# ON A CELL TO MEASURE DIFFUSION COEFFICIENTS OF GASES THROUGH CIGARETTE PAPERS

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Abstract - A diffusion cell has been constructed to measure the molecular diffusion coefficient of carbon monoxide in a carbon monoxide-nitrogen mixture through cigarette paper. The cell and its application are briefly described, and the mathematics pertaining to it is presented. The theory enables the required diffusion coefficient to be calculated from measurements of the exit mixing cup concentrations.

# NOMENCLATURE

Ai, Bi,	airy functions of the first and second kinds	
Ь	breadth of compartments and exposed papers	
<i>b</i> ,	concentration of content monoride:	
ι,	concentration of carbon monoxide;	
с <sub>щ</sub> ,	mixing-cup concentration;	
D <sub>g</sub> ,	diffusion coefficient of carbon monoxide	
	through nitrogen in the unrestricted gas phase;	
D	diffusion coefficient of carbon monoxide	
	through nitrogen in the paper;	
е,	depth of the compartments;	
L,	thickness of the paper;	
М,	length of the exposed paper;	
$P_{i}$	eigenfunctions defined by equations (19)-(21);	
T <sub>b</sub>	eigenconstants defined by equation (22);	
и.	gas velocity in compartment:	
<i>V</i> .	volume flow rate in the compartments:	
x. v. z.	Cartesian co-ordinates	
X -	dimensionless variable defined by equation (13)	
<i></i> ,	dimensionless variable defined by equation (15).	
ireek symbols		
$\alpha = 2\rho \Gamma$	//D dimensionless narameter:	

G

$\alpha = 2eD_{\mu}$	/LD <sub>e</sub> , dimensionless parameter;
$\tilde{\alpha} = \lambda_i^{-27}$	3α;
Г.	gamma function:
$\xi = \lambda_i^{2/3} \eta$	······································
<i>n</i>	dimensionless variable defined by equation (13):
θ.	dimensionless variable defined by equation (13);
À.	eigenvalues defined by equations (19)-(21)
$\mu = \mu$	positive roots of $u \tan u = \alpha$ :
$\frac{\mu}{2} = \frac{\mu}{2^{2}/3}$	(1  n)
$\chi - \chi_i$ ,	$(1 - \eta)$ .

Subscripts

<i>p</i> ,	cigarette paper;
1,	upper compartment;
2.	lower compartment.

#### INTRODUCTION

As SMOKE is drawn through a cigarette, the concentration of carbon monoxide falls due to diffusion out of the cigarette and also due to dilution by air drawn in through the paper [1-10]. In some cigarettes more than half the carbon monoxide formed in the burning zone can be lost by outward diffusion as the

wrapper, and finally away from the outer surface of the paper. It is the first two of these phases that to a major extent limit the mass transfer rates [8]. Thus in studying mass transfer in cigarettes it is crucial to know the molecular diffusion coefficient in the cigarette wrapper. To this end, a diffusion cell has

been constructed to measure the diffusion coefficient through the paper of carbon monoxide in a carbon monoxide-nitrogen mixture. It is the purpose of this paper to describe briefly the cell, its usage and the mathematical theory pertaining to it.

gases are drawn through the cigarette. The carbon

monoxide escapes by a three-stage diffusion process: through the tobacco bed, then through the cigarette

# **DESCRIPTION OF CELL**

The arrangement of the cell, shown schematically in Fig. 1, consisted of two parallel compartments each of depth e separated by the paper of thickness L. The two halves of the cell, which were made of brass, were sealed by means of a rubber "O" ring. The paper was placed between the two sections and the whole unit was held together by clamps. For clarity, the rubber "O" ring and securing clamps are omitted from the diagram in Fig. 1. The depth of each half of the cell was altered by the insertion of a brass plate into the bottom of each compartment. In order to expose different lengths (M) of paper to the gas streams, identical cells of differing lengths were used. The breadth of the exposed paper (b), which was equal to the breadth of the compartments, was held fixed.

