

CHAPTER 5

Column Calibration Factor to Study the Composition Dependence of the Thermal Diffusion Factors of Inert Gas Mixtures.

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5.1. Introduction

Nowadays the study of thermal diffusion is mostly directed to measuring the equilibrium separation factor q_e defined by

$$q_e = (C_i/C_j)_{\text{top}} / (C_i/C_j)_{\text{bottom}} \quad \dots\dots\dots (5.1)$$

for isotopic and nonisotopic binary gas mixtures of different compositions at a fixed temperature, or for a binary gas mixture of fixed composition at different temperatures, in a thermal diffusion (TD) column first introduced by Clusius and Dickel¹⁾. Here C_i and C_j are the mass fractions of the lighter and heavier components of a gas mixture respectively, and the subscripts 'top' and 'bottom' denote the values at the top and bottom of a TD column. The experimental values of q_e are measured usually at different pressures below and around one atmosphere with the help of a precision type mass spectrometer and hence the TD factor α_T of a binary gas mixture as a function of the composition or the temperature is estimated by using various molecular models such as those of Maxwell and Lennard-Jones²⁾ and sometimes by using Sliker's model-independent method³⁾, which is, however, a crude one. In an attempt to get the actual α_T , we,⁴⁻⁶⁾ however, introduced a factor F_s called the column calibration factor of a TD column, by the relation

$$\ln q_{\text{max}} = \alpha_T F_s (r_c, r_h, L \text{ and } \bar{T}) \quad \dots\dots\dots (5.2)$$

where q_{max} is the maximum value of q_e measured experimentally in a TD column, r_c and r_h are the cold wall and hot wall radii of the column of geometrical length L and \bar{T} is the mean temperature in K of the gas mixture in the column, defined by $\bar{T} = (T_h + T_c)/2$, T_h and T_c being the temperatures of the hot and cold walls of the column, respectively. The column calibration factor F_s which is purely an apparatus quantity, is supposed to be independent of a molecular interaction model and depends only on the geometry of the column at any mean temperature \bar{T} .

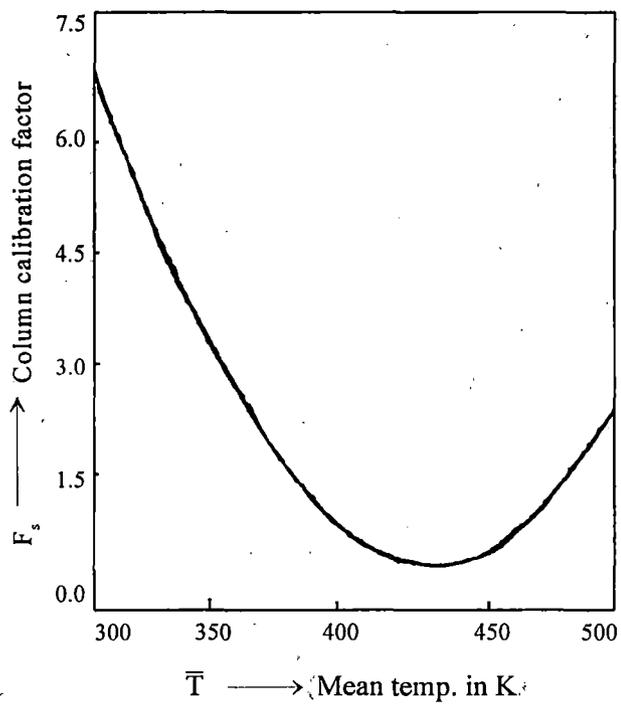


Fig. 5.1. Plot of the column calibration factor against \bar{T} , the mean temperature in K.

