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ON THE DISSOLVING RATE OF A SOLID WITH CHEMICAL REACTION*

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Abstract—An investigation on dissolving rate of a solid in a liquid flowing between two flat plates, with the contribution of a second-order reversible chemical reaction, has been carried out. The process was analyzed as an isothermal one, taking place within a transition region at a stationary laminar flow. From the differential momentum and mass balances a process model was formulated under suitable initial and boundary conditions and a numerical solution was obtained for the benzoic acid–methanol reaction. According to the proposed model it is possible to calculate concentration profiles for any cross-section of a slit reactor. The presented model has been experimentally proved. Average difference between experimental values and theoretical calculations ranged to ±5.3%. Longitudinal diffusion has been shown to have a negligible effect on the results. The presented method of calculation was found to be sufficiently accurate for the problem under consideration.

NOMENCLATURE

c,	concentration [mol/l];
C,	dimensionless concentration, $C = \frac{C}{C_1^s}$;
Da^{II},	II Damköhler's number;
D,	diffusion coefficient in solution [m^2/h];
J,	diffusional stream [mol · m/h · l];
k,	reaction rate constant [1/h] or [l/mol · h];
L,	reactor length [m];
Pe,	Peclet diffusion number, $Pe = \frac{2 \cdot y_0 \cdot \bar{w}}{D}$;
r,	rate of reaction [mol/h · l];
$\bar{w}$,	mean linear velocity [m/h];
x, y, z,	Cartesian coordinates;
y_0,	half distance between two parallel plates [m];
X, Y,	dimensionless coordinates.

Greek symbols

μ,	function dependent on chemical reaction process (+1 or −1);
τ,	time [h].

Subscripts

e.	estrification;
eq,	equilibrium;
h,	hydrolysis;
i,	$i = 1, 2, 3$ benzoic acid, methanol, methyl benzoate respectively;
0,	initial values (except for y_0—as shown above);
s,	at saturation state.

1. INTRODUCTION

THE PROCESS of mass penetration from the surface of a solid into liquid, though rather frequently encountered in practice, appears to be, in fact, relatively little known. General information concerning the rate of this process is normally taken either from analogy between the transfer of heat and mass, or from calculation based on the Hatta's model, Danckwert's model, or other models of this kind. Investigation of the mass-transfer models has been usually based on experimental determination of the transfer coefficients. Such an approach to the problem causes the proved models, and relationships describing the process in question, to be obtained only for some specified conditions and systems. On the other hand, taking the differential equations for mass, momentum and heat transfer as a starting point and by defining suitable initial and boundary conditions for the problem under consideration, more general relationships can be deduced and with the use of a computer one can solve problems considered otherwise as impossible or very difficult for analytical solution. The way of solving transfer equations with the aid of computers appears to be much more economical than experiments, especially where time is concerned. This applies particularly to industrial processes. It will be of crucial importance in the development of an optimization system for such processes, both for the design of equipment and assessment of it.

The aim of this paper is to formulate, on the basis of differential balances of momentum and mass, a model for the dissolution of solid, with chemical reaction in a flow-type flat slit reactor, along with a numerical solution of the problem and its experimental confirmation.

A process of dissolving a solid in the liquid, with the second-order reversible chemical reaction, has been assumed. The benzoic acid–methanol reaction has been chosen, benzoic acid being the solute, and a 1% aqueous methanol solution being the solvent. The reaction was carried out in a flow-type reactor with a rectangular horizontal slit, the upper and the lower wall of the slit were formed by flat benzoic acid plates. The whole process was carried out at isothermal stationary laminar flow conditions.

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2. MASS TRANSFER FROM SOLID SURFACE

The problem of mass transfer from the solid surface into laminar liquid flow has already been the object of studies, however, the amount of information available about this type of phenomenon is still far from being satisfactory. Considerations included bodies and systems varying in their geometry, but without the contribution of chemical reactions. The rate of dissolving of balls, cylinders, tubes and plates without chemical reaction was investigated. The problem of mass transfer with a not very rapid second-order chemical reaction has not been analyzed so far for the flat system and laminar flows. The flat system has been the object of a few papers only [1–3, 5–11, 13]. A small number of research workers have investigated the problem complicated by the presence of a chemical reaction, and only very few of them found an analytical solution for the problem.

