INTERNAL DIFFUSION IN THE EXTRUDED CATALYST PARTICLE WITH TETRALOBE CROSS SECTION

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The problem of internal diffusion in the catalyst particle of geometrical cross profile in the tetralobe shape is solved numerically for the case of the 1st order reaction taking place under isothermal conditions. The calculated values of the effectiveness factor of internal diffusion for an infinite long particle prove favourable effect of the catalyst particle shaping on its activity in comparison with the cylindrical extrudates.

One of the possibilities of partial supression of the effect of internal diffusion on the course of catalytic reaction on the particle of heterogeneous catalyst is provided by the preparation of profiled extrudates with geometrical cross section in the shape of ellipse, three-, tetra-, or polylobes[1]. The advantage of such catalyst particles is a higher ratio of the external surface to the catalyst particle volume, a certain drawback is then a lower bulk density of catalyst bed. With respect to the overall reactor output is therefore necessary, in each concrete case, to judge the suitability of applying the catalyst in the shape of extrudates with shaped cross section.

Catalyst with noncircular section were hitherto applied in large-scale hydrorefining processes in petrochemistry. For instance, the data[1] were reported making it possible to imagine qualitatively the advantages and disadvantages of applying such catalysts.

The aim of this work is to provide a quantitative picture of behaviour of the single catalyst extrudate with cross section in the shape of tetralobe under isothermal conditions and for the case of simple irreversible 1st order reaction taking place in steady state. The problem was solved on using the mathematical model describing two-dimensional diffusion in the infinite long catalyst particle.

THEORETICAL

Effectiveness of catalyst particle under the reaction conditions depends on the concentration field of reacting component in its internal volume[2]. Let us assume an extruded catalyst particle with cross section in the shape of tetralobe, assuming the circular shape of single lobes of the profile. Let us introduce the coordinate system represented in Fig. 1. With respect to the symmetry of such a particle, the problem can be solved just for one lobe or for its half as it is designated by hatching in the figure mentioned.

From the steady mass balance inside the catalyst particle for the 1st order reaction, the following elliptic partial differential second-order equation follows, given in the dimensionless form as

$$\frac{\partial^2 C}{\partial X^2} + \frac{\partial^2 C}{\partial Y^2} - F^2 C = 0\,. \tag{1}$$

Thiele modulus F is in the given case defined by the relation

$$F = r_p (k_0 / D_{eff})^{0.5}\,. \tag{2}$$

For the sake of simplicity let us consider further the perfect mass transfer outside the catalyst particle or the unit concentration of reacting component let be assumed on the external surface of this particle. The respective boundary condition for Eq. (*1*) has then the following form

$$C(X_s, Y_s) = 1\,. \tag{3}$$

the coordinates of the particle external surface being bound by the condition

$$X_s^2 + Y_s^2 - X_s - Y_s + \tfrac{1}{4} = 0\,. \tag{4}$$

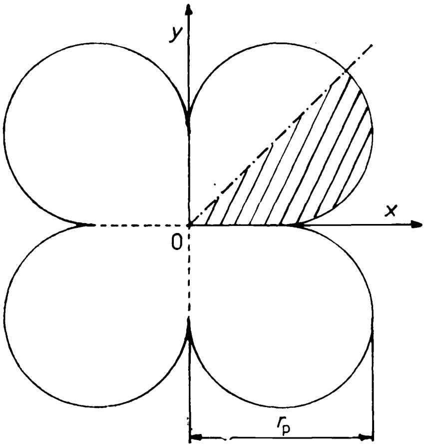

FIG. 1

Cross section of the catalyst extrudate in the shape of tetralobe with designated coordinate system

The remaining boundary conditions necessary for solving diffusion equation (*1*) follow from the symmetry of the given system

– with respect to the extrudate axis

$$X = 0,\ Y = 0,\ \partial C/\partial X = \partial C/\partial Y = 0 \tag{5}$$

– with respect to the planes normal to the extrudate cross section

$$X = 0,\ Y > 0,\ \partial C/\partial X = 0 \tag{6}$$

$$X > 0,\ Y = 0,\ \partial C/\partial Y = 0 \tag{7}$$

$$X = Y,\ \partial C/\partial X = \partial C/\partial Y = 0\,. \tag{8}$$

CALCULATIONS

Partial differential equation (*1*) with the given boundary conditions was solved by the finite difference method[3], the external circular edge of the particle cross section being replaced by the rectangular broken line consistently with the network used, as it is evident from Fig. 3. The integration step in the direction of both coordinates X and Y was chosen to be equal to 0·05, by which the concentration field in a quarter of the extrudate cross section was sought in 331 points.