The gases flowed into and out of the cell via funnelled tubes so that the flow over the exposed paper was well developed. The Reynolds number for flow through the cell, based on cell depth as the characteristic dimension of the cell, varied between 20 and 80 for the range of experimental conditions used. This is sufficiently small for the flow to be taken as laminar.



FIG. 1. Schematic arrangement and cross-section of the diffusion cell.

A gaseous mixture of c% vol./vol. carbon monoxide in nitrogen (about 10% vol./vol., known accurately) flowed into one compartment of the cell, while nitrogen flowed at the same rate into the other compartment. The steady state mixing-cup concentrations of carbon monoxide in the exit gases from each compartment of the cell were measured in turn with a Bosch infra-red analyser type EFAW 215. While the mixingcup concentration in the exit gas from a given compartment was being determined, the gas from the other compartment passed through a length of glass tube chosen so that its impedance to flow was equivalent to that of the infra-red analyser. A pressure transducer (Ether type UP1) placed across the diffusion cell confirmed that when the gases flowed with equal rates into the two halves of the cell, there was always zero pressure difference (<0.01 cm water) across the paper  $(1 \text{ cm water} = 98 \text{ N m}^{-2}).$ 

Gas flow rates were measured with rotameters which had previously been calibrated for the appropriate gas mixture using accurate bubble meters. The outlet flow rates from each compartment were always found to be identical to the inlet rates, indicating that there is no net gas flow through the paper, as would be expected since the molecular weights of nitrogen and carbon monoxide are equal. The steady state mixingcup concentrations in the exit gas streams were reached in less than half a second.

### FORMULATION OF THE MATHEMATICAL MODEL

The x- and z-co-ordinate axes are as shown in Fig. 1, the y-axis (not shown) lies perpendicular to these axes. The cell contains three regions of interest: the lower and upper compartments into which pure nitrogen and the weak mixture of carbon monoxide and nitrogen flow, respectively, and the paper for which the diffusion coefficient is sought.

In the mathematical analysis it is assumed that the molecular diffusion coefficients and the total density are constant, and that axial diffusion may be neglected. Identical flows with volume flow rate V are assumed in both compartments, so that there is no net mass flow across the paper at any station. The flows are assumed steady, laminar and fully developed before the gas contacts the paper. Furthermore, the compartments' aspect ratio b/e is sufficiently large to justify the neglect of the effect of the side walls (at y = 0 and y = b) on the velocity distributions.

Subject to these assumptions, the governing equations for the concentrations of carbon monoxide in the compartments  $(c_1, c_2)$  and paper  $(c_p)$  are

compartments: 
$$D_g \frac{\partial^2 c_i}{\partial z^2} = u_i \frac{\partial c_i}{\partial x}, \quad i = 1, 2$$
 (1)

paper: 
$$\frac{\partial^2 c_p}{\partial z^2} = 0.$$
 (2)

Here the suffices 1, 2 and p refer to the upper and lower compartments and paper, respectively, and for fully developed laminar flow

$$u_1 = \frac{6V}{be^3} \left( z - \frac{L}{2} \right) \left( e + \frac{L}{2} - z \right)$$
(3)

and

$$u_{2} = -\frac{6V}{be^{3}}\left(z + \frac{L}{2}\right)\left(e + \frac{L}{2} + z\right).$$
 (4)

The boundary conditions at the impermeable surfaces  $z = \pm [e + (L/2)]$  are

$$\frac{\partial c_1}{\partial z} = 0$$
 at  $z = e + \frac{L}{2}$ ,  $x \ge 0$  (5)

$$\frac{\partial c_2}{\partial z} = 0$$
 at  $z = -e - \frac{L}{2}$ ,  $x \ge 0$  (6)

whilst at the paper we require

$$c_{1} = c_{p}$$

$$D_{g} \frac{\partial c_{1}}{\partial z} = D_{p} \frac{\partial c_{p}}{\partial z}$$
at  $z = \frac{L}{2}, \quad x \ge 0$ 
(7)

$$C_{2} = c_{p}$$

$$D_{g} \frac{\partial c_{2}}{\partial z} = D_{p} \frac{\partial c_{p}}{\partial z}$$
at  $z = -\frac{L}{2}, \quad x \ge 0.$ 
(8)

Finally, at the leading edge of the paper no mass transfer has yet taken place and the boundary conditions are

$$c_1 = c$$
 at  $x = 0$ ,  $\frac{L}{2} \le z \le \frac{L}{2} + e$  (9)

$$c_2 = 0$$
 at  $x = 0$ ,  $-\frac{L}{2} \ge z \ge -\frac{L}{2} - e$  (10)

where c is the concentration of carbon monoxide at the inlet to the upper compartment.

