Journal of Engineering Physics and Thermophysics, Vol. 80, No. 4, 2007

A MACROKINETIC MODEL OF AEROSOL DEPOSITION OF A DISSOLVED COMPONENT ON THE SURFACE OF CRYSTAL STRUCTURES UNDER ISOTHERMAL CONDITIONS

V. I. Ryazhskikh and K. S. Petrov

UDC 621.1.016.4

Based on kinetic ideas on the phenomena of transfer under isothermal conditions in the crystal–film–aerosol system, a mathematical model of mass transfer that accounts for the change in phase interfaces is suggested.

Obtaining of products with given quality characteristics is of primary importance in the chemical and food industries. Especially promising is the imparting of new properties to disperse substances by depositing a satellite component on their surface [1]. Technically this can be implemented by different means, for example, in a fluidized bed by pumping a dispersion component through a porous fixed block of the substance, etc.

In order to impart new qualitative tints to a base substance, usually a very small amount of the component is needed, which preferably should be deposited on the surface and near-surface layers (e.g., into the liquid film) of disperse particles. However, it is evident that purely mechanical deposition of a dry component on a substance is ineffective, since the adhesion forces are unable to keep the component on the surface [2]. Therefore, mass transfer of the component through the film of a dissolved crystalline substance with subsequent removal of a solvent solves the set problem in principle. Judging by its physical and chemical characteristics, the technology considered relates closest to the newest trend — nanotechnology — but the scale of the proceeding processes relative to molecular dimensions still remains applicable for the use of the phenomenological strategy, too. This fact imparts the leading role to the method of mathematical simulation in research, in identification of rational operating conditions, and in predicting required qualitative characteristics.

The physical picture of the modeled process is such: the crystal on which there is a mass of undersaturated dissolved crystalline substance in a film has a known and constant initial temperature; the crystal is blown by a gas flux which contains aerosol particles with the component dissolved in them and has the same temperature; a heat- and mass-transfer process — dissolution — takes place, which determines the change in the geometric parameters of the system [3].

Based on the kinetic notions of the process of dissolution and material balance, the differential equation that interrelates the mass of the dissolved substance of the crystal $m_{\rm c}$, its concentration c in the film, and the surface area of the crystal $s_{\rm c}$ has the form

$$\frac{dm_{\rm c}}{d\tau} = k\,(c_{\rm sat} - c)\,\tau\,\frac{ds_{\rm c}}{d\tau} - k s_{\rm c}\tau\,\frac{dc}{d\tau} + k\,(c_{\rm sat} - c)\,s_{\rm c}\,. \tag{1}$$

From the definition of the mass concentration $c = m_{\rm c}/V_{\rm h}$, where $V_{\rm h} = V_0 + \tau u - V_{\rm c}$ is the volume of the film, it follows that

$$c = \frac{m_{\rm c}}{V_0 + \tau u - k_{\rm v}\left(\dfrac{s_{\rm c}}{k_{\rm s}}\right)^{3/2}}\,. \tag{2}$$

Voronezh State Technological Academy, 19 Revolyutsiya Ave., Voronezh, 394000, Russia. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 80, No. 4, pp. 20–24, July–August, 2007. Original article submitted December 5, 2005; revision submitted May 4, 2006.

In a differential form, the relationship between the change in the volume of the crystal and the mass of the dissolved substance in the film has the form

$$\frac{dm_{\mathrm{c}}}{\rho} = -\,dV_{\mathrm{c}}\,,$$

where $dV_{\mathrm{c}} = k_{\mathrm{v}} l^3 - k_{\mathrm{v}}(l - dl)^3$; therefore a decrease in the linear dimension

$$dl = \frac{dm_{\mathrm{c}}}{3k_{\mathrm{v}} l^2 \rho}$$

will lead to a reduction in the surface area of the crystal:

$$ds_{\mathrm{c}} = 2k_{\mathrm{s}} l dl\,.$$

Then

$$\frac{ds_{\mathrm{c}}}{d\tau} = -\left(\frac{2k_{\mathrm{s}}^{3/2}}{3k_{\mathrm{v}}\rho\,\sqrt{s_{\mathrm{c}}}}\right)\frac{dm_{\mathrm{c}}}{d\tau}\,. \tag{3}$$

System (1)–(3) is supplemented with the initial conditions

$$m_{\mathrm{c}}\,(0) = m_{\mathrm{c}}^0\,, \quad s_{\mathrm{c}}\,(0) = s_{\mathrm{c}}^0\,. \tag{4}$$