By replacing the derivatives in diffusion equation (*1*) and in boundary conditions (*5*)–(*8*) by differences, a system of linear algebraic equations was obtained whose number amounted to 165 for an eighth of the extrudate profile. For the internal network knots, the single algebraic equations have the following form

$$\frac{C_{i-1,j} - 2C_{i,j} + C_{i+1,j}}{h^2} + \frac{C_{i,j-1} - 2C_{i,j} + C_{i,j+1}}{h^2} - F^2 C_{i,j} = 0\,. \tag{9}$$

The comparatively extensive system of equations was solved by the Gauss–Seidel iteration algorithm[3], the new calculated value of concentration at the given network knot, $C'_{i,j}$, being determined according to the recurrence relation derived from Eq. (*9*)

$$C'_{i,j} = (C_{i-1,j} + C_{i+1,j} + C_{i,j-1} + C_{i,j+1})/(4 + F^2 h^2)\,. \tag{10}$$

Boundary conditions (*5*)–(*8*) were replaced by simple two-point differences according to the relations

$$C'_{0,0} = C_{1,0} \tag{11}$$

$$C'_{0,j} = C_{1,j} \tag{12}$$

$$C'_{i,0} = C_{i,1} \tag{13}$$

$$C'_{i,j} = C_{j,i} . \tag{14}$$

The iteration calculation was started for values $C_{i,j} = 1$ and ended on satisfying the condition

$$\left|(C'_{i,j} - C_{i,j})/C_{i,j}\right| < 0{\cdot}001 \tag{15}$$

at all network knots. The number of iterations needed to achieve the accuracy required depends on the value of the Thiele modulus as it is illustrated in Fig. 2.

The calculated concentration field inside the catalyst particle was represented for a quarter of the extrudate cross section by printing the first significant digit of concentration at the given network knot. It made it possible to distinguish the boundaries of regions of the same concentration of reacting component in the internal space of catalyst particle.

From the concentration field of reacting component of the catalyst particle can also easily be determined the effectiveness factor of internal diffusion which corresponds to the arithmetic mean of dimensionless concentrations of reacting component at all the network points, or

$$E = \frac{1}{m} \sum_{i=0}^{19} \sum_{j=0}^{19} C'_{i,j} , \quad \text{where} \quad m = 331 . \tag{16}$$

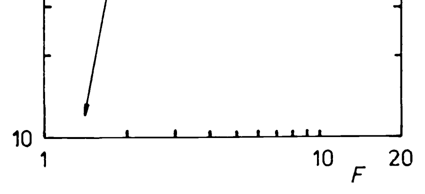

FIG. 2

Number of iterations necessary for solving the system of difference equations as a function of the Thiele modulus

RESULTS AND DISCUSSION

Concentration Field in Catalyst Particle

Visualization of the effect of internal diffusion on the concentration field of reacting component in profiled extrudate with cross section in the shape of tetralobe is yielded by Figs 3 – 5 in which the regions of the same concentration are depicted as expressed

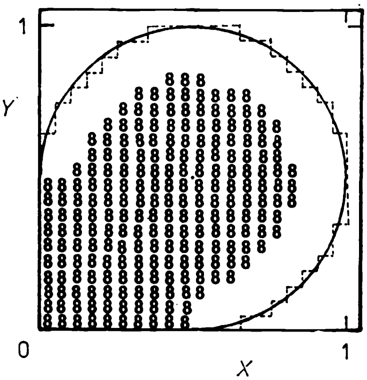

FIG. 3

Concentration field of reacting component in a quarter of section of catalyst particle (nearly kinetic region) $F = 2$; figure 8 denotes the region of values of dimensionless concentration $C = 0{\cdot}8$

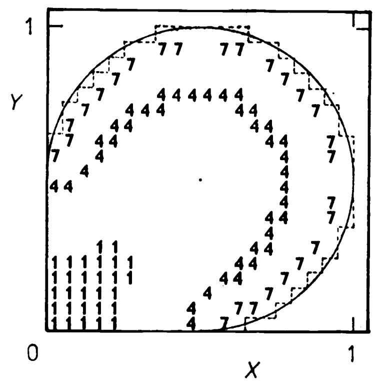

FIG. 4

Concentration field of reacting component in a quarter of section of catalyst particle (transition region) $F = 5$; figures 7, 4, and 1 denote regions of values of dimensionless concentrations $C = 0{\cdot}7$, $0{\cdot}4$, and $0{\cdot}1$, respectively

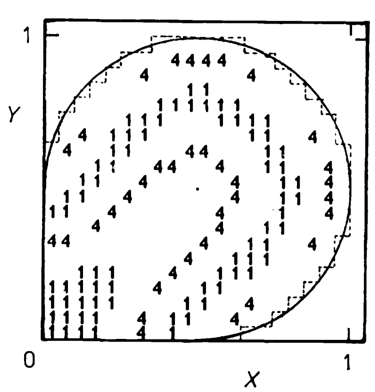

FIG. 5

Concentration field of reacting component in a quarter of section of catalyst particle (diffusion region) $F = 10$; concentration contours from the edge of particle are $C = 0{\cdot}4$, $0{\cdot}1$, $0{\cdot}04$, $0{\cdot}01$

by printing its first digit. The results were obtained for the Thiele modulus 2, 5, and 10. The growing influence of internal diffusion on increasing the value of the Thiele modulus which leads among others to lower values of concentration of component in the axis of extrudate (the minimum component concentration decreases from the values of 0·8 or 0·1 down to the value of 0·01 for the highest Thiele modulus). In the above-mentioned figures is further distinct the effect of the catalyst particle shape on the curvature of zones of the same component concentration.

