

## ISOTOPIC THERMAL DIFFUSION IN NEON

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*SUMMARY* — Isotopic thermal diffusion in neon was observed in the temperature range from 335.6 up to 507.4 °K, which provides a sensitive way to determine the parameter  $\alpha$  for the exponential-six model.

According to the values obtained and the viscosity data available, a new set of parameters,  $\varepsilon/h = 33.4$  °K,  $r_m = 3.144$  Å and  $\alpha = 15.5$ , is proposed for neon.

### 1 — INTRODUCTION

Considerable efforts have been made in recent years to study intermolecular forces by means of transport phenomena, namely viscosity, thermal conductivity, diffusion and thermal diffusion.

From those properties, thermal diffusion has been considered as the most sensitive one to the nature of intermolecular potentials. However, some restrictions have to be pointed out to that common statement if one has to deal with a somewhat sophisticated potential model. Indeed, let us consider the case of the exponential-six potential which is now believed to fit reasonably well the experimental data of transport phenomena:

$$\phi(r) = \frac{\varepsilon}{1 - 6/\alpha} \left\{ (6/\alpha) \exp[\alpha(1 - r/r_m)] - (r_m/r)^6 \right\}$$

$$\text{for } r > r_{max}$$

$$\text{and } \phi(r) = \infty \text{ for } r \leq r_{max}$$

where

- $\phi(r)$  — potential at a distance  $r$  from the center of the molecule;
- $r_{max}$  — value of  $r$  for which  $\phi(r)$  has a «spurious» maximum;
- $\varepsilon$  — depth of the potential well;
- $r_m$  — value of  $r$  for which  $\phi(r)$  is minimum;
- $\alpha$  — additional parameter depending on the slope of the repulsive part of the potential.

The exp.-six potential has therefore three parameters,  $\varepsilon/k$  ( $k$  — Boltzmann's constant),  $r_m$ , and  $\alpha$ , which have to be chosen according to experimental data. Isotopic thermal diffusion depends only on  $\varepsilon/k$  and  $\alpha$ ; viscosity, thermal conductivity and self-diffusion depend, furthermore, on parameter  $r_m$ .

Let us consider the kind of information which isotopic thermal diffusion may provide about  $\varepsilon/k$  and  $\alpha$ . In fig. 1 we present the family of curves for the reduced isotopic thermal diffusion factor, according

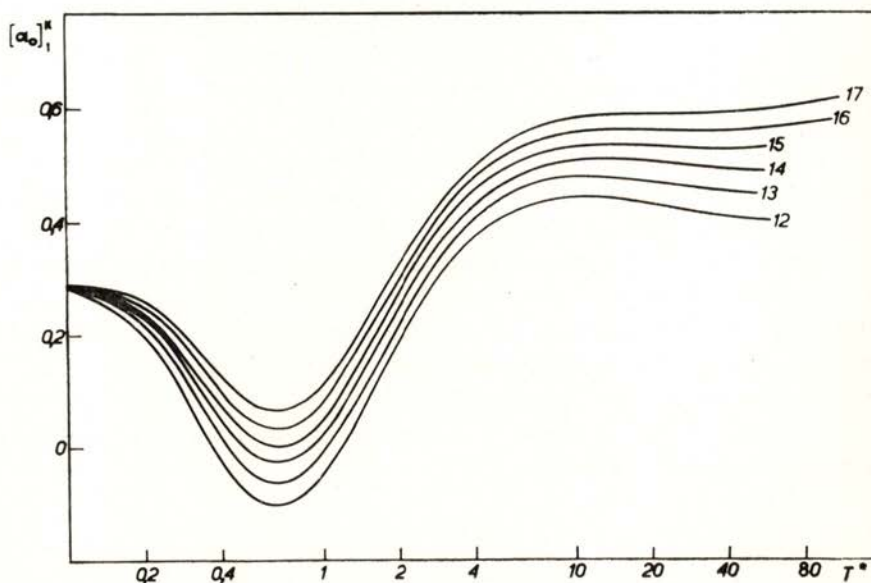


Fig. 1 — Kihara's first approximation for the reduced isotopic thermal diffusion factor,  $[\alpha_0]_1^K$ , for the exp.-6 model with parameter  $\alpha$  equals to 12, 13, 14, 15, 16 and 17.

to the first Kihara's approximation,  $[\alpha_0]_1^K$ , against the reduced absolute temperature  $T^* = T/(\epsilon/k)$ , for values of the potential parameter  $\alpha$  equal to 12, 13, 14, 15, 16 and 17.

In those curves we may consider three temperature ranges:

1 —  $T^*$  below 1. This range is experimentally difficult since  $\epsilon/k$  is of the order of the critical temperature for most gases and some spurious effects may interfere with thermal diffusion.

2 —  $T^*$  between 1 and 4. As fig. 1 shows, the different curves of  $[\alpha_0]_1^K$  are practically parallel to each other. Since parameters  $\epsilon/k$  and  $\alpha$  are obtained by shifting experimental data of the reduced isotopic thermal diffusion factor over the theoretical family of curves, those parameters remain undetermined, bearing in mind the magnitude usually reported for experimental errors.