It is straightforward to show from equations (1)-(10) that the concentrations in the two compartments are connected by the simple relationship

$$c_2(x,z) = c - c_1(x,-z)$$
 (11)

and that  $c_1(x, z)$  is fully determined by equations (1), (3), (5), (9) together with the condition

$$\frac{\partial c_1}{\partial z} = \frac{2D_p}{LD_p} \left( c_1 - \frac{c}{2} \right) \quad \text{at } z = \frac{L}{2}, \quad x \ge 0.$$
 (12)

With the change of variables

$$\theta = \frac{2c_1}{c} - 1$$

$$X = \frac{bD_g x}{6Ve}$$

$$\eta = \frac{1}{e} \left( e + \frac{L}{2} - z \right)$$
(13)

the governing equations become

$$\frac{\partial^{2}\theta}{\partial\eta^{2}} = \eta(1-\eta)\frac{\partial\theta}{\partial X}$$
(14)  
$$\frac{\partial\theta}{\partial\eta} = 0 \quad \text{at } \eta = 0, \quad X \ge 0$$
  
$$\frac{\partial\theta}{\partial\eta} + \alpha\theta = 0 \quad \text{at } \eta = 1, \quad X \ge 0$$

and

$$\theta = 1$$
 at  $X = 0$ ,  $0 \le \eta \le 1$  (16)

where

$$\alpha = \frac{2eD_p}{LD_g}.$$
 (17)

Solution

The solution of equation (14) obtained by separation of variables is

$$\theta = \sum_{i} T_{i} P_{i}(\eta, \lambda_{i}) e^{-\lambda_{i}^{2} X}$$
(18)

where  $P_i(\eta, \lambda_i)$  and  $\lambda_i$  are the eigenfunctions and eigenvalues of

$$P_i'' + \lambda_i^2 \eta (1 - \eta) P_i = 0$$
 (19)

subject to the conditions

$$P'_i(0, \lambda_i) = 0, \quad P_i(0, \lambda_i) = 1$$
 (20)

and

$$P'_{i}(1,\lambda_{i}) + \alpha P_{i}(1,\lambda_{i}) = 0 \qquad (21)$$

where the dashes denote differentiation with respect to  $\eta$ . The eigenconstants  $T_i$  are obtained by applying the initial condition (16) and using orthogonality relations. Thus,

$$T_{i} = \frac{\int_{0}^{1} \eta(1-\eta)P_{i}(\eta,\lambda_{i})\mathrm{d}\eta}{\int_{0}^{1} \eta(1-\eta)P_{i}^{2}(\eta,\lambda_{i})\mathrm{d}\eta}$$
(22)

which may be manipulated into a more useful form as follows: the numerator may be simplified by integrating the differential equation (19) from  $\eta = 0$  to  $\eta = 1$  to obtain

$$\int_0^1 \eta(1-\eta) P_i(\eta,\lambda_i) \mathrm{d}\eta = -\frac{1}{\lambda_i^2} P_i'(1,\lambda_i). \quad (23)$$

Now for the denominator we first introduce the system

$$P'' + \lambda^2 \eta (1 - \eta) P = 0$$
 (24)

) subject to

$$P(0, \lambda) = 0$$
 and  $P'(0, \lambda) = 0$ , (25)

which is the same as the  $(P_i, \lambda_i)$ -system except that the eigenvalue equation (21) is dropped. By subtracting (24) multiplied by  $P_i$  from (19) multiplied by P and then integrating the result between  $\eta = 0$  and 1, we obtain

$$\int_0^1 \eta(1-\eta) P_i(\eta,\lambda_i) P(\eta,\lambda) d\eta$$
$$= \frac{1}{\lambda^2 - \lambda_i^2} \left\{ P_i'(1,\lambda_i) P(1,\lambda) - P'(1,\lambda) P_i(1,\lambda_i) \right\}.$$