Generally, most considerations were related to simple, very rapid and irreversible reaction. Potter [10] has developed a theory of boundary layer for the case of mass transfer from the solid to liquid accompanied by a simultaneous irreversible chemical reaction occurring at a very high rate in the reaction zone which lies in the laminar boundary layer. Fridlander and Litt [5] gave an analytical solution to the problem, based on Potter's theory, by making use of the flow equation and their solutions as proposed by Blasius. Later, Acrivos [1] suggested a solution to the case of mass transfer with a rapid homogeneous chemical reaction. For the case of equality of Schmidt numbers for both reagents, relationships proposed by Acrivos resembled those of Hatta's equation.

3. MATHEMATICAL MODEL OF THE PROCESS

From the principle of mass conservation, differential mass balances of benzoic acid, methyl alcohol and methyl benzoate can be derived.

$$\nabla(\mathbf{w}\cdot c_1)+\nabla(\mathbf{J}_i)+\frac{\partial c_i}{\partial \tau}+r_i=0; \qquad \text{for} \quad i=1,2,3. \tag{1}$$

Where $i = 1, 2, 3$ corresponds to benzoic acid, methanol and methyl benzoate respectively. Streams $\mathbf{J}_i$ may be due to molecular diffusion, ionic diffusion, pressure diffusion or thermal diffusion. Taking into account molecular diffusion only, we get:

$$\mathbf{J}_i = -D_i\cdot\nabla(c_i). \tag{2}$$

Diffusion coefficients will be assumed equal unary aqueous solutions acid, methanol or ester. Such an assumption, is possible on the basis of papers by Curtis and Hirschfelder [4], and by Wilke [12]. Those authors give relationships describing the influence of other components of the mixture on the value of diffusion coefficients. In the case under consideration assuming small concentrations of soluble components in the system, differences in the diffusion coefficients, calculated as for mixture and a simple solution, will be almost identical. The following other assumption has been made. Since in the case of constant density the principle of continuity takes the form:

$$\nabla(\mathbf{w}) = 0, \tag{3}$$

assuming that the flow proceeds in one direction—one gets:

$$w_y = w_z = 0. \tag{4}$$

If, moreover, diffusion in the direction of axes $0X$ and $0Z$ is omitted in the balances and the process is considered as stationary, then equation (1) could be written in the following way:

$$w_x\frac{\partial c_i}{\partial x^2}-D_i\frac{\partial^2 c_i}{\partial y^2}+r_i=0 \qquad \text{for} \quad i=1,2,3. \tag{5}$$

Since we analyze a laminar flow between two parallel flat plates, then:

$$w_x=\tfrac{3}{2}\cdot\bar{w}\cdot\left[1-\left(\frac{y}{y_0}\right)^2\right]. \tag{6}$$

The chemical reaction under consideration can be expressed by the equation:

$$C_6H_5COOH+CH_3OH \rightleftarrows C_6H_5COOCH_3+H_2O \tag{7}$$

From the principle of chemical kinetics:

$$r_1=r_2=-r_3. \tag{8}$$

For the reaction under consideration, we can assume that:

$$r_1=k_e\cdot c_1\cdot c_2-k_h\cdot c_3. \tag{9}$$

Assuming that water concentration $c_4 = \text{const}$

$$k_h=k_h'c_4=\text{cost}. \tag{10}$$

This product is the reaction velocity constant for hydrolysis considered as a pseudofirst-order reaction.

3.1. *Initial and boundafy conditions*

For the analyzed system, initial conditions can be written as:

$$x=0;\quad c_2=c_2^0;\quad c_3=0; \tag{11}$$

$$x=0;\quad -y_0<y<+y_0;\quad c_1=0; \tag{12}$$

$$x=0;\quad y=\pm y_0;\quad c_1=c_1^s. \tag{13}$$

The boundary conditions can be expressed in the form of following relationships:

$$y=\pm y_0;\quad c_1=c_1^s;\quad c_2=c_2^0;\quad c_3=0; \tag{14}$$

$$\left(\frac{\partial c_i}{\partial y}\right)_{y=0}=0 \qquad \text{for} \quad i=1,2,3. \tag{15}$$