3 —  $T^*$  higher than 4. In this case, if experimental values are available over a sufficient temperature range, say in between 4 and 10, hence both  $\epsilon/k$  and  $\alpha$  may be determined. The information may be limited to  $\alpha$  if only high temperatures have been observed.

From the above discussion it follows that the sensitivity of isotopic thermal diffusion to calculate potential parameters for the exp-six model depends markedly on the reduced temperature range covered and, as a consequence, of the gas considered.

In the case of the rare gases, if we assume that the apparatus may cover the mean absolute temperature range,  $\bar{T}$ , between 50 and 1200 °K, and, furthermore, the minimum reduced temperature which may be achieved is of the order of  $T^* = 1$ , hence the range experimentally accessible for each gas is given in table I.

TABLE I

*Workable range of  $T^*$  for thermal diffusion in rare gases.*

gas	$\epsilon/k$ (°K)	Reference	Reduced temp. range $T^*$
He	9.16	M 1	5.5 — 131
Ne	33.4	this work	1.4 — 10
Ar	123.2	M 2	1 — 10
Kr	158.3	M 2	1 — 8
Xe	231.2	M 2	1 — 5



Therefore, experimental values of the isotopic thermal diffusion factor may provide, at most, the following information about the parameters  $\varepsilon/k$  and  $\alpha$  of the exp-six potential:

Helium: practically only  $\alpha$  may be determined;

Neon:  $\alpha$  and  $\varepsilon/k$  may be determined if a sufficiently temperature range is covered;

Argon: only rough values of  $\alpha$  and  $\varepsilon/k$ ;

Krypton and Xenon: determination of  $\alpha$  and  $\varepsilon/k$  uncertain. At most, correlated intervals for those parameters may be chosen.

It follows, therefore, that in case of argon, krypton and xenon the experimental data of thermal diffusion may only be used to testify the agreement of the potential model, by using the values of  $\alpha$  and  $\varepsilon/k$  determined by other transport phenomena.

Neon is obviously an interesting case, since it is the only rare gas which may determine both potential parameters by thermal diffusion with some confidence. Unfortunately, experimental values differ markedly from author to author, as fig. 2 shows.

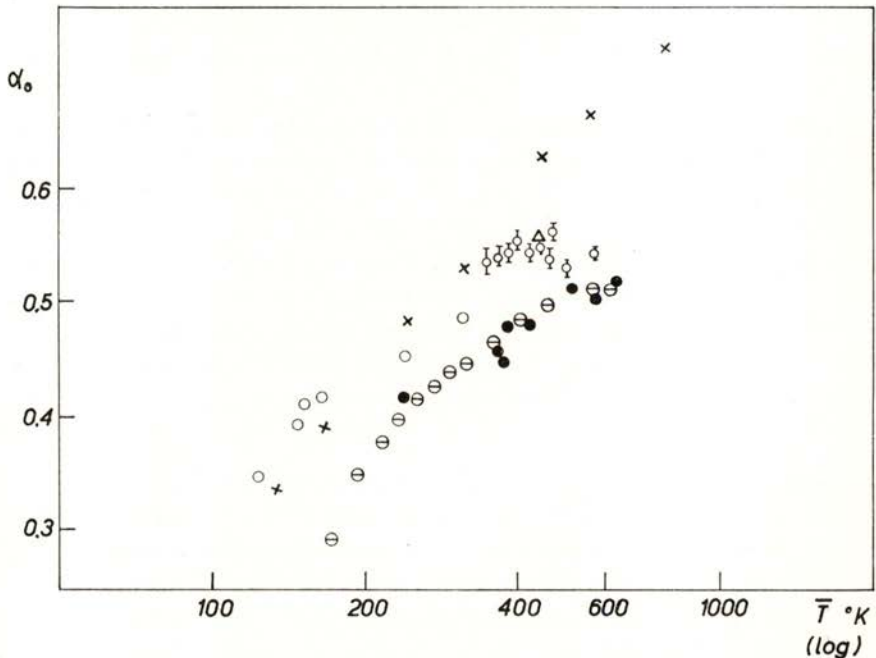


Fig. 2 — *Experimental reduced isotopic thermal diffusion factor,  $\alpha_0$ , for  $^{20}\text{Ne} - ^{22}\text{Ne}$*

$\Phi$  Present work;  $\Delta$  LARANJEIRA & KISTEMAKER (L-1);  $\times$  STIER (S-1);  $\ominus$  SAXENA *et al.* (S-2);  $\bullet$  MORAN & WATSON (H-3);  $\circ$  WATSON *et al.* (W-2);

From the above discussion it follows that further experiments of thermal diffusion with neon isotopes will be of importance.

We have in this Laboratory a running program working with three types of swing separators which cover the following temperature ranges in neon:

- 1 — Low temperature range, from  $T^* \approx 2$  up to 9.
- 2 — Intermediate range, from  $T^* \approx 9$  up to 18.
- 3 — High temperature range, for  $T^* > 18$ .

