and the lepton interactions on the other hand. We note further that the quantities $\psi^{\mathrm{T}} C \gamma_\lambda \psi$ and $\psi^{\mathrm{T}} C \gamma_\lambda \gamma_5 \psi$ are covariants not only under real but also under special complex transformation as can be proved easily by using relation (8 c). As to the quantization, we have already remarked above, that one can perhaps make use of the linearity of the field equation in the "intrinsic" system by writing down the commutatorfunction in that system and then changing over to the "physical" system by means of local transformations.

The author is indebted to Prof. Dr. Scherzer whose critical remarks on a previous paper caused this paper and to Mr. Fuchsteiner, Mr. Walter and Mr. Winter for many helpful and stimulating discussions.

Performance of Thermal Diffusion Columns for Gas Mixtures

A. Youssef, M. M. Hanna, and M. D. Migahed

Nuclear Physics Department, UAR, Atomic Energy Establishment, UAR

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A systematic study of the performance of thermal diffusion columns is carried out in order to examine the limits of validity of the theory of isotopic mixtures in case of mixtures of gases. Nitrogen-carbon dioxide mixtures are used. It is found that the pressure dependence of the separation factor has the same qualitative form as that derived from the simplified theory valid for isotopic mixtures.

The theoretical hydrodynamics of the performance of the Clusius–Dickel thermal diffusion column has been examined by Clusius and Dickel [1], Jensen [2], and Waldmann [3] of the German School; by Furry, Jones, and Onsager [4], Jones and Furry [5], and McInteer and Reisfeld [6] of the American School; and by Srivastava and Srivastava [7], and Saxena and Raman [8] of the Indian School.

The only well established theory known until now is based on some simplifying conditions [8], the most important of which is that the dependence of the coefficient of viscosity (η), thermal conductivity (λ), diffusion (D), density (ϱ), and thermal diffusion factor (α_T) on the composition of the gas mixture is negligible; only temperature dependence of these quantities is taken into consideration. In other words, the only important variation of these quantities with position is that due to the existence of temperature gradients. This condition is justified only in case of isotopic mixtures where the fractional difference of the molecular weight is small. Another important restriction imposed on the theory is that, in limited temperature ranges, the inverse power model is capable of effectively representing the interaction among molecules for the calculation of η and D.

Under these simplifying conditions, it can be shown that for closed column operation at the steady state

$$\ln q = H L / (K_{\mathrm{c}} + K_{\mathrm{d}}) \tag{1}$$

where q is the equilibrium-separation factor and is the ratio of (x_1/x_2) at the top end of the column to that at the bottom end, x_i being the molar concentration of species i; L is the length of the column. The theoretical expressions for H, K_{c}, and K_{d} (which are functions of the transport coefficients of the gas mixture as well as the geometry of the column) indicate that these quantities are proportional to p^2, p^4, and p^0, respectively. Therefore, equation (1) can be put in the form

$$\ln q = \frac{a/p^2}{1 + b/p^4} \tag{2}$$

where a and b are given by

$$a = H L p^2 / K_{\mathrm{c}}, \qquad b = K_{\mathrm{d}} p^4 / K_{\mathrm{c}} \tag{3}$$

and p is the pressure of the gas.

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If asymmetries exist in the geometry or temperature distribution in the column, parasitic remixing occurs which causes a reduction in the separation. JONES and FURRY [5] showed that this effect can be taken into consideration by adding a term K_p in the denominator of equation (1) which has the same dependence on p as K_c. In this case equation (2) is modified to read

$$\ln q = \frac{a'/p^2}{1+b'/p^4} \tag{4}$$

where

$$a = a'(1 + K_p/K_c), \qquad b = b'(1 + K_p/K_c). \tag{5}$$

Equation (2) shows that q has a maximum at

$$p = p(q_{\max}) = \sqrt[4]{b},$$

the value of this maximum being given by

$$\ln q_{\max} = \frac{a}{2\sqrt{b}}.$$

The validity of the theory of performance of the thermal diffusion column is usually examined by plotting $p^2/\ln q$ against p^4. The relation should be a straight line from whose slope and intercept a and b can be obtained to be compared with their theoretical values. The theory was verified experimentally in case of isotopes by a number of authors such as NIER [9], WHALLEY et al. [10], HORIBE [11], and GROTH et al. [12].

The object of the present work is to carry out a systematic study of the performance of the thermal diffusion column in order to examine the limits of validity of the existing theory in case of gas mixtures.

