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MASS TRANSFER IN THE MECHANICAL MIXING OF HETEROGENEOUS SYSTEMS

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The influence of mixing on the kinetics and intensity of mass transfer is examined for dissolution and extraction in fluid-solid systems.

Among the mixing and mass transfer processes in apparatus with agitators, the phenomena of mass transport in heterogeneous fluid–solid systems are greatly widespread. However, hydrodynamics investigations only touch upon the "equilibrium" distribution of solid particles over the height of the vessel [1]. Mass transfer questions are also studied inadequately, and are limited to an analysis of the mass elimination coefficients during mixing under stationary conditions [2]. Practice shows that the duration of such processes as extraction or dissolution can be sufficiently large [3]; consequently, it is necessary to know the kinetic regularities of mass transfer during mixing. An attempt is made below to develop a model of mass transfer processes for the mechanical mixing of heterogeneous systems that would permit the investigation of the transport kinetics of solid particles and their mass elimination to the carrying fluid phase, with the change in particle size during dissolution, the system properties, and the geometric characteristics and operating conditions of the equipment taken into account.

Earlier we examined the hydrodynamics models [4] and mixing processes [5] of homogeneous systems. It was shown that the application of cellular circulation material transport models with a computation of the fluxes through the cell on the basis of an analytic spatial model of the hydrodynamics of apparatus with agitators will permit sufficiently efficient investigation of the dynamics of macromixing processes for liquid media in industrial apparatus. The apparatus working space can, when modeling the mixing of fluid–solid systems, also be divided into m cells between which the exchange process for k cells can be written in the form

$$\frac{dc_k}{d\tau} = \frac{Q_k^{\text{in}}}{V_k} c_k^{\text{in}} - \frac{Q_k^{\text{out}}}{V_k} c_k^{\text{out}}, \tag{1}$$

where the magnitudes of the discharges through the cell faces are computed by using the hydrodynamic model [4]. When a solid phase is present in the fluid, it will be transported together with the fluid phase in transport motion, additional inertial motion, and also because of mass elimination on the solid–fluid boundary. The first transfer is described by the model (1). The additional transfer is due to the influence of inertial fields and the difference in the phase densities, where the additional flux in the direction of rotational motion of the carrying phase can be neglected for subsonic speeds. The axial velocity of the "additional" particle deposition or levitation motion can be defined [6, 7] as

$$w_{\text{add}}^z = \frac{10^{[-1.82+(1-c)]}\,\text{Ar}\,\mu c^2}{(36+1.22\sqrt{\text{Ar}})\,\rho_p r_p}, \quad c > 0.7, \tag{2}$$

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$$w^z_{add} = \frac{0.123\,Ar\,\mu c^3}{(36 + 1.22\sqrt{Ar})(1-c)\,\rho_p r_p}, \quad c \leqslant 0.7, \tag{3}$$

whereupon the additional axial flux of the solid phase can be computed

$$Q^z_{add,i} = w^z_{add,i} F_{z,i}. \tag{4}$$

The additional radial flux of the solid phase is determined analogously

$$Q^r_{add,i} = w^r_{add,i} F_{r,i}, \tag{5}$$

but the magnitude of the centripetal acceleration

$$a = w^2_\varepsilon(r)/r \tag{6}$$

is used instead of g in the expression for the Archimedes criterion (see (2)). The change in the quantity of substance in the cell because of mass transfer can be estimated by using the Shchukarev law

$$dM_i = \beta_i F_i (c_{sat} - c_i)\, d\tau. \tag{7}$$

For instance, if $P_{sub,i}$ dissolved particles of radius r_p and density ρ_p each are in the cell at the time under consideration, then their total surface is

$$F_i = \frac{3P_{sub,i}}{\rho_p r_p}. \tag{8}$$

The mass elimination coefficient to the spherical particle can be expressed, in conformity with [8], in the form

$$\beta = \frac{D_A}{r_p} + \frac{0.42 D_A}{r_p}\left(\frac{2\rho w_{rel} r_p}{\mu_c}\right)^{0.5} Pr_d^{0.33}, \tag{9}$$

where the magnitudes of the relative velocity are defined by using the model [4] and the expressions (2) and (3), as

$$w_{rel} = \sqrt{(w_r - w^r_{add})^2 + (w_z + w^z_{add})^2}, \quad \rho_p > \rho;$$

$$w_{rel} = \sqrt{(w_r + w^r_{add})^2 + (w_z - w^z_{add})^2}, \quad \rho_p < \rho.$$