On using (21) this may be written

$$\int_{0}^{1} \eta(1-\eta)P_{i}P \,\mathrm{d}\eta = \frac{P_{i}'(1,\lambda_{i})}{\alpha(\lambda+\lambda_{i})} \\ \times \left\{ \frac{P'(1,\lambda) - P_{i}'(1,\lambda_{i}) + \alpha[P(1,\lambda) - P_{i}(1,\lambda_{i})]}{\lambda-\lambda_{i}} \right\}$$

which on taking the limit as  $\lambda \rightarrow \lambda_i$  yields

$$\int_{0}^{1} \eta(1-\eta)P_{i}^{2}(\eta,\lambda_{i})d\eta$$
$$=\frac{P_{i}^{\prime}(1,\lambda_{i})}{2\lambda_{i}}\left\{\frac{1}{\alpha}\frac{\partial P^{\prime}}{\partial\lambda}(1,\lambda_{i})+\frac{\partial P}{\partial\lambda}(1,\lambda_{i})\right\}.$$
 (26)

Finally substituting (23) and (26) into (22) yields

$$T_{i} = \frac{-2}{\lambda_{i} \left\{ \frac{\partial P}{\partial \lambda}(1, \lambda_{i}) + \frac{1}{\alpha} \frac{\partial P'}{\partial \lambda}(1, \lambda_{i}) \right\}}.$$
 (27)

The mixing-cup concentrations  $c_{1m}$  and  $c_{2m}$  across any section of the upper and lower compartments, respectively, are given by

$$c_{1m} = \frac{c}{2}(1+\theta_m),$$
$$c_{2m} = \frac{c}{2}(1-\theta_m),$$

where

$$\theta_{m} = \frac{\int_{0}^{1} \eta(1-\eta)\theta(\eta)d\eta}{\int_{0}^{1} \eta(1-\eta)d\eta}$$
$$= 6\alpha \sum_{i} \frac{P_{i}(1,\lambda_{i})T_{i}}{\lambda_{i}^{2}} e^{-\lambda_{i}^{2}X}.$$
 (28)

The quantity of practical interest in the diffusion cell is the difference in the mixing-cup concentrations i.e.

$$\Delta c_m = c_{1m} - c_{2m} = c\theta_m. \tag{29}$$

The eigenvalue problem posed by equations (19)-(21) may be solved by transforming equation (19) to Whittaker's equation, which then may be solved in terms of the confluent hypergeometric function. For example, reference may be made to [11, 12, 13] which concern a related 3-cell liquid diffusion problem that has an application to the mass transfer process associated with haemodialysis. These authors have all ultimately resorted to numerical methods to treat the eigenvalue problem.

In the present investigation we have used a more direct approach and have solved the differential equation by a Runge-Kutta method together with a shooting technique. This was carried out as follows: an approximation to the eigenvalue  $\lambda_1^{(0)}$  was obtained by use of the asymptotic formulae (A5, A6) and a solution to (19) subject to (20) computed by using Gill's modification to the Runge-Kutta technique. An improved estimate to the eigenvalue was obtained by applying the Newton-Raphson root-finding technique to  $f(\lambda) = P'(1, \lambda) + \alpha P(1, \lambda) = 0$ ; the iteration scheme was

$$\lambda_{i}^{(n+1)} = \lambda_{i}^{(n)} - \frac{P'(1,\lambda_{i}^{(n)}) + \alpha P(1,\lambda_{i}^{(n)})}{\frac{\partial P'}{\partial \lambda}(1,\lambda_{i}^{(n)}) + \alpha \frac{\partial P}{\partial \lambda}(1,\lambda_{i}^{(n)})}$$

where  $\partial P/\partial \lambda$  satisfies

$$\frac{\partial P''}{\partial \lambda} + \lambda^2 \eta (1-\eta) \frac{\partial P}{\partial \lambda} + 2\lambda \eta (1-\eta) P = 0$$

subject to

$$\frac{\partial P'}{\partial \lambda}(0,\lambda) = 0, \quad \frac{\partial P}{\partial \lambda}(0,\lambda) = 0.$$

This improved estimate was used to run off a new solution and the iteration continued until convergence was obtained. The average time to obtain an eigenvalue and its corresponding eigenfunction was under two seconds on an ICL 2970 computer.

