NUMERICAL EXPERIMENT ON MASS TRANSFER IN BINARY MIXTURES, USING THE MATHEMATICAL MODEL OF CONVECTIVE DIFFUSION FOR THE TURBULENT FLOW OF GAS IN A TUBE

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A numerical experiment is performed in relation to mass transfer in binary mixtures. The results of the calculation agree satisfactorily with experimental data.

The most rigorous presentation of the mass-transfer problem demands the use of the following well-known equations of mathematical physics for every phase [1, 2]: the equations of motion, the continuity equations for the mixture as a whole, (n − 1) equations of material balance covering each component, (n − 1) diffusion equations, and the energy equations. All the equations of the system are interlinked, reflecting the objective relationship between hydrodynamics and heat and mass transfer.

In general this system cannot be solved. Furthermore, under the complicated conditions of processes taking place in chemical apparatus, the initial and boundary conditions cannot be accurately formulated. Hypotheses and models simplifying the problem are therefore extensively used. In order to solve the fundamental problems it is preferable to use fairly realistic models based on the equations of hydrodynamics and convective diffusion, and to exploit the analogy between momentum transfer and mass (or heat) transfer involving turbulent pulsations [3-8]. Taking all this into consideration, we shall now formulate a model problem for convective diffusion in the turbulent flow of a many-component gas through a tube, and shall discuss the results of a numerical experiment relating to binary mixtures on the basis of this model.

We decided to simulate a film column. In addition to the hypothesis as to the analogy between momentum and mass transfer, we had to make a number of other assumptions in order to simplify the problem to a form capable of numerical solution in computers of the M-220 type.

1. The principal assumption is that of neglecting the influence of mass transfer on the hydrodynamics; this greatly simplifies the problem, so that we may first solve the equations of motion for the velocity profile separately (or use experimental results), and then solve the mass-transfer problem. This assumption does not lead to any serious errors [4-6] if we exclude the case of very high rates of mass transfer [9-11].

2. We assume that the turbulent viscosity coefficients calculated by means of the molar-average and mass-average velocities are equal. This is strictly only valid in the absence of mass transfer, but is practically valid when the rate of mass transfer is much smaller than the rate of the main flow. Actually, the mass-average velocity is

$$\mathbf{w} = \frac{1}{\rho} \sum_i m_i n_i \mathbf{v}_i, \text{ and the molar-average velocity } \mathbf{v} = \frac{1}{n} \sum_i n_i \mathbf{v}_i, \text{ where } \mathbf{v}_i = \frac{1}{n_i} \int \mathbf{c} f_i(\mathbf{c}, \mathbf{r}) d\mathbf{c} \cdot$$

is the average velocity of the i-th component in the laboratory system of coordinates. On calculating the difference $\mathbf{w} - \mathbf{v} = \dfrac{1}{\rho} \sum_i m_i N_i$, by means of these equations we see that $\mathbf{v} = \mathbf{w}$ when $N_i = 0$. The latter equation is also practically valid for equimolar diffusion $\left(\sum_i N_i = 0 \right)$.

<u>3.</u> The turbulent Schmidt criterion is taken as constant over the tube cross section.

<u>4.</u> We assume that the axial velocity profile in the gas phase of the film column will be the same as in a tube with solid walls.

<u>5.</u> We only consider processes in which the resistance to mass transfer is concentrated in the gas phase.

<u>6.</u> We neglect diffusion in the axial and azimuthal directions.

For the steady-state process expressed in dimensionless coordinates (x = 1 − r/R, Z = z/L) we then obtain the equation of convective diffusion

$$\frac{R}{L} v_z \frac{\partial y_i}{\partial Z} = v_x \frac{\partial y_i}{\partial x} + \frac{1}{1-x} \frac{\partial}{\partial x} \left[(1-x) \left(N_i + \frac{E}{Sc_T} \frac{\partial y_i}{\partial x} \right) \right], \tag{1}$$

$$\frac{\partial y_i}{\partial x} = R \sum_{j \neq i} \frac{N_i y_j - N_j y_i}{D_{ij}}, \qquad 0 = \sum_j N_j \tag{2}$$

and the boundary conditions

$$y_i (0, \ x) = y_{i0}, \tag{3}$$

$$y_i (Z, \ 0) = y_{iw}, \tag{4}$$

$$\left. \frac{\partial y_i}{\partial x} \right|_{x=1} = 0 \quad \text{for all} \quad Z. \tag{5}$$

Here y_{iw} is found from the condition of equilibrium between the vapor and the liquid in a specific cross section of the tube. Condition (3) reflects the constancy of the concentration of incoming gas over the tube cross section, and (5) reflects the absence of a concentration gradient along the tube axis.

