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Abstract

Full Text

PHYSICAL CHEMISTRY

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ON THE STATIONARY COMBUSTION OF CONDENSED SYSTEMS

(Presented by Academician V. N. Kondrat'ev, 7 IV 1960)

Let us consider the following questions: 1) the dependence of the burning rate of the c-phase (condensed phase) on the experimental conditions; 2) the temperature and substance-concentration profiles in the c-phase; 3) the influence of the d-phase (smoke-gas phase) and the g-phase (gas phase) on thermal processes in the c-phase; 4) the heat and mass balance of the c-phase⁽¹⁾; 5) the critical conditions for stationary burning of the c-phase. We proceed from the equations:

$$\begin{aligned} \lambda T'' - cmT' + Q_1\Phi + kpq_\ell e^{k\rho x} &= 0, & mN'_1 + \Phi &= 0, & N_1 + N_2 &= 1, \\ 1/\rho &= N_1/\rho_0 + N_2RT/PM, & \rho_1 &= N_1\rho, & \rho_2 &= N_2\rho, & (1) \\ \text{with } T(-\infty) &= T_0, & N_1(-\infty) &= 1, & T(0) &= T_s, & N_1(0) &= \mu, \end{aligned}$$

where λ is the coefficient of thermal conductivity; T is temperature (T_0 , initial; T_s , on the surface of the c-phase; T_1 , in the d-phase; T_2 , in the g-phase); c is specific heat; m is the mass burning rate; Q_1 is the thermal effect of complete gasification of the c-substance (cal/g); Φ is the mass rate of transformation of the c-substance (condensed substance) (g/cm³ sec); N_1 and N_2 are the relative concentrations of the c-substance and gas.

$q_\ell(P) = 2\varepsilon^* \sigma T_2^4 [E_3(k_1 P M_1 x_1 / RT_1) - E_3(k_2 P M_2 \Delta x / RT_2)]$ is the radiative heat transfer from the flame of the g-phase^(2,3); ε^* is the degree of blackness of the powder with respect to the flux⁽³⁾; σ is Stefan's constant; $E_3(x) = \int_1^\infty e^{-xy} \frac{dy}{y^3}$; k , k_1 , and k_2 are absorption coefficients in the c-, d-, and g-phases; P is pressure; M , M_1 , and M_2 are the molecular weights of the gas in the c-, d-, and g-phases, respectively; R is the gas constant; x is the coordinate; x_1 is the width of the d-phase; Δx is the width of the flame zone; μ is the relative concentration of c-substance on the surface of the c-phase (for colloidal powders⁽¹⁾, the depth of dispersion)*; ρ is the density of the c-phase (ρ_0 , initial); ρ_1 and ρ_2 are the weight concentrations of c-substance and gas, respectively.

It is necessary to find m , $T(x)$, $N_1(x)$, $N_2(x)$, $\rho(x)$, $\rho_1(x)$, and $\rho_2(x)$. A solution exists (and is unique) only for a discontinuous distribution of heat sources in the c-phase (^{4,5}): in the region $T_0 \leq T < T^* < T_s$ we set $\Phi \equiv 0$; otherwise it is impossible to satisfy the boundary conditions at $x = -\infty$.

From (1) we obtain the equation of conservation of the energy flux and the equations of the heat and mass balance of the c-phase:

$$-\lambda T' - q_\ell e^{k\rho x} + mH = \text{const} = H_0 = -q_\tau - q_\ell + mH_s, \quad (2)$$

$$q = cm(T_s - T_0) = q_{\text{chem}} + q_\tau + q_\ell, \quad m(1 - \mu) = \int_{-\infty}^0 \Phi dx,$$

* The dispersion effect was first comprehensively studied by P. F. Pokhil, who placed it at the foundation of ideas about the mechanism of combustion of ballistic powders and dispersed high explosives. The phenomenon of dispersion had been observed earlier by A. V. Belyaev (⁶), K. K. Andreev (⁷), and others.

where $H = cT + Q_1 N_1 + Q_2$ is the specific enthalpy (cal/g); Q_2 is the heat effect of combustion of the gas formed during decomposition of the k-substance; φ'_s is the temperature gradient at the burning surface; q is the rate of heat absorption by the k-phase (cal/cm² · sec); $q_{\text{chem}} = mQ_s$ is the chemical heat release (cal/cm² · sec); $Q_s = Q_1(1 - \mu)$ is the heat effect of the reaction in the k-phase (cal/g); $q_\tau = \lambda\varphi_s = m(cT_s - cT_0 - Q_1 + \mu Q_1) = m(Q_2 + \mu Q_1 - cT_2 + cT_s)$ is the conductive heat transfer (cal/cm² · sec).

