations which include chemical reactions, structural and phase transitions, and the heat- and mass-transfer processes in a synthesis wave have different kinetic characteristics and, as a consequence, are extended in time and space. A multiphase product, whose composition, internal structure, and properties are not known *a priori*, is usually formed behind the combustion wave front. Obviously, combustion of such systems is heterogeneous due to their heterogeneous nature. In our opinion, a promising line of development of the SHS theory is to use properly the basic principles of the mechanics of heterogeneous media [14, 15].

We assume that the initial internal structure of a powder mixture consists of elementary cells (EC), each containing a particle of one component surrounded by a layer of particles of another component. Such a structure is typical of Ni + Al, Zr + Al, Ti + C, Ta + C, and other mixtures [1]. The chemical interaction between the substances occurs through the contact surface in the regime of reactive diffusion whose model is based on a powder mixture of two metals *A* and *B*. In most cases, the particles of one reagent melt, and it spreads over capillaries of the system and encapsulates the particles of the other reagent, thus increasing the contact surface and intensifying the process of reactive diffusion. The synthesis reaction gives rise to the formation of a final product in a liquid or solid state. The temperature profile in the combustion wave is characterized by the thickness of the reaction zone and displays all the specific features of the structural, phase, and chemical transformations of substances.

The number of particles *A* per particle of *B* is calculated as

$$N_{AB} = \alpha_A \rho_B R_B^3 / (\alpha_B \rho_A R_A^3).$$

For example, for a Ni+Al system taken in the stoichiometric ratio [16] with weight fractions $\alpha_A = 0.315$ and $\alpha_B = 0.685$, with densities $\rho_A = 2.7 \text{ g/cm}^3$ for Al and $\rho_B = 8.4 \text{ g/cm}^3$ for Ni, and with particle radii $R_A = 5 \text{ }\mu\text{m}$ and $R_B = 20 \text{ }\mu\text{m}$, the calculated value is $N_{AB} = 91$. For a specified initial porosity m_1^0 , the volume of EC in a mixture V_s can be assumed to consist of the sum of the volumes of the *A* and *B* particles and the volume of pores: $V_s = V_B + N_{AB}V_A + m_1^0V_s$. For the cell radius R_s , we then obtain $R_s = (1 - m_1^0)^{-1/3}(1 + \alpha_A\rho_B/\alpha_B\rho_A)^{1/3}R_B$ and, hence, $R_s \approx 2R_B$. According to [1], in most SHS processes the combustion-wave thickness is $l_* \sim 4\text{--}5 \text{ mm}$ and, hence, $R_s \ll l_*$.

We shall study the evolution of the structure of a heat-insulated dispersed mixture in the course of chemical and phase transformations and heat- and mass-transfer processes in the combustion wave at the EC level on the basis of the behavior of the system components with an increase in temperature. The spatial temperature profile is assumed to consist of three zones. The first zone has a granular structure, and the temperature in it changes from the initial temperature T_0 up to the melting point T_A ($T_0 \leq T \leq T_A$) of low-melting particles of *A*. The second zone is responsible for the interval of temperature variation within the melting points of the *A* and *B* components ($T_A \leq T \leq T_B$) and consists of a melt of *A* with suspended solid-state particles of *B* and gas bubbles. At the boundary of the first and second zones, for $T = T_A + \Delta T$ (ΔT is a small quantity) there is an intermediate liquid phase of metal *A* that encapsulates high-melting particles of *B*. The time of complete transition of the particles of *A* into a liquid state depends on the particle size and on the heat-front velocity. A new porosity in EC is formed as the *A* particles melt.

We assume that the gaps between the particles of *A* are transformed into isolated spherical pores filled with the gas. Assuming the number of gaps to be roughly equal to the number of particles of *A*, we can calculate the initial radius of the bubbles formed: $a_0 = R_A(m_1^0/(1 - m_1^0))^{1/3}$. When the entire component *A* melts, an EC consists of one particle of *B* surrounded by the melt of *A* with gas bubbles. The chemical interaction of components *A* and *B* in the reactive-diffusion regime begins at the stage of melting of *A* when the solid particles of *B* dissolve in the melt of *A*. A solid-phase layer *AB* is formed on the surface of the *B* particles. A process that hinders burning is the growth of a layer of a new phase which is determined by mutual diffusion of the atoms of *A* and *B* substances through the layer *AB*.

The third zone corresponds to the variation in temperature from the melting point of the component *B* to the burning temperature T_* ($T_B \leq T \leq T_*$). New-phase grains are formed in this zone, and the centers

of these grains are the particles of B . The grains can first consist of several layers: an upper layer of the new phase, a liquid layer of melted B , and the solid core of the B particle which is not yet melted. As the layer AB grows, the liquid component A is utilized and the contacts between the new-phase grains appear. As the liquid phase reduces, the medium is transformed from a liquid suspension into a porous solid body with definite granularity. Isolated pores-bubbles merge in the gaps between the new-phase grains, forming a bound porous structure filled partly with the liquid component A or with the gas. After some of the components are completely utilized, the synthesis terminates. It is assumed that the gas in the three zones is inert and can be involved only in the heat-transfer processes. Thus, to each temperature value corresponds a definite structural state of the substance.