The relationships (14) are equivalent with an assumption that on the solid–liquid interface concentration of the benzoic acid is equal to this of saturated solution, concentration of methanol to its initial concentration, and this of ester to zero. An assumption that $c_1 = c_1^s$ is used in a number of papers, and it found its experimental confirmation. In the system under consideration, it would be very difficult to find the transformation degree of conversion at the surface, and therefore, the problem of the determination of ester and methanol concentration is difficult to solve. As it was impossible to determine the actual surface concentration, two extreme cases have been analysed with c_3 taken once as 0 and, alternatively as c_3^{eq}. When a molecule is formed at the interface, its energy state would be increased by a suitable energy excess resulting from its location in the field of surface forces. In addition, because of high polarity of water, the benzoic acid–water complexes are forced on to the solid surface, and only after the decomposition of such complexes is a normal chemical reaction likely to take place. Accordingly, the probability of ester formation on the benzoic acid surface seems to be very small. One can suppose that the actual concentration of ester in the interface would be near zero. The assumption that $c_3 = 0$ is, of course, an approximation. Whether it is acceptable for the accuracy of technical calculations, should be found by experimentation. Calculations have been made for both assumptions concerning surface concentrations of ester. Experimental data have shown only the $c_3 = 0$ approximation to be admissible. Consequently, the interface methanol concentration has been taken as equal to initial concentration, this concentration being equal to that in the liquid core.

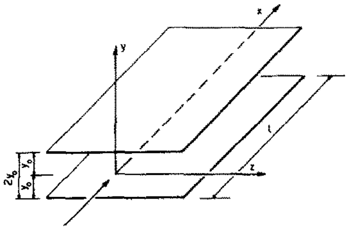

FIG. 1. Flow between two parallel plates ($2y_0 = 3 - 9.7$ mm—slit width; $L = 600$ mm—slit length).

Considering all the above assumptions, the system of balance equations (5) will have the following form:

$$\tfrac{3}{2}\cdot\bar{w}\cdot\left[1-\left(\frac{y_0}{y}\right)^2\right]\cdot\frac{\partial c_i}{\partial x}+D_i\frac{\partial^2 c_i}{\partial y^2}$$
$$+\mu\cdot k_e\cdot c_1\cdot c_2+\mu\cdot k_h\cdot c_3=0;$$
$$\text{for}\quad i=1,2,3. \tag{16}$$

where $\mu = +1$ for $i = 1, 2$ and $\mu = -1$ for $i = 3$.

System (16) can be reduced to a dimensionless form using substitutions, as follows:

$$\frac{x}{L}=X;\quad \frac{y}{y_0}=Y;\quad \frac{c_1}{c_1^s}=C_1;\quad \frac{c_2}{c_1^s}=C_2;$$
$$\frac{c_2^0}{c_1^s}=C_2^0;\quad \frac{c_3}{c_1^s}=C_3. \tag{17}$$

Then initial conditions will have the form:

$$X=0;\quad C_1=C_1^0;\quad C_3=0; \tag{18}$$
$$X=0;\quad -1<Y<+1;\quad C_1=0; \tag{19}$$
$$X=0;\quad Y=\pm 1;\quad C_1=0. \tag{20}$$

The boundary conditions will thus run:

$$Y=\pm 1;\ C_1=1;\ C_2=C_2^0;\ C_3=0; \tag{21}$$
$$\left(\frac{\partial C_i}{\partial Y}\right)_{Y=0}=0;\quad \text{for}\quad i=1,2,3. \tag{22}$$

Having taken into account all above designations and relationships, equations (16) will take the form:

$$\frac{3\cdot\bar{w}\cdot c_1^s}{2\cdot L}\cdot(1-Y^2)\cdot\frac{\partial C_i}{\partial X}-\frac{D_i\cdot c_1^s}{(y_0)^2}\cdot\frac{\partial^2 C_i}{\partial Y^2}$$
$$+\mu\cdot k_e\cdot(c_1^s)^2\cdot C_1\cdot C_2+\mu\cdot k_h\cdot c_1^s\cdot C_3=0;$$
$$\text{for}\quad i=1,2,3. \tag{23}$$

