

DIFFUSION OF DISINTEGRATION PRODUCTS OF INERT GASES IN CYLINDRICAL TUBES †

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Abstract—Presented herein is the problem of steady-state diffusion of the decaying products resulting from the disintegration of an inert gas flowing through a cylindrical tube. The disintegration products of the gas diffuse radially to the walls of the tube where they are absorbed. Three velocity distributions (uniform-, parabolic-, and Langhaar-) are considered and the respective solutions for the concentration distributions are obtained. Also presented are the $F(\mu)$ values for the respective velocity distributions. These values are needed in the determination of the diffusion coefficients of the disintegration products.

NOMENCLATURE

- $h_{n,m}$ defined by equation (10);
- B_n given below equation (14);
- c , mass concentration of the disintegration product;
- D , coefficient of diffusion;
- $F(\mu)$, defined by equation (5);
- q , rate of formation of disintegration product per unit volume of gas;
- r_0 , inner radius of the tube;
- Re , Reynolds number, defined as Vr_0/ν ;
- Sc , Schmidt number, defined as ν/D ;
- S_n given below equation (7);
- x, r , axial and radial coordinates, respectively;
- X_n eigenvalues of equation (9);
- v_x, v_r , axial and radial velocity components, respectively;
- V , mean flow velocity;
- α_n positive roots of $J_0(\alpha) = 0$;
- γ , known function of μ in Langhaar-velocity distribution;
- η , defined as r/r_0 ;
- μ , defined as Dx/Vr_0^2 ;
- ν , kinematic viscosity;

- A , dimensionless velocity distributions (v_x/V), with subscripts u, p and L for uniform-, parabolic- and Langhaar-profiles respectively;
- Φ , defined in equations (8) and (9);
- Ψ , dimensionless concentration distribution, defined as Dc/qr_0^2 .

STATEMENT AND FORMULATION OF PROBLEM

THE EQUATION (1) describing the steady-state mass diffusion of a constituent in a generating but nonreacting binary gas mixture flowing through a cylindrical tube, assuming azimuthal symmetry and constant coefficient of diffusion, can be written as

$$v_x \frac{\partial c}{\partial x} + v_r \frac{\partial c}{\partial r} = D \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial c}{\partial r} \right) + \frac{\partial^2 c}{\partial x^2} \right] + q. \tag{1}$$

The problem of diffusion of the decaying products of an inert gas flowing under the influence of forced convection, such as the diffusion of solid radium-*A* radioelements resulting from the disintegration of radon gas in its flight

† This work was done, in part, at the request of the Health and Safety Laboratory, United States Atomic Energy Commission (1968).

through a cylindrical tube, is mathematically depicted by such an equation [1]. In this situation, v_x and v_r are, respectively, the axial and radial velocity components of the gas; c the mass concentration of the radioelements; D their coefficient of diffusion, and q the rate of formation of these elements per unit volume of the flowing gas.†

For an isothermal system undergoing simultaneous momentum and mass diffusion processes, the Schmidt number Sc is a significant parameter since it expresses the relative speeds within the flow system for the transport of mass and that of momentum by the carriers. For high Schmidt numbers, $Sc > 1$, assumption of a fully developed parabolic-velocity profile does not lead to significant error because the velocity profile is established much more rapidly than the concentration profile. On the other hand, when the concentration profile outgrows the velocity profile at the inlet region (such is the case for $Sc < 1$), solutions based on the uniform-velocity profile provide at least a first-order approximation to the actual transport phenomena. For $Sc \approx 1$, however, both the velocity and concentration profiles develop at a similar rate along the tube, and neither the assumption of a parabolic- nor uniform-velocity profile is satisfactory. Instead, a developing velocity in the form of v_x and $v_r = f[(x/r_0)/Re, r/r_0]$ should be employed, which changes continuously within the transition length of the tube from the uniform-velocity at the tube inlet to the parabolic-velocity far downstream from the entrance. Among the various developing velocity distributions available, such as the works by Schiller [2], Boussinesq [2], Langhaar [3], Campbell and Slattery [4], Hornbeck [5], Christiansen and Lemmon [6], and Vrentas, Duda and Bargeron [7], the velocity given by Langhaar will be used in this analysis since it is given in a

closed analytical form more easily employed in the analysis of the diffusion problem. It should be noted, however, that the choice of this velocity profile limits it only to high Reynolds number flow.

