

## EFFICIENCY OF RECIRCULATION IN MASS-TRANSFER PROCESSES

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*The efficiency of recirculation in mass-transfer processes has been analyzed using the apparatus of transmitting capacities. It is shown that at fixed transmitting capacities of the stages recirculation can only degrade the process. However, in a packed absorber recirculation can turn out to be advantageous, since it increases the active mass-transfer surface. Regions where the use of recirculation can be appropriate have been identified.*

The technological process called recirculation has gained wide acceptance in the chemical industry. Processes of absorption, extraction, rectification, drying [1, 2], and even precipitation occur with recirculation. Recirculation is used when it is necessary to conserve a rather expensive substance or for other purposes.

However, is the use of recirculation always justified? Its introduction does lead to a reduction in the motive force of the process. To answer this question, we conducted the investigation whose results are presented in what follows.

The method of transmitting capacities (TCs) applied to the stages of mass transfer forms the basis of the investigation [3, 4]. In accordance with this method, a mass-transfer process is described (in the "language" of the phase  $X$ ) by three transmitting capacities of successive stages: supply of the substance to the mass exchanger ( $DR$ ), transfer of the substance through the mass-transfer surface ( $K_x F$ ), and withdrawal of the substance ( $W$ ). Here  $D$  and  $W$  are the flows of the inerts;  $R$  is the constant of equilibrium (only processes with a straight line of equilibrium are considered);  $K_x$  is the coefficient of mass transfer in the "language" of the phase  $X$ ;  $F$  is the mass-transfer surface in the apparatus. The transmitting capacity of the entire counterflow apparatus (in the "language" of the phase  $X$ ) for transfer of the substance from the phase  $Y$  to the phase  $X$  is  $M/(X_{in}^{eq} - X_{in})$ , where  $M$  is the interphase flow of the substance, and  $X_{in}$  and  $X_{in}^{eq}$  are the initial concentration of the phase  $X$  and the concentration that is equilibrium to the initial one for the phase  $Y$ , respectively. With allowance for this notation, the transmitting capacity of the apparatus is [3]:

$$\frac{M}{X_{in}^{eq} - X_{in}} = \frac{\exp(-a) - \exp(-b)}{b \exp(-a) - a \exp(-b)} K_x F, \quad (1)$$

where  $a \equiv K_x F / (DR)$ ;  $b \equiv K_x F / W$ .

If we consider (Fig. 1) a process with recirculation of one of the phases (here the phase  $X$ , a degree of recirculation  $n$ ), then two equations are added to this equation, and the system of equations takes the form

$$\frac{M}{X_{in}^{eq} - X^*} = \frac{\exp(-a) - \exp(-b)}{b \exp(-a) - a \exp(-b)} K_x F, \quad (2)$$

$$WX_{in} + nWX_f = (1+n)WX^*, \quad M = W(1+n)(X_f - X^*).$$

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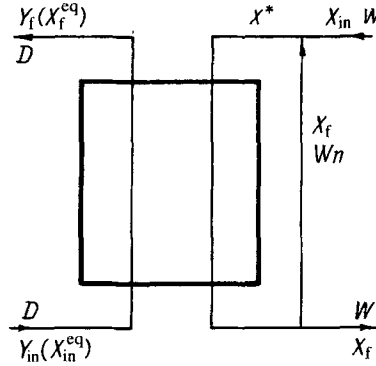


Fig. 1. Schematic of the process of mass transfer with recirculation of the phase X.

Here  $a \equiv K_x F / (DR)$ ,  $b \equiv K_x F / (W(1+n))$ ,  $X^*$  is the concentration of the substance in the phase X after mixing of the flows (at the inlet to the apparatus), and  $X_f$  is the final concentration of the substance in the phase X.

The expression

$$\frac{M}{X_{in}^{eq} - X_{in}} = \frac{\exp(-a) - \exp\left(-\frac{b}{n+1}\right)}{b \exp(-a) - \left(\frac{bn}{n+1} + a\right) \exp\left(-\frac{b}{n+1}\right)} K_x F \quad (3)$$

is the solution of the closed system of equations (2) with three unknowns ( $M$ ,  $X_f$ , and  $X^*$ ). To check formula (3), we can assume  $n$  to be equal to 0. Then formula (3), naturally, converts to (1). If we let  $n$  tend to infinity, then (3) becomes the formula for the apparatus with a completely mixed phase X [3], which also indicates the correctness of this formula.

