TEMPERATURE ASSIGNMENT TO THERMAL DIFFUSION FACTOR (*)

M. F. LARANJEIRA, A. MOUTINHO and M. H. VASCONCELOS

(Laboratório Calouste Gulbenkian de Espectrometria de Massa
Comissão de Estudos de Energia Nuclear (I. A. C.) — Instituto Superior Técnico — Lisbon)

SUMMARY — A new formula to approach the temperature dependence of the isotopic thermal diffusion factor is proposed.

It can be successfully applied to theoretical data of Lennard-Jones (12-6) and Exp.-Six models, as well as to the available observations with isotopes of *Ne*, *Ar*, *Kr*, and *Xe*. This enables us to make a reasonable comparison between theory and experiments, showing that the above models are not satisfactory to explain, simultaneously, thermal diffusion and viscosity data of the referred noble gases.

The assignment of a mean temperature associated with the experimental thermal diffusion factor is discussed, from the point of view of the formula proposed.

RÉSUMÉ — Une nouvelle formule pour approximer la dépendence du facteur de diffusion thermique isotopique est proposée.

Cette formule peut être appliquée aux resultats théoriques des modèles de Lennard-Jones (12-6) et Exp.-Six, aussi bien qu'aux observations avec les isotopes de *Ne*, *Ar*, *Kr* et *Xe*, ce qui nous permet d'établir une raisonnable comparaison entre la théorie et les expérimentations.

Les modèles cités ne sont pas satisfaisants pour expliquer simultanément la viscosité et la diffusion thermique dans les nobles gaz cités.

L'association d'une température moyenne au facteur de diffusion thermique expérimentale est discutée du point de vue de la formule proposée.

1. — INTRODUCTION

The experimental thermal diffusion factor determined by common methods, i. e. the two bulb apparatus ⁽¹⁾ and the swing apparatus ⁽²⁾, is indeed a temperature-average value, $\overline{\alpha_T (T)}$, which is given by the following equations, according to the usual definition (1):

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$$ln\, q = \int_{T_1}^{T_2} \alpha_T(T)\, d\, ln\, T = \overline{\alpha_T(T)}\, ln(T_2/T_1) \qquad (1)$$

where q is the measured separation factor of the elementary effect, and T_1 and T_2 are, respectively, the absolute temperatures of cold and hot regions of the apparatus.

If in a run of measurements of the separation factor both T^1 and T_2 are allowed to change, a mean temperature $\overline{T}$ has to be assigned to $\overline{\alpha_T(T)}$, such that $\overline{\alpha_T(T)} = \alpha_T(\overline{T})$, in order to make reliable comparisons between theoretical and experimental thermal diffusion factors.

Of course equations (1) must be fulfilled. Therefore, the expression for $\overline{T}$ as a function of T_1 and T_2, depends on the assumption of a temperature dependence of $\alpha_T(T)$. Such dependence is very complex, even in the simplest case of isotopic mixtures, so that it has been approximated, in practice, by various simple formulas (1, 3, 4, 5, 6, 7), namely

$$\alpha_T = a - b/T \quad ; \quad \text{ref. } 3 \quad ; \quad \overline{T}_B = \frac{T_1 T_2}{T_2 - T_1}\, ln(T_2/T_1) \qquad (2)$$

$$\alpha_T = a\, ln(T/b) \quad ; \quad \text{ref. } 4{,}5 \quad ; \quad \overline{T}_D = (T_1 T_2)^{1/2} \qquad (3)$$

$$\alpha_T = a + bT \quad ; \quad \text{ref. } 6 \quad ; \quad \overline{T}_P = \frac{T_2 - T_1}{ln\,(T_2/T_1)} \qquad (4)$$

$$\alpha_T = a - b/T^2 \quad ; \quad \text{ref. } 7 \quad ; \quad \overline{T}_L = T_1 T_2 \left[\frac{2\, ln(T_2/T_1)}{T_2^2 - T_1^2}\right]^{1/2} \qquad (5)$$

In the foregoing formulas, a and b are constants. We have

$$\overline{T}_D^2 = \overline{T}_B\, \overline{T}_P = \overline{T}_L^2\, (\overline{T}_m / \overline{T}_B), \text{ with } \overline{T}_m = (T_1 + T_2)/2 \qquad (6)$$

The formula (2) for the mean temperature $\overline{T}_B$, has frequently been used, also in cases where the corresponding temperature dependence for α_T has not been stated or, even worse, not experimentally observed (8).

Temperature dependence expressed by eq. (5) has been applied by LONSDALE and MASON (7) to fit experimental data but no reference has been given to the corresponding mean temperature, $\overline{T}_L$, which we have determined. MORAN and WATSON (9) seem to assign the arithmetic mean temperature, $\overline{T}_m$, to the experimental isotopic thermal diffusion factor for the noble gases. This may be safely used only if α_T is practically constant ⁽¹⁰⁾.

The differences among mean temperatures defined above, are by no means negligible (see table V) taking in mind the temperature stabilisations which are achieved in experimental apparatus. Therefore, it seems to us that the problem of a temperature assignment has frequently been over simplified.