By transforming equations (23), we finally arrive at:

$$\tfrac{3}{4}\cdot Pe_i\cdot\left(\frac{y_0}{L}\right)\cdot(1-Y^2)\frac{\partial C_i}{\partial X}-\frac{\partial^2 C_i}{\partial Y^2}$$
$$+\mu\cdot Da_{ei}^{II}\cdot C_1\cdot C_2+\mu\cdot Da_{hi}^{II}\cdot C_3=0;$$
$$\text{for}\quad i=1,2,3. \tag{24}$$

Such a system of equations represents an isothermal process of dissolving horizontal walls of the rectangular slit, with the contribution of a reversible second-order chemical reaction, but without considering the effect of the longitudinal diffusion. By solving equations (24) one can arrive at concentration distribution profiles for any cross-section of the slit reactor for the acid, methanol and ester; it is also possible to calculate the reagent concentrations at the reactor outlet. Equations (24) have been solved numerically by the grid method using an electronic computer.

4. EXPERIMENTS

Experiments have been carried out at room temperature and normal pressure. The experimental apparatus is shown in Figs. 2 and 3 (the cross-sections). The slit reactor, built of two rectangular horizontal, parallel plates, spaced 3.0–9.7 mm apart, forms the main part of the experimental apparatus. The walls of the middle part of the reactor were formed of benzoic acid. The spacing between plates could be changed by insertion of distance gaskets made of hard rubber, of different thickness. Measurements were taken for four slit widths ($2y_0$): 3.0; 4.6; 7.4; 9.7 mm. The reactor assembly was positioned on a levelled table. Aqueous methanol solution flowed into the reactor from a 100 l tank (1) via "Mariotte's" bottle (2) and graduated capillary (3). Eight different capillaries providing flow rate in the range from 1.1 to 29.0 l/h (which corresponded to Reynolds number from 0.62 to 187.5, $Re = (2\cdot y\cdot \bar{w}/v)$, have been used. Downstream of the reactor inlet, provision has been made for a plate with 2 mm holes located on the 4 mm equilateral triangle

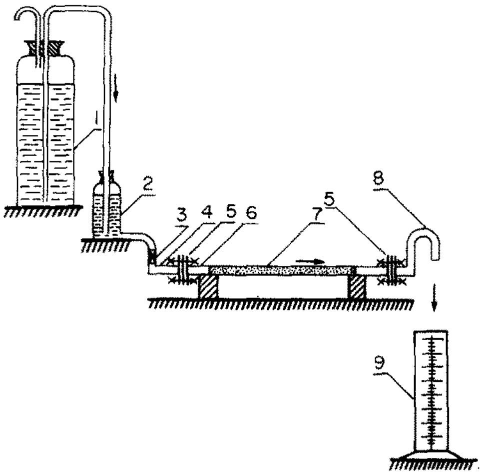

FIG. 2. Scheme of apparatus. 1—Tank for solution; 2—"Mariotte's" bottle; 3—Capillary; 4—Reactor inlet part; 5—Perforated baffle to equalize liquid flow; 6—Part of reactor in which liquid velocity profile is formed; 7—Part of reactor with soluble walls; 8—Hydraulic seal; 9—Measuring vessel.

pattern. This served to equalize the velocity profile. In the inlet part of the reactor (6) is a chamber 200 x 300 x $2y_0$ mm in which the process of velocity pattern build-up took place. In the main part of reactor (7) (300 x 600 x $2y_0$ mm) benzoic acid was dissolved by the flowing solution and reacted with it. The outlet part of reactor (200 x 300 x $2y_0$ mm) had the same task as the inlet part (6). The fluid leaves the reactor through an hydraulic seal. The fluid flow rate was determined by measuring the volume of liquid leaving the reactor during 6 min (three times for each flow rate). The benzoic acid and ester concentrations at the reactor outlet were determined by the spectrophotometric analysis (three samples for each flow rate). Seventeen series of experiments have been realized for four slit thicknesses (6–8 experiments per series for different flow rates). Much attention has been given to obtain even and smooth surfaces on the reactor walls. To this end, the grooves furrowed in the central part of both reactor plates were filled with molten benzoic acid, and the acid layer smoothed by means of a specially designed heated tool.