System (1)–(4) is nondimensionalized as

$$\frac{dM}{d\Theta} = \frac{(1 - C)\,\Theta dS}{d\Theta} - \frac{S\Theta dC}{d\Theta} + (1 - C)\,S\,, \tag{5}$$

$$C = \frac{M}{\left(A_2 + U\Theta - A_3 S^{3/2}\right)}\,, \tag{6}$$

$$\frac{dS}{d\Theta} = -\left(\frac{A_1}{\sqrt{S}}\right)\frac{dM}{d\Theta}\,, \tag{7}$$

$$M\,(0) = S\,(0) = 1\,, \tag{8}$$

where

$$M = \frac{m_{\mathrm{c}}}{m_{\mathrm{c}}^0}\,; \quad S = \frac{s_{\mathrm{c}}}{s_{\mathrm{c}}^0}\,; \quad \Theta = \frac{k c_{\mathrm{sat}} s_{\mathrm{c}}^0 \tau}{m_{\mathrm{c}}^0}\,; \quad C = \frac{c}{c_{\mathrm{sat}}}\,; \quad U = \frac{u}{k s_{\mathrm{c}}^0}\,;$$

$$A_2 = \frac{c_{\mathrm{sat}} V_0}{m_{\mathrm{c}}^0}\,; \quad A_3 = \frac{c_{\mathrm{sat}} k_{\mathrm{v}}}{m_{\mathrm{c}}^0}\left(\frac{s_{\mathrm{c}}^0}{k_{\mathrm{s}}}\right)^{3/2}\,; \quad A_1 = \frac{2m_{\mathrm{c}}^0}{3k_{\mathrm{v}}\rho}\left(\frac{k_{\mathrm{s}}}{s_{\mathrm{c}}^0}\right)^{3/2}\,.$$

System (5)–(8) is reduced to the Cauchy problem for S:

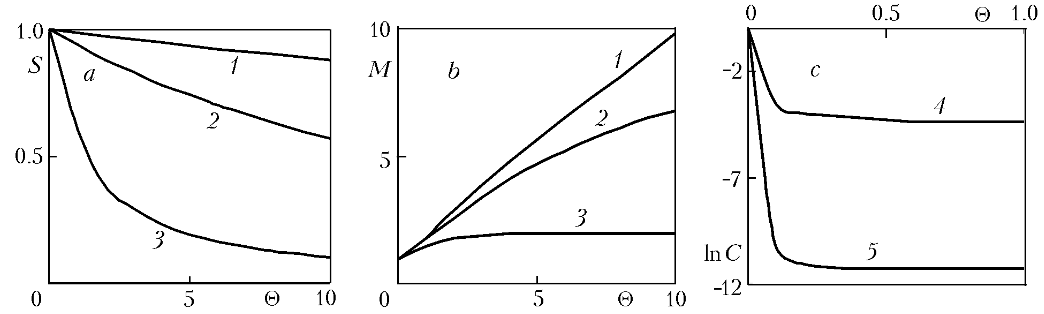

Fig. 1. Kinetics of the process of aerosol deposition of a component on the surface of a crystal at $A_2 = 30.48$, $A_3 = 29.48$ [1) $A_1 = 1.28 \cdot 10^{-2}$; 2) $6.4 \cdot 10^{-2}$; 3) 0.64]: a) relative surface of a crystal; b) relative mass of a crystalline substance in a film; c) relative concentration of a crystalline substance in a film at $A_1 = 1.28 \cdot 10^{-2}$ [4) $U = 85$; 5) $8.5 \cdot 10^4$].

$$\frac{dS}{d\Theta} = \left\{ \frac{\left(U + \Theta \frac{dU}{d\Theta}\right) S\Theta \left[1 + \frac{2}{3A_3}\left(1 - S^{3/2}\right)\right]}{\left(A_1 + U\Theta - A_2 S^{3/2}\right)^2} + \left[1 - \frac{1 + \frac{2}{3A_3}\left(1 - S^{3/2}\right)}{A_1 + U\Theta - A_2 S^{3/2}}\right] S \right\} \times$$

$$\times \left\{ \frac{3}{2} A_2 S^{3/2} \Theta \frac{1 + \frac{2}{3A_3}\left(1 - S^{3/2}\right)}{\left(A_1 + U\Theta - A_2 S^{3/2}\right)^2} - \frac{\sqrt{S}}{A_3} - \left[1 - \frac{1 + \frac{2}{3A_3}\left(1 - S^{3/2}\right)}{A_1 + U\Theta - A_2 S^{3/2}}\right] \Theta - \frac{S^{3/2}\Theta}{A_3} \frac{1}{A_1 + U\Theta - A_2 S^{3/2}} \right\}^{-1}, \quad (9)$$

$$S(0) = 1, \quad (10)$$

from the solution of which we determine

$$M = 1 - \frac{2}{3A_3}\left(S^{3/2} - 1\right) = 1 + \frac{2}{3A_3}\left(1 - S^{3/2}\right) \quad (11)$$

and C by Eq. (6).