In this paper we present the values of the isotopic thermal diffusion factor corresponding to the intermediate range and a new set of potential parameters for the exp-six model is proposed, by using thermal diffusion and viscosity data.

Absolute temperature range observed extends from 335.6 up to 507.4° K. This range has previously been observed by STIER (S-1) in 1942, MORAN & WATSON (M-3) in 1958 and SAXENA, KELLY & WATSON (S-2) in 1961 (see fig. 2).

STIER's results are believed to be extremely high and masked by eventual low pressure effects due to the technique involved. On the other hand, WATSON (W-1) stated that the values of ref. M-3 were systematically low because «mass spectrometer analyses for neon were in error because of an appreciable superposition of a peak from double-ionized  $CO_2$  with that of singly ionized  $^{22}Ne$ .»

SAXENA and coworker's results follow the general trend of those of MORAN and WATSON, so that we may say that they are rather low as well.

## 2 — THEORETICAL.

According to CHAPMAN-ENSKOG's kinetic theory (C-1, H-1), the velocity of mutual diffusion in a binary mixture  $i, j$  at uniform pressure, is given by

$$C_i - C_j = -D_{ij} [\text{grad } \ln (x_i/x_j) - (\alpha_T)_{ij} \text{ grad } \ln T] \quad (1)$$

where

$C_i$  and  $C_j$  — mean values of the peculiar velocities of the molecules of kind  $i$  and  $j$ , respectively.

$D_{ij}$  — concentration diffusion coefficient of the mixture.

$x_i$  and  $x_j$  ( $x_i + x_j = 1$ ) — mole fractions.

$(\alpha_T)_{ij}$  — thermal diffusion factor of the mixture, with appropriate choice of signal.

When equilibrium has been reached, so that the velocity of mutual diffusion is zero, eq. 1 reduces to

$$\text{grad } \ln (x_i/x_j) = (\alpha_T)_{ij} \text{ grad } \ln T \quad (2)$$

If the elementary thermal diffusion effect is observed between temperatures  $T^I$  and  $T^{II}$  ( $T^{II} > T^I$ ), eq. 2 is usually integrated assuming that the thermal diffusion factor is independent of temperature and the mole fractions. The later simplification can be assumed since separation effect is generally very small indeed; the former is usually overcome by choosing a mean temperature,  $\bar{T}$ , which the experimental thermal diffusion factor is referred to.

Making the above assumptions, the integration of eq. 2 gives

$$\ln q = \alpha_T(T) \ln (T^{II}/T^I) \quad (3)$$

where subscripts  $i, j$  referring to gases has been omitted and  $q$  is the separation factor given by

$$q = (x_i/x_j)^{II} / (x_i/x_j)^I$$

Upperscripts  $II$  and  $I$  refer to the high and low temperatures,  $T^{II}$  and  $T^I$ , respectively.

Several formulas have been proposed for the mean temperature,  $\bar{T}$ . This problem has been considered by one of us (L-2). In the temperature range we have dealt in this paper, it is safe to use DAVENPORT'S formula (D-1), namely

$$\bar{T} = (T^{II} T^I)^{1/2}$$

The theoretical expression of the thermal diffusion factor for isotopes of molecular masses  $M_1$  and  $M_2$  close enough, can be written as



$$\alpha_T = \alpha_0 \frac{M_2 - M_1}{M_2 + M_1} \quad (M_2 > M_1) \quad (5)$$

where  $\alpha_0$  is the so called reduced isotopic thermal diffusion factor. According to CHAPMAN-ENSKOG'S theory,  $\alpha_0$  is given as a ratio of two infinite determinants, to which two schemes of serial solutions have been presented, namely, by KIHARA-MASON (K-1, M-4) and CHAPMAN-COWLING (C-1, M-5).

In this paper we will only use the first KIHARA'S approximation,  $[\alpha_0]_1^K$ , for the reduced isotopic thermal diffusion factor:

$$[\alpha_0]_1^K = \frac{15}{16} \cdot \frac{6 C^* - 5}{A^*} \quad (6)$$

where

$$C^* = \Omega^{(1,2)*} / \Omega^{(1,1)*}; \quad A^* = \Omega^{(2,2)*} / \Omega^{(1,1)*}$$

and  $\Omega^{(1,1)*}$ ,  $\Omega^{(1,2)*}$  and  $\Omega^{(2,2)*}$  are reduced collision integrals.

The formulas for CHAPMAN-COWLING'S scheme and higher order of KIHARA-MASON'S approximations are very complex and theoretical data for  $\alpha_0$  can only be obtained by means of computers.

Tabulated values of different approximations for the reduced isotopic thermal diffusion factor according to common potential models, are not yet complete. In our case we have to deal with exp-six model with the potential parameter  $\alpha$  equals to 15 and 16.