The column calibration factor F_s of a column with $L = 149$ cm, $r_c = 1.37$ cm and $r_h = 0.6$ cm has already been studied elsewhere⁵⁾, and the empirical relation of F_s to \bar{T} has been found to be

$$F_s = 68.94796 - 0.3174514 \bar{T} + 3.71383 \times 10^{-4} \bar{T}^2$$

yielding the value of $F_s = 3.946$ at $\bar{T} = 340$ K. The F_s for this column is plotted against \bar{T} in Fig 5.1. Fortunately J. M. Saviron et al⁷⁾ have recently studied the pressure dependence of the reduced logarithmic separation factors for two binary inert gas mixtures of $\text{He}^4 - \text{Ar}^{40}$ and $\text{Ne}^{20} - \text{Xe}^{132}$ and also an isotopic natural mixture of $\text{Ne}^{20} - \text{Ne}^{22}$ for different compositions of the lighter components in the respective mixtures in this column at the experimental mean temperature $\bar{T} = 340$ K, the hot wall and cold wall temperatures being $\bar{T}_h = 380$ K and $T_c = 300$ K respectively. From these results we estimate $\ln q_e$ of the mixtures for different concentrations of the lighter components at different pressures in atmosphere and the data thus obtained are found to satisfy the hydrodynamical part of the column theory, as developed by Furry and Jones⁸⁾ and Furry, Jones and Onsager⁹⁾, so excellently shown in Figs 5.2 and 5.3 that we have at once the following relations of $p^2/\ln q_e$ to p^4 for these mixtures at different concentrations:

$$p^2/\ln q_e = 3.09752 + 177.30496 p^4$$

$$p^2/\ln q_e = 8.52467 + 66.65334 p^4$$

$$p^2/\ln q_e = 17.20000 + 41.70213 p^4$$

$$p^2/\ln q_e = 19.65238 + 16.78669 p^4$$

$$p^2/\ln q_e = 18.74249 + 12.92235 p^4$$

for the $\text{He}^4 - \text{Ar}^{40}$ mixture for the concentrations 8.76%, 21.63%, 30.14%, 49.30% and 64.46% of He^4 respectively;

$$p^2/\ln q_e = 0.59848 + 3156.6 p^4$$

$$p^2/\ln q_e = 2.39745 + 327.3 p^4$$

$$p^2/\ln q_e = 3.03368 + 133.3 p^4$$

for the $\text{Ne}^{20} - \text{Xe}^{132}$ mixture for 15%, 66% and 90% of Ne^{20} , and finally

$$p^2/\ln q_e = 13.3 + 89.06 p^4,$$

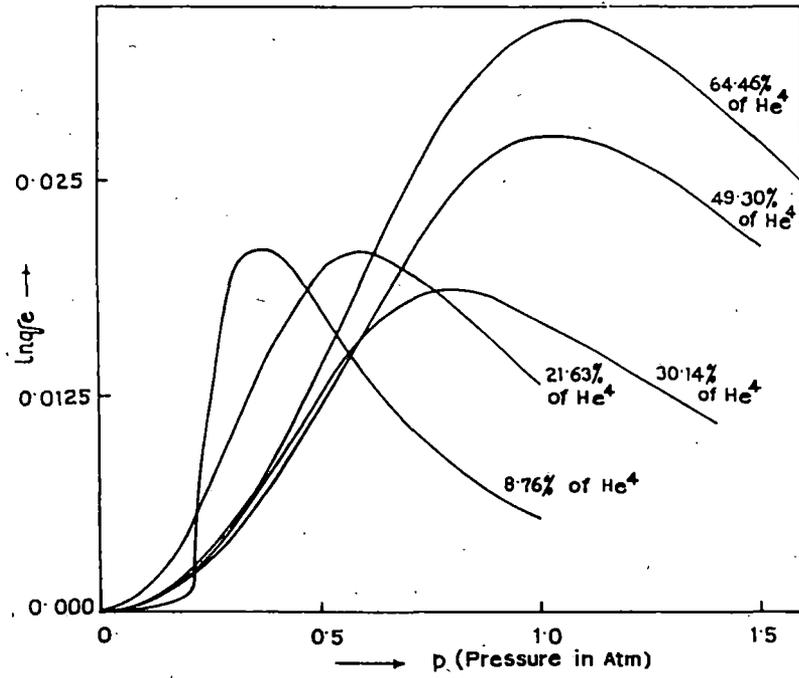


Fig. 5.2. Plot of $\ln q_e$ of He^4 - Ar^{40} (binary inert gas mixture) against pressure p in atm at different concentrations of He^4 at temperature $\bar{T}=340$ K.

Table 5.1. Composition dependence of thermal diffusion factors α_T of inert gas at $\bar{T} = 340$ K in a column of $L =$ geometrical length = 149 cm. $r_c =$ the cold wall radius = 1.37cm and $r_h =$ the hot wall radius = 0.6 cm.