The effect of internal diffusion on the concentration conditions in the catalyst particle is evident, in addition, from Fig. 6 in which the component radial concentration profiles along the plains of symmetry, therefore, concentrations $C_{0,j}$ or $C_{i,i}$, represented by dashed or solid curves, respectively, are plotted. It is apparent from the profiles that in case of stronger effect of internal diffusion on the course of reaction, i.e. for higher values of the Thiele modulus, the concentration profiles in the vicinity of the particle surface are steeper, and simultaneously, the value of the reacting component concentration in the axis of extruded particle decreases.

Effectiveness Factor of Internal Diffusion

The interaction between the diffusion transfer of reacting component into the internal volume of particle and the surface reaction proper is decisive for the rate of catalytic reaction taking place inside the actual particle. The real rate of process is then

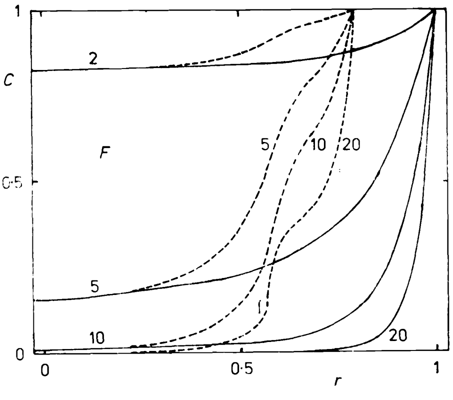

FIG. 6

Radial concentration profiles of reacting component in planes of symmetry of catalyst particle: ——— $C_{i,i}$, – – – $C_{0,j}$

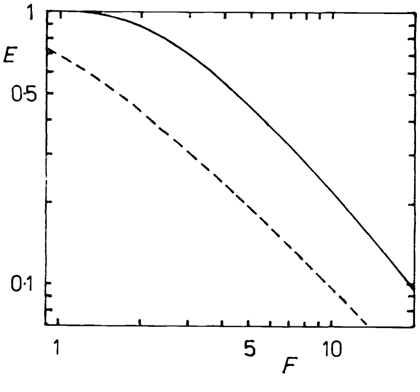

FIG. 7

Effectiveness factor of internal diffusion as a function of the Thiele modulus: ——— extrudate with tetralobe profile with lobes of diameter r_p, – – – cylinder[2] of radius r'_p, with $r_p = r'_p$

directly proportional to the effectiveness factor of internal diffusion which in the given case was evaluated according to Eq. (*16*). The resulting values are represented in Fig. 7 as a function of the Thiele modulus. It is apparent that the effect of mass transfer on the course of reaction may be neglected for $F < 0{\cdot}2$, and on the contrary, for values of the Thiele modulus $F > 10$, the particle performs in the region of strong effect of internal diffusion. This dependence is of similar character which corresponds to the cylindrical extrudates for which the dashed curve was taken from the monograph by Aris[2]. When comparing both the dependences, it is necessary to respect the choice of the particle characteristic dimension for the Thiele modulus definition. The data represented in Fig. 7 for cylindrical geometry correspond to the Thiele modulus definition according to Eq. (*2*) on condition that r'_p (the cylinder radius of unprofiled cylindrical particle) is used instead of r_p.

Fig. 7 proves the favourable effect of shaping the section of extruded catalyst particles leading to higher values of effectiveness factor of internal diffusion. Consequently in the diffusion region it is possible to reach a considerable increase of reaction rate (approximately by 100%). This conclusion corresponds to the experimental results[1] obtained in hydrodesulphurization of vacuum oil on using the Haldor Topsøe TK 551 catalyst, even though in the shape of three-lobe particle profile. From the calculated dependences of effectiveness factor on the Thiele modulus, it is therefore possible to estimate the "equivalent" radius of the tetralobe extrudate which has the same effectiveness factor as the cylindrical extrudate. In the region of strong effect of internal diffusion, such a radius approaches the radius of the extrudate lobe.

SYMBOLS

C	dimensionless concentration of reacting component related to its concentration on particle surface
D_{eff}	effective diffusion coefficient, $m^2 s^{-1}$
E	effectiveness factor of internal diffusion
F	Thiele modulus, (Eq. (*2*))
h	integration step
k_0	rate constant, s^{-1}
m	number of network points
n	number of iterations
r_p	lobe diameter in tetralobe cross section of catalyst (Fig. 1), m
r'_p	radius of cylindrical particle of catalyst, m
X	coordinate
Y	coordinate

Subscripts

i	designation of network knot in direction of coordinate X
j	designation of network knot in direction of coordinate Y
s	external surface of extrudate

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