KIHARA'S first approximation,  $[\alpha_0]_1^K$ , has been already tabulated for  $\alpha = 16$ , (M-2), but not for higher approximation. On the other hand, for  $\alpha = 15$  a complete table has been given for the second approximation,  $[\alpha_0]_2^K$ , (S-3), but only partially for the first one (M-5, I<sub>r</sub>-2, I<sub>r</sub>-3).

Our discussion on the isotopic thermal diffusion factor for neon has, for that reason, been limited to  $[\alpha_0]_1^K$ , which is the only one readily workable and computed using tabulated values of  $C^*$  and  $A^*$ , eq. 6. This is not a serious drawback, since for practical proposes the improvement achieved by higher orders of approximation may be disregarded, bearing in mind the magnitude of experimental errors.

### 3 — EXPERIMENTAL.

Experiments of isotopic thermal diffusion factors for neon have been carried out with a swing separator of CLUSIUS' type (C-2) con-

sisting of 8 stainless steel tubes with an inner diameter of 15 mm and 9 cm long, connected in series by stainless steel capillary tubes with an inner diameter of 0,8 mm (see fig. 3).

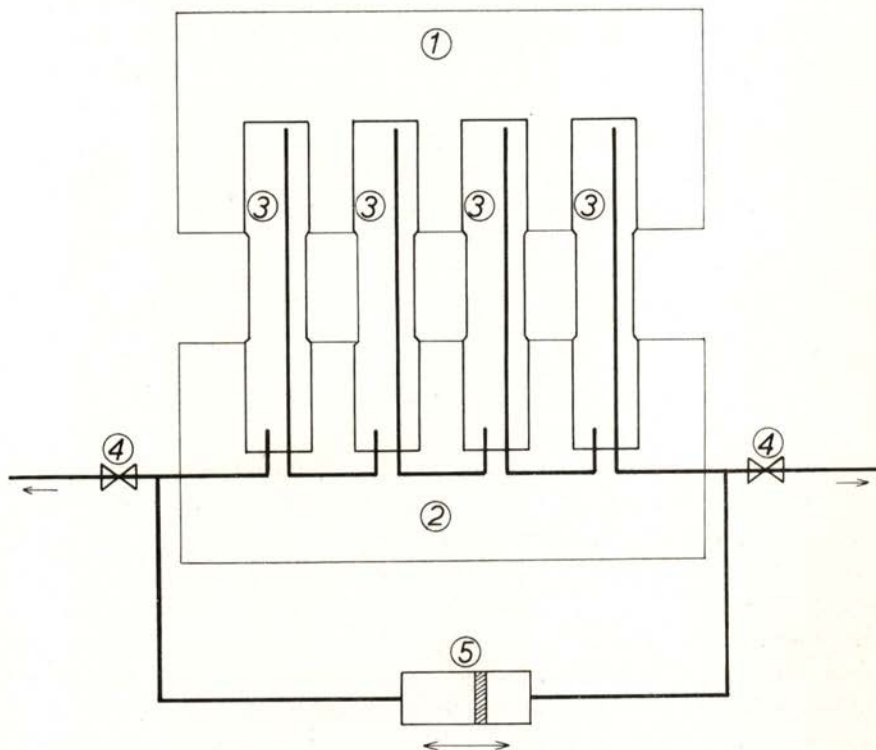


Fig. 3 — *Diagram of the swing separator. 1 — copper block; 2 — oil bath; 3 — separation stainless steel tubes; 4 — to inlet and sampling system; 5 — mercury U tube pump.*

The upper 3 cm of the tubes were forced in a copper block which was heated up thermostatically by a heating element; the lower 3 cm were cooled by a thermostatic oil bath fed by a cold oil tank by means of a small electrical pump.

The transition part of the tubes was isolated with asbestos.

In fig. 4 we present schematically the control system for the heating element and temperature measurements. A chromel-constantan thermocouple is used via a strip chart recorder which registers the temperature and commands by an on-off system a change-over relay. The power for the heating element is furnished through that relay by the variac which has two output voltages,  $V_1$  and  $V_2$ , being



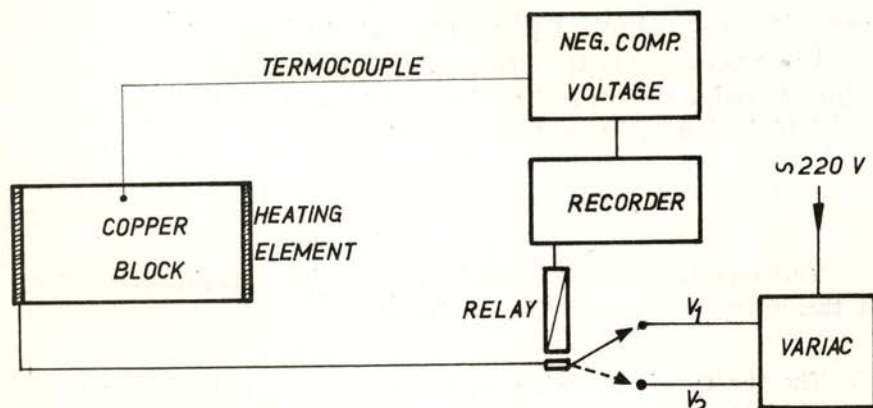


Fig. 4 — Control system for the heating element and for temperature measurements.

one slightly lower and the other slightly higher than the mean voltage necessary to maintain the desired temperature.