Expression (3) is suited for calculating the transmitting capacity of the apparatus with recirculation of the phase X. However, in cases where the value of  $a$  is close to the value of  $b/(n+1)$  and where  $a$  and  $b$  are large (the value of  $K_x$  is large), calculation difficulties arise that are associated with an indeterminacy of the 0/0-fraction type. These difficulties can easily be eliminated by a technique suggested earlier [5]. We consider two cases:

1)  $a > b/(n+1)$ . If we multiply the numerator and denominator in (3) by  $\exp(b/(n+1))$ , then after transformations we obtain

$$\frac{M}{X_{in}^{eq} - X_{in}} = \frac{1 - \exp(-(a - b/(n+1)))}{a + bn/(n+1) - b \exp(-(a - b/(n+1)))} K_x F.$$

Having added  $b/(n+1)$  to and subtracted it from the denominator, we multiply and divide the numerator and denominator by  $a - b/(n+1)$ . Then (3) acquires the form

$$\frac{M}{X_{in}^{eq} - X_{in}} = \frac{\left(1 - \exp\left(-\left(a - \frac{b}{n+1}\right)\right)\right) / \left(a - \frac{b}{n+1}\right)}{1 + b \left(1 - \exp\left(-\left(a - \frac{b}{n+1}\right)\right)\right) / \left(a - \frac{b}{n+1}\right)} K_x F.$$

2)  $a < b/(n+1)$ . We multiply the numerator and denominator in (3) by  $\exp(a)$  and after similar transformations obtain

TABLE 1. Algorithm for Calculation of the Total TC

Calculated quantity	Calculation relations for	
	$a > b/(1+n)$	$a < b/(1+n)$
$d$	$a - b/(1+n)$	$b/(1+n) - a$
$H(d > 0.1)$	$(1 - \exp(-d))/d$	
$H(d < 0.1)$	$1 - d/2 + d^2/3! - d^3/4! + \dots$	
$s$	$b$	$a + bn/(n+1)$
$M/(X_{in}^{eq} - X_{in})$	$(H/(1+sH)) K_x F$	

$$\frac{M}{X_{in}^{eq} - X_{in}} = \frac{\left(1 - \exp\left(-\left(\frac{b}{n+1} - a\right)\right)\right) / \left(\frac{b}{n+1} - a\right) K_x F}{1 + \left(\frac{bn}{n+1} + a\right) \left(1 - \exp\left(-\left(\frac{b}{n+1} - a\right)\right)\right) / \left(\frac{b}{n+1} - a\right)}$$

It is easily seen that the last two expressions are very similar. The differences are in the exponent and the pre-exponential factor of the denominator. Both expressions can be represented as one by introducing the following notation:  $d = abs(b/(n+1) - a)$ ,  $H = (1 - \exp(-d))/d$ , and  $s = b$  for the first case (in the second case  $s$  will be equal to  $bn/(n+1) + a$ ). Now, the whole indeterminacy is concentrated in the expression for  $H$  at small  $d$  when the quantity  $H$  tends to zero in the numerator and denominator of the fraction. To eliminate the indeterminacy, we expand the exponent in a power series, and then  $H = 1 - d/2! + d^2/3! - d^3/4! + \dots$ . An analysis shows that when  $d < 0.1$ ,  $H$  should be calculated by the expansion, and when it is greater than or equal to 0.1, by the initial formula. This algorithm is presented in the form of Table 1.

Using the algorithm obtained, we performed calculations within a wide range of variation of  $b$  and  $c = DR/W$  (in other words, of variation of  $K_x F$ ,  $DR$ , and  $W$ ) - from 0.00001 to 100,000. At any  $b$  and  $c$  use of recirculation (its use with degrees of recirculation varying within the same range was checked) led to deterioration of the process (a decrease in the amount of the substance transferred and, naturally, in the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$ ). Thus, use of recirculation at specified transmitting capacities of the stages of mass transfer can lead only to deterioration of the process, so its use in this respect is inappropriate.