Mathematically, the processes in an SHS wave are difficult to simulate because of a number of factors associated with the evolution of the structure of a heterogeneous medium. A description of the gas flow and phase interaction is complicated by the change in porosity. The difficulties in describing of the stress-strain state are due, on one hand, to vanishing, at the particle melting point, of the fictitious stress tensor responsible for intergranular interaction in the granular layer and, on the other hand, to the appearance of the contact between the grains of the new phase and to the formation of a porous product.

The phase transitions and the synthesis reaction can be formally represented as $A_s \xrightarrow{J_1} A_l - Q_1^0$, $B_s \xrightarrow{J_2} B_l - Q_2^0$, $\nu_1 A_l + \nu_2 B_l \xrightarrow{J_3} \nu_3 AB_s + Q_3$, where J_j and Q_j are the velocities and heats of the processes ($j = 1$ and 2 refer to melting of the A and B particles and $j = 3$ refers to the reaction of AB synthesis), ν_j are the stoichiometric coefficients; the subscripts s and l indicate the solid and the liquid state of the substance.

We consider a two-velocity and two-temperature model of the medium in which ρ_i are the mean densities, \mathbf{v}_i the mean velocities, and T_i the mean temperatures of the gas ($i = 1$) and of the c-phase ($i = 2$).

For the gas and for five components of the c-phase, the equations of conservation of mass have the form

$$\frac{\partial}{\partial t} \rho_1 + \frac{\partial}{\partial x_k} \rho_1 v_{1k} = 0; \quad (3)$$

$$\frac{\partial}{\partial t} \rho_{2j} + \frac{\partial}{\partial x_k} \rho_{2j} v_{2k} = \varphi_j, \quad j = \overline{1, 5} \quad (4)$$

$$[\varphi_1 = -\mu_1 J_1, \quad \varphi_2 = \mu_1(J_1 - \nu_1 J_3), \quad \varphi_3 = -\mu_2 J_2, \quad \varphi_4 = \mu_2(J_2 - \nu_2 J_3), \quad \varphi_5 = \mu_3 \nu_3 J_3],$$

where summation is performed over the repeat subscript k , v_{1k} and v_{2k} ($k = 1, 2, 3$) are the components of the velocity vectors \mathbf{v}_1 and \mathbf{v}_2 , $\rho_{2j} = m_{2j} \rho_{2j}^0$, ρ_{2j}^0 , and m_{2j} are the mean densities, the densities of material of the c-phase components, and their volumetric fractions ($j = 1$ and 2 refer to the solid and liquid components of A , $j = 3$ and 4 , to the solid and liquid components of B , and $j = 5$, to the solid product AB); μ_1 , μ_2 , and μ_3 are the molecular masses of substances A , B , and AB , respectively. The materials of the initial particles and of the product satisfy the incompressibility condition: $\rho_{2j}^0 = \text{const}$.

Let us introduce into consideration the parameters

$$\alpha = \begin{cases} \rho_{22}/(\rho_{21} + \rho_{22}), & \rho_{21} + \rho_{22} \neq 0, \\ 1, & \rho_{21} + \rho_{22} = 0, \end{cases} \quad \beta = \rho_{25}/(\rho_{22} + \rho_{25})$$

that vary from 0 to 1 and characterize the degree of melting of the A particles and the formation of the product AB , respectively. The choice of these parameters satisfies the limiting transitions discussed below. Using the parameters α and β , we write the equations for the radius a and the number of bubbles n :

$$\alpha(1 - \beta)m_1 = (4/3)\pi a^3 n; \quad (5)$$

$$n = \alpha(1 - \beta)N_A. \quad (6)$$

Here m_1 and ρ_1 are the volumetric concentration and mean density of the gas and N_A is the number of the initial particles of A in the mixture. Relations (5) and (6) describe variation in the number and radius of the bubbles when the medium's structure is changed from a granular structure with bound porosity to a liquid one as the particles of A melt, and then again to the porous product.