Apparatus

Two CLUSIUS–DICKEL thermal diffusion columns are constructed in the present work, which represent the "extreme cylindrical" and "nearly plane" cases.

The first column consists of a vertical pyrex glass tube of effective length 285 cm and inner diameter 2.00 cm which forms the cold surface of the column. It is maintained at a constant temperature by means of a stream of water which flows continuously through a glass jacket. The hot surface of the column consists of a tungsten wire of diameter 0.03 cm which is stretched along the glass tube and which is centered in the axis by means of horizontal nickel cross-wires of 0.1 cm diameter, spot-welded along the tungsten wire at equal intervals of about 5 cm. These spacers are fixed in such a manner that they alternatively fall in two perpendicular vertical planes passing through the tungsten wire. The wire is heated electrically and its temperature can be changed by means of a variable transformer. The mean temperature of the wire is calculated by measuring its thermal elongation and knowing the coefficient of thermal expansion of tungsten. The lower end of the wire is attached to a stainless steel weight which keeps the wire taut and which allows its expansion into a mercury cup in the lower joint providing the required electrical connection. The mercury cup is also cooled by means of circulating water to keep the vapour pressure of mercury as low as possible (order of 0.001 Torr). The end connections to the sampling system are made as short and as narrow as possible to minimize the end volumes.

The second thermal diffusion column, which represents the nearly plane case, is a compact all-metal one of the vertical concentric cylinder type. Copper is chosen for the construction of this column because of its good heat conductivity. Along the axis of the column, there are two nichrome heating wires each of 0.3 mm diameter. Each wire is surrounded by a porcelain insulator to separate it from the copper tube which constitutes the inner hot surface of the annular space confining the gas mixture. This inner cylinder has the following dimensions: length = 235 cm, outer diameter = 1.28 cm, wall thickness = 0.1 cm. The corresponding dimensions of the outer cylinder are 211.5 cm, 2.83 cm, and 0.1 cm, respectively. It constitutes the cold surface of the column and is maintained at a constant temperature by means of a stream of water which flows continuously through a copper jacket. A steel sylphon bellow is fixed in a compressed state at the lower end of the cold pipe to allow the expansion of the hot tube. In order to minimize the heat losses, connection between the bellow and the lower end of the hot tube is made by means of thin-walled stainless steel tube of about 5.5 cm length. A similar stainless steel tube of 6 cm length connects the top of the cold tube to the hot one. Moreover, four thin stainless steel centering pins, tapered nearly to a point, are fixed at 40 cm intervals along the hot tube. The temperature of the hot surface is measured by means of an iron-constantan thermocouple which is introduced into the hot cylinder and is extended along its axis more than 150 cm. As in case of the first column, the end connections to the sampling system are made as short and as narrow as possible to minimize the end volumes.

The mixture of gases to be separated is introduced into the previously evacuated column by means of expansion from a 20-liter vessel which serves as a mixing reservoir. In the present case of N_2/CO_2 mixtures, it is

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found that equilibrium separation is essentially attained in 5 hours after which the mixture at either end of the column is withdrawn through a capillary tap and is removed to the mass spectrometer for analysis.

Transport Coefficients of N_2/CO_2 Mixtures

In the theoretical calculations of $p(q_{\max})$ and $q_{\max}$, experimental values of the coefficient of diffusion, the coefficient of viscosity, and the thermal diffusion factor should be used, whenever possible. If these are not available, the values of the coefficients are computed at the relevant temperature according to the LENNARD–JONES (12-6) model. ϱ values are evaluated according to the perfect gas equation.

a) Ordinary Diffusion

The data available (Fig. 1) cover the range of temperature between 270 °K and 1600 °K. They are given by KLIBANOV et al.[13], WALKER and WESTENBERG[14], and EMBER et al.[15]. A least squares analysis of these data yields the following relation[16]

$$D \times 10^5 = 0.9936 \times T^{1.7156}\ \mathrm{cm^2 \cdot sec^{-1}} \qquad (6)$$