For both the uniform- and parabolic-velocity distributions, the radial velocity component is zero, thus eliminating the term involving v_r in equation (1). In the transition length of the tube, however, this component is of considerable importance, but its significance diminishes rapidly away from the entrance. Since equation (1) is mathematically analogous to that of heat transfer in a cylindrical tube,‡ the analyses of Kays [8] and Lemmon [9] may be readily extended to the present diffusion problem. Thus for $\mu > 0.04$, which is the range of study in [13, 14], the effect of the v_r term is quite small compared to the other term on the left-hand side of equation (1) and will be neglected in this analysis.‡

For the typical transport problem, it may be assumed further that the diffusion in the direction of flow is negligible, and thus the $\partial^2 c / \partial x^2$ term is eliminated.† The criteria for which this term may be neglected, as in the case of convective heat transfer, have been established by Singh [10]. For gases with higher diffusivities, however, this assumption becomes less satisfactory.

Assuming that q is constant, the final equation to be considered, in non-dimensional parameters, is thus

$$\frac{1}{\eta} \frac{\partial}{\partial \eta} \left(\eta \frac{\partial \Psi}{\partial \eta} \right) - A \frac{\partial \Psi}{\partial \eta} + 1 = 0, \quad (2)$$

† The problem of diffusion undertaken here differs from the work done in convective heat transfer [8-12] only in the boundary conditions. The differential equation of diffusion and the boundary conditions ($c = 0$ at $x = 0$ and at $r = r_0$) do not describe a trivial system only because of the presence of the constant source term in the governing equation.

‡ The inclusion of axial diffusion and the term involving v_r in equation (1) is currently being studied by this author.

† The transit time within the tube is sufficiently short so that decay of radium-A is negligible. For example, a typical tube of 500 cc might be operated at a flow rate of 10 l./m. The transit time would then be 3 sec, compared to the 3.05 min half-life of radium-A [13].

with

$$\Psi = \frac{Dc}{qr_0^2}, \quad \Lambda = \frac{v_x}{V}$$

$$\eta = \frac{r}{r_0}, \quad \mu = \frac{Dx}{Vr_0^2}$$

where r_0 is the inner radius of the tube and V the mean flow velocity. Equation (2) is to be solved analytically whenever feasible (otherwise numerically) with three velocity profiles, namely a uniform-velocity A_u , a parabolic-velocity A_p and a Langhaar-velocity profile A_L [3, 8], or

$$\left. \begin{aligned} A_u &= 1, & A_p &= 2(1 - \eta^2), \\ A_L &= \frac{I_0(\gamma) - I_0(\gamma\eta)}{I_2(\gamma)}, \end{aligned} \right\} (3)$$

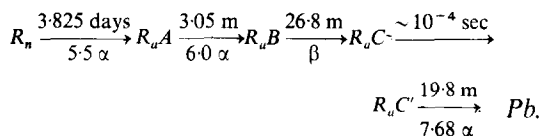
where γ , a known function of μ , is tabulated in Langhaar's paper.

The boundary conditions associated with this particular diffusion problem are based on the following observation and experimental technique. Since the radioelements are completely annihilated at the walls, $\dagger c = 0$ at $r = r_0$. Furthermore, if perfect filtration to remove any particulate matter is accomplished at the tube inlet, for example with a high efficiency inlet filter used in [13], then $c = 0$ at $x = 0$. The pertinent boundary conditions to equation (2), therefore, are

$$\Psi(1, \mu) = 0, \quad \Psi(\eta, 0) = 0. \quad (4)$$

Defining a new parameter F as the ratio of the total particle flux over a cross section at distance x from the tube inlet to the rate of formation of the radioelements in the same element of the tube, we have

\dagger The radium-*A* particle in colliding with the tube walls gives off an α -particle and decays into a radium-*B* particle which has a half-life of about 26.8 min. The chain of activity is shown from radon gas to lead as



$$F(\mu) = \frac{\int_0^{r_0} v_x c 2\pi r dr}{\pi r_0^2 x q} = \frac{2}{\mu} \int_0^1 \Lambda(\eta, \mu) \Psi(\eta, \mu) \eta d\eta. \quad (5)$$

