

THEORETICAL ANALYSIS OF COUNTER-CURRENT ABSORPTION OF A POORLY SOLUBLE GAS BASED ON THE PDE--AD MODEL

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Expressions have been derived in the paper for all four possible transfer functions between the inlet and the outlet gas and liquid streams under the counter-current absorption of a poorly soluble gas in a packed bed column. The transfer functions have been derived for the axially dispersed model with stagnant zone in the liquid phase and the axially dispersed model for the gas phase with interfacial transport of a gaseous component (PDE--AD). Calculations with practical values of parameters suggest that only two of these transfer functions are applicable for experimental data evaluation.

The existence of flow non-idealities in both phases in packed bed columns and other industrial equipment and their negative impact on the efficiency of operation of this equipment has been generally acknowledged. In order to describe axial mixing, provided that an attempt to account for this effect is made at all, the axially dispersed model¹ has been mostly used. Experimental evidence, however, shows that although this model may suit well for the flow of the gas phase, the response curves of the liquid phase indicate that at least part of the liquid passes through the bed by a mechanism different from the one embodied in the axially dispersed model². New models describing the flow of liquid have been therefore proposed with the dynamic and the stagnant liquid zone (PE — piston exchange)³, the axially dispersed model with the dynamic and the stagnant zone (PDE — piston diffusion exchange)⁴ and others. Both just mentioned models describe the reality better than the axially dispersed model but contain more parameters.

The problem of the real flows in irrigated beds of solid particles has been so far studied separately in the gas and the liquid phase with the aid of tracers that do not pass the interface. A tracer capable of crossing the interfacial surface permits simultaneous investigation of the dynamics of one or both phases in the presence of interfacial transport. Works of this kind, however, are so far rather rare^{5,6}.

Models with the dynamic and the stagnant zone, PE and PDE, have not been so far used for research of the flow dynamics in the presence of interfacial transfer. One of the reasons for this situation is no doubt the high number of parameters involved in the model under the two phase flow. For instance, using the PDE model

for both the liquid and the gas phase the total number of parameters amounts to 12. This is, of course, too much from the stand point of subsequent parameter evaluation from experimental data obtained by currently available methods. The number of parameters is somewhat diminished provided that the PDE or PE models are applied to only one phase.

Solution of such models in the time domain, although possible, is awkward. The difficulty of the solution, however, can be reduced by solving the underlying model equations in the Laplace or the frequency domain, for there is no need for the transformation back into the time domain.

The aim of this paper has been to formulate the model of the two-phase counter-current flow with interfacial transfer utilizing the concept of the stagnant zones in the liquid phase and its solution in the frequency domain. From these results one can further derive, by relatively simple means, simpler asymptotic solutions. Organic part of the aim has also been a numerical test of the obtained transfer functions from the stand point of their eventual utilization for evaluation of individual parameters for the process of absorption of a poorly soluble gas. The obtained results should serve in turn for designing experiments to verify the validity of the presented models.

THEORETICAL

Upon confining the following considerations to the case of absorption of a poorly soluble gas in a counter-current column, the flow rates of the gas and the liquid phase will remain constant throughout the column and the resistance to mass transfer will be concentrated on the side of liquid. The overall mass transfer coefficients may then be replaced by the gas side mass transfer coefficients. In addition, we shall assume independence of all quantities and parameters on the radial coordinate which reduces the model to a unidimensional one.

Axially Dispersed Model with the Stagnant Zone in the Liquid Phase

The model for the two-phase flow is constructed from the axially dispersed model (AD) for the gas phase and the axially dispersed model with the stagnant zone (PDE) for the liquid phase. The scheme of the model is shown in Fig. 1. The model contains eight parameters. These are: the gas, the dynamic liquid and the stagnant liquid hold-ups, h_G , h_D and h_S , the volume mass transfer coefficient between the gas and the dynamic liquid hold-up, $k_{LD}a_D$, the mass transfer coefficient between the gas and the stagnant liquid hold-up, $k_{LS}a_S$, the coefficient of liquid exchange between the dynamic and the stagnant hold-up, q , and the axial dispersion coefficients in the gas and the dynamic liquid hold-ups E_G and E_D .

A balance on mass of the absorbed component in individual phases yields:

$$h_G E_G \frac{\partial^2 C_G}{\partial z^2} - G \frac{\partial C_G}{\partial z} - k_{LD} a_D \left(\frac{C_G}{m} - C_D \right) - k_{LS} a_S \left(\frac{C_G}{m} - C_S \right) = h_G \frac{\partial C_G}{\partial t} \quad (1)$$

$$h_D E_D \frac{\partial^2 C_D}{\partial z^2} + L \frac{\partial C_D}{\partial z} + k_{LD} a_D \left(\frac{C_G}{m} - C_D \right) + q(C_S - C_D) = h_D \frac{\partial C_D}{\partial t} \quad (2)$$

$$k_{LS} a_S \left(\frac{C_G}{m} - C_S \right) - q(C_S - C_D) = h_S \frac{\partial C_S}{\partial t} \quad (3)$$

The concentrations C_G , C_D , and C_S are taken to be the deviations of the appropriate concentrations from their steady state values. This definition is advantageous in view of the subsequent solution of the set of Eqs (1)–(3) by the Laplace transform.

The aim of the solution of the set of Eqs (1)–(3) is to derive the transfer functions of the model. The transfer function is generally the ratio of the Laplace transforms

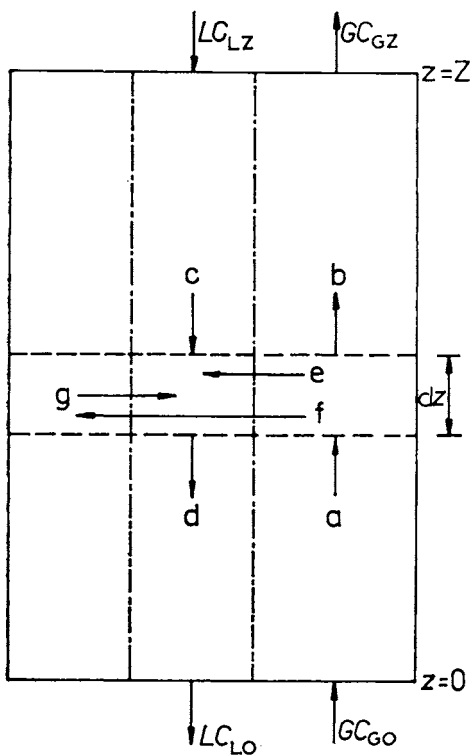


FIG. 1

Balance scheme of the PDE-AD model. **a** $GC_G - h_G E_G (\partial C_G / \partial z)$, **b** $GC_G + G (\partial C_G / \partial z) dz - h_G E_G (\partial C_G / \partial z) - h_G E_G (\partial^2 C_G / \partial z^2) dz$, **c** $LC_D + L (\partial C_D / \partial z) dz + h_D E_D \cdot (\partial C_D / \partial z) + h_D E_D (\partial^2 C_D / \partial z^2) dz$, **d** $LC_D + h_D E_D (\partial C_D / \partial z)$, **e** $k_{LD} a_D (C_G / m - C_D)$, **f** $k_{LS} a_S (C_G / m - C_S)$, **g** $q(C_S - C_D)$