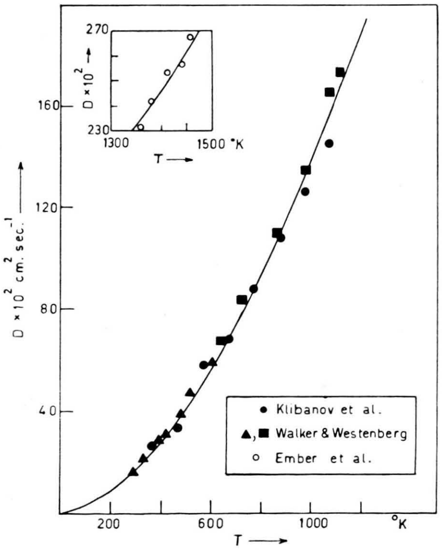

Fig. 1. Ordinary Diffusion of N_2/CO_2 Mixtures.

from which the coefficient of diffusion of N_2/CO_2 mixtures can be calculated at 1 atmosphere. At other pressures, D-values can be obtained from the fact that $(D\,p)$ is pressure independent.

b) Viscosity

The only available systematic measurements are those of KESTIN and LEIDENFROST[17] at 20 °C. In the present work, therefore, we use the values of η computed at 300 °K and 460 °K according to the LENNARD–JONES (12-6) model[18], using the interaction parameters

$$\varepsilon_{12}/k = 130.35\ ^\circ\mathrm{K} \quad \text{and} \quad \sigma_{12} = 3.823\ \text{Å}$$

as derived from the empirical combining rules[19]. These calculations are given in Fig. 2 which shows that our choice of the η-values is justified, at least at the lower temperature.

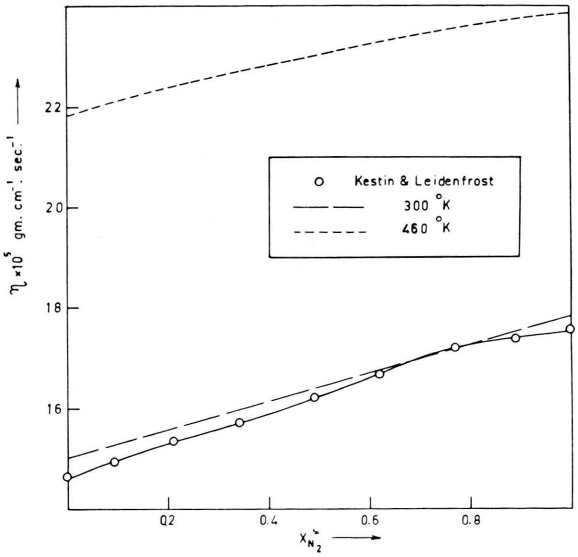

Fig. 2. Viscosity Coefficient of N_2/CO_2 Mixtures at 300 °K and 460 °K.

c) Thermal Diffusion

The only available systematic data are due to IBBS and WAKEMAN[20], WALDMANN[21], and GREW et al.[22] who measured the thermal diffusion factor for

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a 1 : 1 mixture over a range of temperature between 190 °K and 1200 °K. These experimental results can be fitted to the simple relation [16]

$$\alpha_T = 0.1143 - 22.2372/T\,. \qquad (7)$$

Youssef and Hanna [16] could show that, for 1 : 1 mixtures, the values of α_T calculated from equation (7) are in excellent agreement with the values obtained from the Chapman–Cowling relation [23] when the latters are calculated according to the Lennard–Jones (12-6) potential with parameters

$$\varepsilon_{12}/k = 156.26\ °\mathrm{K} \quad \text{and} \quad \sigma_{12} = 3.823\ \text{Å}\,,$$

and are then multiplied by a correction factor 0.73. This state of affairs is shown in Fig. 3. Whether this value of ε_{12}/k and this correction factor still hold for other compositions cannot unfortunately be established until further experimental data become available. In the present work, therefore, we shall assume that these parameters are composition independent. Fig. 4 shows the variation of α_T with composition at 300 °K and 460 °K according to the Lennard–Jones (12-6) model using the Chapman–Cowling relation after being multiplied by the previously mentioned correction factor.

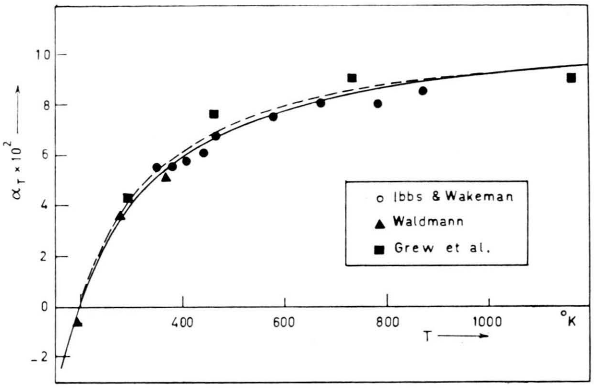

Fig. 3. Thermal Diffusion Factor of 1 : 1 N_2/CO_2 Mixtures. (– – – –) α_T calculated from eq. (7). (———) α_T calculated from modified Chapman–Cowling relation. ● α_T values of Ibbs and Wakeman.