The $F(\mu)$ values are needed in the experimental determination of the coefficient of diffusion, such as the two-filter method described in [13]. It can readily be shown that $F(0) = 1$ for any $\Lambda = \Lambda(\eta)$ and that

$$\mu F(\mu) = (D/qr_0^2) c_b,$$

where

$$c_b = \int_0^{r_0} v_x c r dr / \int_0^{r_0} v_x r dr$$

is the local bulk concentration. It is also apparent that c_b is dependent on the velocity distribution over the tube cross-section. For the uniform velocity, c_b is just the average concentration at any cross-section.

ASYMPTOTIC SOLUTIONS FOR LARGE μ

It is first noted that when the concentration distribution is fully established, $\partial c / \partial \mu \rightarrow 0$, equation (2) yields an asymptotic solution of $\Psi = (1 - \eta^2)/4$, irrespective of the velocity distributions involved. It is based on this asymptotic result that solution of the form of equation (8) is sought for the parabolic-velocity profile.

The asymptotic values of $F(\mu)$, however, depend on the associated velocity distributions. It can easily be shown that for the uniform-velocity distribution, i.e. $\Lambda = A_u$, the asymptotic values of $F(\mu) \rightarrow 1/(8\mu)$; and for the parabolic-velocity $\Lambda = A_p$, $F(\mu) \rightarrow 1/(6\mu)$. As $A_L \rightarrow A_p$ for large μ , it becomes apparent that with the Langhaar-velocity distribution, the $F(\mu)$ values approach those given for the parabolic-velocity case.

SOLUTIONS FOR UNIFORM-VELOCITY PROFILE

With a uniform-velocity profile, $\Lambda = A_u = 1$, the solution to equation (2), subject to the boundary conditions (4), can be found in a

straightforward manner by Laplace transforms to be:

$$\Psi(\eta, \mu) = \frac{1}{4}(1 - \eta^2)$$

$$- 2 \sum_{n=1}^{\infty} \exp(-\alpha_n^2 \mu) \cdot \frac{J_0(\alpha_n \eta)}{\alpha_n^3 J_1(\alpha_n)} \quad (6)$$

where $\pm \alpha_n, n = 1, 2, 3, \dots$, are the positive roots of $J_0(\alpha) = 0$.

The parameter F is then given by:

$$F(\mu) = \frac{1}{\mu} \sum_{n=1}^{\infty} S_n [1 - \exp(-\alpha_n^2 \mu)], \quad (7)$$

where $S_n = 4/\alpha_n^2$. For small values of μ , $\exp(-\alpha_n^2 \mu) = \sum_{m=0}^{\infty} (-1)^m (\alpha_n^2 \mu)^m / m!$, it is then apparent that $\lim_{\mu \rightarrow 0} F(\mu) = \sum_{n=1}^{\infty} 4/\alpha_n^2 \rightarrow 1$. For large μ , equation (7) gives an asymptotic expression of $F(\mu) = 1/(8\mu)$, as expected. The lower values of α_n^2 and S_n and the $F(\mu)$ values are listed respectively in Tables 1 and 2. The functions Ψ and F of equations (6) and (7) are shown respectively in Figs. 1 and 2.

SOLUTIONS FOR PARABOLIC-VELOCITY PROFILE

For the fully developed Poiseuille pipe flow, $A = A_p = 2(1 - \eta^2)$, we seek solution to equation (2) of the following form:

$$\Psi(\eta, \mu) = \frac{1}{4}(1 - \eta^2) + \sum_n \Phi_n(\eta) \exp(-x_n \mu / 2). \quad (8)$$