A further simplification to this problem is to assume that the flow is inviscid, so that the flows are uniform across the compartments. The resulting problem may easily be solved by Laplace transforms and the following results obtained:

$$\theta = -2\alpha \sum_{n=1}^{\infty} \frac{\mu_n \cos \mu_n (1-\eta) + \alpha \sin \mu_n (1-\eta)}{\mu_n [\mu_n^2 + \alpha^2 + \alpha]} e^{-6\mu_n^2 X}$$

and

$$\theta_m = 2\alpha^2 \sum_{n=1}^{\infty} \frac{1}{\mu_n^2 [\mu_n^2 + \alpha^2 + \alpha]} e^{-6\mu_n^2 X}$$

where  $\mu = \mu_n$ , n = 1, 2, 3, ..., are the positive roots of the transcendental equation

$$\mu \tan \mu = \alpha$$
.

For a given  $\alpha$ , the roots of this equation may be found by any of the standard root-finding procedures.

#### **RESULTS AND DISCUSSION**

Figures 2 and 3 illustrate typical output that may be obtained from the theoretical model. The concentration profiles at various axial stations in the two compartments are shown in Fig. 2 for a representative value of  $\alpha$  equal to 4. Within the restriction of the chosen scale there is no discernible difference in the profiles given by the inviscid and viscous models. However on plotting graphs of  $\alpha$  vs  $\Delta c_m/c$  at various axial stations (Fig. 3), it is apparent that there is considerable variation in the two sets of results for the larger values of  $\alpha$ . Thus the inviscid model seems to be appropriate only for the lower values of  $\alpha$ .

One might use Fig. 3 to determine the diffusion coefficient  $D_p$  for a given paper from a single experimental observation. However, in order to minimise experimental error, a least squares procedure was



FIG. 2. Concentration profiles in the upper and lower compartments for  $\alpha = 4$ .



FIG. 3. The dependence of  $\Delta c_m/c$  on the dimensionless groups  $\alpha$  and X, where  $\alpha = 2eD_p/LD_q$  and  $X = bD_p x/6Ve$ .

adopted. In practice N observed values of  $\Delta c_m/c$  in the exit gas are obtained by varying the experimental parameters and the value of  $D_p$  in the mathematical model adjusted so as to minimise

$$S = \sum_{i=1}^{N} \{ (\Delta c_m/c)_i^{obs} - (\Delta c_m/c)_i^{calc} \}^2.$$

Now for a given set of experimental parameters, the slope of the graph of  $\alpha$  against  $\Delta c_m/c$  (see Fig. 3) is dependent on the value of  $D_p$ , and approaches infinity for large values of  $D_p$ . Consequently, if the experimentally determined value of  $\Delta c_m/c$  lies in the neighbourhood of the steepest section of the graph, any small error in the measurements leads to a large error in the value of  $D_p$ . Thus in the determination of  $D_p$ , the experimental parameters should be chosen to avoid the steep part of the curve.

In the experiments the compartment breadth was fixed at 18 mm, the depths were 4.81, 3.22, 1.59 mm and exposure lengths of 50 and 100 mm were normally used and occasionally 20, 40, 60, 140 mm. The gas flow rates through each compartment were varied between 300 and  $1300 \text{ cm}^3 \text{ min}^{-1}$ , depending on the paper under test.