The velocity profile v_Z, the radial velocity, and the turbulent viscosity coefficient may be found by means of the three-layer Karman scheme. However, it is more convenient and no less accurate to use the two-layer approximation of Wasan and Wilke [5, 6]; we shall use this in this paper.

The problem was set up for solution in the M-220 computer, a three-layer implicit difference scheme being used as a basis for the program [12, 13].

The numerical experiment for binary mixtures was carried out with the aim of subsequently using the results in order to provide an engineer's description of many-component mass transfer. We shall discuss the results obtained from this experiment, since these are of independent interest.

Preliminary calculations showed that the Nusselt number Nu[d] only depended on the Re and Sc numbers and the ratio of the tube length to its diameter. This also follows from a consideration of the invariance of the original equations for transformations of this kind.
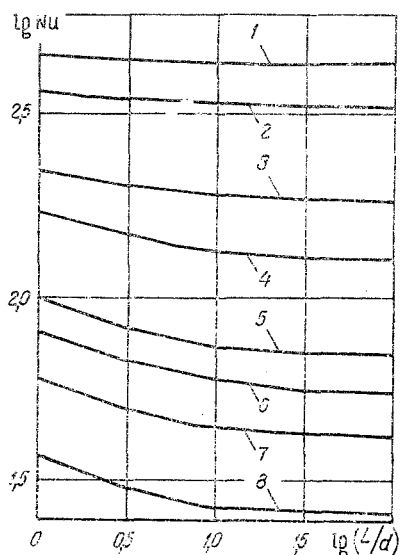
Fig. 1. Dependence of the Nusselt diffusion number $Nu^{(d)}$ on L/d: 1) Re = 10^5 Sc = 8; 2) Re = 10^5, Sc = 4; 3) Re = 10^5, Sc = 1; 4) Re = 10^5, Sc = 0.5; 5) Re = $4 \cdot 10^4$, Sc = 0.5; 6) Re = $3 \cdot 10^4$, Sc = 0.5; 7) Re = $2 \cdot 10^4$, Sc = 0.5; 8) Re = 10^4, Sc = 0.5.

The defining criteria (numbers) were taken in the following ranges:

$$Re = 10^4 - 10^5; \quad Sc = 0.5 - 8; \quad \frac{L}{d} = 0 - 100.$$

It was found that the Nusselt diffusion number fell with increasing L/d; this relationship was not expressible in power form (Fig. 1). The value of L/d has the greatest influence on $Nu^{(d)}$ for small values of the Sc number (and constant Re values), since in this case the thickness of the diffusion layer in which D >> E/Sc_T increases, and the creation of a developed concentration profile takes place over a greater length of tube. For L/d > 30 the $Nu^{(d)}$ number may be regarded as practically independent of L/d for all values of the Schmidt number under consideration. The use of tubes with L/d > 30 enables us to study the dependence of $Nu^{(d)}$ on Re and Sc in pure form.