Now, to determine the mass change because of mass elimination it is necessary to take account of the change in particle radius, which can be written as

$$\frac{dr_p}{\beta} = \frac{c_{sat} - c_0}{\rho_p} d\tau, \tag{10}$$

and we obtain after expansion in a Taylor series and taking account of (9)

$$r_p = r_{p,0} + \left[\frac{D_A}{r_{p,0}} + 0.42\left(\frac{\rho w_{rel}}{\mu_c}\right)^{0,5} \frac{D_A Pr_d^{0,33}}{\sqrt{r_{p,0}}}\right] \frac{c_{sat} - c_0}{\rho_p} \Delta\tau$$
$$- \frac{1}{2}\left[\frac{D_A}{r^2_{p,0}} + 0.21\left(\frac{\rho w_{rel}}{\mu_c}\right)^{0,5} \frac{D_A Pr_d^{0,33}}{r_{p,0}\sqrt{r_{p,0}}}\right] \frac{c_{sat} - c_0}{\rho_p} (\Delta\tau)^2$$
$$+ \frac{1}{6}\left[\frac{2D_A}{r^3_{p,0}} + 0.31\left(\frac{\rho w_{rel}}{\mu_c}\right)^{0,5} \frac{D_A Pr_d^{0,33}}{r^2_{p,0}\sqrt{r_{p,0}}}\right] \frac{c_{sat} - c_0}{\rho_p} (\Delta\tau)^3 + \ldots \tag{11}$$

Therefore, the additional terms

$$+ \frac{Q^{r,in}_{add,i}}{V_i} c^r_{in,i}; \quad - \frac{Q^{r,out}_{add,i}}{V_i} c_{out,i}; \quad + \frac{Q^{z,in}_{add,i}}{V_i} c^z_{in,i};$$
$$- \frac{Q^{z,in}_{add,i}}{V_i} c_{out,i}; \quad - \frac{\beta_i F_i (c_{sat} - c_{out,i})}{V_i}$$

should be introduced in the right side of each equation in the system (1) when investigating mass transfer processes with mixing for heterogeneous systems. Finally, the mathematical model of mixing processes in apparatus with agitators that permits taking account of the influence of the type and size of the agitator, the geometry of the apparatus, the operating conditions and mode, and properties of the media being reworked and can be used to investigate the processes of mixing of homogeneous and heterogeneous systems, suspension, extraction,

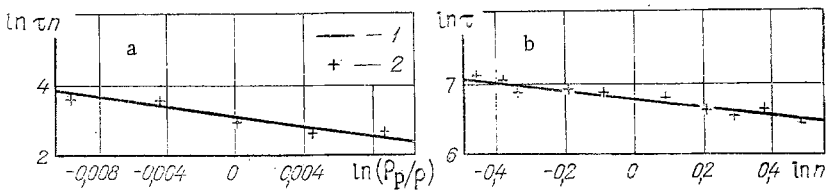

Fig. 1. Influence of the relationship between the solid and fluid phase densities on the suspension time (a) and number of agitator revolutions during dissolution (b) [1) computation, and 2) experiment].

and dissolution in fluid–solid systems has the form

$$\frac{dc_1}{d\tau} = \frac{Q_1^{in}}{V_1} c_{in,1} - \frac{Q_1^{out}}{V_1} c_{out,1} + \frac{V_{add,1}^{r,in}}{V_1} c_{in,1}^{r} - \frac{Q_{add,1}^{r,out}}{V_1} c_{out,1} +$$
$$+ \frac{Q_{add,1}^{z,in}}{V_1} c_{in,1}^{z} - \frac{Q_{add,1}^{z,out}}{V_1} c_{out,1} - \frac{\beta_1 F_1 (c_{sat} - c_{sat,1})}{V_1};$$

$$\frac{dc_i}{d\tau} = \frac{Q_i^{in}}{V_i} c_{in,i} - \frac{Q_i^{out}}{V_i} c_{out,i} + \frac{Q_{add,i}^{r,out}}{V_i} c_{in,i}^{r} - \frac{Q_{add,i}^{r,out}}{V_i} c_{out,i} +$$
$$+ \frac{Q_{add,i}^{z,in}}{V_i} c_{in,i}^{z} - \frac{Q_{add,i}^{z,out}}{V_i} c_{out,i} - \frac{\beta_i F_i (c_{sat} - c_{out,i})}{V_i}; \qquad (12)$$

