Int. J. Heat Mass Transfer. Vol. 13, pp. 871–878. Pergamon Press 1970. Printed in Great Britain

UNSTEADY-STATE LIQUID DIFFUSION IN LAMINAR FLOW IN A CIRCULAR TUBE

M. G. WILLIAMS and J. K. FERRELL
North Carolina State University, Raleigh, N.C., U.S.A.

(*Received* 24 *October* 1969 *and in revised form* 27 *October* 1969)

Abstract—A technique was developed for solving the mathematical model describing unsteady-state diffusion in a Newtonian liquid in steady-state laminar flow in a circular tube. The process of interest in this work was one in which the Péclét numbers were very high and axial diffusion effects could, therefore, be neglected. The mathematical model describing this process was a partial differential equation with three independent variables and its associated boundary conditions. The method of solution used the Laplace transform, followed by either separation of variables or finite difference calculations, depending on the magnitude of the parameters involved.

This method has the advantage that it eliminates the complex finite difference techniques and their associated stability and convergence problems which must otherwise be employed for exact numerical solutions of boundary value problems in three independent variables. The solution obtained in this work showed excellent agreement with experimental data collected in the present work and in previous research. This technique can be applied to other mathematical models of similar form describing transport problems in unsteady-state laminar flow and should make exact numerical solutions to this class of problems more easily obtainable.

NOMENCLATURE

C, dye concentration;
C_0, initial dye concentration;
D, diffusion coefficient;
G, dimensionless concentration;
g, Laplace transform of G;
N_{Pe}, Péclét number;
R, tube radius;
r, radial distance from tube axis;
S, unit step function;
s, Laplace transform parameter;
t, elapsed time;
V_0, liquid velocity at tube axis;
z, axial distance from the interface.

Greek letters
α, scale factor for Laplace transforms;
Δ, difference operator;
ζ, dimensionless axial distance from the interface;
Θ, dimensionless time;
ξ, dimensionless radial distance from tube axis.

THE PROBLEM of diffusion in laminar flow in tubes has received a considerable amount of attention by many people in the last fifteen years. The problem was first discussed by G. I. Taylor [22–24] in terms of the dispersion of a soluble material in a flowing solvent. Since Taylor's original work, the problem has been examined in different ways by other investigators [2–4, 7, 16]; and the understanding of the physical situation has been greatly increased. Of particular interest is a series of papers by Gill and co-workers [1, 12, 13, 17, 18] who have obtained solutions to the problem for a wide range of Péclét numbers and values of dimensionless time.

A practical example of diffusion and convection in laminar flow in a circular tube was discovered by Ferrell *et al.* [9–11] in the development of a technique for measuring velocity profiles of liquids in laminar flow in circular tubes. This technique involved displacing from rest a liquid containing a small amount of dissolved dye by the identical colorless liquid, continuously monitoring the displacement of the dyed liquid at a point downstream from the initial interface, and calculating from this data the velocity profiles. When the technique was applied to Newtonian liquids of low viscosity in laminar flow, it was discovered that the measured profiles showed large deviations from the theoretical laminar profile in the wall-adjacent portion of the tube. This deviation of the measured profiles was thought to be a result of radial diffusion of the dye toward the center of the tube causing it to be displaced more rapidly.

The objective of the work reported here was to verify radial diffusion as the source of error in the dye displacement technique. This required a knowledge of how the mean dye concentration changed with time after the beginning of flow in the tube. It was, therefore, necessary to derive, solve and verify a mathematical model for the particular case of laminar diffusion and convection encountered in the dye displacement technique. It was also an objective of this work to develop a technique for solving the mathematical model which eliminates the complex finite-difference methods which would otherwise be necessary for solving this three independent variable model. Such a method was employed by Gill *et al.* [1] in obtaining solutions to a similar model and was reported to involve trial and error selections of the finite difference increments in order to determine the convergence and stability of the solutions.