of the outlet and the inlet signals. For the two-phase flow with interfacial mass transfer one can derive altogether four different transfer functions. These are: the outlet gas to inlet gas stream transfer function – X_{GZ}/X_{G0} , the outlet liquid to inlet gas stream transfer function – X_{L0}/X_{G0} , the outlet liquid to inlet liquid stream transfer function – X_{L0}/X_{LZ} and the outlet gas to inlet liquid stream transfer function – X_{GZ}/X_{LZ} .

The Laplace transform of the original set of partial differential equations (1) and (2) and the ordinary differential equation (3) with the initial condition:

$$t = 0, \quad C_G = C_D = C_S = 0 \quad (4)$$

yields two ordinary differential equations and a single algebraic equations in the form

$$h_G E_G \frac{d^2 X_G}{dz^2} - G \frac{dX_G}{dz} - k_{LD} a_D \left(\frac{X_G}{m} - X_D \right) - k_{LS} a_S \left(\frac{X_G}{m} - X_S \right) = h_G s X_G \quad (5)$$

$$h_D E_D \frac{d^2 X_D}{dz^2} + L \frac{dX_D}{dz} + k_{LD} a_D \left(\frac{X_G}{m} - X_D \right) + q(X_S - X_D) = h_D s X_D \quad (6)$$

$$k_{LS} a_S \left(\frac{X_G}{m} - X_S \right) - q(X_S - X_D) = h_S s X_S \quad (7)$$

The choice of the initial condition in the form (4) affects in no way the statement of the problem for it only means that all three concentrations are measured as deviations from the steady state values. Any problem can be transformed to this case by a simple transformation while the controlling equations remain unchanged.

The set of Eqs (5)–(7) may be solved analytically, for instance, by eliminating the concentrations X_S and X_D , which transforms the set (5)–(7) to a fourth order differential equation with constant coefficients:

$$A_1 \frac{d^4 X_G}{dz^4} + A_2 \frac{d^3 X_G}{dz^3} + A_3 \frac{d^2 X_G}{dz^2} + A_4 \frac{dX_G}{dz} + A_5 X_G = 0, \quad (8)$$

where

$$A_1 = -H_1 H_5 / H_4 \quad (9)$$

$$A_2 = -(H_2 H_5 + H_1 H_6) / H_4 \quad (10)$$

$$A_3 = -(H_3 H_5 + H_2 H_6 + H_1 H_7) / H_4 \quad (11)$$

$$A_4 = -(H_3 H_6 + H_2 H_7) / H_4 \quad (12)$$

$$A_5 = (H_4 H_8 - H_3 H_7) / H_4. \quad (13)$$

Subscripted H 's in Eqs (9)–(13) have the following meaning:

$$H_1 = h_G E_G \quad (14)$$

$$H_2 = -G \quad (15)$$

$$H_3 = \frac{(k_{LS} a_S)^2}{m(k_{LS} a_S + q + h_S s)} - \frac{k_{LD} a_D}{m} - \frac{k_{LS} a_S}{m} - h_G s \quad (16)$$

$$H_4 = k_{LD} a_D + \frac{k_{LS} a_S q}{k_{LS} a_S + q + h_S s} \quad (17)$$

$$H_5 = h_D E_D \quad (18)$$

$$H_6 = L \quad (19)$$

$$H_7 = \frac{q^2}{k_{LS} a_S + q + h_S s} - q - k_{LD} a_D - h_D s \quad (20)$$

$$H_8 = \frac{k_{LS} a_S q}{m(k_{LS} a_S + q + h_S s)} + \frac{k_{LD} a_D}{m}. \quad (21)$$

The characteristic equation of the differential equation (8) reads as follows:

$$A_1 \lambda^4 + A_2 \lambda^3 + A_3 \lambda^2 + A_4 \lambda + A_5 = 0. \quad (22)$$

It is a fourth order algebraic equation with complex coefficients. Solution of this equation by standard means is impossible. If, however, Eq. (22) is multiplied by a fourth order equation with the complex conjugate coefficients, an eighth order equation results:

$$\begin{aligned} & A_1 B_1 \lambda^8 + (A_1 B_2 + A_2 B_1) \lambda^7 + (A_1 B_3 + A_2 B_2 + A_3 B_1) \lambda^6 + \\ & + (A_1 B_4 + A_2 B_3 + A_3 B_2 + A_4 B_1) \lambda^5 + (A_1 B_5 + A_2 B_4 + A_3 B_3 + A_4 B_2 + \\ & + A_5 B_1) \lambda^4 + (A_2 B_5 + A_3 B_4 + A_4 B_3 + A_5 B_2) \lambda^3 + (A_3 B_5 + A_4 B_4 + \\ & + A_5 B_3) \lambda^2 + (A_4 B_5 + A_5 B_4) \lambda + A_5 B_5 = 0, \end{aligned} \quad (23)$$

where B_i , $i = 1, 2, 3, 4, 5$, are coefficients complex conjugate to A_i . Eq. (23) has real

coefficients and may be solved by standard algorithms. Its solution yields four pairs of complex conjugate roots while always only one out of each pair of roots satisfies Eq. (22). Selection of the roots $\lambda_1, \lambda_2, \lambda_3$ and λ_4 can be made on the basis of the test of validity of Eq. (22). Solution of the set of Eqs (5)–(7) can then be written in the form:

$$X_G = k_1 \exp(\lambda_1 z) + k_2 \exp(\lambda_2 z) + k_3 \exp(\lambda_3 z) + k_4 \exp(\lambda_4 z) \quad (24)$$

$$X_D = a_1 k_1 \exp(\lambda_1 z) + a_2 k_2 \exp(\lambda_2 z) + a_3 k_3 \exp(\lambda_3 z) + a_4 k_4 \exp(\lambda_4 z), \quad (25)$$

where

$$a_i = -H_1 \lambda_i^2 / H_4 - H_2 \lambda_i / H_4 - H_3 / H_4, \quad i = 1, 2, 3, 4. \quad (26)$$

It would be also possible to write an analogous expression for the concentration X_S . Since, however, only the outlet and the inlet concentrations are practically interesting, these expressions shall not be presented here.

