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MASS TRANSFER IN A SOLID PARTICLE WITH COMPETING REACTIONS WITH A MULTICOMPONENT GAS MIXTURE

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A macrokinetic model of the transformation of a solid particle, reacting with a multicomponent gas mixture, is constructed for arbitrary ratios between the rates of the mass-transfer stages of the transformation process (sorption, dissolution, and diffusion of the starting and final products).

Processes for working solid dispersed materials with multicomponent gaseous mixtures are widely used in modern technology. In the general case a macrokinetic model of the transformation of a solid particle reacting in the atmosphere of a gaseous mixture must take into account all elementary mass-transfer stages of the reaction: sorption–desorption of reagents and reaction products from both phases and their dissolution and diffusion in the solid particle. Well-known theoretical studies [1, 2] usually presume that there exists one limiting stage of mass transfer, which is insufficient for describing reactions of practical interest. The model of the solid-phase transformation, constructed in [3, 4] and presuming that the rates of several stages are comparable, must be generalized to the case of the interaction of solid spherical particles with gaseous mixtures. Modeling such processes enables the calculation and optimization of different states as well as the intensification of the interaction of solid particles with the gas phase by increasing the partial pressures of gaseous reagents or by changing the composition of the gaseous mixture.

In studying a gas mixture we assume that we have N gaseous reagents and that correspondingly, N gaseous reaction products form. Under the assumptions made in [3, 4], we assume that the chemical reaction involved in the interaction of the solid reagent with each gaseous reagent itself proceeds much more rapidly than the mass transfer processes, and the reaction front separates the region of the starting reagent and the solid product of the reaction. Analogously to [3, 4] we shall formulate the equations of kinetics of all stages of the process.

Defining S_i and S_i' as the relative fractions of the area of the surface layer filled with

the i-th reagent and the corresponding reaction product, we introduce the free surface area ΔS, which is not filled with molecules of the reagents and reaction products. Assuming that the adsorption is independent (Langmuir adsorption) the equations describing the kinetics of filling of the surface layer have the form

$$\frac{dS_i}{dt} = \alpha_i \Delta S - \beta_i S_i + \frac{\delta S_i}{\delta t}, \tag{1}$$

$$\frac{dS_i'}{dt} = -\beta_i' S_i' + \frac{\delta S_i'}{\delta t}, \quad i = 1, \ldots, N,$$

$$\Delta S = 1 - \sum_{i=1}^{N} (S_i + S_i'). \tag{2}$$

The surface concentrations f_i and f_i' of the reagents and reaction products equal S_i/l_i^2 and $S_i'/l_i'^2$, respectively. The equations describing the kinetics of transfer between the surface layer and the solid phase are

$$\frac{\delta f_i}{\delta t} = -k_{1i} f_i + k_{2i} l_i c_{0i} \Delta S = -D_i \left. \frac{\partial c_i}{\partial r} \right|_{r=R_0},$$

$$\frac{\delta f_i'}{\delta t} = -k_{1i}' f_i' + k_{2i}' l_i' c_{0i}' \Delta S = -D_i' \left. \frac{\partial c_i'}{\partial r} \right|_{r=R_0}, \tag{3}$$

$$c_{0i} = c_i |_{r=R_0}, \quad c_{0i}' = c_i' |_{r=R_0}, \quad i = 1, \ldots, N.$$

The boundary conditions on the reaction front are

$$c_i = 0, \quad -D_i \frac{\partial c_i}{\partial r} = v_i D_i' \frac{\partial c_i'}{\partial r}, \quad r = R(t), \quad i = 1, \ldots, N, \tag{4}$$

where v_i are the corresponding stoichiometric coefficients. From the conditions of material balance on the reaction front we have

$$\varphi \frac{dR(t)}{dt} = -\sum_{i=1}^{N} D_i \left. \frac{\partial c_i}{\partial r} \right|_{r=R(r)}, \tag{5}$$

where φ is the concentration of the reagent present in the solid phase multiplied by the sum of the stoichiometric coefficients of separate chemical reactions. Taking into account the fact that $\varphi \gg c_{0i}$, we find that the relaxation time of the concentration fields of the reagents and reaction products is much shorter than the displacement time of the reaction front (see [3]). Therefore in the quasistationary representation

$$c_i = c_{0i} F(r, R), \quad c_i' = c_{0i}' + (D_i/D_i') c_{0i} F(r, R),$$

$$F(r, R) = \left(\frac{1}{R} - \frac{1}{r} \right) \left(\frac{1}{R} - \frac{1}{R_0} \right), \quad i = 1, \ldots, N. \tag{6}$$