Dependences expressed by equations 2, 3, 4 and 5, are indeed too much simple to fit thermal diffusion data, so that they may only be applied to rather short temperature ranges.

The problem of choosing a mean temperature may be avoided in theory, if one of the temperatures — say T_1 of the cold region of the apparatus — is kept constant over the entire run of measurements of the separation factor, q. From eq. (1) we have, then:

$$\frac{d\,ln\,q}{d\,ln\,T_2} = \alpha_T\,(T_2) \tag{7}$$

Therefore, the thermal diffusion factor at any temperature T_2, is given by the slope of the curve $ln\,q$ against $ln\,T_2$ or, as usually done in practice, against $ln\,(T_2/T_1)$ using log-log scales ⁽¹¹, ¹²⁾.

Besides the experimental requirement of a constant temperature, the slope method involves the disadvantage of a graphical differentiation which in practice gives rise to appreciable loss of accuracy specially at the ends of the curve.

In this paper another formula for the temperature dependence of α_T is proposed. Though not so simple as eqs. (2-5), it can be rather easily handled, having the advantage of covering either theoretical or experimental thermal diffusion data over large temperature ranges.

For the sake of simplicity, only binary isotopic mixtures will be considered. In this case the thermal diffusion factor, α_T, is theoretically given by

$$\alpha_T = \alpha_0\,\frac{m_1 - m_2}{m_1 + m_2} \tag{8}$$

when m_1 and m_2 are the molecular masses of the isotopes and α_0, the so called reduced isotopic thermal diffusion factor, is an universal function of temperature — for each intermolecular model — which expresses the assumption of corresponding states.

Our formula may, however, be also applied to non-isotopic mixtures by an immediate generalization, as we shortly show.

2 — PROPOSED FORMULA. FITTING OF THEORETICAL DATA

Any reliable equation to approach the temperature dependence of experimental data of the reduced isotopic thermal diffusion factor defined by eq. (8), should have a behaviour similar to that predicted by «realistic» theoretical models as, for example, the well known Lennard-Jones and Exponential-Six potentials.

Having this in mind, we first show that our formula

$$\alpha_0 = a - b\ e^{-c\ ln^2 (T^*/\delta)} \tag{9}$$

where a, b, c and δ are constants and $T^* = kT/\varepsilon$ is the reduced temperature ($k \equiv$ Boltzmann's constant; $\varepsilon \equiv$ depth of potential minimum), reproduces appreciably well the theoretical values of α_0, as given by L. J. (12-6) and Exp. - Six potentials, over a very wide temperature range.

In spite of its form, equation (9) can rather easily be handled. The constants a, b, c and δ have the following simple meanings:

— a is the value of α_0 at very high temperatures ($T^* \rightarrow \infty$);
— $(a\text{-}b)$ is the minimum value of α_0, which occurs at $T^* = \delta$;
— b is the difference between maximum and minimum values of α_0;
— c is the slope of the straight line $ln\,(a - \alpha_0)$ againts $ln^2\,(T^*/\delta)$:

$$ln\,(a - \alpha_0) = ln\,b - c\,ln^2\,(T^*/\delta) \tag{10}$$

In table I we give the values of a, b, a-b, c and δ for L. J. (12-6) and Exp.-Six potentials with the parameter $\alpha = 12, 13, 14, 15, 16, 17$.

TABLE I

Values of the constants in Eq. (9) *for L. J.* (12-6) *and Exp.*- 6 *Models*

Model	a	b	a-b	c	δ
L. J. (12-6)	0.575	0.631	— 0.056	0.515	0.62
Exp.-6, $\alpha = 12$	0.445	0.550	— 0.105	0.61	0.65
13	0.483	0.547	— 0.064	0.62	0.66
14	0.518	0.545	— 0.027	0.58	0.66
15	0.540	0.540	0.000	0.58	0.66
16	0.564	0.533	+ 0.031	0.56	0.66
17	0.590	0.527	+ 0.063	0.55	0.66

For the reduced isotopic thermal diffusion factor we have used Kihara's first approximation

$$[\alpha_0]_1^K = \frac{15}{16}\,\frac{6\,C^* - 5}{A^*} \tag{11}$$

which is easier to handle and almost as good as the very complex Chapman's second approximation (13). The numerical tables of $[\alpha_0]_1^K$ for the Exp.-Six model with $\alpha = 16{,}17$ were obtained from Mason and Rice (14).

Values of $[\alpha_0]_1^K$ for L. J. (12-6) and Exp.-Six with $\alpha = 12$, 13, 14, 15, have been computed by using numerical data of A^* and C^* given by HIRSCHFELDER *et al.* (15).