The constants $k_1 - k_4$ in Eqs (24) and (25) may be determined, using Danckwerts' boundary conditions for the gas inlet and the liquid outlet cross section

$$z = 0, \quad X_G = X_{GO} + \frac{E_G h_G}{G} \frac{dX_G}{dz}, \quad \frac{dX_D}{dz} = 0 \quad (27)$$

and the liquid inlet and the gas outlet cross section:

$$z = Z, \quad X_D = X_{LZ} - \frac{E_D h_D}{L} \frac{dX_D}{dz}, \quad dX_G/dz = 0 \quad (28)$$

in the form:

$$k_1 = \frac{\lambda_1 (h_4 X_{LZ} + h_8 X_{GO})}{h_1 h_6 - h_2 h_5} \quad (29)$$

$$k_2 = \frac{\lambda_1 (h_3 X_{LZ} + h_7 X_{GO})}{h_1 h_6 - h_2 h_5} \quad (30)$$

$$k_3 = \frac{\lambda_1 (h_2 X_{LZ} + h_6 X_{GO})}{h_1 h_6 - h_2 h_5} \quad (31)$$

$$k_4 = \frac{\lambda_1 (h_1 X_{LZ} + h_5 X_{GO})}{h_1 h_6 - h_2 h_5}, \quad (32)$$

where

$$h_1 = [a_3 b_2 \lambda_1 \lambda_3 \exp(\lambda_1 Z) - a_2 b_3 \lambda_1 \lambda_2 \exp(\lambda_1 Z) +$$

$$+ a_1 b_3 \lambda_1 \lambda_2 \exp(\lambda_2 Z) - a_3 b_1 \lambda_2 \lambda_3 \exp(\lambda_2 Z) + \\ + a_2 b_1 \lambda_2 \lambda_3 \exp(\lambda_3 Z) - a_1 b_2 \lambda_1 \lambda_3 \exp(\lambda_3 Z)]/J \quad (33)$$

$$h_2 = [a_2 b_4 \lambda_1 \lambda_2 \exp(\lambda_1 Z) - a_4 b_2 \lambda_1 \lambda_4 \exp(\lambda_1 Z) + \\ + a_4 b_1 \lambda_2 \lambda_4 \exp(\lambda_2 Z) - a_1 b_4 \lambda_1 \lambda_2 \exp(\lambda_2 Z) + \\ + a_1 b_2 \lambda_1 \lambda_4 \exp(\lambda_4 Z) - a_2 b_1 \lambda_2 \lambda_4 \exp(\lambda_4 Z)]/J \quad (34)$$

$$h_3 = [a_4 b_3 \lambda_1 \lambda_4 \exp(\lambda_1 Z) - a_3 b_4 \lambda_1 \lambda_3 \exp(\lambda_1 Z) + \\ + a_1 b_4 \lambda_1 \lambda_3 \exp(\lambda_3 Z) - a_4 b_1 \lambda_3 \lambda_4 \exp(\lambda_3 Z) + \\ + a_3 b_1 \lambda_3 \lambda_4 \exp(\lambda_4 Z) - a_1 b_3 \lambda_1 \lambda_4 \exp(\lambda_4 Z)]/J \quad (35)$$

$$h_4 = [a_3 b_4 \lambda_2 \lambda_3 \exp(\lambda_2 Z) - a_4 b_3 \lambda_2 \lambda_4 \exp(\lambda_2 Z) + \\ + a_4 b_2 \lambda_3 \lambda_4 \exp(\lambda_3 Z) - a_2 b_4 \lambda_2 \lambda_3 \exp(\lambda_3 Z) + \\ + a_2 b_3 \lambda_2 \lambda_4 \exp(\lambda_4 Z) - a_3 b_2 \lambda_3 \lambda_4 \exp(\lambda_4 Z)]/J \quad (36)$$

$$h_5 = [a_1 a_3 d_1 \lambda_2 \lambda_3 \exp(\lambda_1 Z + \lambda_2 Z) - a_2 a_3 d_2 \lambda_1 \lambda_3 \exp(\lambda_1 Z + \lambda_2 Z) + \\ + a_2 a_3 d_3 \lambda_1 \lambda_2 \exp(\lambda_1 Z + \lambda_3 Z) - a_1 a_2 d_1 \lambda_2 \lambda_3 \exp(\lambda_1 Z + \lambda_3 Z) + \\ + a_1 a_2 d_2 \lambda_1 \lambda_3 \exp(\lambda_2 Z + \lambda_3 Z) - a_1 a_3 d_3 \lambda_1 \lambda_2 \exp(\lambda_2 Z + \lambda_3 Z)]/J \quad (37)$$

$$h_6 = [a_2 a_4 d_2 \lambda_1 \lambda_4 \exp(\lambda_1 Z + \lambda_2 Z) - a_1 a_4 d_1 \lambda_2 \lambda_4 \exp(\lambda_1 Z + \lambda_2 Z) + \\ + a_1 a_2 d_1 \lambda_2 \lambda_4 \exp(\lambda_1 Z + \lambda_4 Z) - a_2 a_4 d_4 \lambda_1 \lambda_2 \exp(\lambda_1 Z + \lambda_4 Z) + \\ + a_1 a_4 d_4 \lambda_1 \lambda_2 \exp(\lambda_2 Z + \lambda_4 Z) - a_1 a_2 d_2 \lambda_1 \lambda_4 \exp(\lambda_2 Z + \lambda_4 Z)]/J \quad (38)$$