We shall define the surface of the k-phase by the condition $\mu = \text{const}(P)$, i.e., μ does not depend on P . For simplicity, put $\Phi = \rho_0 Z_s e^{-E_s/RT_s}$. To solve (1) we use the elegant method of D. A. Frank-Kamenetskii, developed by him for the stationary theory of thermal explosion (⁸):

$$v'' - v' + \delta e^v + \delta_l \psi e^{\psi\xi} = 0, \quad u'' + \delta e^v = 0, \quad (3)$$

$$v(-\infty) = v_0, \quad u(-\infty) = u_0, \quad v(0) = 0, \quad u(0) = 0,$$

where

$$v = (T - T_s)E_s/RT_s^2, \quad u = (N_1 - \mu)Q_1 E_s/cRT_s^2, \quad \xi = cmx/\lambda,$$

$$\delta_l = \frac{q_l E_s}{cmRT_s^2}, \quad \psi = \frac{kp\lambda}{cm}, \quad \delta = \frac{\lambda\rho_0 E_s Q_1 Z_s}{c^2 m^2 RT_s^2} e^{-E_s/RT_s}. \quad (4)$$

Equations (2) take the form

$$-v' - \delta_l e^{\psi \xi} + h = \text{const},$$

$$-v_0 = u_0 + \omega_s + \delta_l, \quad u_0 = \delta \int_{-\infty}^0 e^v d\xi,$$

where

$$h = (H - H_s)E_s/cRT_s = u + v, \quad \omega_s = \lambda \varphi_s E_s / cmRT_s^2.$$

The parameter δ does not contain T_0 ; therefore we shall also use a second, sometimes more convenient, parameter

$$\delta^0 = \delta(\min v_0)^{-2} = \delta(4RT_0/E_s)^2.$$

Solving (3) approximately, according to the Zel' dovich scheme ⁽⁵⁾, we obtain (for $q_l \simeq 0$)

$$\xi = \int_0^v \frac{dv}{v + \sqrt{\omega_s^2 + 2\delta(1 - e^v)}}, \quad u = -\omega_s + \sqrt{\omega_s^2 + 2\delta(1 - e^v)},$$

$$\delta = \frac{1}{2}(v_0^2 - \omega_s^2) = -u_0 \left(v_0 + \frac{1}{2}u_0 \right) =$$

$$= \frac{E_s^2 Q_1^2 (1 - \mu)(\mu - \mu_0)}{2c^2 R^2 T_s^4}, \quad (5)$$

$$m^2(T_s, T_0, \mu) = \frac{2\lambda \rho_0 R T_s^2 Z_s}{E_{sQ} 1(1 - \mu)(\mu - \mu_0)} e^{-E_s/RT_s},$$

where $\omega_s = -v_0 - u_0$, $\mu > \mu_0 = 1 - 2c(T_s - T_0)/Q_1$.

To determine $v(\xi)$, numerical integration is necessary. The temperature T_s is one of the most important characteristics of the k-phase, since the burning (decomposition) rate of the k-phase depends primarily on T_s . With increasing T_s or T_0 , the burning rate increases. With increasing μ (at $T_s, T_0 = \text{const}$) the burning rate at first decreases, and then, after passing through a minimum, increases.

Equation (3) can be solved approximately analytically. In the case of Arrhenius kinetics the temperature within the chemical layer changes

only by 3-5% ⁽³⁾. Therefore $v(\xi)$ near the surface is approximated by the parabola $v^0 = \omega_s \xi - \frac{1}{2}(\delta - \omega_s)\xi^2$. Substituting v^0 into e^v and solving (3), we

obtain the second approximation (for the region where the parabola accumulates a substantial error, heat release decreases so much that it practically ceases to affect the profiles of temperature and concentrations):

$$v = -(\omega_s + \delta_l)(1 - e^\xi) - \delta_l(e^{\psi\xi} - e^\xi)/(\psi - 1) - (\sqrt{\pi_s}\delta\beta/\omega_s) [e^{\beta^2}(\operatorname{erf}(\beta - \gamma\xi) - \operatorname{erf}\beta) - e^{\xi + \beta_0^2}(\operatorname{erf}(\beta_0 - \gamma\xi) - \operatorname{erf}\beta_0)], \quad (6)$$

$$u = (\sqrt{\pi}\delta\beta/\omega_s)e^{\beta^2}(\operatorname{erf}(\beta - \gamma\xi) - \operatorname{erf}\beta),$$

$$\sqrt{\pi}\delta\beta(1 - \operatorname{erf}\beta)e^{\beta^2} = -\omega_s(v_0 + \omega_s),$$

where

$$\beta^2 = \frac{\omega_s^2}{2(\delta - \omega_s)}, \quad \beta_0^2 = \frac{(\omega_s - 1)^2}{2(\delta - \omega_s)}, \quad \gamma^2 = \frac{\delta - \omega_s}{2}, \quad \operatorname{erf}\beta = \frac{2}{\sqrt{\pi}} \int_0^\beta e^{-y^2} dy.$$

The parameter m and all functions are obtained analytically from (6). The principle of constancy of enthalpy, valid in gases if the Lewis-Landau number $L = 1$, is never satisfied in the k-phase. As $\psi \rightarrow \infty$ (an absolutely black body), q_l is absorbed at the surface, and the fluxes q_t and q_l are simply added. As $\psi \rightarrow 0$ (transparent powder), the initial temperature is as though increased to $T'_0 = T_0 + q_l/cm$.