5. COMPARISON OF NUMERICAL CALCULATIONS WITH EXPERIMENTAL DATA

Equations (24) for initial conditions (18)–(20) and boundary conditions (21), (22) have been solved for the same flow rates as those used in the experiments. Table 1 shows the percentage error of concentrations of acid C_1 and ester C_3 obtained experimentally vs the theoretically calculated values. It is seen that the mean error amounts to ± 5.3% whereas the highest value reaches − 15.76%. These discrepancies are caused by the errors of experimentation, the error of spectrophotometric determination and also by the approximations of numerical computations. In order to show differences between calculations and experiments the dependences of concentrations of benzoic acid C_1 and C_3 ester on linear flow velocity for the $2y_0 = 3$ mm slit are shown in Fig. 4. Continuous lines on the figure show theoretical relationships and the points show the experimental results. As may be seen from Fig. 4, experimental points are close to theoretical lines. The results of calculations corresponding to the range of parameters used in experiments are shown in Fig. 5. Surfaces shown in this figure visually present the character of relationships existing between the acid and ester concentrations, linear flow velocity and thickness of reactor slit. The intersection of these surfaces is ascribed to the transition of the process from the kinetic to the diffusion region with diminishing linear flow rate. On the basis of presented models, 200 profiles of reagent concentration have been calculated every 3 mm of the reactor length (6–8 different flow velocities for each of the 4 slits). For example, Fig. 6 shows the concentration profiles of the three reagents for 3.0 mm slit at the reactor outlet. The curves

Table 1

y_0	C_1 (error %)		C_3 (error %)	
	max.	average	max.	average
1.50	− 13.46	± 6.4	+ 13.54	± 6.9
2.30	− 15.76	± 6.2	− 13.03	± 8.0
3.70	− 13.23	± 4.7	− 11.33	± 3.5
4.85	− 12.18	± 5.2	+ 14.39	± 2.7
for all slits	− 15.76	± 5.6	+ 14.39	± 5.0

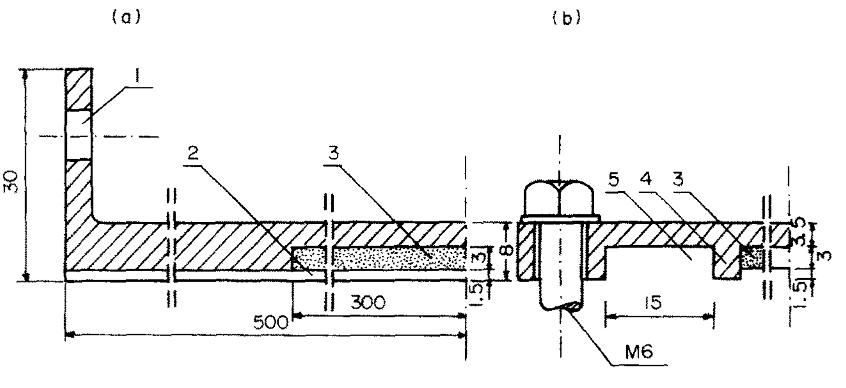

FIG. 3. The section of reactor: (a) longitudinal section; (b) cross-section, 1—flange, 2—slit, 3—groove filled with benzoic acid, 4—projection of guide, 5—space on the gasket.

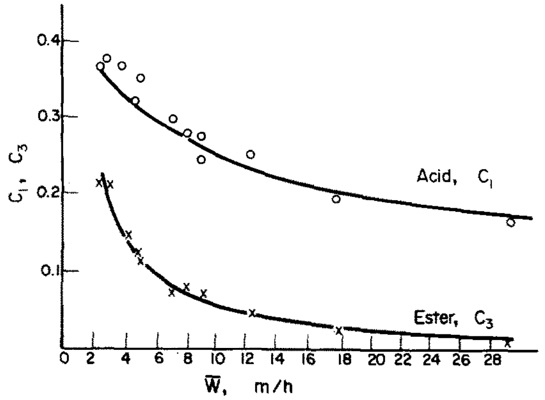

FIG. 4. The concentrations of benzoic acid—C_1 and C_3—ester in dependence on linear flow velocity for the $2y_0 = 3$ mm slit.

presented show full concurrence with the assumptions of the adopted model. The extremum of ester concentration in Fig. 6, can be ascribed to the transition of the process from the diffusional (near the wall) to the kinetic regime.