In order to increase the sensibility of the system, we have chosen a full scale of 0 — 5 *mV* in the recorder. When the e.m.f. of the thermocouple was higher we introduced a negative compensation voltage, as shown on fig. 4.

By this way the temperature stability in the copper block was of about 0.2 °K over the entire range of the experiments.

The temperature of the oil bath was stabilized in a similar way. A variable flow of oil from a cold tank was maintained by a small electrical pump.

The temperature stabilisation was of about 0.3 °K.

About 1 cm<sup>3</sup> of the gas was pumped to and fro periodically by means of a U tube, partially filled with mercury, which was swung by an electromotor and an eccentric wheel. The pumping period can change within large limits (10-120 s) without appreciable difference in the isotopic separation achieved. We have chosen  $t_p = 19$  s.

The swing separator is a convection-free apparatus and an elementary thermal diffusion process should be achieved in each tube. If  $q$  represents the elementary separation factor in each tube in the temperature range  $T^I - T^{II}$ , hence the total separation factor,  $Q$ , obtained with a swing separator of  $n$  tubes should be

$$Q = (X_i/X_j)^{II} / (X_i/X_j)^I = q^n \quad (7)$$

where  $(X_i/X_j)^I$  and  $(X_i/X_j)^{II}$  are the mole fraction ratio of the gases  $i$  and  $j$ , respectively in the cold region of the first tube at the temperature  $T^I$  and in the hot region of the  $n$ th tube at the temperature  $T^{II}$ .

Introducing equation 7 into equation 3, we have

$$\ln Q = n \alpha_T(\bar{T}) \ln (T^{II}/T^I) \quad (8)$$

This equation has been used to determine experimental values of the isotopic thermal diffusion factor.

The theory of the swing separator has been given by VAN DER WAERDEN (W-3). The relaxation time of separation,  $t_R$ , is approximately

$$t_R = \left( \frac{n+1}{\pi} \right)^2 (t_D + t_P V/\Delta V) \quad (9)$$

where

- $n$  — number of the tubes of the swing separator.
- $t_D$  —  $L^2/D$  where  $L$  is the length of each tube and  $D$  is the diffusion coefficient of the gas mixture.
- $t_P$  — pumping period of the U tube.
- $V$  — volume of each tube of the swing separator.
- $\Delta V$  — pumping volume of the U tube.

In our case, the relaxation time for neon isotopes is of the order of 38 minutes, in the temperature range observed.

In practice every experiment has been runned for at least 24 hours. Runs of 48 hours have been performed from time to time. Therefore, stationary conditions have been secured.

Analyses of neon isotopes were made in our laboratory with a mass spectrometer type MS 2-SG, from AEI, Manchester, England.

Normal neon was used in experiments and was delivered by L'Air Liquide, Lisbon. Purity was 99,8 % having helium as the main impurity. No traces of argon and  $CO_2$ , which might interfer with mass spectrometric analyses of neon, have been found.

For the experiments the swing separator was filled to about 76 cm Hg.

To control eventual inleakage the peak at mass 28 corresponding to  $N_2^+$ , was always measured before and after each experiment. For

all the observations reported in this paper, the peak height of  $N_2^+$  in the samples could not be distinguished from/or was very slightly above the normal background of the mass spectrometer.

#### 4 — RESULTS.

Common neon consists of isotopes  $^{20}\text{Ne}$ ,  $^{21}\text{Ne}$  and  $^{22}\text{Ne}$ . Isotopic composition of the neon used was determined by our mass spectrometer giving the following percentages with an error of the order of 0.2 %:

$$^{20}\text{Ne} — 90.9 \quad ; \quad ^{21}\text{Ne} — 0,251 \quad ; \quad ^{22}\text{Ne} — 8.90$$

This results agree satisfactorily with tabulated abundances.

Since  $^{21}\text{Ne}$  was present in small amount, the neon used was regarded as a binary mixture of  $^{20}\text{Ne}$  -  $^{22}\text{Ne}$ . Furthermore, the theory of thermal diffusion for complex isotopic mixtures supports this kind of approximation.

In table III, column 5, we present the experimental values of the isotopic thermal diffusion factor,  $\alpha_T$  (exp), and in column 6 and figs. 2 and 5, the reduced isotopic thermal diffusion factor,  $\alpha_o$  (exp), by using eq. 5.

Each presented value of  $\alpha_T$  (exp) corresponds to a mean of at least 4 observations. The errors reported in the table take into consideration only the deviations to the mean value of  $\alpha_T$  (exp). The magnitude of those errors are of the order of 1 - 2 %.

The mean temperature to which the experimental thermal diffusion factor has been referred is given by DAVENPORT'S formula, eq. 4.