The d-phase affects the thermal processes in the k-phase through T_s and φ_s , and the g-phase—through T_s and q_l (3):

$$T_s = T_0 + Q_s/c + \lambda\varphi_s/cm + q_l/cm, \quad T_1 = T_2 - Q_2/c + \lambda\varphi_1/cm + q_l/cm,$$

$$\lambda\varphi_s = (Q_2 + \mu Q_1 - cT_2 + cT_s)B_2P^{n_2/2}e^{-E_s/2RT_s},$$

$$\lambda\varphi_1 = (Q_2 - cT_2 + cT_1)B_2P^{n_2/2}e^{-E_2/2RT_2}, \quad (7)$$

$$T_s \simeq C_1(C_2 - \ln P)^{-1}, \quad T_1 \simeq C_1^*(C_2^* - \ln P)^{-1},$$

where φ_1 is the temperature gradient at the boundary between the d- and g-phases; B_2 is the Zeldovich factor (3) for the g-phase; $C_1 = E_s/n_2R$; $C_2 = E_s/n_2RT_s^0$, $T_s^0 = T_s$ (1 atm); $C_1^* = E_1/R(n_2 - n_1)$; $C_2^* = E_1/RT_1^0(n_2 - n_1)$;

$T_1^0 = T_1$ (1 atm); n_1 and n_2 are the reaction orders, respectively, in the d- and g-phases.

The formula $T_s(P)$ agrees with experiments ⁽¹⁾, $\varphi_s(P)$ was confirmed experimentally ⁽⁹⁾; $T_1(P)$ and $\varphi_1(P)$ are a theoretical prediction.

Introduce the parameters of the heat balance of the k-phase $\alpha_{\text{chem}} = q_{\text{chem}}/q$; $\alpha_t = q_t/q$; $\alpha_l = q_l/q$; $\alpha_{\text{chem}} + \alpha_t + \alpha_l = 1$. From (2) and (7) we obtain the heat balance of the k-phase as a function of pressure

$$\alpha_{\text{chem}}(P) = \frac{Q_1(1-\mu)}{c[T_s(P) - T_0]}, \quad \alpha_l(P) = \frac{q_l(P)P^{-n_2/2}}{cB_2[T_s(P) - T_0]} e^{E_2/2RT_2},$$

$$\alpha_t(P) = \frac{\mu Q_1 + Q_2 - cT_2 + cT_s(P)}{c[T_s(P) - T_0]} - \alpha_l(P). \quad (8)$$

The result (8) was confirmed experimentally by P. F. Pokhil, V. M. Mal' tsev, and G. V. Lukashenya. For stable burning of fuel in rocket chambers under various conditions, different regimes of the heat balance of the k-phase (8) are advantageous.

According to (5), the values of δ at which the solution can satisfy the boundary conditions do not exceed a certain critical value

$$\delta \leq \delta_{\text{cr}}(T_0) = E^2/32R^2T_0^2, \quad \delta^0 \leq \delta_{\text{cr}}^0 = 0.5. \quad (9)$$

If only T_s is varied, then

$$\delta^0 \leq \delta_{\text{cr}}^0(T_0, \mu) = c^2T_0^2Q_s/2 \left(\frac{2}{3}cT_0 + \frac{1}{3}Q_s \right)^3. \quad (10)$$

If the characteristic scale, the width of the thermal layer $x_T = \lambda/cm$, assumes a value greater than the critical one, then, according to (9) and (10), breakdown of the stationary regime occurs. The parameter δ determines the ratio of two most important scales: the width of the thermal layer to the width of the chemical layer x_{chem} ⁽³⁾. Stationary combustion is possible only for definite relations between the two scales, $x_T/x_{\text{chem}} \leq E_s/4RT_0$. To determine strictly what exactly will occur—the transition of combustion to detonation or extinction—one must solve the nonstationary problem. However, for flameless combustion, when $\delta^0 > 0.5$, extinction occurs, explained by the dissipation in the bulk of the k -phase of heat that had been concentrated in a thin layer at the burning surface of the k -phase ⁽¹⁰⁾.

Considering, by the Frank-Kamenetskii method, the problem of combustion of a cylinder of diameter D with heat losses to the environment, we obtain another criterion for stationary combustion,

$$D \geq D_{\text{cr}} = \frac{2\lambda r_{\text{cr}}(T_0, \text{Bi}, \delta_{\text{cr}})}{cm_{\text{cr}}} = \sqrt{\frac{4\lambda r_{\text{cr}}^2(T_0, \text{Bi}, \delta_{\text{cr}})\delta_{\text{cr}}RT_{\text{scr}}^2 e^{E_s/2RT_{\text{scr}}} P^{-n_1/2}}{\rho Q_1 E_s Z S}} \quad (11)$$

where $r = cmD/2\lambda$; $\text{Bi} = \eta/cm$ is the Biot criterion; η is the heat-transfer coefficient. Stationary combustion is possible only for definite relations among three scales: D , x_T , and x_{chem} . The result $D_{\text{cr}}m_{\text{cr}} = \text{const}$ was previously obtained from dimensional considerations by A. D. Margolin*. Formula (11) agrees with experiments^(1,11).

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* Unpublished.

Note: Figure translations are in progress. See original paper for figures.

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