In table II we compare theoretical and approximated values (eq. 9) of $[\alpha_0]_1^K$ for L. J. (12-6) and Exp.-Six with $\alpha = 12$, 13, 14, 15. As a whole, the agreement is satisfactory (and also for the values of $\alpha = 16$, 17 which have not been presented in the table for the sake of brevity). For the case of the Exp.-Six model the agreement

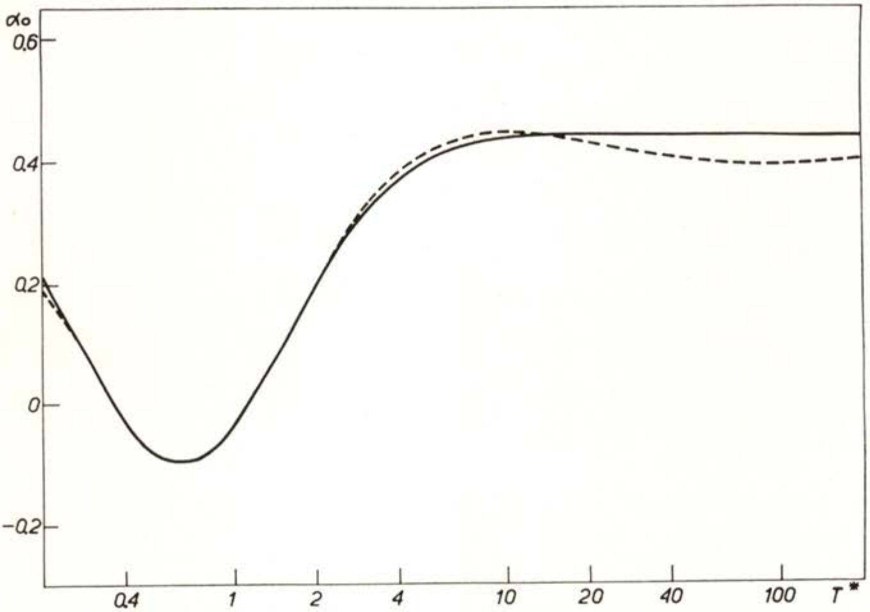

Fig. 1 — Theoretical and approximated curves for $[\alpha_0]_1^K$

---- Th. — Exp.-Six with $\alpha = 12$.

—— App. — Our equation (9) with $a=0.445$, $b = 0.550$, $c = 0.61$ and $\delta = 0.65$.

is not so good at high temperatures, especially for $\alpha = 12$. This is due to the fact that $[\alpha_0]_1^K$ first decreases with the reduced temperature, T^*, in the range 10-100 and then increases again, whereas according to our equation (9) the reduced thermal diffusion factor is a monotonic slowly increasing function of T^* in that range (see fig. 1, for $\alpha = 12$).

3 — APPLICATION TO EXPERIMENTS

3.1 — *Procedure*

To fit experimental data of the reduced isotopic thermal diffusion factor we write equation (9) in the convenient form:

$$\alpha_0 = a - b\ e^{-c \ln^2 [T/(T_c d)]} \tag{12}$$

where T_c is the critical temperature of the isotopic mixture and d is a new constant. The formal identification is therefore made: $\delta/k \equiv T_c\, d$.

TABLE II

Exact and Approximated values of $[\alpha_0]_1^K$

T^*	L. J. (12-6)		Exponential-Six							
			$\alpha = 12$		$\alpha = 13$		$\alpha = 14$		$\alpha = 15$	
	Exact.	Approx.	Exact.	Approx.	Exact.	Approx.	Exact.	Approx.	Exact.	Approx.
0.3	0.076	0.094	0.068	0.063	0.104	0.111	0.130	0.137	0.147	0.164
0.5	-0.042	-0.041	-0.082	-0.082	-0.039	-0.039	-0.004	-0.004	0.022	0.024
0.6	-0.056	-0.056	-0.103	-0.103	-0.061	-0.061	-0.025	-0.025	0.002	0.005
0.7	-0.051	-0.051	-0.104	-0.103	-0.063	-0.063	-0.027	-0.027	0.000	0.001
0.9	-0.009	-0.012	-0.071	-0.071	-0.033	-0.032	0.002	0.002	0.029	0.029
1.2	0.072	0.071	0.007	0.008	0.043	0.044	0.076	0.074	0.101	0.101
1.8	0.223	0.224	0.153	0.153	0.186	0.190	0.215	0.213	0.238	0.239
2.5	0.339	0.343	0.264	0.263	0.295	0.301	0.321	0.323	0.345	0.347
3.0	0.400	0.400	0.315	0.313	0.346	0.351	0.371	0.373	0.394	0.397
3.5	0.436	0.440	0.351	0.347	0.382	0.386	0.407	0.409	0.430	0.433
5	0.503	0.508	0.410	0.402	0.442	0.440	0.465	0.467	0.490	0.490
7	0.543	0.544	0.436	0.428	0.472	0.466	0.498	0.496	0.521	0.519
10	0.566	0.563	0.445	0.439	0.482	0.477	0.511	0.510	0.535	0.533
20	0.574	0.574	0.429	0.445	0.472	0.483	0.507	0.517	0.536	0.539
40	0.574	0.575	0.407	0.450	0.455	0.483	0.495	0.518	0.531	0.540

The use of T/T_c as a pratical reduced temperature, has an obvious advantage. If the assumption of corresponding states was followed by experiments, then a unic set of constants a, b, c, and d might be observed for $\overline{\alpha}_0$.