$$\frac{dc_m}{d\tau} = \frac{Q_m^{in}}{V_m} c_{in,m} - \frac{Q_m^{out}}{V_m} c_{out,m} + \frac{Q_{add,m}^{r,in}}{V_m} c_{in,m}^{r} - \frac{Q_{add,m}^{r,out}}{V_m} c_{out,m} +$$
$$+ \frac{Q_{add,m}^{z,in}}{V_m} c_{in,m}^{z} - \frac{Q_{add,m}^{z,out}}{V_m} c_{out,m} - \frac{\beta_m F_m (c_{sat} - c_{out,m})}{V_m}.$$

The analysis and solution of the model (12) was performed for apparatus with a working volume to 27 m^3, equipped with vane agitators with Γ_D = 1.5-4; Γ_H = 0.4 and a 1-10 rps frequency of rotation.

Experimental investigations were performed on pilot plants and industrial apparatus: suspension of solid particles of from 10^{-4}-10^{-3} m diameter and 812-2700 kg/m^3 density in liquid media of 900-1100 kg/m^3 density and a dynamic viscosity coefficient of 10^{-3}-10^{-2} Pa·sec; dissolution of solid particles in fluids with a diffusion factor of 10^{-9}-10^{-10} m^2/sec and a saturation concentration of 10-370 kg/m^3.

Investigations carried out by using the system (12) exhibited the dynamics of the formation of suspensions in apparatus with agitators. At the initial instant of agitator operation at an angular velocity exceeding the minimal necessary to lift the particles from the bottom, they are gradually distributed over the volume of the apparatus under the influence of circulation streams. Homogeneity of the distribution reaching $(0.8\text{-}1.2)c_{out,i}/c_{av}$ can be achieved here. During further operation of the mixing unit, although the particles are maintained in practically the whole volume of the vessel, the homogeneity of their distribution is reduced because of radial separation up to the formation of an annular space depleted of solid phase in the central part of the apparatus. This phenomenon is especially perceptible for high values of $(\Delta\rho/\rho)$ and large numbers of agitator revolutions in a small vessel. In large size vessels of more than 3 m^3 capacity, particle accumulation is observed along the near-axis zone of the apparatus for $\rho_p < \rho$ and in apparatus of capacity to 1 m^3 particle accumulation occurs near the side walls of the apparatus, in the upper zone, and in the zone of the shaft of the mixing unit.

The relationship between the solid and fluid phase densities (ρ/ρ_p) on the suspension time is represented in Fig. 1a, from which it follows that $\tau n \sim (\rho/\rho_p)^{0.7}$ as is verified by our test data presented in this figure. When utilizing a correlation simplex $(\Delta\rho/\rho)$ the exponent is here reduced to 0.45 and corresponds to that obtained in [9, 10]. In conformity with [12], its value increases to 0.65 during apparatus operation with reflecting baffles.

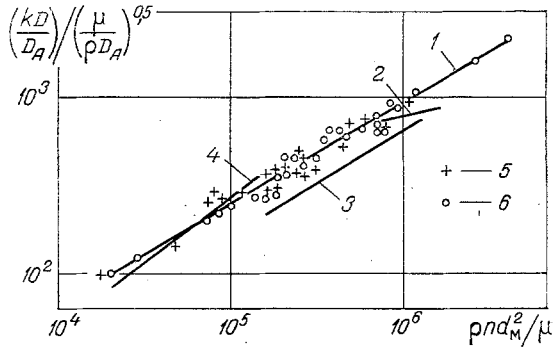

Fig. 2. Mass elimination in apparatus with agitators: 1) dissolution, computation; 2) dissolution, empirical equation [14]; 3) the same [13]; 4) the same [11]; 5) our test data; 6) test data [12].

The influence of the number of revolutions on mixing of the suspension is presented in Fig. 1b, from which $\tau n \sim n^{0.35}$ follows, which is in good agreement with test data.