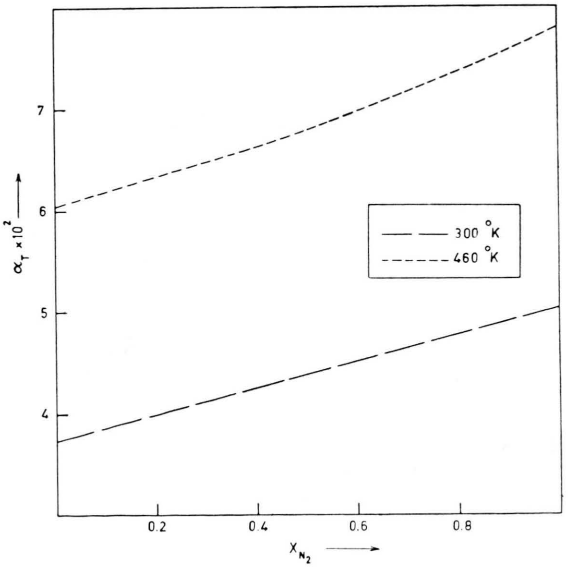

Fig. 4. Thermal Diffusion Factor of N_2/CO_2 Mixtures at 300 °K and 460 °K.

Experimental Results

a) Extreme Cylindrical Case

The equilibrium-separation factor q is measured as a function of pressure for six different N_2/CO_2 mixtures in which the mole fraction of the lighter component ranges from 0.062 to 0.950. The temperatures of the cold and hot surfaces are kept constant at $T_1 = 300$ °K and $T_2 = 693$ °K, respectively. The results are shown in Fig. 5 in which $\ln q$ is plotted against p. In order to examine the functional dependence of $\ln q$ on p, $p^2/\ln q$ is plotted against p^4. This is shown in Fig. 6; from which it can easily be seen that the equation (2) deduced for isotopic mixtures successfully applies to mixtures of gases. By means of a least squares analysis the values of a and b are obtained for each mixture. The values of these constants are then used to draw the curves shown in Fig. 5.

In Fig. 7, $p(q_{\max})$ is plotted as a function of the mole fraction of nitrogen, X_{N_2}. In order to compare these experimental points with the existing theory of isotopic mixtures, $p(q_{\max})$ is calculated as function of X_{N_2} for three models of molecular interaction, namely, rigid sphere model, Maxwellian model, and Lennard–Jones (12-6) model.

In the theoretical calculations, the following expression for $p(q_{\max})$ is used:

$$p(q_{\max}) = \sqrt[4]{\frac{9!}{r_1^6\, g^2}} \cdot \sqrt{\frac{\eta\,(D\,p)}{(\varrho/p)}} \cdot \sqrt[4]{\frac{k_d}{k_c}} \qquad (8)$$

where g is the acceleration of gravity and r_1 is the radius of the cold surface of the column. k_d and k_c are the column shape factors. The gas constants in equation (8) are to be evaluated at the cold temperature (300 °K). In the extreme cylindrical case this choice seems quite reasonable, since most of

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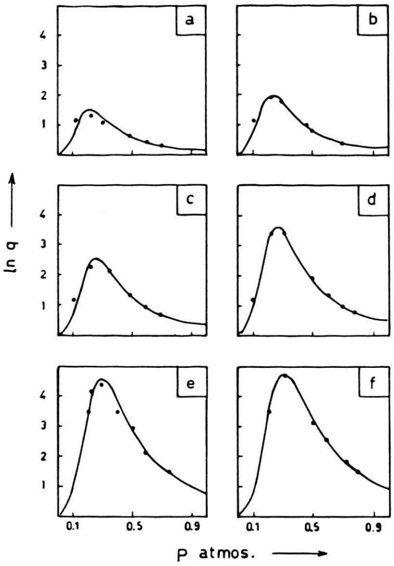

Fig. 5. ln q as a Function of p for Different N_2/CO_2 Mixtures.
(a) $X_{N_2}=0.062$, (b) $X_{N_2}=0.083$,
(c) $X_{N_2}=0.17$, (d) $X_{N_2}=0.50$,
(e) $X_{N_2}=0.91$, (f) $X_{N_2}=0.95$.
(Extreme Cylindrical Case.)