[44]

System	T_h in K	T_c in K	\bar{T} in K	% of lighter component.	$a' \times 10^3$ (atm) ²	$b' \times 10^3$ (atm) ⁴	$\ln q_{\max}$	F_s	Expt $\alpha_T \times 10^3$ present method	Expt $\alpha_T \times 10^3$ based on models			Theo. α_T (elastic)
										Maxwell	L - J	Slieker	
He - Ar	380	300	340	8.76	5.64	17.47	0.0213		5.40	4.20	1.60	3.80	0.226
				21.63	15.00	127.87	0.0210		5.30	3.00	1.20	2.40	0.253
				30.14	23.98	412.46	0.0187	3.946	4.70	2.70	1.00	2.10	0.273
				49.30	59.57	1170.72	0.0275		7.10	3.60	1.40	2.90	0.327
Ne - Xe	380	300	340	64.46	77.39	1295.62	0.0340		8.60	7.20	2.50	5.80	0.340
				15.00	0.32	0.19	0.0115		2.90	4.20	4.50	3.40	0.247
				66.00	3.06	7.32	0.0178	3.946	4.50	7.30	7.50	5.40	0.218
Ne ²⁰ - Ne ²²	380	300	340	90.00	7.50	22.76	0.0249		6.30	13.70	15.50	11.00	0.236
				10.00	11.23	148.87	0.0146	3.946	3.70	5.40	1.70	4.40	0.0248

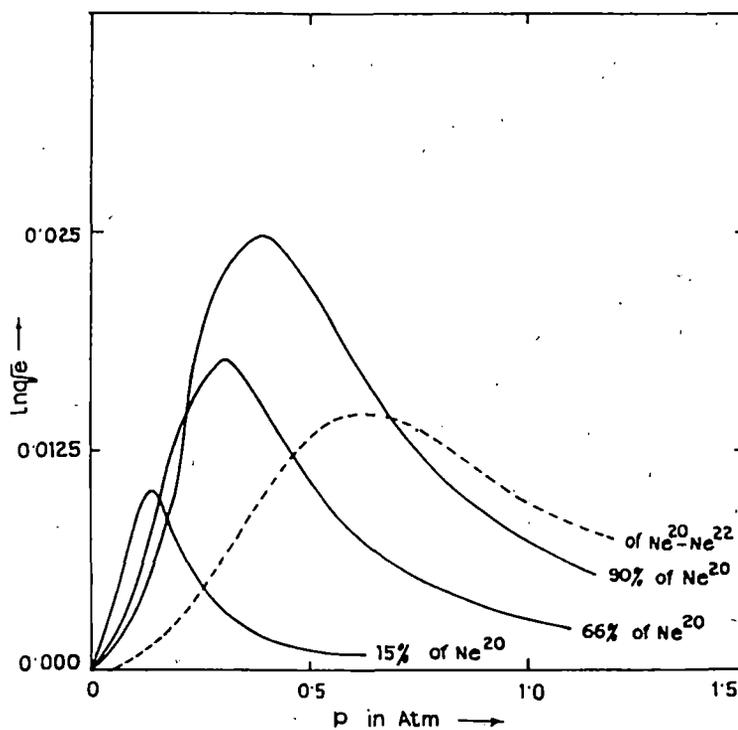


Fig. 5.3. Plot of $\ln q_e$ of $\text{Ne}^{20}\text{-Xe}^{132}$ (binary inert gas mixture) against pressure p in atm at different concentrations of Ne^{20} at temperature $\bar{T}=340$ K. The dotted curve represents that of $\text{Ne}^{20}\text{-Ne}^{22}$.

for the Ne^{20} - Ne^{22} mixture. These establish that the graph of $p^2/\ln q_e$ against p^4 is always a straight line as predicted by the FJO theory⁹⁾. Rutherford and Kaminski¹⁰⁾ have recently shown that the FJO theory⁹⁾ is applicable mainly in the case of an isotopic binary gas mixture and also in the case of a mixture in which one of the components is in trace concentration.