Substitution of the above into equation (2) yields

$$(\eta \Phi_n)' + X_n \eta (1 - \eta^2) \Phi_n = 0, \quad (9)$$

where the prime designates derivative with respect to η . Equation (9) is the familiar Sturm-Liouville equation, the solutions of which form a complete orthogonal set with real eigenvalues X_n . Realizing that Φ_n is finite at $\eta = 0$, and symmetrical about the μ -axis, they may be found by expanding Φ_n in a power series [11]

$$\Phi_n(\eta) = \sum_{m=0}^{\infty} b_{n,m} \eta^{2m}. \quad (10)$$

Substitution into equation (9) yields the following recursion formula for the coefficients

$$b_{n,m} = -\frac{X_n}{4m^2} (b_{n,m-1} - b_{n,m-2}), \quad (11)$$

Table 1. Lower eigenvalues and associated coefficients for equations (7), (13) and (14)

n	α_n^2	S_n	X_n	$b_{n,0}$	B_n
1	5.783186	0.119598	7.3135869	-0.286464	0.1589154
2	30.471262	0.004308	44.609461	0.049572	0.0059974
3	74.887007	0.000713	113.92103	-0.019679	0.0010864
4	139.040284	0.000207	215.24054	0.010483	0.0003401
5	222.932304	0.000080	348.56412	-0.006008	0.0001304
6	326.563353	0.000037	513.8899	0.000118	0.0000018
7	449.933529	0.000020			
8	593.042870	0.000011			
9	755.891395	0.000007			
10	938.479114	0.000004			

Table 2. $F(\mu)$ values

μ	Uniform-velocity	Parabolic-velocity	Langhaar-velocity
0.001	0.9513	0.9020	0.9378
0.01	0.8545	0.7894	0.8342
0.02	0.7974	0.7360	0.7777
0.03	0.7548	0.6990	0.7375
0.04	0.7197	0.6697	0.7055
0.05	0.6895	0.6451	0.6782
0.06	0.6627	0.6238	0.6542
0.07	0.6386	0.6048	0.6331
0.08	0.6165	0.5876	0.6143
0.09	0.5961	0.5719	0.5965
0.10	0.5772	0.5574	0.5800
0.11	0.5595	0.5439	0.5652
0.12	0.5428	0.5312	0.5511
0.13	0.5271	0.5192	0.5378
0.14	0.5122	0.5079	0.5253
0.15	0.4981	0.4972	0.5134
0.16	0.4847	0.4870	0.5021
0.17	0.4719	0.4772	0.4914
0.18	0.4597	0.4679	0.4810
0.19	0.4480	0.4589	0.4712
0.20	0.4368	0.4503	0.4618
0.21	0.4261	0.4420	0.4528
0.22	0.4158	0.4339	0.4441
0.23	0.4059	0.4262	0.4357
0.24	0.3964	0.4187	0.4276
0.25	0.3873	0.4114	0.4198
0.26	0.3785	0.4044	0.4123
0.27	0.3700	0.3976	0.4050
0.28	0.3618	0.3909	0.3979
0.29	0.3539	0.3845	0.3910
0.30	0.3463	0.3782	0.3844
0.32	0.3319	0.3662	0.3718
0.34	0.3184	0.3548	0.3600
0.36	0.3058	0.3440	
0.38	0.2940	0.3338	
0.40	0.2829	0.3241	
0.50	0.2367	0.2818	
0.60	0.2021	0.2479	
0.70	0.1756	0.2202	
0.80	0.1548	0.1974	
0.90	0.1382	0.1783	
1.00	0.1246	0.1623	
1.50	0.0833	0.1105	
2.00	0.0625	0.0832	
2.50	0.0500	0.0666	
3.00	0.0417	0.0555	
3.50	0.0357	0.0476	
4.00	0.0312	0.0416	
4.50	0.0278	0.0370	
5.00	0.0250	0.0333	

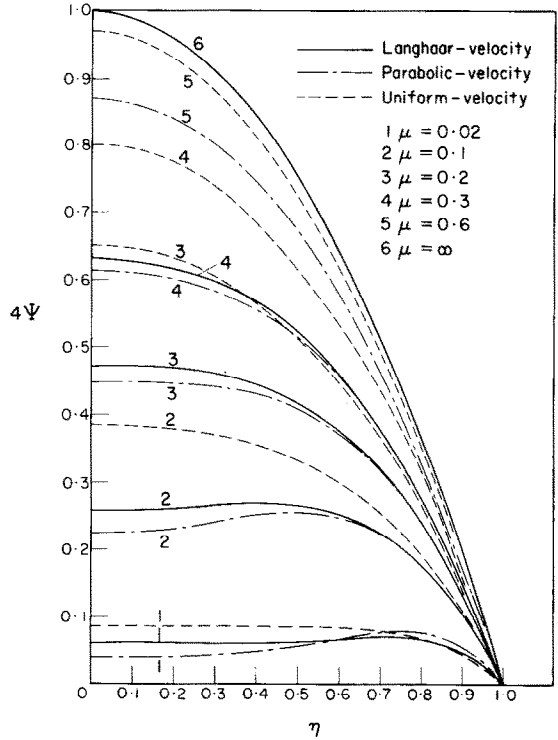


FIG. 1. Development of concentration profiles for various velocity distributions.