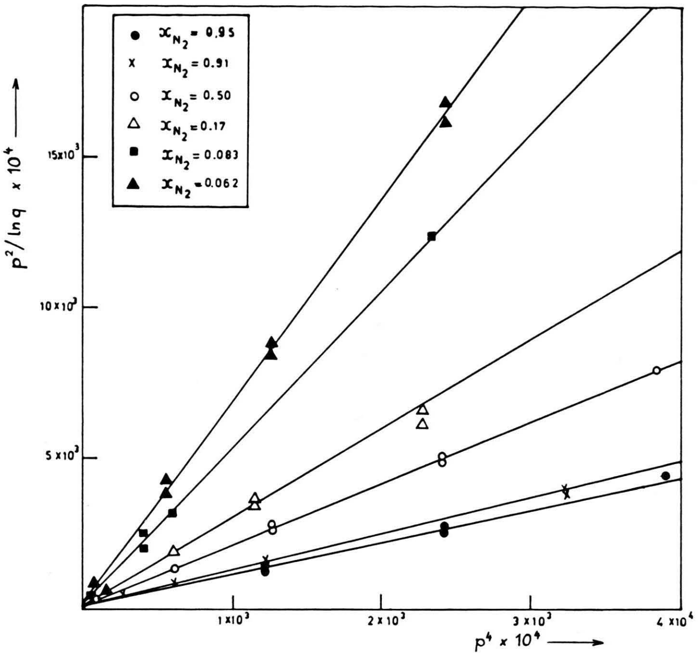

Fig. 6. $p^2/\ln q$ as a Function of p^4 (Extreme Cylindrical Case).

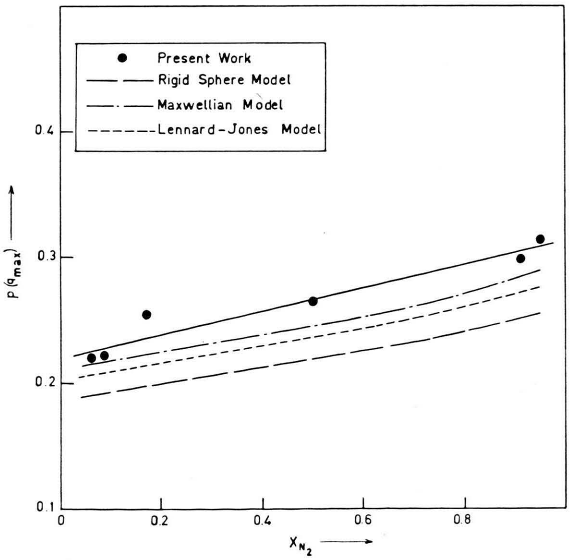

Fig. 7. $p(q_{\max})$ as a Function of X_{N_2} (Extreme Cylindrical Case).

the gas in the column is maintained at this temperature. It is worthwhile to mention that $p(q_{\max})$ is independent of the thermal diffusion factor. Throughout our calculations we have used the tabulated column shape factors of McINTEER and REISFELD [24] for the LENNARD–JONES (12-6) potential, of FURRY and JONES [25], and of RAMAN and SAXENA [26] for the inverse power potential with $n=1$ (MAXWELLian model), and $n=1/2$ (rigid sphere model), respectively.

Fig. 7 shows that the dependence of $p(q_{\max})$ on X_{N_2} is qualitatively in good agreement with the existing theory for isotopic mixtures. For a first look, however, it seems astonishing that the measured values of $p(q_{\max})$ are higher than the calculated ones. This should mean that either we have a negative parasitic effect, which is meaningless; or the intermolecular forces for the given mixture have a longer range than that corresponding to MAXWELLian interaction, which is quite improbable. As a matter of fact, this discrepancy may be accounted for if we bear in mind that the gas constants are all evaluated at the cold temperature (300 °K). If it happens that the temperature of the gas as a whole is somewhat higher than this temperature, which is quite plausible due to the existence of the axial hot wire, then higher values of η and D and lower

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[26] S. RAMAN and S. C. SAXENA, J. Chem. Phys. **36**, 3345 [1962].

values of ϱ should have been inserted in equation (8). As it is clear from this equation, this temperature correction results in an upward shift of the theoretical curves of Fig. 7. It can be actually shown that a temperature rise of about 10 to 15 °K brings the MAXWELLian and LENNARD–JONES curves in good agreement with the experimental points.