We consider small deformations that cannot exert a substantial effect on variation in temperature of the solid and liquid phases, in other words, the work of internal intergranular forces is small, compared with the heat produced by the chemical reaction of synthesis. The changes in the internal energy of the gas and c-phases are determined by heat inflow:

$$\frac{\partial}{\partial t} \rho_1 e_1 + \frac{\partial}{\partial x_k} (\rho_1 e_1 v_{1k} - q_{1k}) = -F_{\mu j} (v_{1j} - v_{2j}) + g_{12}; \quad (7)$$

$$\frac{\partial}{\partial t} \rho_2 e_2 + \frac{\partial}{\partial x_k} (\rho_2 e_2 v_{2k} - q_{2k}) = F_{\mu j} (v_{1j} - v_{2j}) + g_{21}. \quad (8)$$

Here ρ_i , e_i , $q_{ik} = \lambda_i m_i \partial T_i / \partial x_k$, m_i , and λ_i are the mean densities, internal energies, heat fluxes, volumetric concentrations and mean thermal conductivities in the gas phase ($i = 1$) and in the c-phase ($i = 2$):

$$\begin{aligned} \rho_2 &= \rho_2^0 m_2 = \sum_{j=1}^5 \rho_{2j}^0 m_{2j}, \quad e_1 = c_1 (T_1 - T_0) + e_1^0, \quad e_2 = \left(\sum_{j=1}^5 \rho_{2j} e_{2j} \right) / \rho_2, \\ e_{21} &= c_{21} (T_2 - T_0) + e_{21}^0, \quad e_{22} = e_{21} + Q_1^0, \quad e_{23} = c_{23} (T_2 - T_0) + e_{23}^0, \quad e_{24} = e_{23} + Q_2^0, \\ e_{25} &= c_{25} (T_2 - T_0) + e_{25}^0, \quad \lambda_2 = \left(\sum_{j=1}^5 \lambda_{2j} m_{2j} \right) / m_2, \quad m_2 = \sum_{j=1}^5 m_{2j}, \\ g_{21} &= -g_{12} = (1 - \alpha) g_{21}^{(1)} + \alpha (1 - \beta) g_{21}^{(2)} + \alpha \beta g_{21}^{(3)}; \end{aligned}$$

e_{2j} , c_{2j} , and λ_{2j} are the internal energies, specific heats, and thermal conductivities of the c-phase components ($j = 1$ and 2 refer to the solid and liquid components of A , $j = 3$ and 4 to the solid and liquid components of B , and $j = 5$ to the solid product AB); e_{2j}^0 are the specific internal energies at temperature T_0 , and, hence, the heat of the chemical reaction of synthesis is $Q_3^0 = (\mu_1 \nu_1 e_{21}^0 + \mu_2 \nu_2 e_{23}^0 - \mu_3 \nu_3 e_{25}^0) (1 / \mu_3 \nu_3)$, $g_{12}^{(i)}$ and $g_{21}^{(i)}$ are the heat fluxes due to the internal heat exchange between the gas and the c-phase in the granular layer ($i = 1$), in the solid-liquid suspension ($i = 2$), and in the final porous product ($i = 3$), and F_μ are the interphase-interaction forces.

Let us introduce combined average stress tensors in the gas Γ_1^{ke} and in the c-phase Γ_2^{ke} in the form

$$\Gamma_1^{ke} = -m_1 p_1 \delta^{ke}, \quad \Gamma_2^{ke} = (1 - \alpha) [\sigma_{21}^{ke} - m_2 p_1 \delta^{ke}] + \alpha (1 - \beta) m_2 \sigma_{22}^{ke} + \alpha \beta [\sigma_{23}^{ke} - m_2 p_1 \delta^{ke}],$$

where σ_{2j}^{ke} are the fictitious stress tensors in the granular skeleton ($j = 1$) and in the porous solid product ($j = 3$), σ_{22}^{ke} is the stress tensor in the solid-liquid suspension, and p_1 is the gas pressure:

$$\begin{aligned} p_1 &= \rho_1^0 R T_1; \quad (9) \\ \sigma_{2j}^{ke} &= m_2 (\lambda^{(j)} \varepsilon_2 \delta^{ke} + 2\mu^{(j)} \varepsilon_2^{ke}) + m_2 \beta^{(j)} k^{(j)} p_1 \delta^{ke}; \\ \sigma_{22}^{ke} &= - \left[m_1 \left(p_1 - \frac{2\Sigma}{a_0} \right) + m_2 p_2^{(2)} \right] \delta^{ke}; \quad \frac{\partial}{\partial t} \varepsilon_2^{ke} = \frac{1}{2} \left(\frac{\partial v_{2k}}{\partial x_e} + \frac{\partial v_{2e}}{\partial x_k} \right); \end{aligned}$$

R is a gas constant, $\lambda^{(j)}$ and $\mu^{(j)}$ (Lamé coefficients), $k^{(j)}$ (moduli of triaxial compression), and $\beta^{(j)}$ (compressibility coefficients) are the average effective coefficients for the mixture ($j = 1, 3$), Σ is the surface tension coefficient; $p_2^{(1)}$, $p_2^{(2)}$, and $p_2^{(3)}$ indicate the pressure in the granular skeleton, liquid melt, and porous product, ε_2^{ke} is the fictitious stress tensor, and $\varepsilon_2 = (1/3)(\varepsilon_2^{11} + \varepsilon_2^{22} + \varepsilon_2^{33})$.

