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GENERALIZATION OF THE KINETICS OF CHEMICAL DISSOLUTION UNDER CONDITIONS OF GAS FORMATION

G. A. Aksel'rud, A. I. Dubynin, and B. I. Duda

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A generalization has been made of the experimental data on the chemical dissolution of several nonferrous metals and carbonates in solutions of hydrochloric acid in the presence of forced motion of the liquid.

In a number of papers [1-5] it has been shown on the basis of experimental studies that a gaseous phase has a considerable effect on the kinetic relationships for mass transfer, intensifying or retarding the rate of dissolution depending on the particular conditions. These processes have been studied most fully under conditions corresponding to an external problem, i.e., when a solid object dissolves in a stagnant or moving liquid. The same cannot be said for the case of the internal problem, i.e., during the motion of the solvent inside hollow objects of tubular or rectangular cross section. At the same time, chemical dissolution during the motion of a reagent along channels and tubes is quite frequently encountered in processes of chemical milling, in the production of parts for waveguide channels, in the removal of salt incrustations in heat-exchange tubes, etc. This has prompted the authors to consider certain aspects of this problem in more detail on the basis of investigations carried out earlier [6-8].

During the flow of a reagent in a channel or tube a stream is formed whose hydrodynamics are greatly complicated (especially in the layers near the wall) as a result of the gaseous products which are liberated at the reaction surface. The gas bubbles, which are generated on the surface at which the phases interact, grow until some diameter is reached, and then under the influence of the hydrodynamic head of the liquid and the buoyancy forces they break away and are carried off in the core of the flow. The periodic processes of growth and separation leads to a partial disturbance of the diffusion layer and to the turbulization of this layer, a reduction in the resistance to mass transfer, and an intensification of the dissolution process in the case of reactions which are controlled by diffusion. The liquid velocity also shows the same qualitative effects on mass transfer. However, in quantitative terms, the effects of the action of the gas phase and of a forced flow are not the same and differ depending on the concentration. At small concentrations of the reagent the hydrodynamic effects caused by the reduction of the thickness of the diffusion boundary layer predominate. At relatively large concentrations, on the other hand, the effect of turbulization of the layer by the gas bubbles predominates. It is obvious that at some critical concentration the mass flow rate of the gas becomes so large that the rate of the heterogeneous phase interaction

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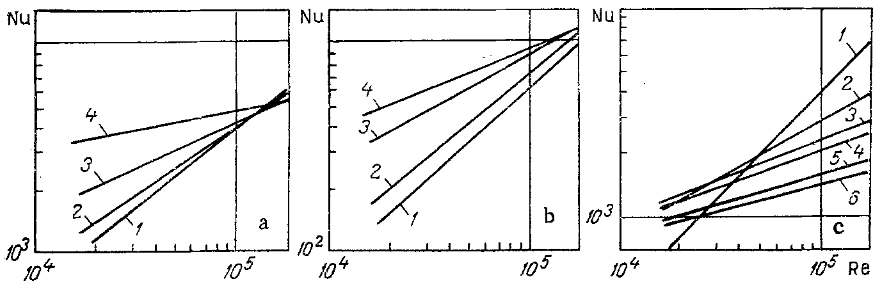

Fig. 1. Mass transfer during the chemical dissolution of magnesium (a), zinc (b), and calcium carbonate (c) in hydrochloric acid solutions of various concentrations during the forced circulation of the reagent: a. Curve 1: C_R = 3.30 kg/m^3; 2) 6.39; 3) 25.5; 4) 50.3. b. Curve 1: C_R = 10.2 kg/m^3; 2) 27.7; 3) 60.2; 4) 90.1. c. Curve 1: C_R = 1.06 kg/m^3; 2) 10.5; 3) 20.1; 4) 40.1; 5) 60.3; 6) 80.0.

no longer depends on the hydrodynamic regime of the liquid flow, but is governed by the rate of production of the gas phase.