#### 5 — DISCUSSION.

In the following discussion we will only consider the exp.-six potential for theoretical comparisons. This potential model is believed to fit reasonably well experimental data of transport phenomena, very probably not only due to theoretical reasons but also because it has three adjustable parameters,  $\epsilon/k$ ,  $\alpha$  and  $r_m$ .

If one intends to determine those potential parameters by means of experimental data of transport phenomena, one has obviously to realize what kind of information may be obtained by each of the phenomena considered.



As we have pointed out in § 1, though thermal diffusion is very sensitive to the nature of molecular potential, the possibility of determining directly the potential parameters  $\varepsilon/k$  and  $\alpha$ , has to be examined considering each specific case. For the temperature range we have covered the thermal diffusion factor of neon isotopes is able to provide a good value for the parameter  $\alpha$ , but  $\varepsilon/k$  is completely uncertain.

On the other hand, if  $\alpha$  is already known, viscosity provides the best way to determine  $\varepsilon/k$  as well as  $r_m$ , since accurate experimental data is available for the case of neon covering a large temperature range.

The coefficient of viscosity,  $\eta$ , can be written as

$$10^7 \eta = 266.93 (M T)^{1/2} \eta_0 \quad (10)$$

where  $M$  is the molecular weight and  $\eta_0$ , which we call the reduced coefficient of viscosity, is given by

$$\eta_0 = f_{\eta}^{(m)}(\alpha, T^*) / r_m^2 \Omega^{(2,2)*}(\alpha, T^*) \quad (11)$$

In the above formulas,  $\eta$  is expressed in poise and  $r_m$  in Angstrom.

Tables for the reduced collision integral  $\Omega^{(2,2)*}$  and the dimensionless function  $f_{\eta}^{(m)}$  can be obtained in ref. M-5, M-2 and H-1 for the second and third order of approximation, ( $m$ ).

By means of equation 10 experimental values of  $\eta_0$  can be obtained. Determinations of  $\varepsilon/k$  and  $r_m$ , once  $\alpha$  fixed, have been made by the usual superposition technique (W-4). By superposition of the log-log plot of  $\eta_0$  versus  $T$ , over the theoretical log-log curve of  $f_{\eta}^{(m)} / \Omega^{(2,2)*}$  versus  $T^*$ , the potential parameters  $\varepsilon/k$  and  $r_m$  are graphically calculated. Further improvement to those values are obtained by the least square method.

For experimental and theoretical comparisons, we have taken as well the self-diffusion coefficient, which is the only transport coefficient directly correlated with the collision integral  $\Omega^{(1,1)*}(\alpha, T^*)$  being therefore of particular interest:

$$10^4 D = 26.280 M^{-1/2} T^{3/2} D_0 \quad (12)$$

with

$$D_0 = f_D^{(m)}(\alpha, T^*) / r_m^2 \Omega^{(1,1)*}(\alpha, T^*) \quad (13)$$

We call  $D_0$  as the reduced self-diffusion coefficient. In principle experimental values of  $D_0$  provides another way to determine  $\varepsilon/k$

and  $r_m$  once fixed the parameter  $\alpha$ . However, since observations with neon isotopes are rather scarce and covering an insufficient temperature range, the self-diffusion coefficient has only been used as a kind of control.

In short, the method we propose to determine the potential parameters of neon is the following:

- 1 — Bearing in mind the temperature range covered by our experiments, the parameter  $\alpha$  is determined by thermal diffusion.
- 2 — Values of  $\epsilon/k$  and  $r_m$  are then calculated from viscosity data.
- 3 — Self-diffusion is only used as a kind of control.

TABLE II

*Exponential-six parameters for neon.*

Parameters	Present work			Ref. M-2
	Values of parameters	Experimental data used	$T^*$ range observed	Values of parameters
$\alpha$	15.5	thermal diff.	10 — 15	14.5
$\epsilon/k$ ( $^{\circ}K$ )	33.4	viscosity	0.6 — 23	38.0
$r_m$ ( $\text{Å}$ )	3.144	viscosity	0.6 — 23	3.147

In table II we give the values of the potential parameters determined by this method. Previous results proposed by MASON & RICE (M-2) using viscosity data and the second virial coefficient are also presented.

In table III, columns 6 and 7, and in the upper part of fig. 5 we compare the experimental reduced thermal diffusion factor with KIHARA'S first approximation given by eq. 6, for our proposed parameters. Values of  $[\alpha_0]_1^K$  for the chosen parameter  $\alpha = 15.5$  have been interpolated by means of the existing tables for  $\alpha = 16$ , ref. M-2, and our computed values for  $\alpha = 15$ , given in table IV, which complete previous tabulations of ref. M-2, I-2 and I-3 in the temperature range of interest.