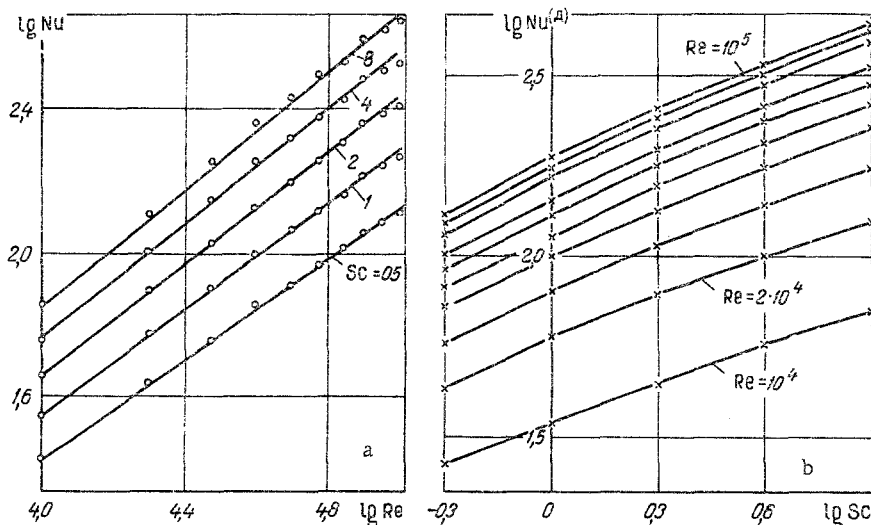The Nusselt number $Nu^{(d)}$ is shown as a function of the Re and Sc numbers in Fig. 2. An important point here is that, if we analyze the results in the most widely accepted form $Nu^d = ARe^m Sc^n$, we find that the power indices m and n themselves depend on the Re and Sc numbers. The greater the value of the Schmidt number, the more does the Reynolds number influence the intensity of mass transfer. This is quite natural, since on reducing the diffusion coefficient or increasing the viscosity of the mixture the coefficient of turbulent diffusion determined by the mode of flow plays a greater and greater part (Fig. 2a). On increasing the Re number, however, the power index of the Sc number (n) also increases (Fig. 2b); this reflects the lessening influence of the diffusion coefficient on the mass-transfer coefficient on increasing the degree of turbulence of the flow. These characteristics of the process are reflected in the well-known equation obtained by using the three-layer Karman scheme:

Fig. 2. Dependence of $Nu^{(d)}$ on Re (a) and Sc (b).

$$Nu^{(d)} = \frac{\frac{f}{2} Re\, Sc}{1 + 5\sqrt{\frac{f}{2}}\left[Sc - 1 + \ln\left(\frac{1 + 5Sc}{6}\right)\right]}, \tag{6}$$

which for Sc = 1 reduces to the Reynolds equation

$$Nu^{(d)} = \frac{f}{2} Re. \tag{7}$$

Subsequent analysis will thereror be based on Eq. (6). First of all, in accordance with Eq. (6), it is convenient to examine the results obtained for Sc = 1.

Analysis based on the method of least squares gave the power index of the Re number in the Sc = 1 case as 0.74. This is close to the value of 0.75 obtained by using the Blasius equation $f = 0.079 Re^{-0.25}$. Substituting the latter in (6), we have

$$Nu^{(d)} = 0.0395\, Re^{0.75}, \quad Sc = 1.$$

However, the relationship illustrated in Fig. 2a corresponds to a slightly different equation

$$Nu = 0.0345\, Re^{0.75} \quad \text{for } Sc = 1. \tag{8}$$

The difference between the values obtained for the critical number from Eq. (8) and from the numberical experiment is less than 1%, except for the values corresponding to Re = 10^5, in which case the error is 4.3%.

Remembering that the thickness of the diffusion layer (determined from the condition $X^+ = 5$ in the Karman scheme) actually depends on the form of motion, subsequent analysis of the results is based on the equation

$$Nu^{(d)} = \frac{0.0345\, Re^{0.75} Sc}{1 + a\, Re^{b}\left[Sc - 1 + 2.3\lg\left(\frac{1 + 5Sc}{6}\right)\right]}. \tag{9}$$

The unknown quantities a and b were determined from the experimental data, not by any of the standard methods, but from only two points: Sc = 8, Re = 10^4 and Sc = 0.5, Re = $8 \cdot 10^4$. The result was

$$Nu^{(d)} = \frac{0.0345\, Re^{0.75} Sc}{1 + 0.75\, Re^{-0.09}\left[Sc - 1 + 2.3\lg\left(\frac{1 + 5Sc}{6}\right)\right]}. \tag{10}$$

The fact that the resultant equation (10) correctly reflects the essentials of the process is confirmed by its excellent agreement with the numerical experiment. This may be seen from Fig. 2a (continuous lines). For most of the points the deviation of the experimental results from Eq. (10) is less than 1%.