In the thermal diffusion phenomenon of isotopic gas mixtures, the elastic collisions are supposed to play a prominent role between the interacting molecules. Again, the thermal diffusion phenomenon is very sensitive to intermolecular interactions of the binary gas molecules, therefore, the study of the composition dependence of the TD factors α_T of any gas mixture is necessary to throw much light on the interactions between like molecules and those between unlike molecules of a gas mixture. The temperature dependence of α_T is, however, entirely governed by the factor $(6C_{ij}^* - 5)$, where C_{ij}^* is the ratio of the collision integrals. Moreover, the effect of inelastic collisions on the thermal diffusion, if any, is practically negligible in gas mixtures containing inert gases. All the facts mentioned above inspired us to study He^4 - Ar^{40} and Ne^{20} - Xe^{132} mixtures at different compositions of the lighter components at a given experimental mean temperature $\bar{T} = 340$ K and also the isotopic mixture Ne^{20} - Ne^{22} with its natural isotopic abundances. The purpose of this work is to observe how far the TD factors α_T as estimated by our column calibration factor method agree with those obtained by the existing methods using the column shape factors involved in the molecular models, and by Sliker's model independent method⁹⁾. The comparison is presented in Table 5.1 and shown graphically in Figs 5.4 and 5.5 for He^4 - Ar^{40} and Ne^{20} - Xe^{132} mixtures, respectively. The molecular parameters as well as the column shape factors used to calculate the experimental α_T by the existing method are shown in Table 5.2. The theoretical α_T 's as functions of molefractions of the lighter components are also calculated from the formula derived from the Chapman-Enskog kinetic theory¹¹⁾ based upon the elastic collisions between the interacting molecules. These are also shown in Table 5.1 and in Figs 5.4 and 5.5 for comparison.

5.2. Mathematical Formulations to Estimate the Experimental α_T 's of the Mixtures.

For a TD column with both ends closed, the hydrodynamical part of the column theory related with the transport of the lighter component up the tube, so far developed by Furry, Jones and Onsager⁹⁾ yields that the net transport of the *i*th component of a binary gas mixture up the column tube is zero, i.e.

$$\tau = Hc_i c_j - (K_c + K_d) \frac{\delta c_i}{\delta z} = 0. \quad \dots\dots\dots (5.3)$$

where *z* is the coordinate along the column, and *H*, *K_c* and *K_d* are called the transport coefficient, the convective remixing coefficient and the diffusive remixing coefficient respectively; they are complicated functions of the wall temperatures, the geometry of the column and also the transport properties of the binary gas mixture and are given by

$$H = \frac{2\pi}{Q^3} \int_{T_c}^{T_h} \frac{\rho D_{ij}}{\lambda} \alpha_T \frac{G(T)}{T} dT \quad \dots\dots\dots (5.4)$$

$$K_c = \frac{2\pi}{Q^7} \int_{T_c}^{T_h} \frac{\rho D_{ij}}{\lambda} G^2(T) dT, \quad \dots\dots\dots (5.5)$$

$$K_d = \frac{2\pi}{Q} \int_{T_c}^{T_h} r^2 \lambda \rho D_{ij} dT \quad \dots\dots\dots (5.6)$$

where ρ is the density, *r* is the radial coordinate, λ and *D_{ij}* are the thermal conductivity and diffusion coefficient of the gas mixtures respectively, *T_h* and *T_c* are the hot and cold wall temperatures, and $2\pi Q$ denotes the radial heat flow per unit length of the column. The function *G(T)* is the solution of the fourth-order differential equation

$$\frac{d}{dT} \left(\frac{1}{\lambda r^2} \right) \frac{d}{dT} \left(\frac{\eta}{\lambda} \right) \frac{d}{dT} \left(\frac{1}{\lambda \rho r^2} \right) \frac{d}{dT} \left\{ \frac{\rho D_{ij}}{\lambda} G(T) \right\} = g \frac{d\rho}{dT} \quad \dots\dots\dots (5.7)$$