TABLE III

*Experimental and theoretical values of thermal diffusion for  $^{20}\text{Ne}-^{22}\text{Ne}$ .*

$T^{II}$ (°K)	$T^I$ (°K)	$\bar{T}$ (°K)	$\bar{T}^*$	$\alpha_T$ (exp) $\times 10^2$	$\alpha_o$ (exp)	$[\alpha_o]_1^K$ (theor.)	Dev.
373.0	302.0	335.6	10.05	$2.56 \pm 0.06$	$0.538 \pm 0.013$	0.547	— 0.009
413.0	314.0	354.3	10.61	$2.59 \pm 0.03$	$0.544 \pm 0.007$	0.548	— 0.004
442.0	303.0	366.0	10.96	$2.61 \pm 0.05$	$0.548 \pm 0.011$	0.549	— 0.001
475.0	308.0	382.1	11.44	$2.66 \pm 0.03$	$0.559 \pm 0.006$	0.550	+ 0.009
536.0	307.6	407.4	12.20	$2.61 \pm 0.04$	$0.548 \pm 0.008$	0.551	— 0.003
563.0	318.0	423.1	12.67	$2.64 \pm 0.01$	$0.554 \pm 0.002$	0.551	+ 0.003
616.0	316.0	441.2	13.21	$2.58 \pm 0.04$	$0.542 \pm 0.008$	0.551	— 0.009
646.0	315.6	451.5	13.52	$2.70 \pm 0.02$	$0.566 \pm 0.004$	0.551	+ 0.015
732.0	313.0	478.7	14.33	$2.64 \pm 0.03$	$0.554 \pm 0.007$	0.551	+ 0.003
773.0	333.0	507.4	15.19	$2.60 \pm 0.02$	$0.547 \pm 0.005$	0.551	— 0.004

TABLE IV

*Values of  $[\alpha_o]_1^K$  for exponential-six and  $\alpha = 15$ .*

$T^*$	$[\alpha_o]_1^K$	Ref.	$T^*$	$[\alpha_o]_1^K$	Ref.
8	0.528	a)	18	0.536	a)
9	0.533	a)	20	0.536	M-5, L-2, L-3
10	0.535	M-5, L-2, L-3	25	0.533	a)
12	0.538	a)	30	0.531	a)
14	0.538	L-3	35	0.531	a)
16	0.538	a)	40	0.531	L-2, L-3

a) Present work.

In table v we compare experimental and theoretical coefficients of viscosity,  $\eta$ , and in fig. 5 the reduced coefficient of viscosity,  $\eta_o$ , eqs. 10 and 11. The agreement between experimental and theoretical data is satisfactory and seems to be better than that reported by MASON & RICE (M-2) using their set of parameters indicated in table II.

As we have pointed out, experimental values of self-diffusion are very scarce and, therefore, insufficient for determinations of potential parameters of the exp.-six model. Comparison between experi-



TABLE V

*Experimental and theoretical values of viscosity for neon.*

$T$ (°K)	$T^*$	$\eta$ (exp) ( $\mu$ poise)	Ref.	$\eta$ (theor.) ( $\mu$ poise)	Dev. ( $\mu$ poise)
20.43	0.6117	33.6	K1	34.47	— 0.9
30.55	0.9147	51.1	M6	51.27	— 0.2
39.52	1.183	64.9	M6	65.86	— 1.0
49.97	1.496	80.4	J1	81.97	— 1.6
59.59	1.784	96.0	M6	95.89	+ 0.1
69.38	2.077	109.4	M6	109.07	+ 0.3
80.00	2.395	119.8	J1	122.61	— 2.8
90.3	2.704	135.2	R1	134.96	+ 0.2
100.0	2.994	143.5	J1	146.03	— 2.5
140.0	4.192	184.1	J1	186.78	— 2.7
160.0	4.79	202.6	J1	205.16	— 2.6
180.0	5.39	220.4	J1	222.5	— 2.1
200.0	5.99	237.6	J1	239.0	— 1.4
229.0	6.86	267.0	R1	261.8	+ 5.2
240.0	7.19	270.8	J1	270.1	+ 0.7
260.0	7.78	286.7	J1	285.0	+ 1.8
273.0	8.17	297.3	R2	294.3	+ 3.0
293.16	8.777	309.2	T1	308.4	+ 0.8
298.0	8.92	313.0	R2	311.7	+ 1.3
323.0	9.67	329.4	R2	328.7	+ 0.7
373.0	11.17	364.6	T1	361.5	+ 3.1
473.0	14.16	424.8	T1	422.2	+ 2.7
523.0	15.66	453.2	T1	450.9	+ 2.3
558.0	16.71	470.8	T2	470.4	+ 0.4
702.0	21.02	545.4	T2	546.8	— 1.4
775.0	23.20	580.2	T2	583.5	— 3.3

mental and theoretical values of  $D$  and  $D_0$ , eqs. 12 and 13, using our set of parameters, is given in table VI and in fig. 5, respectively. The agreement may be considered rather good but, as a general trend, experimental data is about 2 % higher than theoretical values. That difference is, however, of the order of magnitude of experimental errors. If we choose the parameters  $\epsilon/k = 33.4$  °K and  $\alpha = 15.5$  of our set, experimental data of self-diffusion in neon is more consistent with a value of  $r_m = 3.108$  Å whereas from viscosity we have obtained  $r_m = 3.144$  Å.