with

$$b_{n,1} = -\frac{X_n}{4} b_{n,0},$$

$$b_{n,2} = -\frac{X_n}{16} \left(\frac{X_n}{4} - 1 \right) b_{n,0}, \text{ etc.}$$

The eigenvalues X_n are found by invoking the boundary condition of $\Psi(1, \mu) = 0$. It amounts to

$$\sum_{m=0}^{\infty} b_{n,m} = 0. \tag{12}$$

The values of $b_{n,0}$ can then be evaluated from the other boundary condition $\Psi(\eta, 0) = 0$ to be

$$b_{n,0} = -\frac{1}{2} \left\{ \sum_{m=0}^{\infty} \frac{b_{n,m}/b_{n,0}}{(m+1)(m+2)(m+3)} \right\} / \left\{ \sum_{m=0}^{\infty} \sum_{j=0}^{\infty} \frac{(b_{n,m}/b_{n,0})(b_{n,j}/b_{n,0})}{(m+j+1)(m+j+2)} \right\}. \tag{13}$$

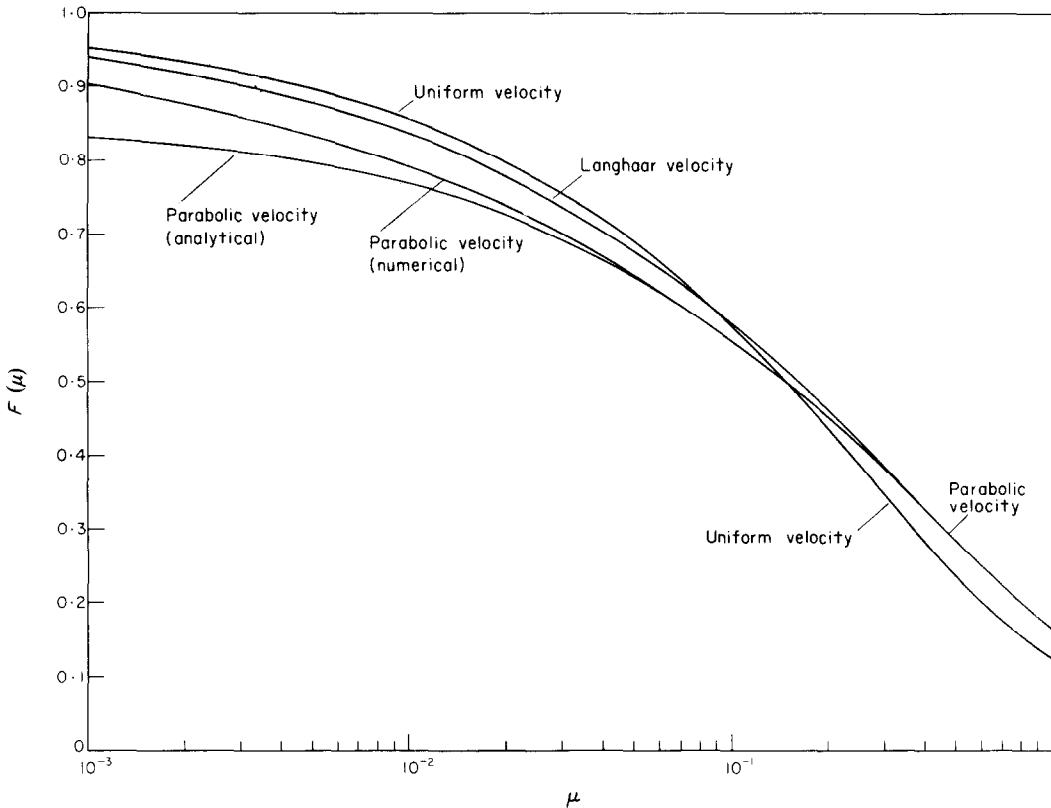


FIG. 2. $F(\mu)$ for various velocity distributions.

