Relative Importance of Mass Transfer and Chemical Reaction in Fixed-bed Converters

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One of the important considerations in the study or operation of a fixed-bed catalytic converter is the relative contribution of mass transfer and chemical reaction at the particle surface to the determination of the over-all reaction rate. It is obvious that, if mass transfer controls, the investigation is not a reaction study but is, in essence, a diffusion study. In industry, under such conditions, an increase in productivity can be obtained through an increase in flow rate. Conversely, if the process is controlled by the reaction occurring at or on the particle surface, then flow rate through the bed is not a significant parameter except insofar as it determines the contact time. Thus it is important in any research study utilizing a fixed-bed catalytic reactor to determine whether the rate of mass transfer between fluid phase and particle surface is approximately the same as the rate of reaction at or on the particle surface.

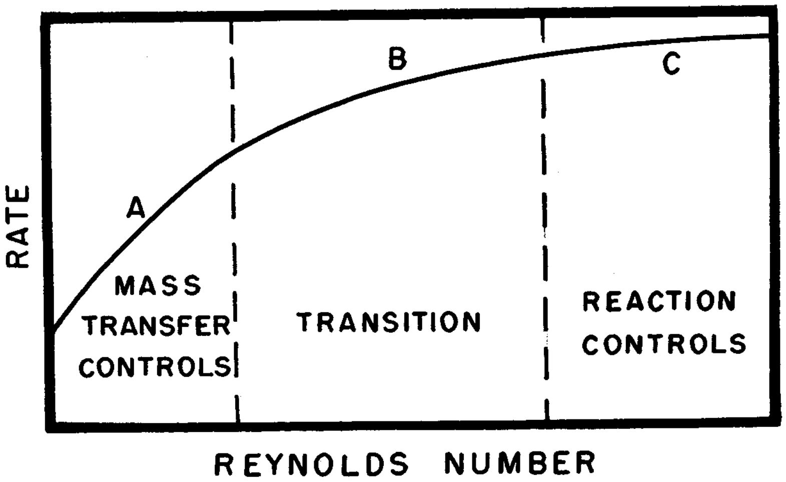

Fig. 1.

The word *control* as used in the previous paragraph designates a situation wherein one step is exceedingly slow in comparison with another and, hence, the over-all observed result depends upon the rate of that controlling step.

At the present time there are generally two methods used by those engaged in fixed-bed catalytic-reaction studies to determine the importance of mass transfer to the process under investigation. The first technique is to calculate the partial pressure drop through the "film" by use of the method of Wilke and Hougen (*1*) and/or that of Yang and Hougen (*2*) and to compare this with the partial pressure of the significant or limiting reactant in the gas phase. Others have varied the flow rate to observe its effect on conversion.

It is apparent that at constant contact time the rate–Reynolds-number relationship would appear as shown schematically in the figure. However, if one operates at a fixed-bed length and varies the Reynolds number and if, simultaneously, one finds oneself in the range designated by *A* in Figure 1, wherein the rate is varying almost linearly with Reynolds number, it is obvious that the effect of Reynolds number on mass transfer is confounded with the effect of contact time on reaction rate. Doubling the Reynolds number would increase the rate of mass transfer but would decrease the contact time—possibly in about the same proportion. Thus only quite sensitive analytical techniques would be expected to pick up the effect of a change

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Dispersed-phase Holdup

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In the paper "Dispersed-phase Holdup in Packed, Countercurrent Liquid-liquid Extraction Columns," by C. E. Wicks and R. B. Beckmann, which appeared in the December, 1955, issue of the *Journal* (page 426), reference is made to a paper on the same subject by Gayler and myself. Thus, on page 427 the following statement is made:

"Gayler and Pratt also present the following equation for normal holdup at flooding:

$$X_{flood} = 0.62 \frac{V_D{}^2 a_v}{gF^3}$$

which must be in error, for a substitution of the proper values for the variables into the equation resulted in values of X_{flood} greater than one, which is an impossibility."

The above expression is quoted incorrectly and should read* as follows:

$$X_{flood} = 0.62\left[\frac{V_D{}^2 a_v}{gF^3}\,\frac{\rho_d}{\Delta\rho}\right]^{0.11}$$

Further, substitution of data from the authors' paper, as well as our original data (*2*), indicates that neither of the above expressions gives values for X_{flood} greater than unity; the incorrect expression in fact gives impossibly *low* values of X_{flood}, owing to omission of the exponent.

The second of the above expressions represented an early attempt of ours to correlate holdup at the flood point for packed columns, and the authors may perhaps be interested in our later work. Thus, Thornton (*7*) has shown that for mechanically agitated columns, where the droplet size is determined entirely by the power input, the following expression for the holdup, derived originally by Gayler, Roberts, and Pratt (*3*) for packed columns, is applicable up to the flood point:

$$V_D + \frac{X}{1 - X} V_c = F\bar{v}_0 X(1 - X)$$

where $\bar{v}_0$ is the "characteristic droplet velocity."

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*The symbols of Wicks and Beckmann are used here.

By differentiating with respect to X, treating V_C and V_D as dependent variables, setting the differentials equal to zero, and eliminating V_C, V_D, and $\bar{v}_0$, the following equation is obtained for the holdup at the floodpoint:

$$X_{flood} = \frac{(L_R^2 + 8L_R)^{0.5} - 3L_R}{4(1 - L_R)}$$

where $L_R = V_D/V_C$

This expression indicates that the holdup at the flooding point is dependent only upon the flow ratio and is independent of the actual flow rates and the physical properties of the phases, and it has been shown to be applicable to spray (*5*), rotary annular (*7*), rotary disk (*6*), and pulsed columns (*6*). (See also reference *4*.) In the case of the packed column, however, hydrodynamical conditions are somewhat different since a transition occurs at the point where flooding would normally be expected to take place and a region of constant holdup sets in, in which the holdup remains constant as the dispersed-phase flow is increased up to the flood point. Consequently in the case of packed columns the above equation applies to the holdup at the upper transition point and not at the flood point. Presumably, however, the holdup at the flood point could in principle be obtained in this case by trial-and-error calculation from the expressions for the throughput and holdup at the upper transition point together with the correlation of flood-point data (*1*).

The authors' concept of three, rather than two, types of holdup is not without interest. It must be remembered, however, that the "permanent" holdup (*2*, *3*) is much higher for solvents such as benzene and toluene, where the interfacial tension is high and the density difference small, than for the majority of other systems (reference *3*, Table V). Consequently the difference between the author's "free" and the "operating" holdups would in most cases be small for packings sizes above the critical.

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