

MATHEMATICAL SIMULATION AND STRUCTURAL MACROKINETICS OF THE HIGH-TEMPERATURE SYNTHESIS OF INTERMETALLIC COMPOUNDS

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UDC 621.7.011

In extending the results of [1], a theoretical study is made of the ignition temperature of a nickel-aluminum powder mixture as a function of the power of the external heating source, the dispersity of the refractory component, and the porosity of the powder mixture in the case of a volume reaction. The initial mixture is modeled by a set of spherical elementary cells whose dimension is determined by the range of nickel particle sizes, the mixture composition, and the porosity.

In solving the problem, we proceed from the following assumptions: 1) the diffusion process is quasistationary; 2) the melt volume is unrestricted, which is valid for small interaction times characteristic of ignition processes; 3) eutectic melting proceeds in a heat conduction regime; 4) there is local thermodynamic equilibrium at the phase boundaries, which allows determination of a heat release function by the method described in [2]; 5) the effect of gas pores in the powder mixture is insignificant. According to the above assumptions the heat balance equation for the system under consideration is

$$C_V \frac{dT}{dt} = NQ\rho_A \frac{dl_n}{dt} + N\Phi_L - NL\rho_B \frac{dV_L}{dt} + \frac{WS}{V} - \alpha \frac{S_V}{V} (T - T_0), \quad (1)$$

where $C_V = (C_L\rho_A V_A + C_L\rho_B V_B)/(V_A + V_B)$ is the volume heat capacity; C_L is the limiting solubility of a refractory component in the melt of a lower-melting component of the powder mixture; V is the volume of the powder mixture; $N = 1/(4/3\pi R_e^3)$ is the number of elementary spherical cells per unit volume; R_e is the radius of an elementary cell; S is the heating area of the external surface of the powder mixture; S_V is an area of the external surface of the sample; α is the heat transfer coefficient; T_0 is the ambient temperature; Q is the thermal effect of the reaction of product formation per unit mass of the refractory component A; ρ_A and ρ_B are the nickel and aluminum densities;

$$I_n = 4\pi \left[\left(C_1 - \frac{C_1 - C_2}{\Gamma_2 - \Gamma_1} \Gamma_2 \right) \left(\frac{\Gamma_2^3 - \Gamma_1^3}{3} + \frac{C_1 - C_2}{\Gamma_2 - \Gamma_1} \Gamma_1 \Gamma_2 \frac{\Gamma_2^2 - \Gamma_1^2}{2} \right) \right], \quad (2)$$

I_n is the amount of element A bound in the A_2B_3 product layer; C_1, C_2 is the nickel concentration at the phase boundaries A- A_2B_3 and A_2B_3 -B, respectively. The coordinates of the phase boundaries A- A_2B_3 (Γ_1) and A_2B_3 - B (Γ_2) are determined by the relations [3]:

$$\frac{d\Gamma_1}{dt} = \frac{D_s}{1 - C_1} \frac{\partial C}{\partial \Gamma} \Big|_{\Gamma_1+0},$$