The solution of system (9)–(11) was done numerically using a fourth-order Runge–Kutta method [4]. The analysis has shown that the influence of the values of dimensionless complexes A_2 and A_3 on the picture of the process is insignificant. The determining complex is A_1, which characterizes, in essence, the specific mass of dissolved crystalline substance per unit crystal surface. As expected, an increase in the complex A_1 leads to a higher intensity of dissolution of the crystalline substance [5] (Fig. 1a). The mass of the dissolved crystalline substance in the film also depends to a greater extent on A_1 (Fig. 1b). An increase in the dissolved-component aerosol flow rate leads to a decrease in the concentration of the crystalline substance in the film (Fig. 1c). Despite the rather simple reasonings, the proposed model adequately, on a qualitative level, estimates the real process.

In order to carry out a quantitative checking of the model, experiments were conducted on a pilot plant (Fig. 2) to evaluate the coating of a vibrating bed of crystalline sugar with stevioside.

Sputtering of a 30–60% solution of stevioside in the form of aerosol was done for 70 sec through a pneumatic nozzle 4 under an air pressure of 0.1 MPa onto a vibrating bed of sugar 3, wetted up to $W \approx 0.8\%$, on a tray 1 in a vibrofluidized state. A heated solution of stevioside was supplied to the nozzle 4 by a peristaltic pump, and the pressure of the compressed air was created by an FX-95 oilless electrical compressor. The sugar–stevioside mixture was dried in a drum drier for 10–15 min at 55–80°C.

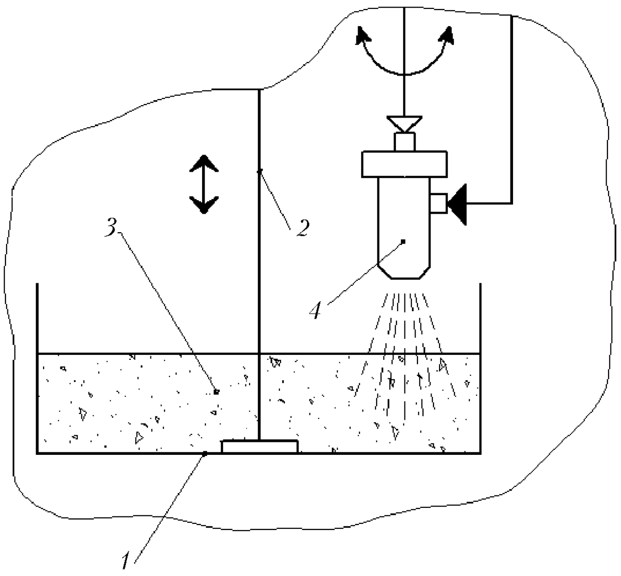

Fig. 2. Technique of aerosol deposition of a coating on a crystalline product: 1) vibrating tray; 2) rod of a vibrodrive; 3) bed of sugar; 4) pneumatic nozzle.

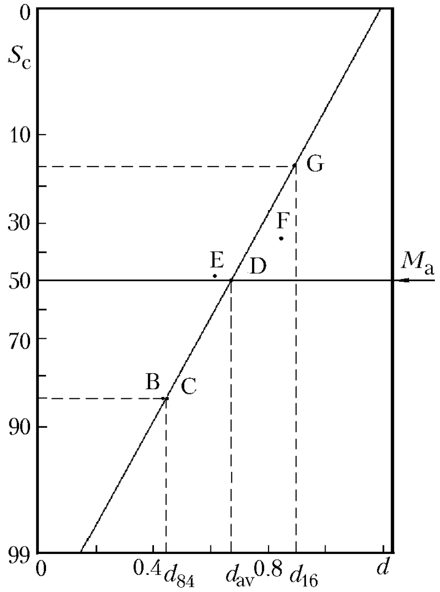

Fig. 3. Probability graph of a dispersion analysis of granulated sugar on sputtering of a 60% solution of stevioside (d_{av} = 0.69 mm): B, E, F) experimental points corresponding to residues on sieves with cells measuring 0.45, 0.63, and 0.85 mm. S_c, %; d, mm.