with the boundary conditions

$$G(T_c) = G(T_h) = G'(T_c) = G'(T_h) = 0. \quad \dots\dots\dots (5.8)$$

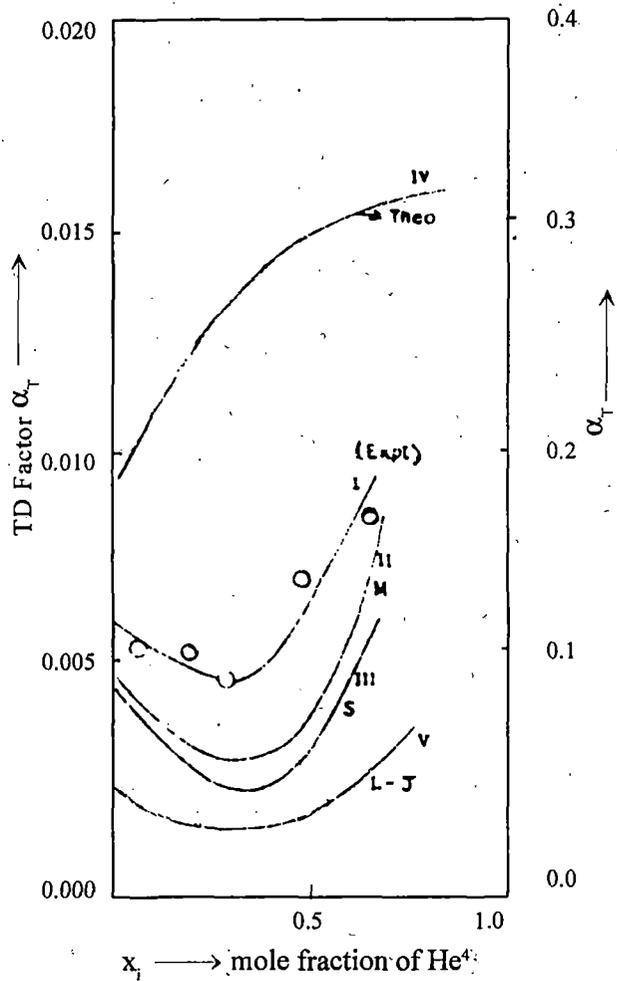


Fig. 5.4. Experimental α_T against mole fraction of He⁴ of He⁴-Ar⁴⁰ mixture at mean temperature $\bar{T}=340\text{K}$. I. Expt α_T (our calibration factor method), II. Expt α_T (Maxwell's model), III. Expt α_T (Slieker), IV. Expt α_T (L-J model).

Here η and g denote the coefficient of viscosity of the gas mixture and the acceleration due to gravity respectively.

Integrating eq (5.3) between the column ends $z=0$ and $z=L$ where L is the geometrical length of the column one gets

$$\ln q_e = \frac{HL}{K_c + K_d} \dots\dots\dots (5.9)$$

The coefficients H , K_c and K_d for an ideal column are proportional to the 2nd, the 4th and the 0th powers of the pressure p in atmosphere respectively for a binary gas mixture and hence eq (5.9) reduces to

$$\ln q_e = \frac{ap^2}{b+p^2} \dots\dots\dots (5.10)$$

However, in a practically constructed column, parasitic remixing of the components of the mixture always occurs. This can be taken into consideration by adding a term K_p , proportional to p^4 , in the denominator of eq (5.9), so that eq (5.9) becomes

$$\ln q_e = \frac{a'p^2}{b'+p^4}$$

or

$$p^2/\ln q_e = \frac{b'}{a'} + \frac{1}{a'} p^4 \dots\dots\dots (5.11)$$

The a' and b' in eq (5.11) are related to a and b in eq (5.10) by $a=a'(1 + K_p/K_c)$ and $b = b'(1 + K_p/K_c)$ respectively. The a and b in eq (5.10) again are expressed in terms of the column coefficients and the pressure of the binary gas mixture as

$$a = (HL/K_c)p^2 \text{ and } b=(K_d/K_c)p^4.$$

Further, since $(1 + K_p/K_c)=K_d/(b'K_c)$ we have

$$H=(a'K_d)/(b'L)=K_d/(LC), \dots\dots\dots(5.12)$$

where $C=b'/a'$ and is given by the intercept of the straight line expressing eq (5.11) for $p^2/\ln q_e$ against p^4 , demanded by the FJO column theory⁹⁾.

The possible values of H , K_c and K_d can be obtained by the following equations :

Table 5.2 : Column Shape factors and other molecular parameters used to calculate α_T of He³ - Ar⁴⁰, Ne²⁰ - Xe¹³² and Ne²⁰ - Ne²² mixtures at $\bar{T} = 340$ and the L - J potential r_{12} in erg.