With the complete orthogonal functions Φ_n determined, $F(\mu)$ can thus be determined:

$$F(\mu) = \frac{1}{\mu} \sum_n B_n [1 - \exp(-X_n \mu / 2)], \quad (14)$$

where

$$B_n = - \sum_{m=0}^{\infty} 2b_{n,m} / (m + 1)(m + 2).$$

With the aid of a digital computer, the lower modes of X_n , $b_{n,0}$, and B_n can be computed. They are tabulated in Table 1. The corresponding $F(\mu)$ values are shown in Fig. 2. For large μ , the series solution technique is adequate, yielding $\lim_{\mu \rightarrow \infty} F(\mu) = 1/(6\mu)$, as expected. As $\mu \rightarrow 0$, however, this technique based on the six lower eigenvalues tabulated in Table 1, yields $F(0) = 0.841$ instead

of the theoretical value of $F(0) = 1$. The principal drawback of this technique for small μ is the difficulty in calculating directly the eigenfunctions Φ_n corresponding to the higher modes of the Sturm-Liouville equation. For these modes, the $b_{n,m}$ grow very large before diminishing, and the summing of the series to zero, equation (12), involves the difference of large numbers with the result that if accuracy is desired many digits must be carried in the computation. Furthermore, as $\mu \rightarrow 0$, the infinite series converges rather slowly necessitating a large number of terms.

The above difficulties may be circumvented by an iterative method described in [12, 15]. Instead, a numerical solution to equation (2) is sought. The non-dimensional concentration distribution 4Ψ is plotted against η and μ in

Fig. 1 and the numerically computed F -values shown in Fig. 2 and Table 2. For the range of our present interest, $\mu > 0.04$, the analytical and numerical solutions are practically identical, to at least the second decimal place.

SOLUTIONS FOR LANGHAAR-VELOCITY PROFILE

The partial differential equation (2) can be expressed in finite difference form, for the interior points of the tube, as follows:

$$\Psi_{\eta, \mu + \Delta\mu} = f_0 + f_1 \Psi_{\eta + \Delta\eta, \mu} + f_2 \Psi_{\eta, \mu} + f_3 \Psi_{\eta - \Delta\eta, \mu}, \tag{15}$$

where

$$f_0 = \frac{\Delta\mu}{\Lambda},$$

$$f_1 = -\frac{1}{\Lambda} \frac{\Delta\mu}{\Delta\eta} \left(\frac{1}{\Delta\eta} + \frac{1}{2\eta} \right),$$

$$f_2 = 1 - \frac{2}{\Lambda} \frac{\Delta\mu}{\Delta\eta^2},$$

$$f_3 = \frac{1}{\Lambda} \frac{\Delta\mu}{\Delta\eta} \left(\frac{1}{\Delta\eta} - \frac{1}{2\eta} \right).$$

When the nodal point (η, μ) is at the origin, $\eta = 0$, we have

$$\Psi_{0, \mu + \Delta\mu} = \frac{\Delta\mu}{\Lambda(0, \mu)} \left\{ 1 + \frac{4(\Psi_{1, \mu} - \Psi_{0, \mu})}{\Delta\eta^2} \right\}.$$

$\Delta\mu$ and $\Delta\eta$ are the arbitrarily chosen increments in the axial and radial directions, respectively. For the present numerical solutions $\Delta\eta$ was specified at $\frac{1}{30}$ and $\Delta\mu$ at 0.0001. As was observed in [8], with $\Delta\eta$ chosen, the choice of $\Delta\mu$ is somewhat restricted, because if $\Delta\mu$ is chosen such that f_2 becomes negative the solution will not converge. The f 's in equation (15) are functions of the velocity Λ and η , and it is here that the Langhaar-velocity $\Lambda = \Lambda_L = [I_0(\gamma) - I_0(\gamma\eta)]/I_2(\gamma)$ is introduced. Since γ is tabulated in [3] in terms of σ , where $\sigma = \mu Sc$, a value of $Sc = 1$ is assumed for a one-to-one

correspondence between σ and μ . Other values of Sc can be used with equal ease. Numerical calculations are carried out to $\mu = 0.34$, at and before which the concentration profile becomes sufficiently near fully developed. The functions Ψ and F for this case are included in Figs. 1 and 2, and the latter listed in Table 2.