MATHEMATICAL MODEL

The mathematical model was derived by simplifying the general diffusion equation for the case of laminar flow in a circular tube and adding the proper boundary conditions. It was assumed in this derivation that the system was of constant mass density, that the diffusion coefficient was a function of temperature only, that diffusion in the axial direction was negligible compared with convection, that the system was at constant temperature and pressure, and that the fluid velocity followed the theoretical parabolic form for laminar flow. The mathematical model in non-dimensional form is

$$\frac{\partial^2 G}{\partial \xi^2} + \frac{1}{\xi}\frac{\partial G}{\partial \xi} = 4(1-\xi^2)\frac{\partial G}{\partial \zeta} + 4\frac{\partial G}{\partial \Theta} \qquad (1)$$

$$G(\xi, \zeta, 0) = 1 \quad \text{for} \quad \zeta \geqslant 0 \qquad (2)$$

$$G(\xi, 0, \Theta) = 0 \quad \text{for} \quad \Theta > 0 \qquad (3)$$

$$G_\xi(0, \zeta, \Theta) = 0 \qquad (4)$$

$$G_\xi(1, \zeta, \Theta) = 0, \qquad (5)$$

where the subscript ξ indicates the partial derivative with respect to ξ, and

$$\xi = r/R \qquad (6)$$

$$\zeta = 4zD/V_0R^2 \qquad (7)$$

$$\Theta = 4Dt/R^2 \qquad (8)$$

$$G = G(\xi, \zeta, \Theta) = C(\xi, \zeta, \theta)/C_0. \qquad (9)$$

In these equations ξ is the dimensionless tube radius, ζ is the dimensionless axial distance from the interface, Θ is the dimensionless time and G is the dimensionless concentration. The factors of four were included to facilitate the solution.

The boundary conditions state in the order of their listing that:

a. The tube is initially filled in the positive z direction with a dye solution of uniform concentration, C_0.
b. The dye at the interface, $z = 0$, is completely displaced at the beginning of the flow.
c. The concentration profile is symmetric about the tube axis.
d. There is no transport of material through the tube wall.

SOLUTION OF SIMPLIFIED MODEL

The normalized model was first solved for the special case of dye displacement by convection only. This meant that the diffusion terms on the left-hand side of equation (1) were set equal to zero and the boundary conditions given by equations (4) and (5) were discarded. A solution in closed form was possible in this case which is shown in equation (10)

$$G(\xi, \zeta, \Theta) = S[\zeta - (1 - \xi^2)\Theta] \quad (10)$$

where $S[\zeta - (1 - \xi^2)\Theta]$ is the so-called unit step function. The value of this function is unity when the expression in the brackets is positive, and it is zero when this expression is negative.

It was found to be convenient to express this solution in terms of the concentration measured by the spectrophotometer. This is a mean concentration based on the diameter of the tube and defined by

$$G_{m'}(\zeta, \Theta) = \int_0^1 G(\xi, \zeta, \Theta)\, d\xi. \quad (11)$$

The subscript m' is used to differentiate this "linear" mean concentration from mean concentrations defined in the usual way. When equation (10) was substituted into equation (11) and the integration was performed the result was

$$G_{m'}(\zeta, \Theta) = 1 \quad \text{for} \quad \Theta \leqslant \zeta \quad (12)$$

$$G_{m'}(\zeta, \Theta) = 1 - \sqrt{(1 - \zeta/\Theta)} \quad \text{for} \quad \Theta > \zeta. \quad (13)$$

This solution was called the "convection" solution to denote the absence of diffusion. Since ζ/Θ is equal to zV_0/t, the experimental data can always be compared with the convection solution without prior knowledge of the dye diffusion coefficient.

SOLUTION OF DIFFUSION–CONVECTION MODEL

The first step in the solution of the complete diffusion–convection model was to take the Laplace transform of the model with respect to the dimensionless axial distance ζ. This variable was selected for transformation because of the zero initial condition given by equation (3).

The next step was to solve the transformed model for desired values of s, the Laplace transform parameter. At this point it should be noted that for values of s less than 1000, the transformed model could be solved by separation of variables. At higher values of s the large magnitudes of the numerical quantities in the separation of variables calculation made it necessary to resort to finite difference techniques for the solution. Complete details of the method of solution may be found in [25].

A digital computer was used to perform the numerical calculations required by the separation of variables and finite difference methods. The results of these calculations were numerical values of $g_{m'}(s, \Theta)$ at discrete values of Θ for the values of s required.

The final step in the solution of the model was to take the inverse transform by means of Salzer's method [19–21]. This is a numerical method for approximating inverse transformations which yields values of the inverse transformations at desired values of ζ. In the present case it was necessary to scale the transform by employing the well-known theorem for Laplace transforms [6] which states that if $g(s, \Theta)$ is the Laplace transform of $G(\zeta, \Theta)$, then

$$L^{-1}\{\alpha g(\alpha s, \Theta)\} = G\left(\frac{\zeta}{\alpha}, \Theta\right). \quad (14)$$

Here α is a scale factor which must be properly selected so that the results of the inversion are valid. Complete details of the numerical inversion may also be found in [25].