A comparison between the results obtained and the generalized Notter relationship (8) for heat transfer shows that the analogy between mass and heat transfer is by no means complete. For Pr = 1 in particular, the Notter equation takes the form

$$Nu = 5 + 0.016\, Re^{0.83},$$

while for Sc = 1 Eq. (10) gives

$$Nu^{(d)} = 0.0345\, Re^{0.75}.$$

A comparison between Nu and $Nu^{(d)}$ for Sc = 1 gives the following

Re	10^4	$3 \cdot 10^4$	10^5
$Nu^{(d)}$	35	79	195
Nu	38	88	231

Let us compare the final result (10) with the equation derived experimentally by É. Sh. Telyakov [14] in connection with the rectification of a mixture of n-heptane and toluene, for which the resistance to mass transfer was concentrated in the vapor phase:

$$Nu^{(d)} = 1.642 \cdot 10^{-2} Re^{0.84} Sc^{0.5}. \tag{11}$$

Since the power index of the Sc number was not determined experimentally in [14], but taken as 0.5 (the value of the Sc number varying from 0.5 to 0.7), the comparison presented below refers to Sc = 0.6. The Re number in the experiments varied from $6 \cdot 10^3$ to $1.5 \cdot 10^4$:

Re	$6 \cdot 10^3$	$9 \cdot 10^3$	$1,2 \cdot 10^4$	$1,5 \cdot 10^4$
$Nu^{(d)}$ from (11)	18,7	26,7	34,2	41
$Nu^{(d)}$ from (10)	19,3	26,2	32,2	37,8
Error %	+3,2	—1,9	—6,2	—8,5

The agreement between the numerical and physical experiments is better than might have been expected in view of the assumptions made in the present investigation.

NOTATION

N is the mass flow; R is the tube radius; L is the tube length; D is the diffusion coefficient; w is the mass-average velocity; v is the molar-average velocity; m is the molecular mass; n is the number of molecules; ρ is the density; c is the velocity; f_i is the distribution function; r and z are the coordinates; y is the mole fraction; E is the turbulent viscosity coefficient; d is the tube diameter; f is the Fanning coefficient of friction; Nu is the Nusselt number; Re is the Reynolds number; Sc is the Schmidt number. Indices: i and j — components of the mixture; z and x — axial and radial directions; T — turbulent; (d) — diffusion.

LITERATURE CITED

1. A. V. Lykov, Heat and Mass Transfer (Handbook) [in Russian], Énergiya, Moscow (1972).
2. A. A. Pomerantsev, Course of Lectures on the Theory of Heat and Mass Transfer [in Russian], Vysshaya Shkola, Moscow (1965).
3. G. L. Behren, W. O. Jones, and D. T. Wasan, AIChE Journal, 18, 25 (1972).
4. C. V. Sechadri and H. L. Toor, Indian Chem. Eng., 95 (1963).
5. D. T. Wasan, C. R. Wilke, and C. L. Tien, AIChE Journal, 9, 565 (1963).
6. D. T. Wasan and C. R. Wilke, Int. J. Heat Mass Transf., 7, 87 (1964).
7. E. M. Sparrow and H. S. Yu, AIChE Journal, 16, 688 (1970).
8. R. H. Notter and C. A. Sleicher, Chem. Eng. Sci., 27, 2073 (1972).
9. L. D. Berman, Izv. VTP, No. 8, 11 (1947).
10. L. D. Berman, Teploénergetika, 5, 25 (1954); 6, 43 (1957).
11. L. D. Berman, Zh. Prikl. Khim., 32, 807 (1959).
12. A. A. Samarskii, Introduction to the Theory of Difference Schemes [in Russian], Nauka, Moscow (1971).
13. W. Wasow and J. Forsythe, Finite Difference Methods for Solving Partial Differential Equations, Wiley (1960).
14. É. Sh. Telyakov, Candidate's Dissertation, Kazan' (1969).