According to the temperature dependence expressed by eq. (12), the mean temperature, $\overline{T}$, associated with the experimental value α_0 is given by

$$e^{-c\, ln^2 (\overline{T}/T_c d)} = \frac{(\pi/c)^{1/2}}{ln\,(T_2/T_1)} (\psi_2 - \psi_1) \qquad (13)$$

with

$$\psi_i = Erf\ t_i = \frac{1}{\sqrt{2\pi}} \int_o^{t_i} e^{-z^2/2}\, dz$$

and

$$t_i = \sqrt{2c}\ ln \left(\frac{T_i}{T_c d} \right)$$

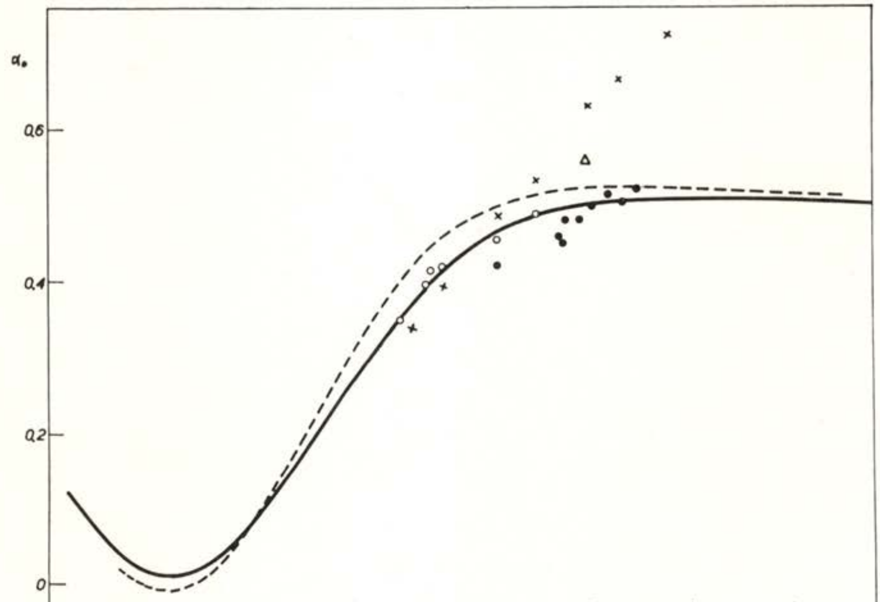

Fig. 2 — Reduced isotopic thermal diffusion factor for ^{20}Ne-^{22}Ne.

× — STIERS, ref. 8 (1942).
● — MORAN and WATSON, ref. 16 (1958).
△ — LARANJEIRA and KISTEMAKER, ref. 17 (1960).
○ — WATSON ET AL., ref. 18 (1963).
—— Experimental smooth curve.
- - - - Exp.-6, $\alpha = 14.5$, $\varepsilon/k = 38.0$ °K

In order to obtain $\overline{T}$ and the most probable values of a, b, c and d, we have adopted the following procedure:

1 — As a first approximation we take $\overline{T} \sim \overline{T}_D = (T_1\, T_2)^{1/2}$. This is nearly correct for observations lying in the region where formula (12) is roughly linear in $ln\overline{T}$ (see fig. 1 and confront eq. (3)).

2 — Experimental data of $\bar{\alpha}_0$ is plotted against $ln\,(\overline{T}/T_c)$, using transparent paper and semi-log scales.

3 — The above plot is shifted over the family of curves

$$y = b\;(1 - e^{-c\; ln^2 x})$$

drawn in identical scales of step 2, with b and c of the same order of magnitude of those given in table I.

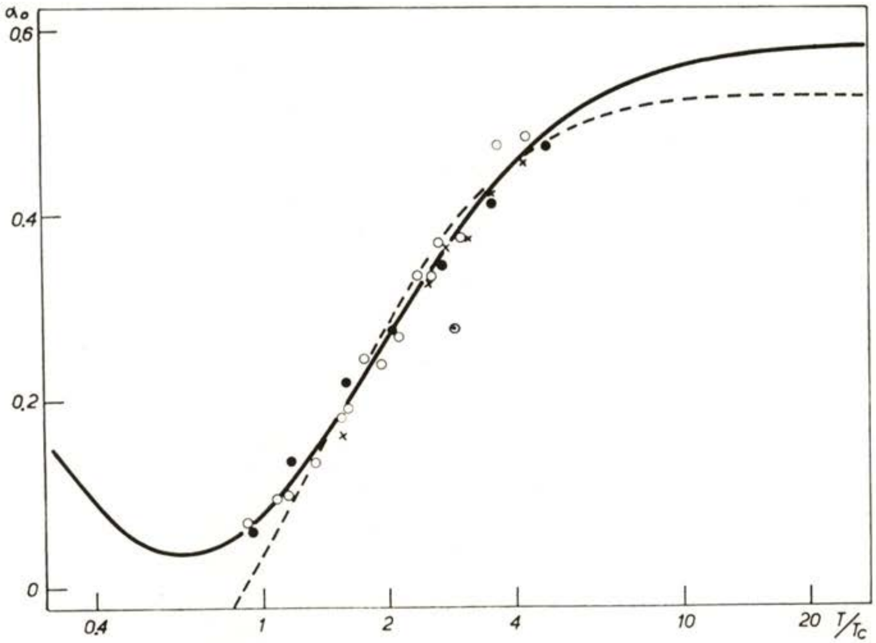

Fig. 3 — Reduced isotopic thermal diffusion factor for ^{36}Ar-^{40}Ar.