$$h_7 = [a_1 a_4 d_1 \lambda_3 \lambda_4 \exp(\lambda_1 Z + \lambda_3 Z) - a_3 a_4 d_3 \lambda_1 \lambda_4 \exp(\lambda_1 Z + \lambda_3 Z) + \\ + a_3 a_4 d_4 \lambda_1 \lambda_3 \exp(\lambda_1 Z + \lambda_4 Z) - a_1 a_3 d_1 \lambda_3 \lambda_4 \exp(\lambda_1 Z + \lambda_4 Z) + \\ + a_1 a_3 d_3 \lambda_1 \lambda_4 \exp(\lambda_3 Z + \lambda_4 Z) - a_1 a_4 d_4 \lambda_1 \lambda_3 \exp(\lambda_3 Z + \lambda_4 Z)]/J \quad (39)$$

$$h_8 = [a_3 a_4 d_3 \lambda_2 \lambda_4 \exp(\lambda_2 Z + \lambda_3 Z) - a_2 a_4 d_2 \lambda_3 \lambda_4 \exp(\lambda_2 Z + \lambda_3 Z) + \\ + a_2 a_3 d_2 \lambda_3 \lambda_4 \exp(\lambda_2 Z + \lambda_4 Z) - a_3 a_4 d_4 \lambda_2 \lambda_3 \exp(\lambda_2 Z + \lambda_4 Z) + \\ + a_2 a_4 d_4 \lambda_2 \lambda_3 \exp(\lambda_3 Z + \lambda_4 Z) - a_2 a_3 d_3 \lambda_2 \lambda_4 \exp(\lambda_3 Z + \lambda_4 Z)]/J. \quad (40)$$

Some of the parameters appearing in Eqs (33)–(40) designate the following:

$$J = a_1 \lambda_2 \exp(\lambda_2 Z) - a_2 \lambda_1 \exp(\lambda_1 Z) \quad (41)$$

$$b_i = 1 - \frac{h_G E_G \lambda_i}{G}, \quad i = 1, 2, 3, 4 \quad (42)$$

$$d_i = 1 + \frac{h_D E_D \lambda_i}{L}, \quad i = 1, 2, 3, 4. \quad (43)$$

Upon substituting for the constants k_1 through k_4 in Eqs (24) and (25) from Eqs (29)–(32) one obtains solutions for the concentrations in the gas and the dynamic part of the liquid phase in the Laplace domain.

If the concentration in the inlet liquid stream is not perturbed, then, according to the understanding that the concentrations are taken as deviations from the steady state values, one can write:

$$X_{LZ} = 0. \quad (44)$$

Under such conditions the expression for the concentration in the outlet gas stream is obtained by substituting $z = Z$ into Eq. (24). The resulting expression is then easily rearranged with the aid of Eqs (29)–(32) to yield the transfer function X_{GZ}/X_{GO} representing the concentration response in the outlet gas stream following a change of the concentration in the inlet gas stream:

$$\frac{X_{GZ}}{X_{GO}} = \frac{\lambda_1 [h_8 \exp(\lambda_1 Z) + h_7 \exp(\lambda_2 Z) + h_6 \exp(\lambda_3 Z) + h_5 \exp(\lambda_4 Z)]}{h_1 h_6 - h_2 h_5}. \quad (45)$$

Under the same conditions, Eq. (44), *i.e.* if the inlet liquid stream is not being perturbed, one obtains after substituting $z = 0$ into Eq. (25), while making use of Eqs (29)–(32), an expression for the outlet liquid concentration. This in turn can be modified by an arithmetic operation to yield the transfer function X_{LO}/X_{GO} . This transfer function represents the response in the outlet liquid stream to a perturbation of the inlet gas stream.

$$\frac{X_{LO}}{X_{GO}} = \frac{\lambda_1 (a_1 h_8 + a_2 h_7 + a_3 h_6 + a_4 h_5)}{h_1 h_6 + h_2 h_5} \quad (46)$$

If, on the contrary, the inlet gas stream is not being perturbed, one writes, again with the understanding regarding the reference value of the concentration scale, that:

$$X_{GO} = 0. \quad (47)$$

With this assumption we can derive two transfer functions. One of them is X_{LO}/X_{LZ} , expressing the concentration changes of the liquid outlet stream in response to a con-

centration change in the inlet liquid stream. This transfer function is obtained from Eq. (25) by substituting $z = 0$ and after some arrangements in the form:

$$\frac{X_{LO}}{X_{LZ}} = \frac{\lambda_1(a_1h_4 + a_2h_3 + a_3h_2 + a_4h_1)}{h_1h_6 + h_2h_5} \quad (48)$$

The other function, X_{GZ}/X_{LZ} , is obtained from Eq. (24) by substituting $z = Z$ and expresses, on the contrary, the concentration changes in the outlet gas stream induced by the perturbations of the inlet liquid stream:

$$\frac{X_{GZ}}{X_{LZ}} = \frac{\lambda_1[h_4 \exp(\lambda_1 Z) + h_3 \exp(\lambda_2 Z) + h_2 \exp(\lambda_3 Z) + h_1 \exp(\lambda_4 Z)]}{h_1h_6 + h_2h_5} \quad (49)$$

Axially Dispersed Model (AD – AD)

The axially dispersed model both in the liquid and the gas phase is a limiting case of the PDE – AD model model when the stagnant liquid hold-up, h_s , is put equal to zero. This reduces the number of parameters involved from eight to five. Apart from the stagnant hold-up also disappear the mass transfer coefficient between the gas and the stagnant liquid phase $k_{LS}a_s$ and the coefficient of liquid exchange between the dynamic and the static liquid, q . The remaining model parameters are the gas and liquid hold-ups h_G and h_L , the axial dispersion coefficients in the gas and the liquid phases E_G and E_L and the mass transfer coefficient $k_L a$. A balance on mass of the absorbed component yields the following equations:

$$h_G E_G \frac{\partial^2 C_G}{\partial z^2} - G \frac{\partial C_G}{\partial z} - k_L a \left(\frac{C_G}{m} - C_L \right) = h_G \frac{\partial C_G}{\partial t} \quad (50)$$

$$h_L E_L \frac{\partial^2 C_L}{\partial z^2} + L \frac{\partial C_L}{\partial z} + k_L a \left(\frac{C_G}{m} - C_L \right) = h_L \frac{\partial C_L}{\partial t} \quad (51)$$