A certain advantage from the use of recirculation can reside in the following. In a packed apparatus, the active mass-transfer surface is not the entire surface of the packing, but only part of its wetted surface. Thus, with increase in the amount of the liquid in the apparatus (due to recirculation) the transmitting capacity ( $K_x F$ ) of the stage of substance transfer through the surface can increase due to the increase in the active contact surface, which, in turn, will lead to an increase in the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$ .

An analysis of the literature data [2] shows that the fraction of the active surface (in the case of a "poorly soluble" gas) is expressed as  $\Psi = AV^m \sigma^{-k}$ , where  $\sigma$  is the surface tension,  $A$  and  $k$  are constants corresponding to the size and shape of the packing,  $V$  is the mass velocity of the liquid, and  $m$  is an exponent equal to 0.455. It should be noted that, in fact, this formula (within the framework of the adopted model of packing wetting) is limited physically by the value of  $\Psi$  equal to unity. We use this formula only to trace the tendency of the change in the mass-transfer rate.

Substituting all the constants into the value for  $K_x F$  and allowing for the fact that in recirculation the mass velocity increases by a factor of  $(n+1)$ , we obtain that the transmitting capacity of the stage of mass transfer through the mass-transfer surface in the process with recirculation is expressed as  $K_x F = K_x F_0 (n+1)^m$ , where  $K_x F_0$  is the transmitting capacity of the mass-transfer surface without recirculation. We emphasize that in this case (in contrast to the previous one), recirculation, first, reduces the motive force and, second, increases the transmitting capacity of one of the mass-transfer stages. Thus, with recirculation (with increase in it), the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  must, on the one hand, increase (due

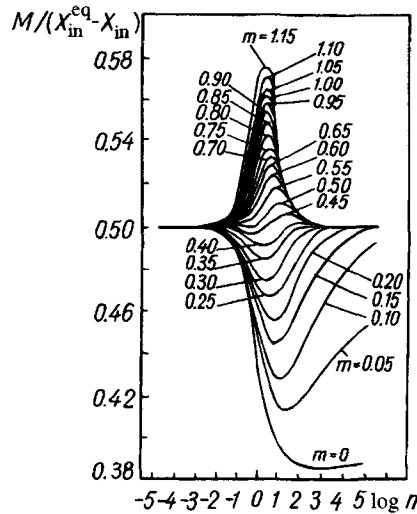


Fig. 2. Influence of the degree of recirculation  $n$  on the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  for different values of the exponent  $m$ .

to the increase in the transmitting capacity of the stage  $K_x F$ , and, on the other, decrease (due to the reduction in the mean motive force of the process). Which of these two factors will prevail must be shown by calculation.

The only difference between this calculation and the previous one was preliminary calculation of the transmitting capacity of the mass-transfer surface  $K_x F$  from the known degree of recirculation and the initial  $K_x F_0$ . An initial assumption lay in the fact that the total transmitting capacity of the apparatus either only increases or only decreases with increase in the degree of recirculation. The program of the calculation compared the value of the transmitting capacity without a recycle and its value at  $n$  tending to infinity. On the basis of a comparison of these values a conclusion was drawn about the advantage of the use of recirculation for a specified set of  $b$  and  $c$ .

However, this assumption turned out to be incorrect. The curves of the dependence of the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  on  $n$  often had an extremum and sometimes had two extrema – a maximum and a minimum. In this connection the question of the character of this dependence for any arbitrary exponent  $m$  arose. Results of this calculation at the point  $b = 1$ ,  $c = 1$  are presented in Fig. 2. In an analysis of these curves the following question can arise: what does the transmitting capacity of the apparatus tend to when  $n$  tends to infinity? To answer this question, we write (3) in the form

$$\frac{M}{X_{in}^{eq} - X_{in}} = \frac{\exp(-a) - \exp\left(-\frac{b}{n+1}\right)}{\frac{1}{W} \exp(-a) - \left(\frac{1}{W} \frac{n}{n+1} + \frac{1}{DR}\right) \exp\left(-\frac{b}{n+1}\right)}. \quad (4)$$

We consider the case  $0 < m < 1$ . Then, as  $n$  tends to infinity,  $\exp(-a)$  will tend to zero and  $\exp(-b/(n+1))$  to unity. As a result, the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  will tend to  $1/(1/(DR) + 1/W)$ , which, incidentally, is characteristic of cells with ideal mixing of at least one flow when the surface tends to infinity [3]. All the curves for these values of  $m$  in Fig. 2 come, in fact, to this value. At  $m = 0$  (a trivial case – the active surface does not increase due to recirculation), the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  tends to the expression  $(1 - \exp(-a))/(1/(DR)) + (1 - \exp(-a))/W$ , which also corresponds to the case where the phase  $X$  is completely mixed [3].