Transforming (1)-(6) we obtain a system of equations which completely describes the process

$$\frac{dS_i}{dt} = \alpha_i \Delta S - \beta_i S_i - \frac{k_{1i} D_i}{k_{2i} l_i \Delta S R_* + D_i} S_i,$$

$$\frac{dS_i'}{dt} = -\beta_i' S_i' + \frac{k_{1i} D_i}{k_{2i} l_i \Delta S R_* + D_i} \frac{l_i'^2}{v_i l_i^2} S_i,$$

$$R_* = R_0 (R_0 R^{-1} - 1), \quad i = 1, \ldots, N,$$

$$\Delta S = 1 - \sum_{i=1}^{N} (S_i + S_i'), \tag{7}$$

$$\frac{dR}{dt} = -\left(\sum_{i=1}^{N} \frac{k_{1i} D_i}{k_{2i} l_i \Delta S R_* + D_i} \cdot \frac{S_i}{l_i^2} \right) \frac{1}{\varphi} \left(\frac{R_0}{R} \right)^2.$$

In order to introduce dimensionless variables we single out the reagent, whose pressure in the gaseous mixture differs from zero; for definiteness we label this gas with the index i = 1. In this case

$$\zeta = \frac{R}{R_0}, \ \lambda_i = \frac{\alpha_i}{\alpha_1}, \ \tau = \alpha_1 t,$$

$$\left\{ \begin{array}{c} b_i \\ b_i' \end{array} \right\} = \frac{1}{\alpha_1} \left\{ \begin{array}{c} \beta_i \\ \beta_i' \end{array} \right\}, \ \varkappa_i = \frac{l_i'^2}{v_i l_i^2},$$

$$K_i = \frac{k_{1i}}{\alpha_1}, \ L_i = \frac{k_{2i} l_i R_0}{D_i},$$

$$M_i = \frac{1}{R_0 \varphi l_i^2}, \ i = 1, \ldots, N,$$

(8)

and the system assumes the form

$$\frac{dS_i}{d\tau} = \lambda_i \Delta S - b_i S_i - \frac{K_i}{L_i (\zeta^{-1} - 1) \Delta S + 1} S_i,$$

$$\frac{dS_i'}{d\tau} = - b_i' S_i' + \frac{\varkappa_i K_i}{L_i (\zeta^{-1} - 1) \Delta S + 1} S_i,$$

$$i = 1, \ldots, N, \ \Delta S = 1 - \sum_{i=1}^{N} (S_i + S_i'),$$

$$\frac{d\zeta}{d\tau} = - \left(\sum_{i=1}^{N} \frac{K_i M_i}{L_i (\zeta^{-1} - 1) \Delta S + 1} S_i \right) \frac{1}{\zeta^2}.$$

(9)

We write the initial conditions

$$S_i = S_i' = 0, \ i = 1, \ldots, N, \ \zeta = 1, \ \tau = \tau_0 \geqslant 0. \tag{10}$$

Let us examine the effect of the composition of the gas phase on the kinetics of the process. Initially for small τ we have

$$S_i \approx \lambda_i \tau, \ S_i' \approx (\lambda_i \varkappa_i K_i / 2) \tau^2,$$

$$\zeta \approx \left[1 - \frac{3}{2} \left(\sum_{i=1}^{N} \lambda_i K_i M_i \right) \tau^2 \right]^{1/3}. \tag{11}$$