Solution of this set of equations may be carried out in a manner analogous to that applied to the PDE – AD model. After the Laplace transform one can modify the resulting set of equations to an ordinary fourth order differential equation with constant coefficients, Eq. (8), with the coefficients $A_1 - A_5$ being in this case defined as follows:

$$A_1 = - \frac{h_L E_L h_G E_G}{k_L a} \quad (52)$$

$$A_2 = \frac{h_L E_L G - h_G E_G L}{k_L a} \quad (53)$$

$$A_3 = \frac{h_L E_L h_{GS} + LG + h_G E_G h_{LS}}{k_L a} + \frac{h_L E_L}{m} + h_G E_G \quad (54)$$

$$A_4 = \frac{L h_{GS} - G h_{LS}}{k_L a} + \frac{L}{m} - G \quad (55)$$

$$A_5 = -h_{GS} - \frac{h_{LS}}{m} - \frac{h_{LS}^2 h_G}{k_L a} \quad (56)$$

The solution may then be written in the form of Eqs (24) and (25) replacing formally the concentration X_D by X_L in Eq. (25). The constants $a_1 - a_4$ are in this case defined by the following formula:

$$a_i = -\frac{h_G E_G}{k_L a} \lambda_i^2 + \frac{G}{k_L a} \lambda_i + \frac{h_{GS}}{k_L a} + \frac{1}{m}, \quad i = 1, 2, 3, 4. \quad (57)$$

The constants $k_1 - k_4$, in Eqs (29)–(32), were determined from the Danckwerts' boundary conditions:

$$z = 0, \quad X_G = X_{G0} + \frac{E_G h_G}{G} \frac{dX_G}{dz}, \quad \frac{dX_L}{dz} = 0 \quad (58)$$

$$z = Z, \quad X_L = X_{LZ} - \frac{E_L h_L}{L} \frac{dX_L}{dz}, \quad \frac{dX_G}{dz} = 0. \quad (59)$$

The transfer functions of the axially dispersed model AD–AD are formally identical with Eqs (44)–(49) for the PDE–AD model. However, the constants $d_1 - d_4$ for the AD–AD model are defined as follows:

$$d_i = 1 + (h_L E_L \lambda_i) / L, \quad i = 1, 2, 3, 4. \quad (60)$$

The PE–AD Model

The PE–AD model is a limiting case of the PDE–AD model if the coefficient of axial dispersion in the dynamic part of liquid E_D is taken equal zero. This causes that the dynamic liquid moves by plug flow, the total number of parameters is reduced to seven and the first term on the left hand side of Eq. (2) disappears. The PE–AD model was formulated and analyzed in an earlier paper⁷ which gives also the complete solution of the balance equations and the derivation of the transfer functions.

Plug Flow in Both Phases

The plug in both phases (PF–PF) as the model may be regarded also to be an asymptotic case of the PDE–AD model. The reduction of the PDE–AD model is effected by putting equal to zero the stagnant hold-up and the axial dispersion coefficients in the gas and the dynamic liquid phase. The model then contains only three parameters: the gas and liquid hold-ups h_G and h_L and the mass transfer coefficient $k_L a$.

The balance equations of the PF–PF model take the form

$$-G \frac{\partial C_G}{\partial z} - k_L a \left(\frac{C_G}{m} - C_L \right) = h_G \frac{\partial C_G}{\partial t} \quad (61)$$

$$L \frac{\partial C_L}{\partial z} + k_L a \left(\frac{C_G}{m} - C_L \right) = h_L \frac{\partial C_L}{\partial t} . \quad (62)$$

Upon performing the Laplace transform and eliminating the concentration X_L one obtains a single second order differential equation with constant coefficients

$$A_1 \frac{d^2 X_G}{dz^2} + A_2 \frac{dX_G}{dz} + A_3 X_G = 0 , \quad (63)$$

where

$$A_1 = \frac{GL}{k_L a} \quad (64)$$

$$A_2 = \frac{L}{m} - G + \frac{s(h_G L - h_L G)}{k_L a} . \quad (65)$$

$$A_3 = - \frac{h_G h_L s^2}{k_L a} - \frac{h_L s}{m} - h_G s . \quad (66)$$

The characteristic equation

$$A_1 \lambda^2 + A_2 \lambda + A_3 = 0 \quad (67)$$

this time is a quadratic equation with its roots being given by the following relation

$$\lambda_{1,2} = \frac{-A_2 \pm (A_2^2 - 4A_1 A_3)^{0.5}}{2A_1} . \quad (68)$$

Solution of Eqs (61) and (62) may then be written in the form

$$X_G = k_1 \exp(\lambda_1 z) + k_2 \exp(\lambda_2 z) \quad (69)$$

$$X_L = a_1 k_1 \exp(\lambda_1 z) + a_2 k_2 \exp(\lambda_2 z), \quad (70)$$

where

$$a_i = \frac{G\lambda_i}{k_{1a}} + \frac{h_{GS}}{k_{1a}} + \frac{1}{m}, \quad i = 1, 2. \quad (71)$$

The constants k_1 and k_2 , determined from the boundary conditions

$$z = 0, \quad X_G = X_{GO} \quad (72)$$

$$z = Z, \quad X_L = X_{LZ} \quad (73)$$

take the form

$$k_1 = \frac{a_2 \exp(\lambda_2 Z) X_{GO} - X_{LZ}}{a_2 \exp(\lambda_2 Z) - a_1 \exp(\lambda_1 Z)} \quad (74)$$

$$k_2 = \frac{X_{LZ} - a_1 \exp(\lambda_1 Z) X_{GO}}{a_2 \exp(\lambda_2 Z) - a_1 \exp(\lambda_1 Z)}. \quad (75)$$

The transfer functions of the PF-PF model are expressed by the following expressions

$$\frac{X_{GZ}}{X_{GO}} = \frac{(a_2 - a_1) \exp(\lambda_1 Z + \lambda_2 Z)}{a_2 \exp(\lambda_2 Z) - a_1 \exp(\lambda_1 Z)} \quad (76)$$

$$\frac{X_{LO}}{X_{GO}} = \frac{a_1 a_2 [\exp(\lambda_2 Z) - \exp(\lambda_1 Z)]}{a_2 \exp(\lambda_2 Z) - a_1 \exp(\lambda_1 Z)} \quad (77)$$

for $X_{LZ} = 0$ and

$$\frac{X_{LO}}{X_{LZ}} = \frac{a_2 - a_1}{a_2 \exp(\lambda_2 Z) - a_1 \exp(\lambda_1 Z)} \quad (78)$$

$$\frac{X_{GZ}}{X_{LZ}} = \frac{\exp(\lambda_2 Z) - \exp(\lambda_1 Z)}{a_2 \exp(\lambda_2 Z) - a_1 \exp(\lambda_1 Z)} \quad (79)$$

for $X_{GO} = 0$.