To evaluate the results of using the suggested procedure to determine  $D_p$ , we have substituted the calculated value of  $D_p$  back into the model and have compared experimental and theoretical results. For example, Fig. 4 illustrates the effect of varying the length of paper exposed to the gas flow on the observed values of  $\Delta c_m/c$  for various gas flow rates. The agreement is seen to be quite satisfactory. Values of  $D_p$  obtained by this method range from 0.001 to 0.020 cm<sup>2</sup> s<sup>-1</sup> for inherently porous papers used on cigarettes and having permeabilities in the range 2.0–200 cm min<sup>-1</sup> (10 cm water)<sup>-1</sup> respectively. The diffusion coefficient for diffusion of a binary mixture of carbon monoxide and nitrogen through the unrestricted gas phase is  $0.21 \text{ cm}^2 \text{ s}^{-1}$  at room temperature and pressure [17]. Consequently, it is seen that the interlocking network of cellulose fibres interspersed with chalk particles, which make up the structure of cigarette paper, offers a substantial hindrance to gaseous diffusion. This is due to the tortuous paths round the cellulose fibres through which the gas molecules must diffuse. The values of  $D_p$  are discussed further elsewhere [8].

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FIG. 4. Effect of length of exposed paper on  $\Delta c_m/c$  for various gas flow rates.  $D_p = 4.0 \times 10^{-3} \text{ cm}^2 \text{ s}^{-1}$ ;  $L = 37 \,\mu\text{m}$ ;  $e = 3.22 \,\text{mm}$ . The curves are theoretical.

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#### APPENDIX

IN THE numerical treatment of the eigenvalue problem (19)-(21) it is extremely important that good estimates of the eigenvalues are available. The standard techniques for obtaining these estimates are the W.K.B. method and, when the differential equation has transition points, the method of matched asymptotic expansions [14-16]. Although the estimates thus obtained are strictly only valid asymptotically, they are often surprisingly good even for the leading eigenvalues.

In the present problem there are transition points at  $\eta = 0$ and  $\eta = 1$ , and so we must find asymptotic expansions in three distinct regions:  $\eta \simeq 0$ ,  $\eta \simeq 1$  and  $0 < \eta < 1$ .

 $0 < \eta < 1$ 

In this region we may employ the straightforward W.K.B. method [14] and obtain

$$P_{i} = \frac{1}{\left[\eta(1-\eta)\right]^{1/4}} \left\{ a_{m} \cos\left(\lambda_{i} \int_{0}^{\eta} \sqrt{t(1-t)} dt\right) + b_{m} \sin\left(\lambda_{i} \int_{0}^{\eta} \sqrt{t(1-t)} dt\right) \right\} + O(a_{m}/\lambda).$$
(A1)

The factor  $[\eta(1-\eta)]^{-1/4}$  causes this solution to break down near  $\eta = 0$  and  $\eta = 1$  and therefore these regions must be studied separately. The arbitrary constants  $a_m$ ,  $b_m$  will be determined by matching.

 $\eta \simeq 0$ 

The appropriate independent variable near  $\eta = 0$  is  $\xi = \lambda_i^{2/3}\eta$  and the problem becomes

$$\frac{\mathrm{d}^2 P_i}{\mathrm{d}\xi^2} + \xi (1 - \lambda_i^{-2/3}\xi) P_i = 0$$

with

$$P_i(0) = 1$$
 and  $\frac{\mathrm{d}P_i}{\mathrm{d}\xi}(0) = 0$ .

To a first approximation in  $\lambda_i$ , we obtain

$$P_{i} = \frac{3^{1/6} \Gamma(2/3)}{2} \{ Bi(-\xi) + \sqrt{3} Ai(-\xi) \} + O(\lambda_{i}^{-2/3})$$
(A2)

and in order that (A1) and (A2) match, we require

$$a_{m} = \frac{(\sqrt{3}+1)\Gamma(2/3)}{2\sqrt{2\pi}} \left(\frac{3}{\lambda_{l}}\right)^{1/6},$$
$$b_{m} = \frac{(\sqrt{3}-1)\Gamma(2/3)}{2\sqrt{2\pi}} \left(\frac{3}{\lambda_{l}}\right)^{1/6}.$$

Here Ai and Bi denote Airy functions of the first and second kind, respectively.