With the momentum and mass balance equations taken as a basis, a system of equations (elliptic second-order partial differential equations) has also been formulated to represent mass transfer for laminar flow in a flat system including the effect of longitudinal diffusion and reversible chemical reaction. Calculated data differed only very slightly from those calculated using the model not accounting for the effect of longitudinal diffusion (24). This means that the effect in question may be considered negligible for the process. A balance equation for benzoic acid only has been formulated (this being equivalent to the assumption that coefficients of diffusion for acid, ester and methanol are equal). The results show that this equation does not represent the process with sufficient accuracy, they differ greatly from those obtained by experiment.

6. CONCLUSIONS

1. The physico-chemical model of the solid dissolving process, with the contribution of a reversible second-order chemical reaction, has been formulated in the form of a system of three parabolic non-linear partial differential equations. The system in question has been numerically solved by the grid method.

2. The computations worked out can be used for laminar flow calculations covering different geometrics of the reactor and other chemical reactions.

3. Making use of the proposed model, one can calculate concentration profiles for any cross-section of a slit reactor.

4. The general character of the model equations (including terms describing convection, diffusion and reaction in both directions) allows their use directly regardless of wherever the process takes place in the diffusional, kinetic or mixed region.

5. Using the calculated concentration profiles one can easily determine the kinetic region in which the process takes place at various flow points.

6. The model presented here has been experimentally verified for the reaction in which benzoic acid is estrified with the methyl alcohol solution. Experimental results confirmed the velocity of assumptions adopted in the model.

7. From the results obtained it appears that in the system under consideration it is not necessary to take into account the effect of longitudinal diffusion.

8. Calculations based on the assumption of equality of diffusion coefficients of acid, ester and methanol (the balance of the benzoic acid only) do not give correct results.

9. The model presented constitutes a good representation of the process in which benzoic acid is dissolved in aqueous methanol solution, with ester interface concentration assumed to be equal to zero. The boundary condition for the ester, assuming the

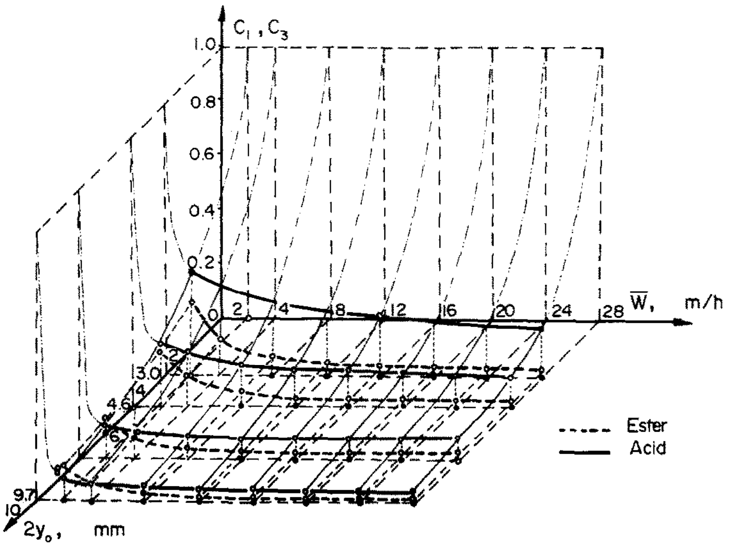

FIG. 5. The concentrations of acid—C_1 and ester—C_3 in dependence on mean linear flow velocity $\bar{w}$ and the dimension of the slit $2y_0$.