In general form, the influence of the parameters investigated on the time of suspension can be represented in the form

$$\tau n \sim n^{0.35} \Gamma_D^{1.5} \left(\frac{\rho}{\rho_p}\right)^{0.7}.$$

The influence of the apparatus dimensions, the viscosity of the fluid medium, and the location of the agitator is not unique, which partially explains the significant discrepancy between the correlation equations of the "governing" number of revolutions obtained by researchers in different apparatus [11]. Thus, as the volume of the apparatus increases to 10 m^3, the change in Γ_H goes to 0.7 $\tau n \sim \Gamma_H^{0.9} V^{0.4}$ while a further change in the parameters results in an abrupt change in the exponent, where for $\Gamma_H > 1$ and $\rho_p > \rho$ the exponent of Γ_H increases to 3.

Numerous researchers have established [11] a mass elimination equation of the form

$$\frac{kD}{D_A} = C \mathrm{Re}_c^{0.32-1.4} \mathrm{Pr}_d^{0.5} \qquad (13)$$

in investigations of mass elimination to solid particles moving and soluble in a miscible fluid medium. However, there is no information about the kinetics of the process of dissolution during mixing in the literature, and the magnitudes of the mass elimination factor were investigated at the initial instant of dissolution when the particle size diminished by not more than 30% and their change was neglected.

The analytic dependence (13), computed for dissolution processes in apparatus of up to 3 m diameter and having the form

$$\frac{kD}{D_A} = 0.04 \mathrm{Re}_c^{0.55} \mathrm{Pr}_d^{0.5}, \qquad (14)$$

is represented in Fig. 2. Our test data as well as the results of other investigations [12-14] presented there exhibit satisfactory agreement.

However, engineers are interested not only in the mass transfer intensity, but also, and perhaps still more, in the dissolution time, the productivity of the technological process. Knowledge of the mass elimination factor is not sufficient in such a case since the particle dimensions vary during dissolution, and therefore, so do the conditions of their motion in the system.

Investigations of the dissolution kinetics during mixing, performed by using the model (12), yielded the dependence

$$\tau n \sim n^{-0.55} D_A^{-0.2} \Gamma_H^{0.9} \Gamma_D^{1.5} \left(\frac{\Delta\rho}{\rho}\right)^{0.4}, \qquad (15)$$

which conforms well to the test data. The apparatus dimensions, along with the reflecting baffles, do not substantially influence the duration of the dissolution, which sometimes even grows in apparatus with agitators for high numbers of revolutions, as has been detected also

in [14]. It should also be noted that the exponent for the simplex $(\Delta\rho/\rho)$ in [15], which equals 0.4,* is somewhat higher than that obtained by other researchers who cite the quantities (0.25-0.33) [15-17]. Taking account of the difficulties in an accurate measurement of the variable size of the particles, such discrepancies are completely explicable.

In conclusion, let us note that solid particle dissolution can last 8-70 min in apparatus of up to 1 m^3 in volume.

NOTATION

$Ar = 8\Delta\rho\rho r_p^3 g/\mu^2$ is the Archimedes criterion; $Pr_d = \mu/\rho D_A$, Prandtl diffusion criterion; $Re_c = \rho n d_m^2/\mu$, Reynolds centrifugal criterion; $\Gamma_D = D/d_m$; $\Gamma_H = H/d_m$, geometric similarity simplexes; C, coefficient; D, apparatus diameter, m; D_A, diffusion factor, m^2/sec; F, mass elimination surface, m^2; H, the height to which the apparatus is filled, m; P_{sub}, mass of solid phase, kg; V, working volume of the apparatus, m^3; a, acceleration, m/sec^2; c, concentration, kg/m^3; c_{sat}, saturation concentration, kg/m^3; d_m, agitator diameter, m; k, mass elimination factor, m/sec; m, number of cells; n, number of agitator revolutions, rps; r, running radius, m; r_p, solid particle radius, m; w, velocity, m/sec; β, mass elimination factor to the particle, m/sec; ρ, density of the liquid phase, kg/m^3; ρ_p, solid phase density, kg/m^3; $\Delta\rho$, difference in phase densities, kg/m^3; μ, dynamic viscosity of the fluid phase, Pa·sec; μ_s, dynamic viscosity of the suspension, Pa·sec; τ, time, sec. Subscripts: in, input; out, output; add, additional; 0, preceding instant; rel, relative; av, averaged with respect to the apparatus volume; ε, tangential; r, radial direction; z, axial direction; i, k, m, cell indices.

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*Strictly speaking, it has the value 0.4 only for $\rho_p > \rho$ for $\Gamma_H < 0.6$. Both $|\Delta\rho/\rho|$ and V affect the dissolution time arbitrarily.