● — STIERS, ref. 8 (1942).
× — MORAN and WATSON, ref. 16 (1958).
⊙ — LARANJEIRA and KISTEMAKER, ref. 17 (1960).
○ — PAUL *et al.*, ref. 19 (1963).
—— Experimental smooth curve.
---- L. J. (12-6) ε/k=124°K.

The curve of the family which gives, by superposition, the best fit for experimental data, automatically determines b and c. Also, the point $(ln\,1{,}0)$ of this curve «reads» in the transparent plot the point $(ln\,d, a - b)$.

4 — To confirm the values of the parameters determined by step 3, we plot $ln\,(a - \alpha_0)$ against $ln^2\,[\overline{T}/(T_c\,d)]$.
According to eq. (12), a straight line should be obtained having a slope equal to c.

5 — By the foregoing procedure, first approximations for $\overline{T}$, and a, b, c and d are determined.

Second approximation may be obtained as follows:

a) for $\overline{T}$, by means of equation (13). This will be necessary especially for observations lying outside the «linear portion» of the curve $\bar{\alpha}_0$ against $ln\ \overline{T}/T_c$.

b) for parameters a, b, c and d, by using now the second approximation for the mean temperature $\overline{T}$. This may be done either graphically or by algebraic methods.

Higher approximations might be obtained in the same way. We have observed that second approximation is satisfactory since the method is quickly convergent.

3.2 — *Fitting of Experimental Data for Ne, Ar, Kr, and Xe*

The proposed method has been applied to fit the available data of reduced thermal diffusion factors for isotopic mixtures of neon (8, 9, 16, 17), argon (8, 16, 17, 19), krypton (9) and xenon (9).

Plots of $\bar{\alpha}_0$ against $ln\,(\overline{T}/T_c)$ are shown in figs. 2, 3, 4 and 5. Theoretical curves which appear to give the best fit of experiments, by using values of ε/k from viscosity data, are also drawn in the figures.

Values of the critical temperature, T_c, and of ε/k which has been used in the computations are given in table III.

TABLE III

Values of ε/k from viscosity and of T_c

Gas	ε / k (°K)	Model	Ref.	T_c (°K)	Ref.
Neon	38.0	Exp.-6 $\alpha = 14.5$	14	44.5	15
Argon	124	L. J. (12-6)	15,19	151	15
Krypton	212	Exp.-6 $\alpha = 14$	20	210	21
Xenon	231.2	Exp.-6 $\alpha = 13$	14	289.81	15

Determinations of $\overline{T}$, and of the constants *a*, *b*, *c* and *d* given in table IV, has been performed up to the second approximation as explained in § 3.1.

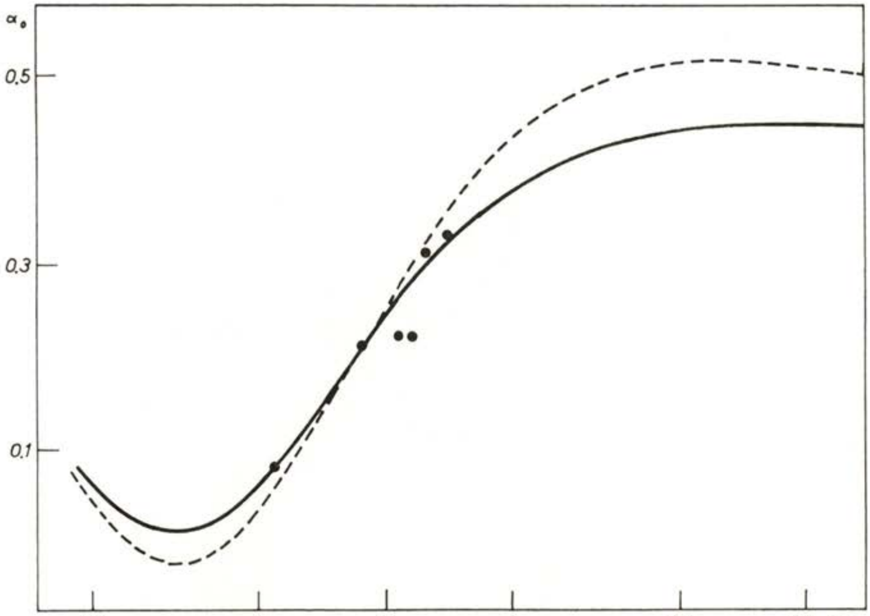

Fig. 4 — Reduced isotopic thermal diffusion factor for ^{80}Kr, ^{82}Kr, ^{83}Kr, ^{84}Kr and ^{86}Kr.

● — MORAN and WATSON, ref. 16, (1958).
—— Experimental smooth curve.
- - - - Exp.-6, $\alpha = 14$, ε/k = 212°K.

Thermal diffusion data of argon provides a good example of the application of the method. Observations have been carried out over large temperature ranges and the scattering of results of different authors appears to be of the order of experimental errors.