RESULTS

Numerical calculations of applicability of the transfer functions for the evaluation of the model parameters in the frequency domain. The transfer function may be transformed into the frequency domain simply by replacing the Laplace parameter by $i\omega$, where i designates the imaginary unit. The dependence of the transfer function on the frequency of the signal is the frequency characteristics of the transfer function which may be used to evaluate the parameters of the investigated model. The Bode plots, *i.e.* the dependences of the amplitude ratio and the phase lag on frequency, are graphical representations of the decay and shift of the input signal passing through the equipment. The amplitude ratio and the phase lag are related to the transfer function as follows

$$P = (R^2 + I^2)^{0.5} \quad (80)$$

$$\phi = \text{arctg}(I/R). \quad (81)$$

Our earlier paper⁷ gives a numerical test of the transfer functions from the standpoint of their applicability to parameter evaluation of the PE-AD model in the frequency domain. In this paper we have generalized the analysis of the PDE-AD model and its two asymptotic cases – the AD-AD and the PF-PF models.

The calculations, similarly as in the previous work, were carried out for the superficial velocities $G = 0.2$ m/s and $L = 0.004$ m/s, the equilibrium constant $m = 30$, the column height 2 m and the void fraction 0.4 (spheres 0.01 m in diameter). The principal set of parameters consisted of the following values: $E_G = E_D = 0.02$ m² s⁻¹, $h_D = h_S = 0.05$, $h_G = 0.3$, $k_{LD}a_D = k_{LS}a_S = 0.05$ s⁻¹, $q = 0.04$ s⁻¹.

Frequency characteristics of the transfer functions X_{GZ}/X_{GO} and X_{LO}/X_{GO} for this set of parameters are plotted in Fig. 2.

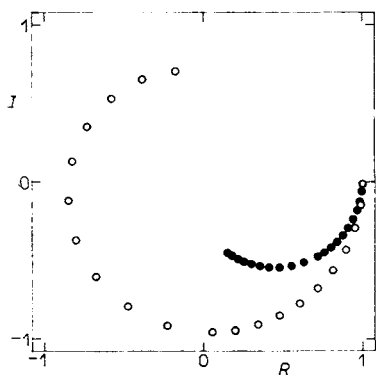


FIG. 2
Frequency dependence of the transfer functions X_{GZ}/X_{GO} and X_{LO}/X_{GO} for the PDE-AD model; \circ X_{GZ}/X_{GO} ; \bullet $m X_{LO}/X_{GO}$

The effect of individual parameters was investigated by constructing the Bode plots for several values of the parameter while keeping values of the remaining parameters constant. If, however, the investigated parameter was one of the hold-ups, the constraint was applied requesting that the sum of all hold-ups be equal to the void fraction of the bed. The courses in the Bode plots were investigated in the range of angular velocities between 0 and 1.5 rad s^{-1} .

From the obtained results it was concluded that for the above defined conditions the transfer functions X_{LO}/X_{LZ} and X_{GZ}/X_{LZ} are inapplicable. Although the former transfer function is significantly affected by all parameters of the model the outlet concentrations are extremely low owing to the practically total desorption of the dissolved gas in the column of the investigated length. Neither does the transfer function X_{GZ}/X_{LZ} supply a measurable response in case of a poorly soluble gas, regardless of the column length.

The remaining two transfer functions may be used for parameter evaluation. From these, however, the transfer function X_{GZ}/X_{GO} is significantly affected only by the parameters pertaining to the gas phase, while the X_{LO}/X_{GO} function is affected by the gas phase parameters only little while responding strongly to a change of the liquid phase related parameters and the mass transfer coefficients.

From these findings it follows that the generalization of the analysis to the PDE-AD model did not bring about any qualitative changes. At the value of $E_D = 0.02 \text{ m}^2 \text{ s}^{-1}$ the transfer function X_{GZ}/X_{GO} practically does not differ from that of PE-AD model and in case of the X_{LO}/X_{GO} transfer function the amplitude ratio is lower by about 5% and the phase lag is about 10 to 15% higher. The effect of the axial dispersion coefficient, E_D , on the amplitude ratio of the transfer function X_{LO}/X_{GO} is plotted in Fig. 3. With the exception of very high, and for a packed bed unrealistic values, its effect is relatively small. The same may be said about the phase lag of this transfer function, Fig. 4.

A very significant effect on the transfer function X_{LO}/X_{GO} exercise the mass transfer coefficients $k_{LD}a_D$ and $k_{LS}a_S$, see Figs 5-8. These figures show a greater effect of the $k_{LD}a_D$ coefficient both on the amplitude ratio and the phase lag. This observation, however, depends on the value of the exchange coefficient between the dynamic and the stagnant liquid phase, q . The effect of the coefficient q on the transfer function X_{LO}/X_{GO} is nevertheless small, equally as in the case of the PE-AD model. The evaluation of this parameter from the transfer function X_{LO}/X_{GO} may therefore encounter difficulties.

A comparison of the Bode plots of the basic model with its three limiting cases for the basic set of data (for the limiting cases of the PDE-AD model the values of the vanishing parameters are set equal to zero) is furnished in Figs 9-11. For the amplitude ratio of the transfer function X_{GZ}/X_{GO} , Fig. 9, the curves of the PDE-AD and the PE-AD models are identical and the AD-AD model differs only very little. The PF-PF model differs from the above three models fundamentally, for it deviates

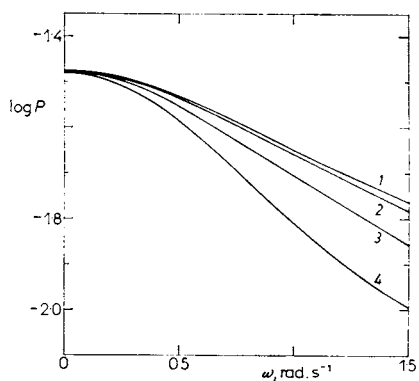


FIG. 3

The effect of the axial dispersion coefficient E_D on the frequency dependence of the amplitude ratio of the X_{LO}/X_{GO} function for the PDE-AD model. 1 $E_D = 0.001 \text{ m}^2 \text{ s}^{-1}$, 2 $E_D = 0.01 \text{ m}^2 \text{ s}^{-1}$, 3 $E_D = 0.1 \text{ m}^2 \text{ s}^{-1}$, 4 $E_D = 0.5 \text{ m}^2 \text{ s}^{-1}$