 $\eta \simeq 1$ 

This problem is similar in character to that for  $\eta \simeq 0$ . The appropriate scaling is  $\chi = \lambda_i^{2/3}(1 - \eta)$  and we shall take  $\alpha = \lambda_i^{2/3}\tilde{\alpha}$ , where  $\tilde{\alpha}$  is O(1), in order to cover both the cases  $\alpha \to 0$  and  $\alpha \to \infty$ . The governing equation becomes

$$\frac{\mathrm{d}^2 P_i}{\mathrm{d}\chi^2} + \chi(1 - \lambda_i^{-2/3}\chi)P_i = 0$$

with

$$\frac{\mathrm{d}P_i}{\mathrm{d}\chi} - \tilde{\alpha}P_i = 0 \quad \text{at } \chi = 0.$$

Again to a first approximation in  $\lambda_i$  we obtain

$$P_{i} = b_{0} \{ Bi(-\chi) - \beta Ai(-\chi) \} + O(\lambda_{i}^{-2/3}b_{0})$$
 (A3)

$$B = \sqrt{3} \left\{ \frac{\tilde{\alpha} \, \Gamma(1/3) + 3^{1/3} \, \Gamma(2/3)}{\tilde{\alpha} \, \Gamma(1/3) - 3^{1/3} \, \Gamma(2/3)} \right\}.$$

On matching (A2) and (A3) we obtain the asymptotic expansion for  $\lambda_i$ 

$$\lambda_i = 8i - \frac{16}{3} + \frac{8}{\pi} \tan^{-1}(1/\beta) + O(i^{-2/3})$$
 (A4)

and the constant  $b_0$ 

$$b_0 = (-1)^{l+1} 3^{1/6} \Gamma(2/3) \{1 + \beta^2\}^{-1/2}$$

where i is a large positive integer.

Thus for moderate values of  $\alpha$  (let  $\tilde{\alpha} \rightarrow 0$ ) we may use

$$\lambda_i = 8i - \frac{20}{3}, \quad i = 1, 2, 3, \dots$$
 (A5)

as an estimate for the eigenvalues, whilst when  $\alpha$  is very large (let  $\hat{\alpha} \rightarrow \infty$ )

$$\lambda_i = 8i - 4, \quad i = 1, 2, 3, \dots$$
 (A6)

serves as a good estimate. Strictly speaking, the estimates (A5), (A6) relate to the cases of zero mass flux and constant concentration at the paper, respectively.

#### UNE CELLULE DE MESURE DU COEFFICIENT DE DIFFUSION DES GAZ A TRAVERS DES FEUILLES DE PAPIER A CIGARETTE

Résumé — Une cellule est construite pour mesurer le coefficient de diffusion moléculaire du monoxyde de carbone dans un mélange monoxyde de carbone/azote, à travers un papier à cigarette. La cellule et son utilisation sont décrites brièvement et le modèle mathématique est présenté. La théorie permet d'atteindre le coefficient de diffusion cherché, à partir des mesures des concentrations.

#### EINE ZELLE ZUR MESSUNG DES DIFFUSIONSKOEFFIZIENTEN VON GASEN DURCH ZIGARETTENPAPIERE

Zusammenfassung – Es wurde eine Diffusionszelle zur Messung des molekularen Diffusionskoeffizienten von Kohlenmonoxid in einer Kohlenmonoxid-Stickstoff-Mischung durch Zigarettenpapier hergestellt. Die Zelle und ihre Anwendung werden kurz beschrieben und die zugehörigen mathematischen Zusammenhänge dargelegt. Die Theorie ermöglicht die Bestimmung des gesuchten Diffusionskoeffizienten aus Messungen der Konzentrationen in der Austritrsmischkammer.

# ЯЧЕЙКА ДЛЯ ИЗМЕРЕНИЯ КОЭФФИЦИЕНТОВ ДИФФУЗИИ ГАЗОВ ЧЕРЕЗ ПАПИРОСНУЮ БУМАГУ

Аннотация — Разработана диффузионная ячейка для измерения коэффициента молекулярной диффузии окиси углерода в смеси окись углерода-азот через папиросную бумагу. Дано краткое описание ячейки и её применение. Развиваемая в работе математическая теория позволяет рассчитать коэффициент диффузии по данным измерений концентраций на выходе.