TABLE IV

Mean values of parameters in our equation 12

Gas	*a*	*b*	*a-b*	*c*	*d*
Neon	0.52	0.50	0.02	0.50	0.60
Argon	0.580	0.543	0.027	0.45	0.66
Krypton	0.442	0.439	0.003	0.50	0.60
Xenon	0.455	0.403	0.052	0.50	0.56

In table v we give a representative set of experimental data for the reduced isotopic thermal diffusion factor, $\bar{\alpha}_0$, and the corresponding smooth values and reduced mean temperatures, $\bar{T}/T_c$, de-

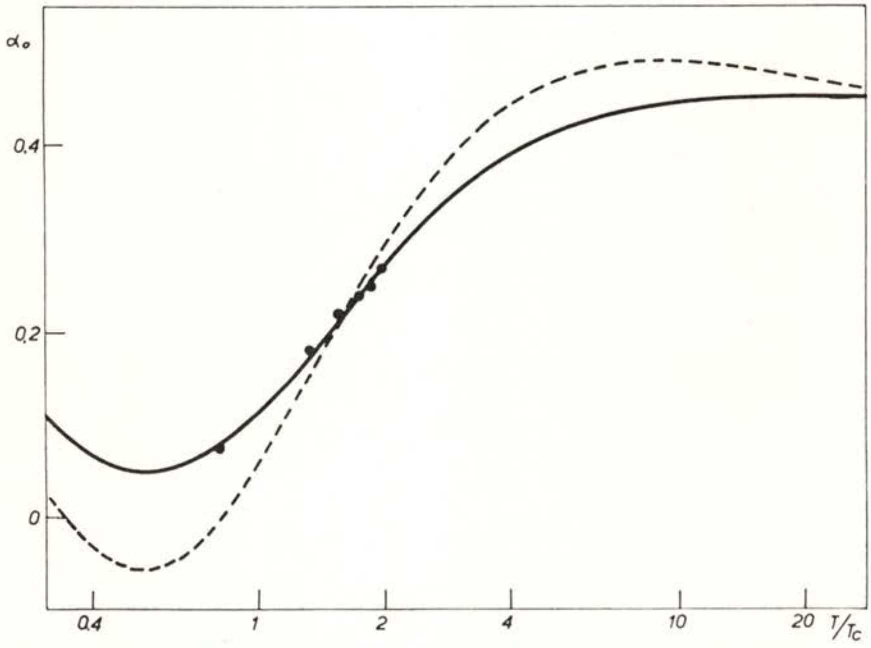

Fig. 5 — Reduced isotopic thermal diffusion factor for ^{128}Xe, ^{129}Xe, ^{130}Xe, ^{131}Xe, ^{132}Xe, ^{134}Xe and ^{136}Xe.

● — MORAN and WATSON, ref. 16, (1958).
—— Experimental smooth curve.
- - - - Exp.-6 $\alpha = 13$; $\varepsilon/k = 231,2$°K.

TABLE V

Experimental data for ^{36}A- ^{40}A and temperature assignement

Reduced T. D. Factor $\bar{\alpha}_0$					Reduced mean temperature. $T_c = 151^o$ K				
Experimental	smooth, this work eq. (12)	T_2 (^{o} K)	T_1 (^{o} K)	Ref.	$\bar{T}_L / T_c$ Lonsdale (eq. 5)	$\bar{T}_B / T_c$ Brown (eq. 2)	$\bar{T}_P / T_c$ Paul (eq. 4)	$\bar{T}_D / T_c$ Davenport (eq. 3)	$\bar{T} / T_c$ This work (eq. 13)
0.068	0.063	195	77	18	0.756	0.783	0.841	0.811	0.920
0.095	0.095	273	77	18	0.846	0.899	1.026	0.960	1.091
0.098	0.106	303	77	18	0.872	0.936	1.093	1.011	1.149
0.133	0.145	417	77	18	0.954	1.056	1.333	1.187	1.336
0.182	0.185	273	195	18	1.514	1.521	1.535	1.528	1.531
0.220	0.197	296	195	8	1.568	1.580	1.603	1.591	1.595
0.245	0.221	346	195	18	1.674	1.697	1.744	1.720	1.723
0.240	0.255	435	195	18	1.830	1.878	1.981	1.929	1.923
0.270	0.284	373	273	18	2.096	2.105	2.122	2.113	2.115
0.336	0.316	463	273	18	2.300	2.327	2.382	2.354	2.342
0.329	0.339	416	351	9	2.525	2.528	2.534	2.531	2.536
0.371	0.351	527	307	18	2.601	2.632	2.696	2.664	2.644
0.377	0.386	594	353	18	2.966	2.999	3.067	3.033	3.003
0.414	0.427	635	455	8	3.528	3.544	3.576	3.560	3.537
0.485	0.468	725	585	18	4.297	4.305	4.319	4.313	4.297
0.475	0.488	835	638	8	4.805	4.819	4.848	4.834	4.816

termined, respectively, by our proposed formulas (12) and (13). For comparison, we also present in the table the values of reduced mean temperatures $\overline{T}_L/T_c$ (LONSDALE and this work, eq. 5), $\overline{T}_B/T_c$ (BROWN, eq. 2), $\overline{T}_P/T_c$ (PAUL, eq. 4) and $\overline{T}_D/T_c$ (DAVENPORT, eq. 3).