RESULTS AND CONCLUSIONS

The development along the tube length of the non-dimensional concentration profiles for the three velocity distributions is shown in Fig. 1. It is seen that as the value of μ increases, the concentration profile based upon the Langhaar-velocity distribution approaches that of the parabolic-velocity distribution. As $\mu \rightarrow \infty$, all three velocity distributions give the same asymptotic expression of $\Psi = (1 - \eta^2)/4$ as they should.

Figure 2 shows the $F(\mu)$ values for the three velocity distributions considered. The analytical solution based on the six eigenvalues listed in Table 1 for the parabolic-velocity distribution is included in the figure together with the corresponding numerical solution for comparison. The Langhaar-velocity solution runs close to the uniform-velocity solution for small μ and merges asymptotically with the parabolic-velocity solution as μ becomes large.

Although the present analysis is applicable for $\mu > 0.04$, values of $F(\mu)$ are extended below this range to display the trend extrapolated from the present theory. It is then observed that in the length of transition the uniform-velocity solution provides an upper limit, while the parabolic-velocity solution gives a lower limit, to the $F(\mu)$ values. This is as expected since the former solution approximates the transport phenomena within the length of transition which would be obtained as the Schmidt number approaches zero, while the latter solution relates that as the Schmidt number becomes very large. The true solution within the length of transition for a fluid with Schmidt number close to unity, therefore, must lie somewhere between these two extreme cases.

Subject to the accuracy of the numerical technique, the value of F at $\mu = 0.04$ based on A_u is about 2 per cent higher, while that based on A_p is about 5 per cent lower, than the corresponding value of A_L . At $\mu = 0.23$,[†] the values of F based on A_u and A_p are about 7 per cent and 2 per cent lower, respectively, than that for A_L . For large μ , e.g. $\mu > 0.23$, the $F(\mu)$ values based on A_L are practically the same as those given by A_p , with an error of not more than 2 per cent. It is thus concluded that for the range $0.04 < \mu < 0.23$, the values of $F(\mu)$ based on A_L should be used, whereas for $\mu > 0.23$, the parabolic-velocity solution is adequate.

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[†] The value of $\sigma = 0.23$, where $\sigma = \mu Sc$, is taken to be the length of transition in [5].

Résumé—On présente ici le problème de la diffusion en régime permanent des produits décroissants provenant de la désintégration d'un gaz inerte s'écoulant à travers un tube cylindrique. Les produits de désintégration diffusent radialement vers les parois du tube où ils sont absorbés. On considère trois distributions de vitesse (uniforme, parabolique et de Langhaar) et les solutions pour les distributions de concentration sont obtenues. On présente aussi les valeurs de $F(\mu)$ pour les distributions respectives de vitesse. Ces valeurs sont nécessaires pour la détermination des coefficients de diffusion des produits de désintégration.

Zusammenfassung—Es wird das Problem der stationären Diffusion von Zerfallsprodukten eines Inertgases behandelt, wobei das Gas in zylindrischen Röhren strömt. Die Zerfallsprodukte des Gases diffundieren radial zur Rohrwand, wo sie absorbiert werden.

Drei Geschwindigkeitsverteilungen (gleichförmige, parabolische und nach Langhaar) werden betrachtet und ihre entsprechenden Konzentrationsverteilungen ermittelt. Daneben werden auch die $F(\mu)$ Werte für die entsprechenden Geschwindigkeitsverteilungen angegeben. Diese Werte sind zur Bestimmung der Diffusionskoeffizienten der Zerfallsprodukte erforderlich.

Аннотация—В статье рассматривается задача стационарной диффузии продуктов радиоактивного разложения инертного газа, текущего в цилиндрической трубе. Продукты распада диффундируют к стенке трубы, где они поглощаются. Рассматриваются три распределения скорости (равномерное, параболическое и распределение Лангхаара) и получены соответствующие решения для распределения концентраций. Представлены также значения $F(\mu)$ для соответствующих распределений скорости. Эти значения необходимы при определении коэффициентов диффузии продуктов разложения.