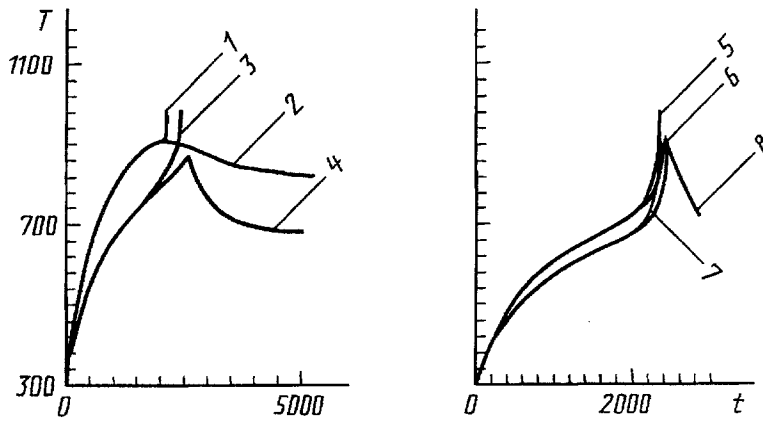


Fig. 1. Calculated thermograms of the initial stage of Ni_3Al synthesis at near-critical powers of the external heating source as a function of the radius of the nickel particles: 1, 2) $W = 30,437$ and $30,435 \text{ W/m}^2$ at $R = 1 \cdot 10^{-5} \text{ m}$; 3, 4) $W = 22,929$ and $22,906 \text{ W/m}^2$ at $R = 1/3 \cdot 10^{-5} \text{ m}$; 5, 6) $W = 19,208$ and $19,187 \text{ W/m}^2$; 7, 8) $W = 18,281$ and $17,312 \text{ W/m}^2$ at $R = 1/9 \cdot 10^{-5} \text{ m}$.

$$\frac{d\Gamma_2}{dt} = \begin{cases} -\frac{D_s}{C_2} \frac{\partial C}{\partial \Gamma} \Big|_{\Gamma_2=0}, & T \leq T_e, \\ \frac{D_L}{C_2 - C_1} \frac{dC}{d\Gamma} \Big|_{\Gamma_2=0} - D_s \frac{\partial C}{\partial \Gamma} \Big|_{\Gamma_2=0}, & T \geq T_e, \end{cases}$$

where $D_s = D_{0s} \exp(-E_s/RT)$ is the diffusion coefficient of the element A in the A_2B_3 layer; $D_L = D_{0L} \exp(-E_L/RT)$ is the diffusion coefficient of the component A in the melt; E_s and E_L are the activation energy of diffusion in the solid and liquid phases, respectively; C_L is the limiting concentration of the element A in the melt; T is the temperature; T_e is the melting point of the eutectic; R is the universal gas constant.

The rates of melting and heat release due to dissolution of the element in the melt are as follows

$$\frac{dV_L}{dt} = \begin{cases} \frac{V_B^* C_L (T - T_e)}{t_L L}, & T \geq T_e, \\ 0, & T \leq T_e; \end{cases} \quad (3)$$

$$\Phi_L = \begin{cases} 4\pi Q_L \rho_B \Gamma_2^2 \left(\frac{D_L(T)}{\Gamma^2} - \frac{d\Gamma_2}{dt} \right), & T > T_e, \\ 0, & T < T_e, \end{cases} \quad (4)$$

where V_B^* is the volume of the component at the moment of heating of the powder mixture to the melting point of the eutectic; t_L is the time of the exothermic reaction; Φ_L is the power of the external heating source.

The initial section of the thermogram of intermetallic synthesis, involving thermal explosion, has been calculated for the following initial parameters: $C_{\text{pNi}} = 462 \text{ J}/(\text{sec} \cdot \text{kg} \cdot \text{K})$, $C_{\text{pAl}} = 900 \text{ J}/(\text{sec} \cdot \text{kg} \cdot \text{K})$, $\rho_{\text{Ni}} = 8800 \text{ kg/m}^3$, $\rho_{\text{Al}} = 2700 \text{ kg/m}^3$, $L_{\text{Al}} = 0.38 \cdot 10^6 \text{ J/kg}$, $\lambda_{\text{Al}} = 94.3 \text{ J}/(\text{sec} \cdot \text{m}^2 \cdot \text{K})$ [4], $Q = 1.38 \cdot 10^6 \text{ J/kg}$, $C_1 = 0.576$, $C_2 = 0.528$, $C_L = 0.05$ [5], $E_T = 100,000 \text{ J/mole}$, $D_{0s} = 6 \cdot 10^{-14} \text{ m}^2/\text{sec}$ [6], $E_L = 58.66 \cdot 10^3 \text{ J/mole}$, $D_{0L} = 0.5 \cdot 10^{-5} \text{ m}^2/\text{sec}$ [7].