Systems	(r_{12}/k) in K	σ_{12} in Å	Shape factors due to Maxwell's model		Slieker's shape factors	
			h'	k_d'	[S.F.] ₁	(1 - a ²)
He - Ar ⁴⁰	34.95	3.191				
Ne ²⁰ - Xe ¹³²	79.36	3.457	0.99425	0.97734	0.7041	1.764
Ne ²⁰ - Ne ²²	27.50	2.858			6!	
Lennard-Jones shape factors						
			h	k_c	k_d	
He ⁴ - Ar ⁴⁰	34.95	3.191	0.00750	0.00224	0.41875	
Ne ²⁰ - Xe ¹³²	79.36	3.457	0.00250	0.00180	0.40700	
Ne ²⁰ - Ne ²²	27.50	2.858	0.00975	0.00240	0.42340	

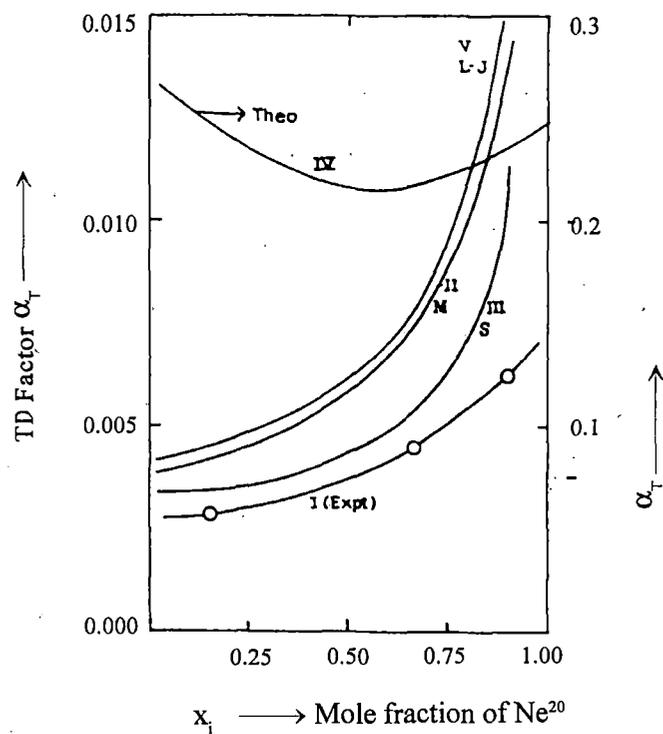


Fig. 5.5. Experimental α_T against mole fraction of Ne^{20} of $\text{Ne}^{20}\text{-Xe}^{132}$ mixture at mean temperature $T=340\text{K}$. I. Expt α_T (our calibration factor method), II. Expt α_T (Maxwell's model), III. Expt α_T (Slieker), IV. Theoretical α_T (elastic), V Expt α_T (L-J model).

i) Maxwell's⁸⁾ model of binary interaction (α_T being assumed to be temperature independent and $n=1$, for Maxwellian molecules).

$$H = \frac{2\pi}{6!} \left(\frac{\alpha_T \rho^2 g}{\eta} \right) \frac{1}{2} (r_c + r_h) (r_c - r_h)^3 (2u)^2 h'$$

$$K_c = \frac{2\pi}{9!} (\rho^3 g^2 / \eta^2 D)_1 \frac{1}{2} (r_c + r_h) (r_c - r_h)^7 (2u)^2 k'_c$$

$$K_d = 2\pi(\rho D)_1 \frac{1}{2} (r_c + r_h) (r_c - r_h) k'_d$$

ii) Sliker's⁹⁾ model-independent method.