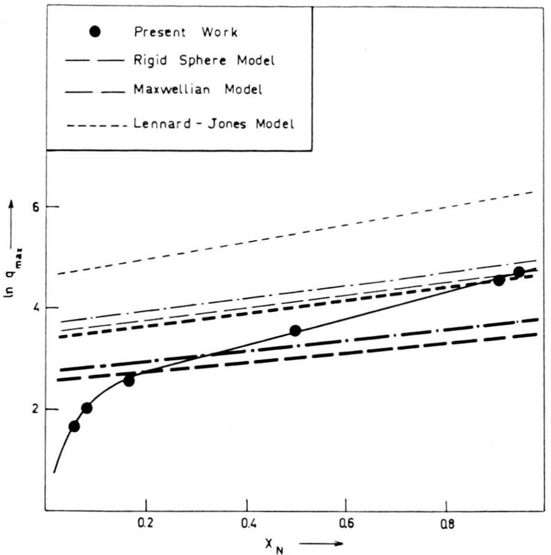

Fig. 8. q_{max} as a Function of Composition According to Rigid Sphere, MAXWELLian, and LENNARD–JONES Models. (Extreme Cylindrical Case.) Thin curves correspond to α_T calculated from the CHAPMAN–COWLING relation. Bold curves correspond to α_T calculated from the modified CHAPMAN–COWLING relation.

Fig. 8 shows the variation of $\ln q_{max}$ as a function of the nitrogen mole fraction X_{N2}. Two theoretical curves are plotted for each model of interaction to be compared with the experimental points. One of these curves is based on the values of α_T calculated according to the LENNARD–JONES (12-6) model with the potential parameters ε_{12}/k and σ_{12} as mentioned above from the CHAPMAN–COWLING relation at 300 °K. The thermal diffusion factor used to plot the second theoretical curve is still calculated from the CHAPMAN–COWLING relation at 300 °K but after introducing the correction factor 0.73 assumed to be composition independent (Fig. 4). The theoretical values of $\ln q_{max}$ are obtained from the following relation:

$$\ln q_{max} = \frac{1}{2} \frac{\sqrt{9!}}{6!} \frac{L}{r_1} \alpha_T \frac{h}{\sqrt{k_c\, k_d}} \tag{9}$$

where L is the length of the column and h, k_c, and k_d are the column shape factors. Throughout the calculations, the shape factor h is evaluated [24–26] under the assumption that α_T is independent of temperature. From equation (9), it is clear that $\ln q_{max}$ is independent of ϱ, η, and D; it depends only on the thermal diffusion factor α_T.

It is clear that the experimental points do not agree with the calculated curves. The discrepancy is principally due to the values of α_T introduced in equation (9). As a matter of fact, if we consider the MAXWELLian model, and if we introduce the previously mentioned temperature correction (i. e. we calculate α_T at 315 °K instead of 300 °K), we find that the theoretical curve corresponding to the modified CHAPMAN–COWLING values of α_T cuts the experimental curve exactly at $X_{N2} = 0.5$. Of course, this is to be expected, since it is for this composition that the experimental value of α_T is perfectly known (Fig. 3). Therefore, a possible explanation of the observed discrepancy is that the correction factor, which is to be multiplied with α_T of CHAPMAN and COWLING to give the correct value of the thermal diffusion factor, is not composition independent as it was previously assumed. If this point of view were true, then the multiplying correction factor, which has the value 0.73 at $X_{N2} = 0.5$, should change from about 0.5 to about 1 as X_{N2} increases from 0.05 to 0.95. Further experimental data of α_T for N_2/CO_2 mixtures are required to investigate this problem.

b) Nearly Plane Case

The equilibrium-separation factor q is measured as a function of pressure for eight different N_2/CO_2 mixtures in which the mole fraction of nitrogen ranges from 0.05 to 0.95. The temperatures of the cold and hot surfaces are kept constant at $T_1 = 300$ °K and $T_2 = 620$ °K, respectively. Similar results (Fig. 9) are obtained as those shown in Fig. 5 for the extreme cylindrical case. By plotting $p^2/\ln q$ against p^4, the straight lines shown in Fig. 10 are obtained which prove that equation (2) deduced for isotopic mixtures still successfully holds for mixtures of gases in coaxial-cylinder thermal diffusion columns.