The considerations outlined above are clearly confirmed by the graphical relationships shown in Fig. 1. Here use has been made of results from investigations [6-8] into the dissolution of tubular samples of diameter 40 mm and length 90 mm over a wide range of flow velocities of the reagent (0-4 m/sec) at a constant temperature of 30°C.

The dimensionless groups which govern the kinetics of the process are the following:

$$\mathrm{Nu} = \frac{Kd}{D_R}, \quad \mathrm{Re} = \frac{Wd}{\nu}, \quad \mathrm{Ja} = \frac{nC_R}{\rho_g}\left(\frac{D_R}{D_g}\right)^{2/3}.$$

The nature of the simultaneous effect of the gas phase and the reagent velocity on the dissolution process, which is of the same type for all three systems, makes it possible to carry out a further generalization of the results in the form of the relationship

$$\mathrm{Nu} = A(\mathrm{Ja})\,\mathrm{Re}^{m\,(\mathrm{Ja})}. \tag{1}$$

The Jacob number which appears in Eq. (1) defines the role of the gas phase in the mass transfer kinetics in accordance with the conditions mentioned earlier.

After making the necessary numerical calculations on a computer, the correlation assumed the forms:

for magnesium:

$$\mathrm{Nu} = P_3(\mathrm{Ja})\,\mathrm{Re}^{0{,}761\exp(-0{,}1\,\mathrm{Ja})}, \tag{2}$$
$$P_3(\mathrm{Ja}) = 0.087 + 1.957\,\mathrm{Ja} - 1.026\,\mathrm{Ja}^2 + 0.245\,\mathrm{Ja}^3,$$
$$\mathrm{Ja} = 0.7 - 13.2,\ \mathrm{Re} = (1{,}6 - 20)\cdot 10^4;$$

for zinc:

$$\mathrm{Nu} = P_3(\mathrm{Ja})\,\mathrm{Re}^{0{,}917\exp(-0{,}037\,\mathrm{Ja})}, \tag{3}$$
$$P_3(\mathrm{Ja}) = 0.092 - 0.0295\,\mathrm{Ja} + 0.00072\,\mathrm{Ja}^2 + 0.00075\,\mathrm{Ja}^3,$$
$$\mathrm{Ja} = 2.2 - 24.8,\ \mathrm{Re} = (1{,}7 - 17)\cdot 10^4;$$

for calcium carbonate:

$$\mathrm{Nu} = A\,\mathrm{Re}^{0{,}625\exp(-0{,}29\,\mathrm{Ja}) - 0{,}003\,\mathrm{Ja} + 0{,}38}, \tag{4}$$
$$A = \left|\begin{array}{ll} 0.0561\exp(0.933\,\mathrm{Ja}) & 0.24 \leqslant \mathrm{Ja} \leqslant 4.73 \\ 23.336\exp(0.029\,\mathrm{Ja}) & 9.36 \leqslant \mathrm{Ja} \leqslant 44{,}5 \end{array}\right. \quad \mathrm{Re} = (1.6 - 18)\cdot 10^4.$$

Equations (2)-(4), which have been obtained, are of interest in that, in addition to their practical use in engineering calculations, they make it possible to evaluate the quantitative effect of gas liberation on the mass-transfer coefficient.

Let us consider Eq. (2), for example. When Ja = 0 (i.e., for $C_R = 0$), it assumes the form

$$\mathrm{Nu} = 0.087\,\mathrm{Re}^{0.761}. \tag{5}$$

The mass-transfer coefficient determined from this relationship is a function of the hydrodynamic conditions only. Such an equation can be used in the case of ordinary physical dissolution.

NOTATION

K, mass-transfer coefficient with respect to the solid phase; d, inside diameter of sample; W, liquid velocity; D_R, D_g, diffusion coefficients of reagent and gas; ν, viscosity of reagent; C_R, concentration of reagent in solution; ρ_g, gas density; n, stoichiometric coefficient.

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