The small numerical values of ζ for which inverse transforms were required in this work made the use of large scale factors, α, necessary. Inverse transforms were calculated with α equal to 100 and 1000. The latter value produced results for values of ζ ranging from $1{\cdot}0 \times 10^{-4}$ to $1{\cdot}50 \times 10^{-3}$, while the former produced results for ζ's ranging from $1{\cdot}50 \times 10^{-3}$ to $2{\cdot}0 \times 10^{-2}$. This meant that the transformed model had to be solved at $s = \alpha k$ equal to 100,200, . . . 1000 for α equal to 100 and at s equal to 1000, 2000,. . . 10000 for α equal to 1000.

The results of the inverse transformation were numerical values of $G_{m'}(\zeta, \Theta)$ versus Θ for discrete values of ζ. These data were then plotted and smooth curves drawn through them to produce continuous solutions.

RESULTS

The comparison of the diffusion–convection solution with the convection solution at $\zeta = 1{\cdot}0 \times 10^{-3}$ is shown in Fig. 2. In this figure the dimensionless mean concentration, $G_{m'}(\zeta, \Theta)$, is plotted as the ordinate and the dimensionless time, Θ, as the abscissa on logarithmic co-ordinates. The concentration measurements in a run always start when the value of Θ becomes equal to the value of ζ. This value of Θ is the time at which dye displacement begins at the monitoring location. At the start of the run the convection solution shows a rapid decrease in concentration from the initial dimensionless concentration of 1·0 as shown in Fig. 1. This idealized solution then gradually becomes linear with time with a slope of minus one. The diffusion-convection solution follows the convection solution for the early part of the run. It then rises slightly above the convection solution, and finally, shows a large downward deviation from it. Both of these deviations of the diffusion–convection solution from the convection solution result from the diffusion of the dye.

Figure 2 shows a comparison of the diffusion–convection solution with some experimental data on potassium permanganate in water. The experimental value of ζ was calculated using a value of the diffusion coefficient of potassium permanganate in water which was found in the literature [22]. It is obvious from this figure and Fig. 1 that a mathematical model including only convection does not represent the physical process. The model must take into account the diffusion effect as well.

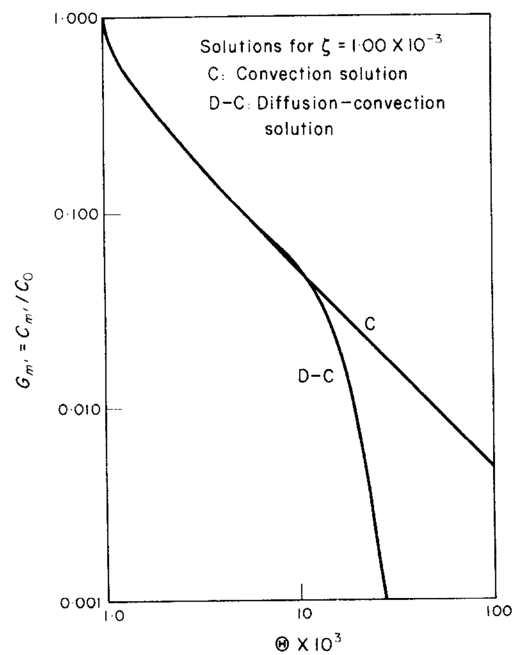

FIG. 1. Comparison of the diffusion–convection solution with the convection solution for $\zeta = 1{\cdot}00 \times 10^{-3}$.

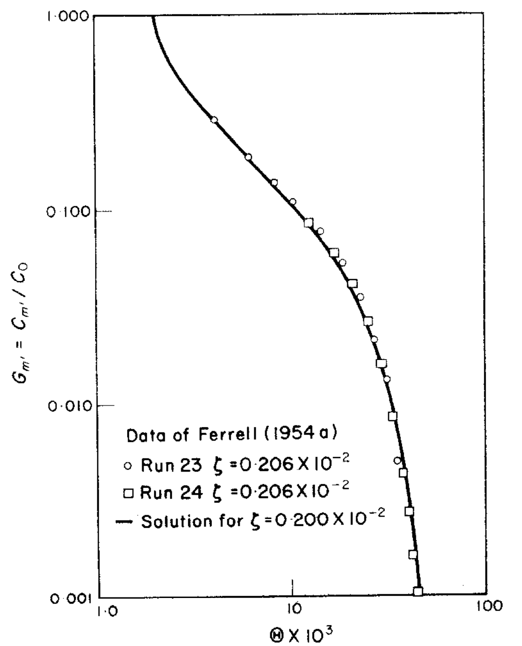

FIG. 2. Comparison of theoretical solution for $\zeta = 0{\cdot}200 \times 10^{-2}$ with experimental data for potassium permanganate.