In Fig. 11, $p(q_{max})$ is plotted as a function of X_{N2}. In a similar manner as in case of the extreme cylindrical column, $p(q_{max})$ is calculated for the MAXWELLian and LENNARD–JONES models of molecular interaction. The theoretical curves are also shown in Fig. 11. For the MAXWELLian model, the

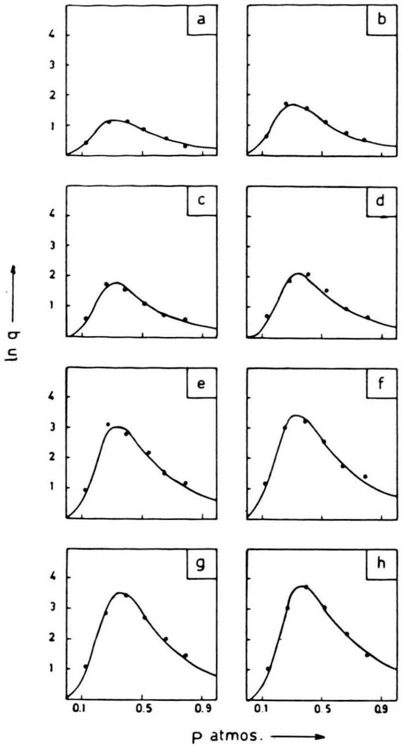

Fig. 9. ln q as a Function of p for Different N_2/CO_2 Mixtures.
(a) $X_{N_2}=0.05$, (b) $X_{N_2}=0.07$,
(c) $X_{N_2}=0.1$, (d) $X_{N_2}=0.2$,
(e) $X_{N_2}=0.3$, (f) $X_{N_2}=0.5$,
(g) $X_{N_2}=0.7$, (h) $X_{N_2}=0.95$
(Nearly Plane Case).

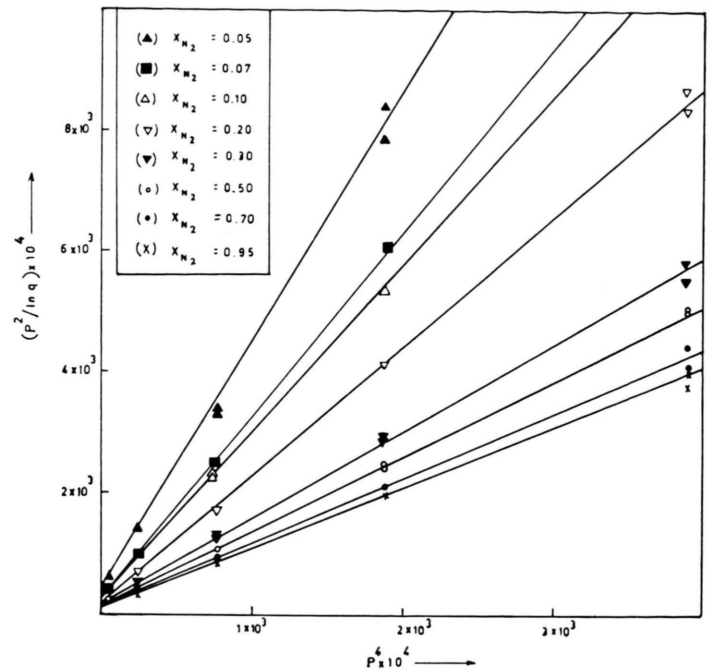

Fig. 10. $p^2/\ln q$ as a Function of p^4 (Nearly Plane Case).

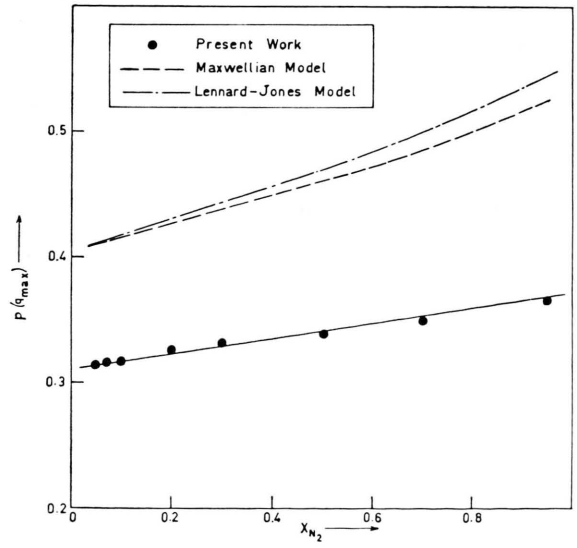

Fig. 11. $p(q_{\max})$ as a Function of X_{N_2} (Nearly Plane Case).