$$C_{,1} = H = [S.F.]_1 r_c^4 \left(\frac{\rho^2 g \alpha_T}{\eta} \right) \left(\frac{\Delta T}{\bar{T}} \right)^2$$

$$C_2 = K_d = \pi(1-a^2) r_c^2 (\rho D)_1$$

$$C_3 = K_c = [SF]_3 r_c^8 (\rho^3 g^2 / \eta^2 D)_1 \left(\frac{\Delta T}{\bar{T}} \right)^2$$

iii) Lennard-Jones' molecular model of binary interaction,

$$H = \frac{2\pi}{6!} \left(\frac{\alpha_T \rho^2 g}{\eta} \right) r_c^4 h,$$

$$K_c = \frac{2\pi}{9!} (\rho^3 g^2 / \eta^2 D)_1 r_c^8 k_c,$$

$$K_d = 2\pi(\rho D)_1 r_c^2 k_d.$$

The dimensionless quantities h' , k'_c and k'_d , $[S.F.]_1$, $[S.F.]_3$ and $\pi(1 - a^2)$ and h, k_c , and k_d called the column shape factors due to Maxwell, Sliker and Lennard-Jones respectively are presented in Table 5.2, the factors in (ii) are free from a molecular interaction model, $u = (T_h - T_c) / (T_h + T_c)$, $a = r_h / r_c$ and $\Delta T = T_h - T_c$. The experimental α_T 's of He⁴ - Ar⁴⁰, Ne²⁰ - Xe¹³² and Ne²⁰-Ne²² can thus be evaluated by three methods, the former two at the mean temperature $\bar{T} = (T_h + T_c) / 2$ and the last one using L - J model at the cold wall temperature T_c of the column. Equation (5.12) is usually employed to calculate the experimental α_T 's in terms of H , $K_c L$ and the experimentally observed intercept $C (= b/a')$ for a given experimental temperature by the existing three methods. The experimental values, thus obtained, are presented in Table 5.1 and shown

graphically in Figs. 5.4 and 5.5 for comparison with the experimental α_T 's due to our method and the theoretical α_T 's also.

Now, the parameters a' and b' in eq. (5.10) influence the manner of variation of $\ln q_0$ for any binary gas mixture with the pressure at any experimental temperature. The graph of $\ln q_0$ against the pressure for He⁴-Ar⁴⁰ and Ne²⁰-Xe¹³² mixtures are shown in Figs 5.2 and 5.3 respectively, at different compositions of the lighter components, along with that for Ne²⁰-Ne²² isotopic mixture. As in most cases of experimental observation, Fig. 5.2 and 5.3 show that, as the pressure increase $\ln q_0$ increases for a given composition of the binary mixtures mentioned above and becomes maximum at the pressure $p=(b')^{1/4}$, where $\delta \ln q_0/\delta p = 0$. From eq.(5.11) the value $\ln q_0$ at the point of maximum is

$$\ln q_{max} = a'/2\sqrt{b'} \quad \dots\dots\dots (5.13)$$

The reliable value of $\ln q_{max}$ of the inert gas mixture of a certain composition and at a given temperature can thus be measured from eq (5.13), in terms of the experimental parameters a' and b' which are obtained by fitting the experimental data of $\ln q_0$ and pressure p in atmosphere. Using eqs (5.2) and (5.13) with the knowledge of the value of the column calibration factor, the TD factors α_T of the inert gas mixtures at different concentrations of the lighter components are then estimated, and shown in Table 5.1 and in Figs. 5.4 and 5.5.

5.3. Theoretical Formulations

The theoretical α_T 's can be calculated from

$$\alpha_T = \frac{1}{6[\lambda_i]_1} \frac{S^{(0)}x_i - S^{(0)}x_j}{[x_\lambda + y_\lambda]_1} (6C_{ij}^* - 5) \quad \dots\dots\dots (5.14)$$

where the symbols are defined by Chapman and Enskog⁽¹⁾. Here, $(6C_{ij}^*-5)$ depends strongly on the temperature alone, and the other factor in eq. (5.14) is not only a function of the temperature but also a complicated function of the compositions and the thermal conductivities of gases and gas mixtures. The α_T 's thus computed from eq (5.14) are presented in the last column of Table

5.1 and shown graphically in Figs 5.4 and 5.5 respectively, for the gas mixtures He⁴-Ar⁴⁰ and Ne²⁰-Xe¹³². A sample calculation was also made from

$$\alpha_{ij} = \frac{(6C_{ij}^* - 5)\mu_{ij}}{5nk[D_{ij}]} \left(\frac{\lambda_{j \text{ trans}}^\alpha}{x_j m_j} - \frac{\lambda_{i \text{ trans}}^\alpha}{x_i m_i} \right) \dots\dots\dots (5.15)$$

as deduced by Monchick, Munn and Mason¹²⁾ by assuming elastic collisions between the molecules and the results obtained are found to be much higher than those computed from eq (5.14).