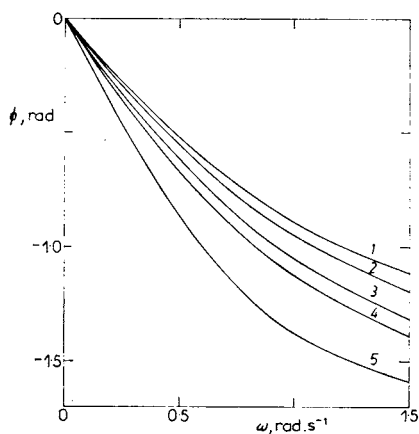


FIG. 4

The effect of the axial dispersion coefficient E_D on the frequency dependence of the phase lag of the X_{LO}/X_{GO} function for the PDE-AD model. 1 $E_D = 0.001 \text{ m}^2 \text{ s}^{-1}$, 2 $E_D = 0.01 \text{ m}^2 \text{ s}^{-1}$, 3 $E_D = 0.05 \text{ m}^2 \text{ s}^{-1}$, 4 $E_D = 0.1 \text{ m}^2 \text{ s}^{-1}$, 5 $E_D = 0.5 \text{ m}^2 \text{ s}^{-1}$

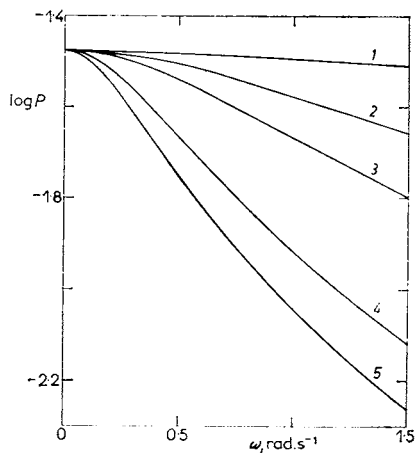


FIG. 5

The effect of the mass transfer coefficient $k_{LD}a_D$ on the frequency dependence of the amplitude ratio of the X_{LO}/X_{GO} function for the PDE-AD model. 1 $k_{LD}a_D = 0.5 \text{ s}^{-1}$, 2 $k_{LD}a_D = 0.1 \text{ s}^{-1}$, 3 $k_{LD}a_D = 0.05 \text{ s}^{-1}$, 4 $k_{LD}a_D = 0.01 \text{ s}^{-1}$, 5 $k_{LD}a_D = 0.001 \text{ s}^{-1}$

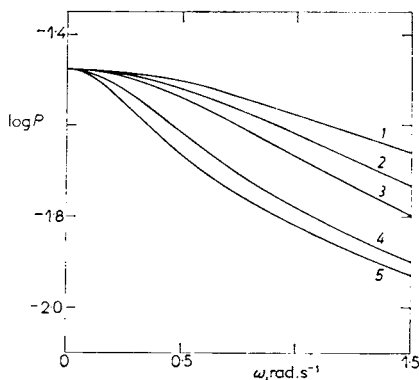


FIG. 6

The effect of the mass transfer coefficient $k_{LS}a_S$ on the frequency dependence of the X_{LO}/X_{GO} function for the PDE-AD model. 1 $k_{LS}a_S = 0.5 \text{ s}^{-1}$, 2 $k_{LS}a_S = 0.1 \text{ s}^{-1}$, 3 $k_{LS}a_S = 0.05 \text{ s}^{-1}$, 4 $k_{LS}a_S = 0.01 \text{ s}^{-1}$, 5 $k_{LS}a_S = 0.001 \text{ s}^{-1}$

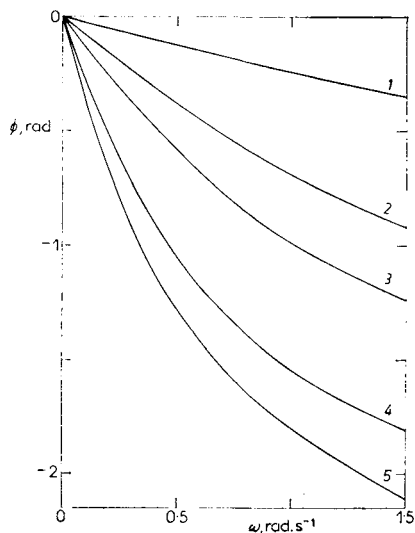


FIG. 7

The effect of the mass transfer coefficient $k_{LD}a_D$ on the frequency dependence of the phase lag of the X_{LO}/X_{GO} function for the PDE-AD model. 1 $k_{LD}a_D = 0.5 \text{ s}^{-1}$, 2 $k_{LD}a_D = 0.1 \text{ s}^{-1}$, 3 $k_{LD}a_D = 0.05 \text{ s}^{-1}$, 4 $k_{LD}a_D = 0.01 \text{ s}^{-1}$, 5 $k_{LD}a_D = 0.001 \text{ s}^{-1}$

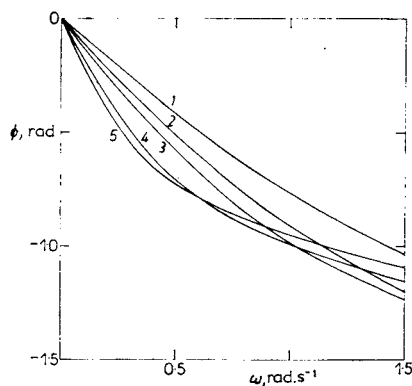


FIG. 8

The effect of the mass transfer coefficient $k_{LS}a_S$ on the frequency dependence of the phase lag of the X_{LO}/X_{GO} function for the PDE-AD model. 1 $k_{LS}a_S = 0.5 \text{ s}^{-1}$, 2 $k_{LS}a_S = 0.1 \text{ s}^{-1}$, 3 $k_{LS}a_S = 0.05 \text{ s}^{-1}$, 4 $k_{LS}a_S = 0.01 \text{ s}^{-1}$, 5 $k_{LS}a_S = 0.001 \text{ s}^{-1}$