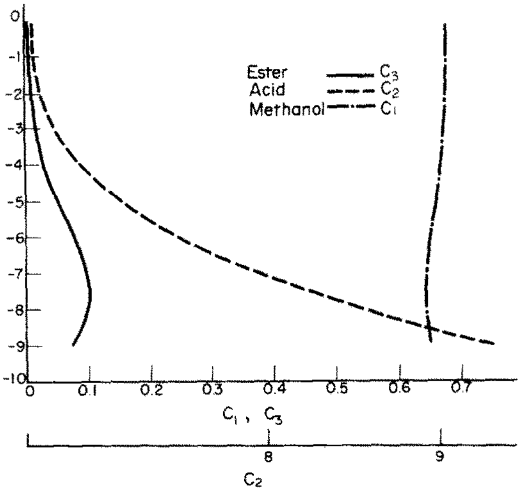

FIG. 6. The halfs of the concentrations profiles of: acid—C_1; methanol—C_2; and ester—C_3 at the reactor outlet. In the y-axis i indicates the distance from the axis of the reactor (for i = 10 the dissoluble wall of the reactor made from the benzoic acid).

ester concentration on the solid boundary being at equilibrium, gives results far apart from the experimental data.

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VITESSE DE DISSOLUTION D'UN SOLIDE AVEC REACTION CHIMIQUE

Résumé—On étudie la vitesse de dissolution d'un solide dans un liquide s'écoulant entre deux plaques planes en présence d'une réaction chimique réversible du second ordre. Le processus a été considéré isotherme et se produisant dans un écoulement stationnaire laminaire. A partir des équations différentielles de bilan de quantité de mouvement et de masse pour chaque composant du système, et avec des conditions initiales et aux limites convenables, un modèle physico-chimique du processus a été formulé et résolu numériquement dans le cas particulier de la réaction de l'acide benzoïque sur l'alcool méthylique. Le modèle proposé permet de calculer les profils de concentration dans le réacteur à section rectangulaire allongée. Le modèle a été vérifié expérimentalement. L'erreur relative moyenne entre expériences et calculs théoriques atteint ±5,3 pour cent. On a démontré en outre la possibilité de négliger l'influence de la diffusion longitudinale dans le problème traité.

ÜBER DIE AUFLÖSUNGSRATE EINES FESTEN STOFFES UNTER GLEICHZEITIGER CHEMISCHER REAKTION

Zusammenfassung—Es wurde die Auflösungsgeschwindigkeit eines festen Stoffes in einer Flüssigkeit unter gleichzeitiger sekundärer, reversibler chemischer Reaktion untersucht. Der Vorgang wurde bei stationärer Laminarströmung zwischen zwei Platten und isotherm betrachtet. Auf der Grundlage differentieller Impuls- und Stoffbilanzen wurde ein Modell für den Vorgang aufgestellt; geeignete Anfangs- und Randbedingungen wurden formuliert. Für die Reaktion Benzoesäure–Methanol wird eine numerische Lösung angegeben. Mit Hilfe des vorgeschlagenen Modells können die Konzentrationsprofile in jedem beliebigen Querschnitt des Spaltreaktors ermittelt werden. Das vorgeschlagene Modell wurde experimentell überprüft. Die mittlere Abweichung zwischen Meß- und Rechenwerten liegt in den Grenzen ±5,3%. Es wurde nachgewiesen, daß die Längsdiffusion nur einen vernachlässigbar kleinen Einfluß auf das Ergebnis ausübt. Die vorgeschlagene Berechnungsmethode kann als ausreichend genau für das betrachtete Problem angesehen werden.

О СКОРОСТИ РАСТВОРЕНИЯ ТВЕРДОГО ТЕЛА ПРИ НАЛИЧИИ ХИМИЧЕСКОЙ РЕАКЦИИ

Аннотация — Исследуется скорость растворения твердого тела в потоке жидкости между двумя плоскими пластинами при наличии обратимой химической реакции второго порядка. Процесс анализируется как изотермический, протекающий в переходной области стационарного ламинарного потока. На основе дифференциальных уравнений баланса количества движения и массы при соответствующих начальных и граничных условиях сформулирована модель процесса и получено численное решение для реакции бензойной кислоты с метанолом. Предложенная модель позволеет рассчитать профиль концентрации для любого поперечного сечения щелевого реактора. Представленная модель подтверждена экспериментально. Среднее. отклонение между экспериментальными и теоретическими данными составляет ±5,3%. Показано, что продольной диффузией можно пренебречь. Найдено, что предложенный метод расчета является достаточно точным для рассматриваемой задачи.