Let us write the momentum equations in the gas and in the c-phase for the entire range of temperature variation:

$$\frac{\partial}{\partial t} \rho_1 v_{1k} + \frac{\partial}{\partial x_j} (\rho_1 v_{1k} v_{1j} - \Gamma_1^{kj}) = -R_k; \quad (10)$$

$$\frac{\partial}{\partial t} \rho_2 v_{2k} + \frac{\partial}{\partial x_j} (\rho_2 v_{2k} v_{2j} - \Gamma_2^{kj}) = R_k. \quad (11)$$

Here $R_k = (1 - \alpha)R_k^{(1)} + \alpha(1 - \beta)R_k^{(2)} + \alpha\beta R_k^{(3)}$ ($k = 1, 2, 3$), $R_k^{(1)} = F_{\mu k}^{(1)} - p_1 \partial m_1 / \partial x_k$, $R_k^{(2)} = F_{\mu k}^{(2)} - \sigma_{22}^{kj} \partial m_1 / \partial x_j$, and $R_k^{(3)} = F_{\mu k}^{(3)} - p_1 \partial m_1 / \partial x_k$.

In describing the phase interaction, we take into account only the buoyancy and friction forces because of viscosity of the carrier phase, with all the remaining forces ignored. Since the above-mentioned temperature zones differ in the internal structure and in the carrier phases, different friction forces are considered in each of them: $F_{\mu}^{(1)}$, $F_{\mu}^{(2)}$, and $F_{\mu}^{(3)}$.

According to the plastic gas model [17], the fictitious pressure between the grains in a porous solid body under loading depends on the porosity and density:

$$p_f = \varphi(m_1, \rho_2^0) = \begin{cases} \rho_2^0 m_* c_*^2 (m_* / m_1 - 1), & m_1 < m_*, \\ 0, & m_1 \geq m_* \end{cases}$$

(m_* and c_* are the porosity and sound velocity in an extremely compressed porous body).

The equations of common deformation can be represented as

$$m_2(p_2^{(k)} - p_1) = \varphi_k(m_1, \rho_2^0, m_*^{(k)}, c_*^{(k)}), \quad k = 1, 3, \quad (12)$$

where $k = 1$ corresponds to the granular layer and $k = 3$ refers to the porous product.

The equations for bubble deformation are written in the Rayleigh-Lamb form [14]:

$$a \frac{d^2 a}{dt^2} + \frac{3}{2} \left(\frac{da}{dt} \right)^2 = \frac{p_1 - p_2^{(2)} - 2\Sigma/a}{\rho_{22}^0}. \quad (13)$$

Supplementing the resulting equations by the relation

$$m_1 + \sum_{j=1}^5 m_{2j} = 1, \quad (14)$$

we obtain system (3)-(14) with unknown functions:

$$\rho_1^0, m_1, m_{2j} \quad (j = \overline{1, 5}), \quad v_1, v_2, T_1, T_2, p_1, a, n, p_2^{(j)} \quad (j = 1, 2, 3).$$

The kinetics of phase transitions and chemical transformations and of the forces of phase and thermal interaction is a subject of special consideration and is omitted in the present paper.

The choice of the parameters α and β in the above form is due to the necessity of performing the following limiting transitions. First, with $\alpha \rightarrow 0$ and $\beta \rightarrow 0$, system (3)-(14) is transformed into equations of the mechanics of saturated porous media [15], with $\alpha \rightarrow 1$ and $\beta \rightarrow 0$, we obtain equations for an ideal incompressible liquid with suspended solid particles and gas bubbles [14], and with $\alpha \rightarrow 1$ and $\beta \rightarrow 1$, we have a model for a porous solid body [14, 15]. Second, the cited limiting values of the parameters are related to the composition of the final product obtained. If the mixture of powders is taken in the stoichiometric ratio, the components react completely. The final product is pure, contains no admixtures, and has a granular structure and bound porosity between the grains. If the starting mixture is deficient in the particles of A , the final product also has a granular structure, but the grains can consist of the upper layer of substance AB and the internal core of substance B in a liquid or partially solid state. If the particles of A are in excess in the mixture, the final product is a melt of A with suspended AB particles and gas bubbles.

Thus, the model of structural transformations has been mathematically reduced to (3)-(14) without an explicit separation of the temperature regions responsible for structural transformations.

The model allows one to consider the SHS process under pressures which make it possible to assume incompressibility of the materials of components in solid and liquid states. Depending on the SHS conditions, one can calculate the formation of the structure of the final product consisting of isolated (spherical) and bound pores with a certain graininess and chemical composition.

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