Here and below we shall study the degree of transformation $A = 1 - \zeta^3$. We obtain

$$A \approx \frac{3}{2} \left(\sum_{i=1}^{N} \lambda_i K_i M_i \right) \tau^2 = \frac{3}{2} \left(\sum_{i=1}^{N} \frac{k_{1i} \alpha_i}{R_0 \varphi l_i^2} \right) t^2,$$

$$\frac{dA}{dt} \approx 3 \left(\sum_{i=1}^{N} \frac{k_{1i}}{R_0 \varphi l_i^2} \alpha_i \right) t.$$

(12)

Thus at the initial stage we observe the dependence of the rate of the process on the characteristic features of the adsorption (on the magnitude of the coefficient of the rate of adsorption and the dimensions of the area occupied by the adsorbed reagent molecules) and dissolution stages. In the simplest case, when adsorption proceeds without dissociation, α_i is proportional to the partial pressure of the i-th reagent, and therefore the addition of a reagent which is a more active adsorbent, while holding constant the total pressure of the gas phase, enables substantially raising the initial rate of the process. A more complete analysis requires taking into account the dissociation of the adsorbed reagent (using, for example, Siewert's law) and the dependence of the coefficients of the adsorption rate on the pressure of other reagents (using, for example, Lewis' relation). We shall study below in detail the case of dissociation of adsorbed molecules of one of the reagents.

To study the process at subsequent times we shall examine the quasistationary state,

neglecting the rates of change of the relative fractions of the areas of the surface layer occupied by reagents and reaction products, and assuming that the relative fraction of the area which remains free is constant. In this case

$$S_i = \lambda_i \Delta S \left[b_i + \frac{K_i}{L_i \left(\zeta^{-1} - 1 \right) \Delta S + 1} \right]^{-1},$$

$$S_i' = \frac{\varkappa_i K_i}{b_i' \left[L_i \left(\zeta^{-1} - 1 \right) \Delta S + 1 \right]} S_i, \tag{13}$$

$$\frac{d\zeta}{d\tau} = - \left\{ \sum_{i=1}^{N} \frac{\lambda_i M_i \Delta S K_i}{b_i \left[L_i \left(\zeta^{-1} - 1 \right) \Delta S + 1 \right] + K_i} \right\} \frac{1}{\zeta^2}.$$

From (2) and (13) we have

$$\Delta S = \text{const} = 1 - \sum_{i=1}^{N} (S_i + S_i')|_{\zeta=1} = 1 - \left[\sum_{i=1}^{N} \left(1 + \frac{\varkappa_i K_i}{b_i'} \right) \frac{\lambda_i}{b_i + K_i} \right] \Delta S,$$

whence

$$\Delta S = \left[1 + \sum_{i=1}^{N} \left(1 + \frac{\varkappa_i K_i}{b_i'} \right) \frac{\lambda_i}{b_i + K_i} \right]^{-1}. \tag{14}$$

For low degrees of transformation ($\zeta \approx 1$)

$$\frac{d\zeta}{d\tau} = - \left(\sum_{i=1}^{N} \frac{K_i M_i \lambda_i}{b_i + K_i} \Delta S \right) \frac{1}{\zeta^2} =$$

$$= - \left(\sum_{i=1}^{N} \frac{K_i M_i \lambda_i}{b_i + K_i} \right) \left[1 + \sum_{i=1}^{N} \frac{\lambda_i}{b_i + K_i} \left(\frac{\varkappa_i K_i}{b_i'} + 1 \right) \right]^{-1} \frac{1}{\zeta^2}, \tag{15}$$

$$A = 3 \left(\sum_{i=1}^{N} \frac{K_i M_i \lambda_i}{b_i + K_i} \right) \left[1 + \sum_{i=1}^{N} \frac{\lambda_i}{b_i + K_i} \left(\frac{\varkappa_i K_i}{b_i'} + 1 \right) \right]^{-1} \tau =$$

$$= 3 \left(\sum_{i=1}^{N} \frac{k_{1i} \alpha_i}{l_i^2 (\beta_i + K_{1i})} \right) \left[1 + \sum_{i=1}^{N} \frac{\alpha_i}{\beta_i + k_{1i}} \left(\frac{l_i'^2}{v_i l_i^2} \frac{k_{1i}}{\beta_i'} + 1 \right) \right]^{-1} \frac{t}{R_0 \varphi},$$

$$\frac{dA}{dt} = 3 \left(\sum_{i=1}^{N} \frac{k_{1i} \alpha_i}{l_i^2 (\beta_i + k_{1i})} \right) \left[1 + \sum_{i=1}^{N} \frac{\alpha_i}{\beta_i + k_{1i}} \left(\frac{l_i'^2}{v_i l_i^2} \frac{k_{1i}}{\beta_i'} + 1 \right) \right]^{-1} \frac{1}{R_0 \varphi}.$$