The other two cases (more likely exotic) correspond to  $m \geq 1$ . In the first case,  $a > b/(n+1)$ . Here, the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  also tends to  $1/(1/(DR) + 1/W)$ . In the other,  $a < b/$

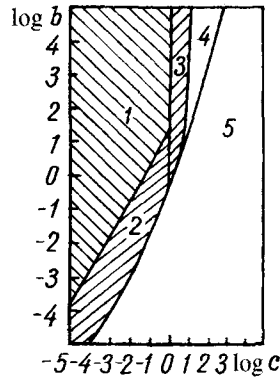


Fig. 3. Analysis of the efficiency of use of recirculation: 1-3) regions where it can be appropriate; 1-5) types of relations.

( $n + 1$ ). Here the transmitting capacity of the apparatus  $M/(X_{in}^{eq} - X_{in})$  tends simply to  $W$ . This modest analysis explains the behavior of the curves  $M/(X_{in}^{eq} - X_{in})$  with change in  $n$  and their occasionally, extremal character at different values of  $b$  and  $c$ .

In this connection we introduced a classification of the relations between  $M/(X_{in}^{eq} - X_{in})$  and  $n$ . Depending on the behavior of the curve with increase in  $n$  we distinguished five types: 1, increases monotonically; 2, increases but has an extremum (a maximum); 3, has both a minimum and a maximum (higher than the initial value); 4, decreases but has an extremum (a minimum); 5, decreases monotonically. Only the first three types correspond to cases where the use of recirculation can be justified (advantageous).

Calculation results for  $m = 0.455$  are presented in Fig. 3. Here, regions can be seen where the use of recirculation is advantageous and where it is not. The first types (1-3) lie on the left-hand side of the figure, which corresponds to small values of  $b$ , i.e., small values of  $K_r F$ . Thus, we can draw the conclusion that the use of recirculation can turn out to be advantageous only in cases where the stage of mass transfer through the mass-transfer surface is limiting.

Similar calculations were made for other values of  $m$ . The character of the regions remained the same, although the separating lines shifted somewhat.

Thus, the analysis of the process of mass transfer with recirculation conducted on the basis of the developed concept of TC shows that the use of recirculation cannot be always advantageous or always disadvantageous. The effects revealed are rather obvious from the physical point of view: if there is no effect of the flow stage on the mass transfer surface ( $K_r F$ ), the process will deteriorate, and if such an effect is present, then the competition of two tendencies occurs, and the analysis conducted makes it possible to determine regions where the use of recirculation is advantageous and vice versa. If we consider the conclusions drawn in relation to other concepts of mass transfer, then in practice the approach from the position of the long-developed method of transfer units will give similar results.

Probably, in the future it is worthwhile to conduct a similar investigation for the case of a poorly soluble gas, where an increase in the transmitting capacity of the flow stage is caused by an increase in the coefficient of mass transfer rather than by surface enlargement.

## NOTATION

$D$  and  $W$ , mass flows of the inerts, kg/sec;  $F$ , mass-transfer surface,  $m^2$ ;  $K$ , coefficient of mass transfer,  $kg/(m^2 \cdot sec)$ ;  $M$ , transferred mass, kg/sec;  $R$ , constant of equilibrium, kg/kg;  $X$  and  $Y$ , relative mass concentrations of the component in the phases  $X$  and  $Y$ , respectively, kg/kg;  $n$ , degree of recirculation;  $\Psi$ , fraction of the active surface of the packing. Superscripts: eq, equilibrium; \*, after mixing. Subscripts: in, initial; f, final;  $x$ , related to the phase  $X$ ; 0, initial (without recirculation).

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