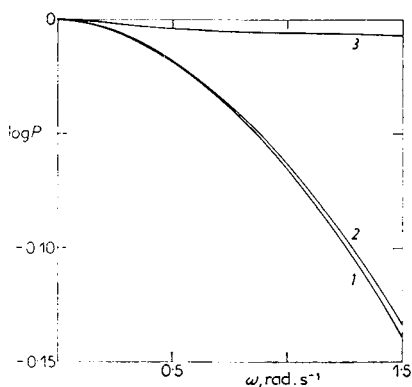


FIG. 9

The dependence of the amplitude ratio of the function X_{GZ}/X_{GO} on the angular velocity (frequency) for the four tested models. 1 PDE-AD, PE-AD; 2 AD-AD; 3 PF-PF

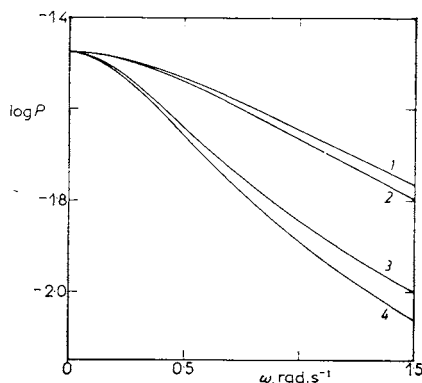


FIG. 10

The dependence of the amplitude ratio of the function X_{LO}/X_{GO} on the angular velocity (frequency) for the four tested models. 1 PE-AD; 2 PDE-AD; 3 PF-PF; 4 AD-AD

only little from unity with increasing frequency. The phase lag of the transfer function X_{GZ}/X_{GO} of the PF–PF model grows linearly with angular velocity. The remaining three models deviate from the plug flow model only little toward lower values of the phase lag.

The amplitude ratio of the transfer function X_{LO}/X_{GO} for all four tested models equals at zero frequency 0.033, which is an equilibrium value with the inlet concentration. With increasing angular velocity the amplitude ratio decreases, this decrease being the slowest for the PE–AD model and in the series PDE–AD, PF–PF and AD–AD the decrease increases (see Fig. 10). The difference between the PE–AD and the PDE–AD models is relatively small, similarly as between the PF–PF and the AD–AD models. Both pairs of models, however, differ one from another far more significantly. The difference between the two pairs of models is apparently due to the different magnitudes of the dynamic part of the hold-up. For the PE–AD and PDE–AD models the hold-up amounts to $h_D = 0.05$, while for the AD–AD and PF–PF models all liquid hold-up is mobile, $h_L = 0.1$. The difference between the PE–AD and the PDE–AD models is caused by the axial dispersion coefficient E_D and between the AD–AD and the PF–PF models by both axial dispersion coefficients, E_G and E_L . The phase lag of the transfer function X_{LO}/X_{GO} in the investigated frequency range increases for individual models in the series PE–AD, PDE–AD, PF–PF and AD–AD, *i.e.* in the opposite sequence than the amplitude ratio.

CONCLUSION

The performed analysis has shown that the transfer functions applicable for the evaluation of parameters of absorption of a poorly soluble gas in a counter-current packed bed column based on the PDE–AD model are X_{GZ}/X_{GO} and X_{LO}/X_{GO} . The transfer function X_{GZ}/X_{GO} may be used to evaluate parameters characterizing

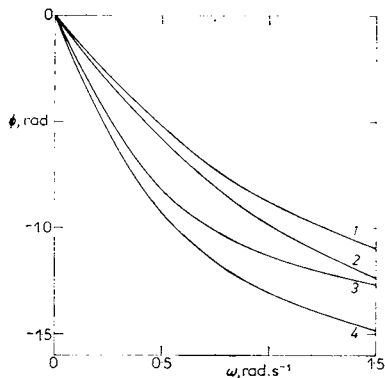


FIG. 11

The dependence of the phase lag of the function X_{LO}/X_{GO} on the angular velocity (frequency) for the four tested models. 1 PE–AD; 2 PDE–AD; 3 PF–PF; 4 AD–AD

the dynamics of the gas phase, while the parameters characterizing the dynamics of the liquid phase and interfacial transfer may be evaluated from the X_{Lo}/X_{Go} transfer function. Both these transfer functions, however, are relatively little sensitive to the coefficient of exchange between the dynamic and the stagnant liquid phase, q . This conclusion is practically identical with the conclusions made earlier⁷ on the basis of the PE—AD model.

LIST OF SYMBOLS

| | |
|-----------|--|
| a | specific interfacial area m^{-1} |
| C | concentration defined as deviation from steady state value $kmol m^{-3}$ |
| E | axial dispersion coefficient $m^2 s^{-1}$ |
| G | superficial gas velocity $m s^{-1}$ |
| h | hold-up |
| I | imaginary part of the transfer function |
| k_L | mass transfer coefficient $m s^{-1}$ |
| L | superficial liquid velocity $m s^{-1}$ |
| m | equilibrium constant |
| P | amplitude ratio |
| q | coefficient of exchange between dynamic and stagnant liquid phase s^{-1} |
| R | real part of the transfer function |
| s | Laplace parameter s^{-1} |
| t | time s |
| X | Laplace transform of concentration C |
| Z | packed height m |
| z | axial coordinate m |
| λ | root of characteristic equation m^{-1} |
| ϕ | phase lag rad |
| ω | angular velocity $rad s^{-1}$ |

SUBSCRIPTS

| | |
|---|-----------------------|
| D | dynamic liquid phase |
| G | gas phase |
| L | liquid phase |
| S | stagnant liquid phase |
| Z | top of the column |
| O | bottom of the column |

REFERENCES

1. Danckwerts P. V.: Chem. Eng. Sci. 2, 1 (1953).
2. Michell R. W., Furzer I. A.: Chem. Eng. J. 4, 53 (1972).
3. Hoogendoorn C. J., Lips J.: Can. J. Chem. Eng. 43, 125 (1965).
4. van Swaaij W. P. M., Charpentier J. C., Villermaux J.: Chem. Eng. Sci. 24, 1083 (1969).
5. Hatton T. A., Woodburn E. T.: AIChE J. 24, 187 (1978).
6. Linek V., Beneš P., Sinkule J., Krivský Z.: Ind. Eng. Chem. Fundam. 17, 298 (1978).
7. Moravec P., Staněk V.: This Journal 47, 2639 (1982).

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