At $\zeta = 1{\cdot}50 \times 10^{-3}$ it was possible to compare the solutions obtained by the two different methods using the two different values of α. This comparison is shown in Fig. 3. Methods I and II refer to solutions of the transformed model by separation of variables and finite difference techniques, respectively. Since the solutions shown in this figure result from numerical methods and an approximate inversion scheme, it was felt that the agreement of these two solutions was excellent.

On the basis of results of the kind shown in Figs. 2 and 3, it was concluded that the theoretical solutions were correct and that the mathematical model was a valid description of the physical process. The high values of the centerline velocity, V_0, which were used in the experimental runs along with the relatively large tube diameter (0·50 in.) produced very high values of the Péclét number, $N_{Pe} = RV_0/D$. It was, therefore, reasonable from the results of previous work [1] to eliminate the axial diffusion term from the general diffusion equation in the derivation of the mathematical model. The assumption that the diffusion coefficient, D, was a function of temperature only and not dependent on concentration also was apparently valid at the low dye concentrations employed in view of the agreement between the theoretical solution and the experimental data. The assumption that the velocity profile followed the theoretical parabolic form for laminar flow can be justified from the results of an example presented by Bird *et al.* [5] for laminar flow starting from rest in a circular tube. These results shown that the centerline velocity attains 95 per cent of its steady-state value in only 20 s for the present problem. The experimental observations in the present work were all at much greater times than 20 s. The process could thus be described as unsteady state convection and diffusion in a steady state laminar flow. The assumption of constant mass density was established by actual density

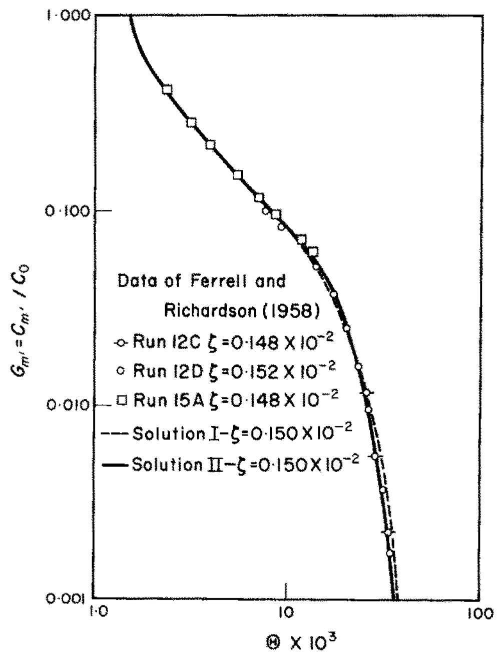

FIG. 3. Comparison of solutions by Method I and Method II for $\zeta = 0{\cdot}150 \times 10^{-2}$

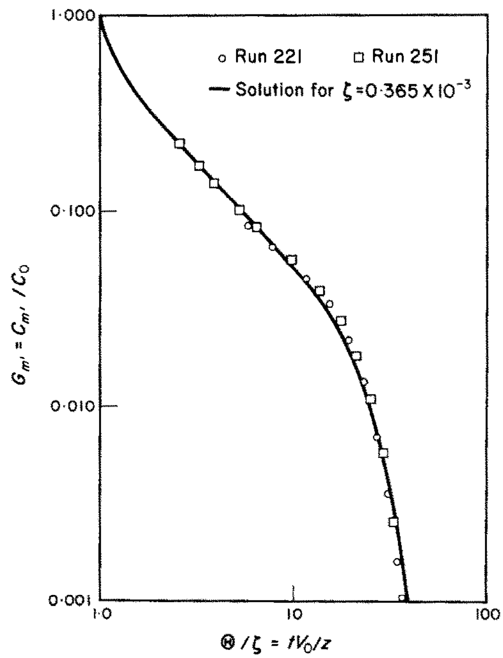

FIG. 4. Comparison of normalized solution and experimental data for runs 221 and 251.

measurements on pure water and the concentrated dye solution. This result also implies that natural convection effects are negligible.