TABLE VI  
*Experimental and theoretical values of self-diffusion for neon.*

$T$ (°K)	$T^*$	$D$ (exp) $\times 10^2$ (cm <sup>2</sup> s <sup>-1</sup> )	Ref.	$D$ (theor.) $\times 10^2$ (cm <sup>2</sup> s <sup>-1</sup> )	Dev. $\times 10^2$ (cm <sup>2</sup> s <sup>-1</sup> )
77.7	2.326	4.92	W 6	4.962	- 0.04
194.7	5.83	25.5	W 6	24.67	+ 0.83
273.2	8.18	45.2	W 6	43.69	+ 1.51
293.0	8.77	48.4	G 1	49.14	- 0.74
298.2	8.93	51.6	W 6	50.61	+ 0.99
353.2	10.57	70.3	W 6	67.22	+ 3.08

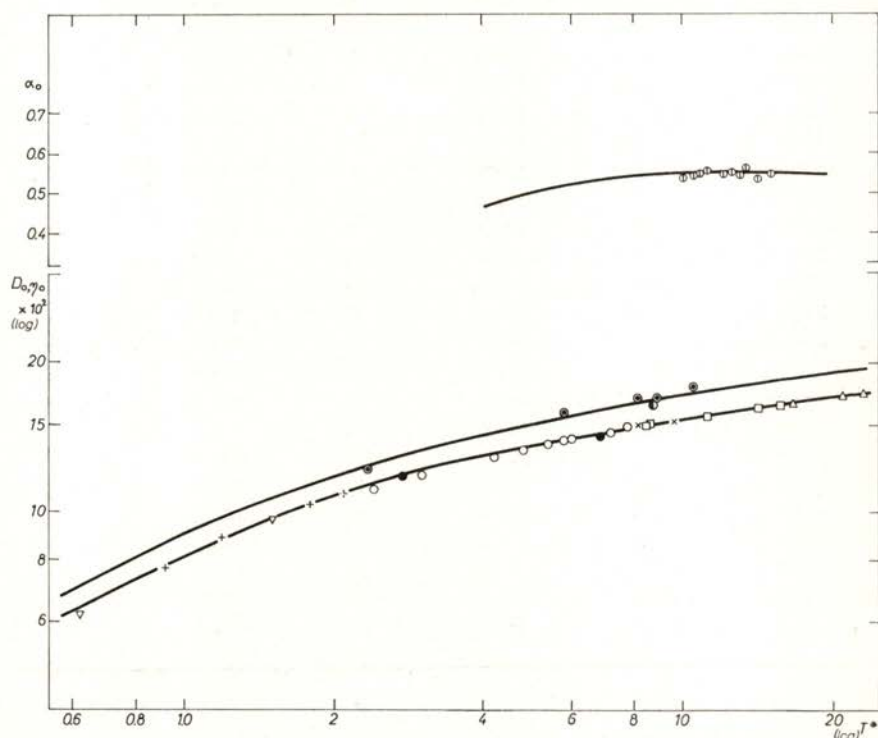


Fig. 5 — *Reduced isotopic thermal diffusion factor,  $\alpha_0$ , for  $^{20}\text{Ne} - ^{22}\text{Ne}$ :  
 ——— Kihara's first approximation.  $\odot$  our experimental data.  
 Reduced self-diffusion coefficient,  $D_0$ :  
 ——— theoretical curve. Experimental:  $\odot$  WINN (W 6);  $\bullet$  GROTH & SUS-SUER (G-1).  
 Reduced viscosity coefficient,  $\eta_0$ :  
 ——— theoretical curve. Experimental:  $\circ$  JOHNSTON & GRILLY (J-1);  
 $\bullet$  RIETVELD & VAN ITTERBEEK (R-1);  $\times$  RANKINE (R-2);  
 $\square$  TRAUTZ & BINKELE (T-1);  $\triangle$  TRAUTZ & ZINK (T-2);  
 $\nabla$  KESTIN & WANG (K-2);  $+$  MARIENS & PAEMEL (M-6).*

It should be pointed out that our set of parameters (as well as those of MASON & RICE) cannot explain experimental thermal diffusion of neon isotopes recently reported by WATSON *et al.* (W-5) and by SAXENA *et al.* (S-2) in the low temperature range from  $\bar{T} \approx 170$  up to 300°K (see fig. 2).

For WATSON's results the exp.-six parameter  $\epsilon/k$  would be of the order of 50 °K and for SAXENA's data of the order of 67 °K (with  $\alpha = 14$ ). Such high values of  $\epsilon/k$  are quite inadequate for experimental viscosity data which is known to be accurately measured.

In principle, we may think that WATSON and SAXENA's data are affected by unknown systematic errors and, therefore, it would be very convenient that further experiments of isotopic thermal diffusion of neon should be realized in that low temperature range.

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