Figure 1 presents the calculated thermograms of the initial stage of Ni_3Al synthesis as a function of the power of the external heating source for various dispersities of the nickel powder and constant porosity of the compacted powder. It is established that under the given heating conditions a deviation of 0.005% from the critical power of the source toward one or the other side drastically changes the character of intermetallide synthesis: the slow reaction of intermetallide formation gives way to the rapid regime of thermal explosion. The departure from

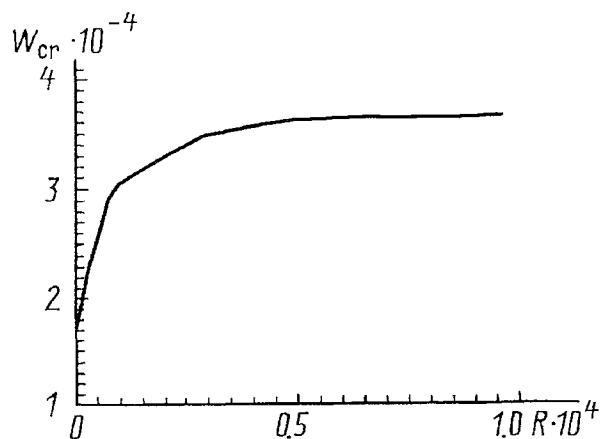


Fig. 2. Critical power, W/m^2 , of the external heating source, required for ignition of a powdered Ni_3Al powder mixture as a function of the radius, m , of the nickel particles at $\alpha = 10 W/(m^2 \cdot K)$.

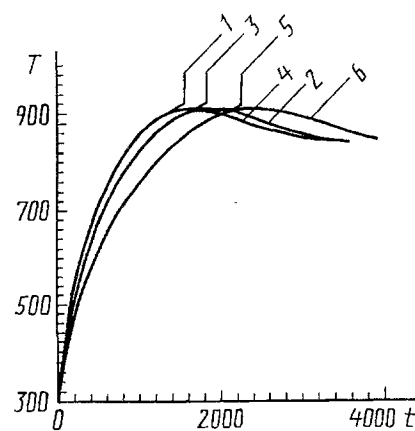


Fig. 3. Calculated thermograms of the initial stage of Ni_3Al synthesis at near-critical powers of the external heating source as a function of the porosity of the a powder mixture of nickel and aluminum (the volume of the compacted powder remains unchanged): 1, 2) $W = 31,288$ and $31,281 W/m^2$ at $P = 0.4$; 3, 4) $W = 30,843$ and $30,812 W/m^2$ at $P = 0.3$; 5, 6) $W = 30,109$ and $30,054 W/m^2$ at $P = 0.1$. T , K ; t , sec .

thermal equilibrium is caused by enhanced heat release due to the appearance of an additional source owing to Ni dissolution in the metallic melt. An increase in the dimension of the powdered refractory component entails an increase in the critical power of the heating source and, correspondingly, in the heating rate of the compacted powder. In this case, the ignition temperature increases as well. Figure 2 shows the critical power of the external heating source as a function of the radius of the nickel particles in a powdered mixture with aluminum. With increasing size of the nickel particles, the critical power of the heating source also increases. This dependence is especially manifested with a change in the diameter of the nickel particles from $1.1 \cdot 10^{-6}$ to $1.0 \cdot 10^{-5} m$, when the rate of increase of the critical power slows down in the particle size range of nickel from $1.0 \cdot 10^{-5}$ to $5 \cdot 10^{-5} m$. A further increase in the nickel particle size exerts a weak influence on the critical power of the heating source. An increase in porosity of the compacted powder entails an increase in the critical power of the heating source and the ignition temperature of the Ni-Al powder mixture (Fig. 3).

To sum up, SHS of an intermetallic compound in a powder mixture of pure elements in continuous heating of the mixture by an external power source may be initiated only at a sufficiently high rate of heating up. The particle size of the refractory component of the powder mixture, exerts the most pronounced influence on the ignition temperature of this mixture. With increasing dispersity of the latter, the ignition temperature and the critical power of the heating source decrease.

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