In fig. 6 we present the plot of $ln(a-\bar{\alpha}_0)$ against $ln^2(\overline{T}/T_c d)$ which, as stated in § 3.1, should be linear and gives a simple graphical confirmation of the goodness of the values determined for parameters a, b, c and d.

4 — DISCUSSION

As it has been shown, our proposed equations (9) and (12) for approaching the temperature dependence of the reduced isotopic thermal diffusion factor, α_0, may be used with confidence over large

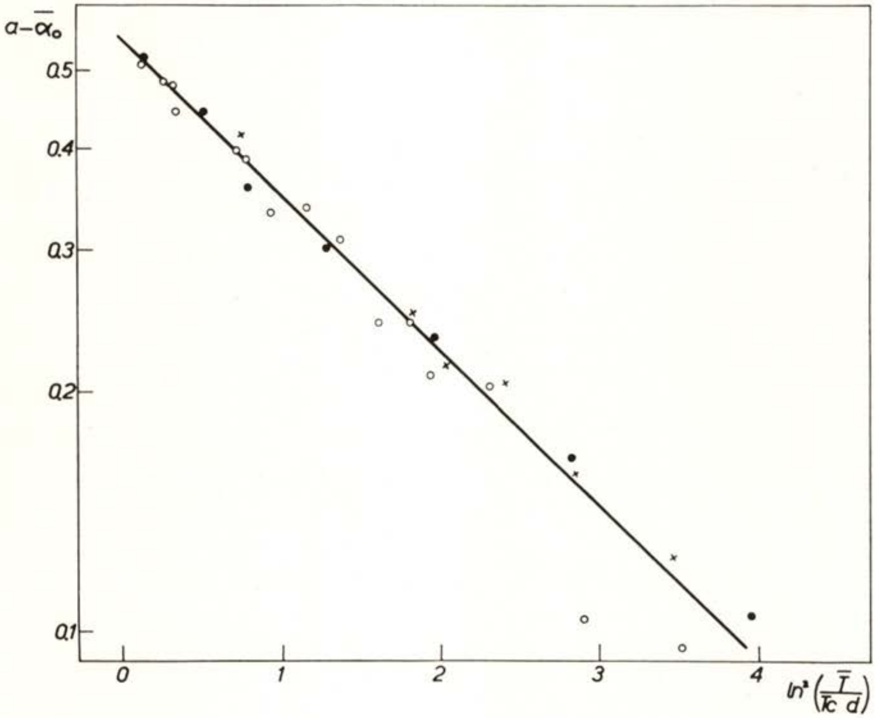

Fig. 6 — Plot of $a-\bar{\alpha}_0$ against $ln^2\left(\frac{\overline{T}}{T_c d}\right)$ for $^{36}A-^{40}A$

● — STIERS, ref. 8, (1942).
× — MORAN and WATSON, ref. 16, (1958).
○ — PAUL, ET AL., ref. 19, (1963).

temperature ranges for fitting either theoretical or experimental data. This enables us to make reliable comparisons.

Therefore, concerning the problem of a mean temperature definition, our value for $\overline{T}$, as given by formula (13), may be safely assigned to experimental data of α_0. As table v shows, the difference of our value, $\overline{T}$, to those previously proposed, $\overline{T}_B$, $\overline{T}_P$ and $\overline{T}_D$, are in general not negligible. Therefore, they may be applied only in restrict temperature ranges, as first approximations: at low temperatures, Paul's formula 4 for $\overline{T}_P$ is preferable; for intermediate temperatures, Davenport's formula (3) for $\overline{T}_D$; and, at higher temperatures, Brown's formula (2) for $\overline{T}_B$.

As it is well known, thermal diffusion is very sensitive to the nature of intermolecular potentials and even the best models available give rather poor agreement with observations. The set of potentials parameters which fits other transport phenomena — like viscosity, concentration diffusion and thermal conductivity — are frequently inadequated to properly explain thermal diffusion. From