Therefore the addition of a more active adsorbent, which increases the rate of the process initially, can at this stage, on the contrary, reduce it, if, for example, the reaction product is characterized by a low rate of desorption into the gas phase.

At the final stages when $\zeta^{-1} \gg 1$

$$S_i' = 0, \quad \lambda_i \Delta S - b_i S_i = 0,$$

$$\frac{d\zeta}{d\tau} = - \left(\sum_{i=1}^{N} \frac{K_i M_i \lambda_i}{b_i L_i} \right) \frac{1}{\zeta}. \tag{16}$$

Since the time for completion of the process is determined primarily by the duration of the last stage, to intensify the process reagent with an optimum combination of the rate coefficients of sorption–desorption, dissolution, and diffusion must be added (see (16)).

To determine the time dependence of the coordinates of the reaction front we shall study a mixture consisting of two gases. We write

$$x_i = 1 + \frac{b_i}{K_i} - \frac{b_i}{K_i} L_i \Delta S,$$

$$y_i = \frac{b_i}{K_i} L_i, \quad z_i = \lambda_i M_i, \quad i = 1, 2, \tag{17}$$

where ΔS is defined in (14). Integrating the last of the equations in (13), we obtain the equation of motion of the reaction front
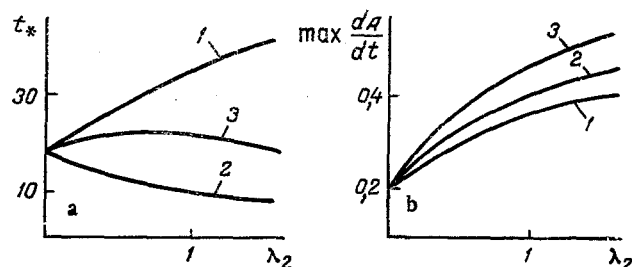
Fig. 1. Effect of the partial pressure of one reacting gas on the kinetics of the process: dependence of the completion time of the process (a) and the maximum of the rate (b) on $\lambda_2 = \alpha_2 / \alpha_1$ with $\alpha_1 = \text{const}$ with $\alpha_1 = 1.0$ min^{-1} it was assumed that $K_1 = 0.03$, $K_2 = 0.07$, $L_1 = 0.75$, $L_2 = 0.75$, $M_1 = 0.33$, $M_2 = 0.33$, $b_1 = 0.03$, $b'_1 = 0.05$, $b_2 = 0.03$, $x_1 = 1.0$; 1-3) values of $b'_2 = 0.01$, 0.1, 0.5; $x_2 = 0.5$, 1.0, 3.0, t_*, min.

$$\frac{1}{3}\,\frac{x_1 x_2}{x_1 z_2 + x_2 z_1}\,(\zeta^3 - 1) + \frac{1}{2}\,\frac{x_1^2 y_2 z_2 + x_2^2 y_1 z_2}{(x_1 z_2 + x_2 z_1)^2}\,(\zeta^2 - 1) -$$

$$- \frac{z_1 z_2 (x_1 y_2 - x_2 y_1)^2}{(x_1 z_1 + x_2 z_1)^3}\,(\zeta - 1) + \frac{z_1 z_2 (x_1 y_2 - x_2 y_1)^2 (z_1 y_2 + z_2 y_1)}{(x_1 z_2 + x_2 z_1)^4}$$

$$\ln\left|\left(\zeta + \frac{z_1 y_2 + z_2 y_1}{x_1 z_2 + x_2 z_1}\right)\left(1 + \frac{z_1 y_2 + z_2 y_1}{x_1 z_2 + x_2 z_1}\right)^{-1}\right| = \Delta S\,(\tau_0 - \tau). \tag{18}$$