5.4. Results and Discussions

The Chapman-Enskog kinetic theory of gases¹¹⁾ is strictly applicable only to spherically symmetric molecules like noble gases, and give a good account of the viscosity, diffusion and heat conductivity of gases so excellently that it has often been used with fair success to interpret the experimental thermal diffusion factors of isotopic mixtures of noble gases¹⁴⁾. But it has been found that for binary mixtures of spherically symmetric molecules the theory is quite unable to explain the composition dependence and probably the temperature dependence of the experimental thermal diffusion factors α_T for mixtures of different noble gases. We computed the theoretical α_T 's from eq (5.14) with the available force parameters as presented in Table 5.2 for the mixtures He⁴-Ar⁴⁰ and Ne²⁰-Xe¹³², the data thus obtained are shown in Table 5.1 and in Figs 5.4 and 5.5 only to see that they do not tally with the experimental ones computed by the present as well as the existing methods. The theoretical α_T 's are found to be two order higher in magnitude than the experimental α_T 's estimated while in the case of Ne²⁰-Ne²² isotopic mixture the theoretical α_T 's agree fairly well with the experimental ones as shown in Table 5.1. This is the reason why most of the authors in this field of research conclude that the FJO column theory is successful in predicting both the temperature and the composition dependence of the experimental α_T 's for isotopic mixtures as they are reproduced by the theoretical α_T 's as computed by the Chapman-Enskog theory of monatomic gases.

In the column theory, to estimate the experimental α_T 's for binary mixtures, molecular models such as Maxwells inverse fifth power and Lennard-Jones

12:6 potential model are commonly used. The method based on Lennard-Jones potential model is applicable to evaluate α_T at the cold wall temperature T_c of the column. The column shape factors and other molecular transport parameters also are required to be determined at the cold wall temperature. As evident from Table 5.1 the α_T 's values for He⁴-Ar⁴⁰ mixture due to the L-J molecular model shape factor as shown in Table 5.2 are very low compared with our α_T 's from the column calibration factor (F_s) method as well as with those due to Maxwell's molecular model and Slieker's model independent method. For He⁴-Ar⁴⁰ mixture, as the percentage of the lighter component, say He⁴, increases, the experimental α_T 's decrease gradually, attain the minimum values and then increase again as shown in Fig. 5.4. This sort of behaviour of the composition dependence of α_T 's for He⁴-Ar⁴⁰ could not be at all explained by the elastic theoretical α_T 's obtained from the Chapman-Enskog kinetic theory of monatomic gases. The magnitudes of the values are found to be two order higher than all the experimental α_T 's as observed in Table 5.1. The similar situation occurs in the case of Ne²⁰-Xe¹³² mixture; the theoretical α_T 's here also could not explain the composition dependence of the experimental α_T 's, which are also two-order less in magnitude than the theoretical ones, as observed from Table 5.1 and Fig. 5.5.

It is interesting to note that our experimental α_T 's due to the column calibration factor method, which is simple, straightforward and free from any binary molecular interaction model, is very close to those due to Slieker's method as well as the method using Maxwell's model, so far as the trends of variation of the α_T 's with mole fraction and their magnitudes are concerned. The existing method using L-J model yields α_T 's of less magnitude for He⁴-Ar⁴⁰ mixture, but in the case of Ne²⁰-Xe¹³² these α_T 's are of about the same magnitudes as the α_T 's due to Slieker's and our method.

Thus it is concluded that the present method, with the use of the column calibration factor F_s and the experimental $\ln q_{max}$ as obtained from the experimental parameters a' and b' , is not only applicable to isotopic gas mixtures, like the existing methods based on molecular models and Slieker's method, but is also a universal method to estimate the experimental α_T 's of

polyatomic gas mixtures. The temperature as well as the composition dependence of the actual and relatively small α_T 's of any binary gas mixture can thus be estimated by our present method. This study finally establishes that the column calibration factor F_s plays a significant role in column measurements. Thus it is wise to study a large number of columns of different r_c , r_h and L and \bar{T} to explore the functional relationship of F_s to r_c , r_h , L and \bar{T} , by choosing the interesting gas pairs forming binary mixtures, so that we can arrive at the unique formulation of the column calibration factor for a given column.

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