For comparison of the theoretical solutions with experimental data, it was convenient to normalize the graphical solutions by plotting the abscissa as Θ/ζ. The ratio Θ/ζ is equal to tV_0/z, all of the parameters of which are experimentally measurable. This normalization of the solutions produces curves which all start at the same point, $\Theta/\zeta = 1{\cdot}0$. Physically this point represents the time at which the tip of the parabolic profile of the displacing liquid reaches the monitoring point and dye displacement begins. Since the mean concentrations were measured by a spectrophotometer and the initial concentration within a given run was known, all of the experimental data could be plotted in this form. The best theoretical solution representing the data could be then drawn in on this plot as shown in Fig. 4. From the value of ζ corresponding to this theoretical solution, an approximate value of the dye diffusion coefficient could be calculated. This technique was not found to be a very accurate method of determining diffusion coefficients, however. Experimental values of $G_{m'}$ greater than 0·3 or 0·4 were difficult to obtain because of the very rapid change of dye concentration with time in this portion of the run.

CONCLUSIONS

A technique was developed for solving the unsteady-state diffusion–convection equation in cylindrical coordinates for the case of negligible axial diffusion and the boundary conditions for a solution being displaced from rest by a pure solvent. Dye diffusion coefficients cannot be reliably measured by comparing theoretical solutions to this problem with experimental data. The technique for solving the mathematical model eliminated the use of complex finite-difference techniques and can be utilized on relatively modest sized computing equipment. Since many other physical problems can be represented by similar mathematical models, it was concluded that the solution technique used here could be very valuable in theoretical treatments of other engineering problems.

REFERENCES

1. A. Ananthakrishnan, W. N. Gill and A. J. Barduhn, *A.I.Ch.E. Jl* **11**, 1063 (1965).
2. Rutherford Aris, *Proc. R. Soc. (London)* **235A**, 67 (1956).
3. Rutherford Aris, *Proc. R. Soc. (London)* **252A**, 538 (1959).
4. H. R. Bailey and W. B. Gogarty, *Proc. R. Soc. (London)* **259A**, 352 (1962).
5. R. B. Bird, W. E. Stewart and E. N. Lightfoot, *Transport Phenomena*. John Wiley, New York (1960).
6. R. V. Churchill, *Operational Mathematics*. McGraw-Hill, New York (1958).
7. J. J. Van Deemter, J. J. Broeder and H. A. Lauwerier, *Appl. Scient. Res.* **A5**, 374 (1956).
8. M. A. Farrell and E. F. Leonard, *A.I.Ch.E. Jl* **9**, 190 (1963).
9. J. K. Ferrell, Ph.D. Thesis, North Carolina State College, Raleigh, N.C. (1954).
10. J. K.Ferrell and F. M. Richardson, Final Report, Office of Ordnance Research Contract No. DA-36-034-ORD-2199. Department of Engineering Research, North Carolina State College, Raleigh, N.C. (1958).
11. J. K. Ferrell, F. M. Richardson and K. O. Beatty Jr., *Ind. Engng Chem.* **47**, 29 (1955).
12. W. N. Gill and V. Ananthakrishnan, *A.I.Ch.E. Jl* **12**, 906 (1966).
13. W. N. Gill and V. Ananthakrishnan *A.I.Ch.E. Jl* **13**, 801 (1967).
14. Leon Lapidus, *Digital Computation for Chemical Engineers*. McGraw-Hill, New York (1962).
15. H. A. Lauwerier, *Appl. Scient. Res.* **8A**, 366 (1959).
16. J. R. Philip, *Aust. Phys.* **16**, 287–99 (1963).
17. N. S. Reejhsinghani, W. N. Gill and A. J. Barduhn, *A.I.Ch.E. Jl* **12**, 916 (1966).
18. N. S. Reejhsinghani, A. J. Barduhn and W. N. Gill, *A.I.Ch.E. Jl* **14**, 100 (1968),
19. H. E. Salzer, *J. Math. Phys.* **37**, 89 (1958).
20. C. J. Shirtliffe and D. G. Stephenson, *J. Math. Phys.* **40**, 135 (1961).
21. J. F. Swanson, M.S. Thesis, North Carolina State College, Raleigh, N. C. (1962).
22. G. I. Taylor, *Proc. R. Soc. (London)* **219A**, 186 (1953).
23. G. I. Taylor, *Proc. R. Soc. (London)* **223A**, 446 (1954).
24. G. I. Taylor, *Proc. R. Soc. (London)* **225A**, 473 (1954).
25. M. G. Williams, Ph.D. Thesis, North Carolina State University, Raleigh, N. C. (1965).