From the formula (18) one can derive a generalization of the Ginstling–Brounshtein formula, presented in [4], by assuming that only one reagent gas is present in the atmosphere (in this case $\alpha_2 = \lambda_2 = z_2 = 0$) or assuming that the reagents have the same kinetic characteristics ($x_1 = x_2$, $y_1 = y_2$). Thus the formula (18) is a generalization of the Ginstling-Brounshtein formula to the case of the interaction of a particle with a gaseous mixture. The optimal composition of the gas mixture can be determined by solving the problem of minimizing the time for completion of the process t_*, obtained from (18) by substituting (8) and (17) with the corresponding restrictions (for example, leaving the pressure of the gas mixture unchanged).

The numerical study of the system (9) enables analyzing the effect of different kinetic coefficients and external conditions, in particular, the particle sizes and the partial pressures of the reacting gases. Figure 1 shows the dependence of the completion time and the maximum rate on the increase in the pressure of one of the reagents (with the partial pressures of the other components of the gas mixture held fixed).

The model constructed explains the behavior of the kinetic curves of the processes involved in the interaction of solid substances with mixtures of gaseous reagents and enables optimizing the composition of the gas mixture. Since in most experimental studies the kinetic parameters are either unknown or are not presented by the authors, it is difficult to perform theoretical calculations which fully reflect real situations. The behavior of the process can nevertheless be analyzed qualitatively and the optimal technological conditions can be conjectured based on the agreement between the computed and experimental curves.

We shall study the problem of reducing the oxides of metals, which is of important practical value. Thus in [5, 6] a great deal of attention is devoted to the study of the effect of the change in the composition of the gas phase (mixture of hydrogen with carbon monoxide) on the kinetics of the reduction of iron-ore pellets. Figure 2 shows the obtained dependence of the kinetics of reduction on the composition of the gas mixture (the broken curves are the computed curves). The authors correctly call attention to the importance of taking into account all mass-transfer factors: adsorption–desorption, dissolution and diffusion of the starting and final products. Since adsorption of hydrogen usually proceeds with dissociation, while the adsorption of carbon monoxide proceeds without it.
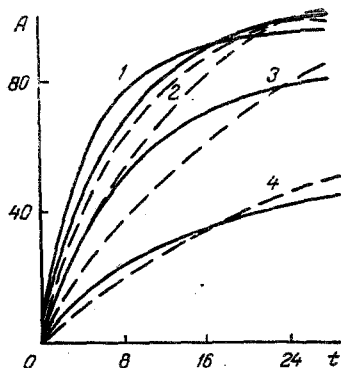
Fig. 2. Effect of the
composition of the gas
phase on the kinetics of
reduction of magnetic
pellets under a pressure
of 1 atm and at a temper-
ature of 1000°C, % H_2 + % CO:
$1 - 100 + 0$; $2 - 75 + 25$; $3 - 50 + 50$;
$4 - 0 + 100$; the solid curves
are the experimental curves
([6]) and the broken curves
are the computed curves.
With $\alpha_1 = 1,0$ min^{-1}, the follow-
ing values were used: $K_1 = 0,1$, $K_2 =$
$0,08$, $L_1 = 0,75$, $L_2 = 0,75$, $M_1 = 0,33$, $M_2 = 0,33$,
$b_1 = 0,5$, $b_2 = 0,01$, $b_1' = 0,1$, $b_2' = 0,03$, $\varkappa_1 = 1,0$,
$\varkappa_2 = 1,0$; $1 - 4$: $\alpha_1 = 30$, $22,5$, $15,0$; $\alpha_2 = 0$,
$7,5$, 15, 30 A, %: t, min.

$$\alpha_1 = a_1 p_1^{1/2}, \quad \alpha_2 = a_2 p_2. \tag{19}$$

Here and below the index 1 refers to hydrogen and the index 2 refers to carbon monoxide.
We write the condition for the pressure of the gas phase to remain constant as follows:

$$p_1 + p_2 = p = \text{const.} \tag{20}$$

We obtain the rate of reduction at the initial stage from (12), substituting (19),

$$\frac{dA}{dt} = \frac{3}{R_0 \varphi} \left(\frac{k_{11} a_1}{l_1^2} p_1^{1/2} + \frac{k_{12} a_2}{l_2^2} p_2 \right) t . \tag{21}$$