The graphical interpretation of the results of Powers' sieve analysis [6] of the disperse composition of the granulated sugar crystals on the surface of which a solution of stevioside was deposited is given on the probability graph (Fig. 3). A comparative estimate of the granulometric composition of a sample was made by determining the average size of the crystals and of the nonuniformity coefficient:

(a) the average size of sugar crystals d_{av} was taken equal to the clear dimension of the sieve cells through which precisely half the mass of the sample taken could pass;

(b) the nonuniformity coefficient C_{var} was defined as the ratio of the average quadratic deviation to the average size of crystals:

$$C_{\text{var}} = \frac{100\,(d_{16} - d_{\text{av}})}{d_{\text{av}}} = \frac{100\,(d_{\text{av}} - d_{84})}{d_{\text{av}}}\,.$$

An analysis of the results reveals a good convergence of the calculated and experimental values of the mass of crystals under the conditions of aerosol deposition of a 50–60% aqueous solution of the satellite component on the crystalline structure in a vibrofluidized state of the latter. It has been established that a decrease in the concentration of the sputtered aqueous solution of stevioside below 30%, which leads to a decrease in the degree of solution saturation by less than 0.7 in the boundary film, favors partial dissolution of sugar crystals.

Using the given mathematical approach and considering the physicochemical properties of various satellite components and crystalline base, it is possible to predict the needed effect of coating by an aerosol technique.

The proposed mathematical model is a tool for determining rational regimes of depositing aerosol particles on the surface of crystals, which allows one to minimize losses in the quality of a crystalline structure because of the possible partial dissolution of the surface layers of crystals.

NOTATION

A_1, specific mass of the substance per unit crystal surface; A_2, relative mass of dissolved substance in a film of saturated solution; A_3, relative concentration of dissolved substance in a film of saturated solution; C, relative concentration of crystalline substance in a film; c, mass concentration of dissolved substance in a surface film, kg/m^3; c_{sat}, mass concentration of a saturated solution of a substance in a surface film, kg/m^3; C_{var}, nonuniformity coefficient, %; d_{16}, abscissa of point G of the intersection of the graph with the line S_{c} = 16%, mm; d_{84}, abscissa of the point C of intersection of the graph with the line S_{c} = 84%, mm; d_{av}, average size of sugar crystals (abscissa of the point D of intersection of the graph with the horizontal line $S_{\text{c}}M_{\text{a}}$), mm; k, coefficient of the rate of dissolution of the crystal substance, m/sec; k_{s}, coefficient of the shape of the crystal surface; k_{v}, coefficient of the shape of the crystal volume; l, characteristic linear dimension of a crystal, m; M, relative mass of the crystalline substance in a film; M_{a}, ordinate corresponding to a 50% mass of a sample of crystals; m_{c}, mass of the dissolved substance of a crystal, kg; S, relative surface of a crystal; S_{c}, cumulative residue of crystals on the sieve, %; s_{c}, surface area of a crystal, m^2; u, unsteady-state integral volumetric flux of aerosol particles to the film surface, m^3/sec; V_0, combined initial volume of the crystal–film system, m^3; V_{c}, volume of a crystal, m^3; U, relative flux of aerosol particles per unit crystal surface; W, moisture content of sugar, %; Θ, dimensionless time; ρ, density of a crystal, kg/m^3; τ, times, sec. Subscripts: a, aperture; c, crystal; h, height; s, surface; sat, saturation; v, volume; var, variation; 0, initial; av, average.

REFERENCES

1. V. D. Karmazin, *Technique and Application of a Vibrating Bed* [in Russian], Naukova Dumka, Kiev (1977).
2. P. A. Rebinder, Surface phenomena in disperse systems, in: *Colloidal Chemistry: Selected Papers* [in Russian], Nauka, Moscow (1979).
3. K. S. Petrov and V. I. Ryazhskikh, A mathematical model of aerosol deposition of solutions on a stochastically oriented surface of a crystal, in: V. S. Balakirev (Ed.), *Proc. 18th Int. Sci. Conf. "Mathematical Methods in Technique and Technologies–MMTT-18"* [in Russian], in 10 volumes, Vol. 9, Sec. 11, Izd. Kazansk. Gos. Tekhnol. Univ., Kazan' (2005), P. 50.
4. L. I. Turchak, *Principles of Numerical Methods* [in Russian], Nauka, Moscow (1987).
5. G. A. Aksel'rud and A. D. Molchanov, *Dissolution of Solids* [in Russian], Khimiya, Moscow (1977).
6. J. Dobzicki, *Analysis in Sugar Production* [Russian translation], Agropromizdat, Moscow (1985).