DIFFUSION INSTATIONNAIRE EN PHASE LIQUIDE DANS UN ÉCOULEMENT LAMINAIRE DANS UN TUBE CIRCULAIRE

Résumé—Une technique de résolution a été élaborée pour le modèle mathématique décrivant la diffusion instationnaire dans un liquide newtonien en écoulement laminaire permanent dans un tube circulaire. Le processus intéressant dans ce travail était celui pour lequel les nombres de Péclét étaient très élevés et oú les effets de la diffusion axiale pouvaient donc être négligés. Le modèle mathématique décrivant ce processus était une équation aux dérivées partielles avec trois variables indépendantes et leurs conditions aux limites associées. La méthode de résolution employait la transformée de Laplace, suivie, soit d'une séparation de variables, soit de calculs de différences finies, selon la grandeur des paramètres en cause.

Cette méthode a l'avantage qu'elle élimine les techniques complexes de différences finies et leurs problèmes associés de stabilité et de convergence qui doivent autrement être employées pour les solutions numériques exactes de problèmes de valeurs aux limites pour les trois variables indépendantes. La solution obtenue dans ce travail montrait un excellent accord avec les résultats expérimentaux dans le travail actuel et dans les recherches antérieures. Cette technique peut être appliquée à d'autres modèles mathématiques de formes semblable décrivant les problèmes de transport dans l'écoulement laminaire en régime transitoire et rendraient plus facile l'obtention des solutions numériques exactes de cette classe de problèmes.

INSTATIONÄRE FLÜSSIGKEITSDIFFUSION BEI LAMINARER ROHRSTRÖMUNG

Zusammenfassung—Es wurde eine Technik entwickelt, ein mathematisches Modell zu lösen, das die instationäre Diffusion in einer Newtonschen Flüssigkeit bei stationärer laminarer Strömung im runden Rohr beschreibt. Da bei dem in dieser Arbeit interessierenden Prozess die Péclétzahlen sehr gross waren, konnten axiale Diffusionseffekte vernachlässigt werden. Das mathematische Modell, das diesen Prozess beschreibt, war eine partielle Differentialgleichung mit drei unabhängigen Variablen und den zugehörigen Randbedingungen. Zur Lösung wurde zunächst eine Laplace-Transformation durchgeführt, die weitere Lösung erfolgte, abhängig von der Grösse der vorkommenden Parameter, durch Trennung der Variablen oder durch Differenzenverfahren.

Diese Methode hat den Vorteil, dass man damit komplexe Differenzenverfahren und die damit verbundenen Stabilitäts- und Konvergenzprobleme umgehen kann, die man sonst zur exakten numerischen Lösung von Grenzwertproblemen mit drei unabhängigen Veränderlichen benötigt. Die in dieser Arbeit erhaltene Lösung zeigt hervorragende Übereinstimmung mit experimentellen Werten, die bei gegenwärtigen Arbeiten und früheren Forschungen zusammengetragen wurden. Diese Methode kann auf andere mathematische Modelle ähnlicher Form angewandt werden, die Transportprobleme in instationärer laminarer Strömung beschreiben. Exakte numerische Lösungen dieser Art von Problemen lassen sich dadurch leichter ermitteln.

НЕСТАЦИОНАРНАЯ ДИФФУЗИЯ ЖИДКОСТИ ПРИ ЛАМИНАРНОМ ТЕЧЕНИИ В КРУГЛОЙ ТРУБЕ

Аннотация—Рассматривается решение математической модели, описывающей нестационарную диффузию в ньютоновской жидкости при стационарном ламинарном течении в круглой трубе. В интересующем нас случае значения критерия Пекле были весьма большими, а поэтому влиянием аксиальной диффузии можно было пренебречь. Математическая модель, описывающая данный процесс—дифференциальное уравнение в частных производных с тремя независимыми переменными и связанными с ними граничными условиями. Для решения использовалось преобразование Лапласа, а затем разделение переменных или вычисления в конечных разностях в зависимости от величины используемых параметров.

Данный метод имеет то преимущество, что он исключает сложную технику расчета в конечных разностях и связанные с ней проблемы устойчивости и сходимости, которые в противном случае необходимо использовать для получения точных численных решений краевых задач с тремя независимыми переменными. Решение, полученное в данной работе, показало прекрасное согласование с экспериментальными данными, упомянутыми в настоящей работе и в предыдущих исследованиях. Такой метод расчета можно применить к другим аналогичным математическим моделям, описывающим перенос при стационарном ламинарном течении, что дает возможность более легко получить численное решение такого класса задач.