The maximum rate at the initial stage (taking into account (20)) is reached when

$$p_2^* = \max \left\{ 0, \; p - \left[\left(2 \frac{k_{11} a_1}{l_1^2} \right) \left(\frac{k_{12} a_2}{l_2^2} \right)^{-1} \right]^2 \right\}. \tag{22}$$

If the total pressure p is not too high, then $p_2^* = 0$, i.e., an atmosphere of pure hydrogen
will be optimal at the initial stage. But if p is greater than some critical value, depending
on the properties of the gases, then a mixture of hydrogen and carbon monoxide with a definite
composition will be best.

At the final stage, from (16) and (19) we have

$$\frac{dR}{dt} = - \left[\frac{k_{11} D_1 a_1}{\beta_1 k_{21} l_1^3} p_1^{1/2} + \frac{k_{12} D_2 a_2}{\beta_2 k_{22} l_2^3} p_2 \right] \frac{R}{R_0^2 \varphi} , \tag{23}$$

and in order to intensify the process it is necessary to select the composition of the mixture
with partial hydrogen pressure equal to

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$$p_1^* = \max\left\{p, \left[\left(2\frac{k_{11}D_1a_1}{\beta_1 k_{21} l_1^3}\right)\left(\frac{k_{12}D_2a_2}{\beta_2 k_{22} l_2^3}\right)^{-1}\right]^2\right\}. \tag{24}$$

In the experiments (in particular, in [6]) a definite optimal composition of the gas phase is indeed observed. In this case

$$p_1^* = \left[\left(2\frac{k_{11}D_1a_1}{\beta_1 k_{21} l_1^3}\right)\left(\frac{k_{12}D_2a_2}{\beta_2 k_{22} l_2^3}\right)^{-1}\right]^2. \tag{25}$$

Since the duration of the process depends primarily on the duration of the final stage, the optimal composition of the gas phase is determined by the formula (25). It should be noted that the optimal partial pressure of hydrogen is independent of the pressure of the gas mixture and the addition of hydrogen after some critical value is attained, determined by (25), only reduces the reaction rate.

In conclusion, we note that from the results of this work it is not difficult to describe the processes accompanying the inhibiting action of one of the reagent gases or reaction products. Such an effect actually occurs, in particular, in cases when the desorption rate of one of the reaction products into the gas phase is low, even if the corresponding reagent gas actively participates in the reaction (for example, it is a good reducer and is rapidly transported to the reaction front). Inert gases present in the starting mixture even in insignificant quantities, can have an inhibiting effect, if they are actively sorbed onto the surface of the particles, thereby poisoning the mixture. It is not difficult to describe these and some other effects quantitatively based on the theory developed above.

NOTATION

A, degree of transformation; a, adsorption rate constant; c, concentration; D, diffusion coefficient; f, surface concentration; b, K, L, M, dimensionless parameters defined in (8); k_1, k_2, rate constants for adsorption and desorption into the solid phase; l, characteristic linear size of a molecule in the surface layer; N, total number of reagents; p, total pressure of the gas mixture; p_i, partial pressure; p_i^*, optimal partial pressure; R and R_0, radius of the reaction front and of the particle, respectively; S, relative filling of the surface layer; t, time; t_*, conversion time; x, y, and z, quantities defined in (17), α, rate constant of adsorption from the gas phase; β, rate constant of desorption into the gas phase; ΔS, unfilled fraction of the surface layer; κ, λ, dimensionless parameters defined in (8); ν, stoichiometric constant; ξ, a radial coordinate; τ, dimensionless time; τ_0, dimensionless duration of the incubation period; φ, concentration of the starting solid reagent. Indices: i, quantities referring to the corresponding reagents from the gas phase, the prime refers to the gaseous reaction products; 0 refers to situations with $R = R_0$; 1 and 2 in (19)–(25) determine quantities referring to hydrogen and carbon monoxide, respectively.

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