column shape factors are calculated after FURRY and JONES [25] and the following expression for $p(q_{\max})$ is used:

$$p(q_{\max}) = \sqrt[4]{\frac{9!}{(r_1-r_2)^6\, g^2}}\; \sqrt{\frac{\eta\,(D\,p)}{(\Delta T/\bar{T})\,(\varrho/p)}}\; \sqrt[4]{\frac{k_d'}{k_c'}} \quad (10)$$

where r_1 and r_2 are the radii of the cold and hot cylinders, respectively, $\Delta T = T_2 - T_1$, $\bar{T} = \frac{1}{2}(T_1 + T_2)$, and k_d' and k_c' are the column shape factors. For the LENNARD–JONES model, the column shape factors are obtained from the tables of MCINTEER and REISFELD [24], and equation (8) is used to calculate $p(q_{\max})$. In the nearly plane case, the gas constants in equations (8) and (10) are to be calculated at the mean temperature $\bar{T}$ (in our present work, $\bar{T} = 460$ °K).

Fig. 12 shows the variation of $\ln q_{\max}$ as a function of X_{N_2}. Four theoretical curves are also plotted in the figure to be compared with the experimental points. For the MAXWELLian model, the column shape factor h' is calculated after FURRY and JONES [25] for α_T temperature independent and α_T proportional to T, and after SRIVASTAVA and SRIVASTAVA [7] for α_T varying as $A - B/T$. The following expression is used to obtain $\ln q_{\max}$:

$$\ln q_{\max} = \frac{1}{2}\,\frac{\sqrt{9!}}{6!}\,\frac{L}{(r_1-r_2)}\,\frac{\Delta T}{\bar{T}}\,\alpha_T\,\frac{h'}{\sqrt{k_c'\,k_d'}}\,. \quad (11)$$

For the LENNARD–JONES model, the tables of MCINTEER and REISFELD[24] and equation (9) are used to calculate $\ln q_{\max}$. The values of α_T introduced in our calculations are taken from Fig. 4 at the mean temperature $\overline{T} = 460\ {}^\circ\mathrm{K}$.

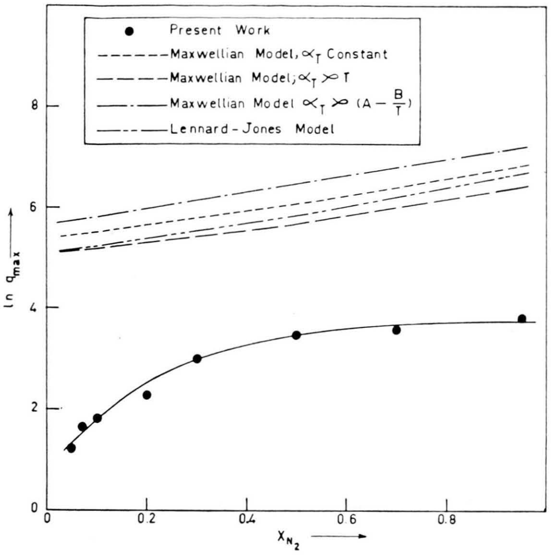

Fig. 12. $q_{\max}$ as a Function of Composition According to MAXWELLIAN and LENNARD–JONES (12-6) Models (Nearly Plane Case).

The general features of the experimental curves are the same as in the extreme cylindrical case. However, in the present case, a parasitic remixing is quite clear. If equation (5) is used to calculate K_p/K_c as a function of X_{N_2}, we find that the $p(q_{\max})$-curve (Fig. 11; MAXWELLian model) yields a value of K_p/K_c which changes from 1.9 to 2.9 as X_{N_2} increases from 0.1 to 0.9. On the other hand the $\ln q_{\max}$-curve (Fig. 12; MAXWELLian model, α_T constant) yields a nearly constant value $K_p/K_c \approx 2$ for values of $X_{N_2} \geqq 0.5$. That the latter value of K_p/K_c is composition independent is in good agreement with the results derived by JONES and FURRY[5] assuming simplified modes of asymmetry.

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