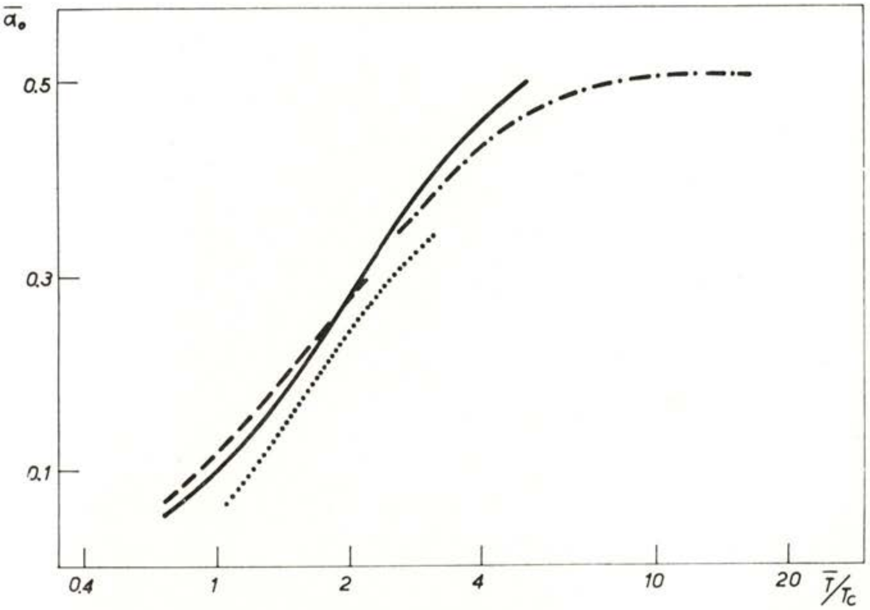

Fig. 7 — Experimental smooth curve of $\overline{\alpha}_0$ for isotopes of *Ne*, *Ar*, *Kr*, and *Xe*, in the temperature ranges observed.

-·-·- ^{20}Ne — ^{22}Ne.
—— ^{36}Ar — ^{40}Ar.
····· ^{80}Kr, ^{82}Kr, ^{83}Kr, ^{84}Kr and ^{86}Kr.
----- ^{128}Xe, ^{129}Xe, ^{130}Xe, ^{131}Xe, ^{132}Xe, ^{134}Xe and ^{136}Xe.

figs. 2, 3, 4 and 5, it follows that: (I) the experimental difference $(\alpha_0)_{Max} - (\alpha_0)_{min} = b$ is always smaller than the corresponding theoretical value; (II) the slope of the curve $\bar{\alpha}_0$ against $ln(\overline{T}/T_c)$ in the intermediate temperature range from $\overline{T}/T_c = 1$ to 4, is also smaller than in theory; (III) the value of $(\alpha_0)_{min} = a\text{-}b$ appears to be always positive for the noble gases considered.

The use of T/T_c as reduced temperature gives rise to somewhat similar trend for thermal diffusion data of *Ne*, *Ar*, *Kr* and *Xe*, but we may not state that the assumption of corresponding states is followed, as fig. 7 shows. Nevertheless, it is interesting to point out that, by making small translations in the $ln(T/T_c)$ direction, the experimental smooth curves are more or less overlapped in the temperature ranges observed.

The method described in this paper may be also applied to non-isotopic mixtures, in principle. Indeed, in this case, the temperature dependence of the thermal diffusion factor is mostly expressed by the quantity $6C^*\text{-}5$, being therefore roughly identical to that of Kihara's first approximation, $[\alpha_0]_1^K$.

REFERENCES

(1) GREW, K. E. and IBBS, T. L. — *Thermal Diffusion in Gases*. Cambridge University Press, (1952).

(2) CLUSIUS, K. and HUBER, M. — *Z. Naturforsch.*, **10 a**, 230 (1955).

(3) BROWN, H. — *Phys. Rev.*, **58**, 661 (1940).

(4) DAVENPORT, A. N. and WINTER, E. R. S. — *Trans. Faraday Soc.*, **47**, 1160 (1951).

(5) HOLLERAN, E. M. — *J. Chem. Phys.*, **21**, 1901 (1953).

(6) PAUL, R., HOWARD, A. J. and WATSON, W. W. — *J. Chem. Phys.*, **39**, 3053 (1963).

(7) LONSDALE, H. K. and MASON, E. A. — *J. Phys. Chem.*, **61**, 1544 (1957).

(8) STIER, L. G. — *Phys. Rev.*, **62**, 548 (1942).

(9) MORAN, T. I. and WATSON, W. W. — *Phys. Rev.*, **109**, 1184 (1958).

(10) VAN DER VALK, F. and DE VRIES, A. E. — *Physica*, **29**, 427 (1963).

(11) GHOZLAN, A. I. — *Thermal Diffusion for Quasi-Lorentzian Mixtures between* 10° *and* 800° *K*. Thesis, Leiden (1963).

(12) SLIEKER, C. J. G. — *Thermal Diffusion of Asymetric Molecules*. Thesis, Delft (1964).

(13) MASON, E. A. — *J. Chem. Phys.*, **22**, 169 (1954).

(14) MASON, E. A. and RICE, W. E. — *J. Chem. Phys.*, **22**, 843 (1954).

(15) HIRSCHFELDER, J. O., CURTISS, C. F. and BIRD, R. B. — *Molecular Theory of Gases and Liquids* (1954).

(16) LARANJEIRA, M. F. and KISTEMAKER, J. — *Physica*, **26**, 431 (1960).

(17) WATSON, W. W., HOWARD, A. J., MILLER, N. E. and SHIFFRIN, R. M. — *Z. Naturforsch*, **18 a**, 242 (1963).

(18) PAUL, R., HOWARD, A. J. and WATSON, W. W. — *J. Chem. Phys.*, **39**, 3053 (1963).

(19) JOHNSTON, H. L. and GRILLY, E. R. — *J. Phys. Chem.*, **46**, 948 (1942).

(20) MASON, E. A. — *J. Chem. Phys.*, **32**, 1832 (1960).

(21) *Handbook of Chemistry and Physics*, Chemical Rubber Publishing Co. Cleveland, Ohio, 36th ed. 1954-55.