From the values in Table II one sees that the agreement is fairly good, but that there is a discrepancy which is definitely outside of the statistical error. The difference is in the sense that the experimentally observed absorption is less dependent upon the atomic number than should be expected according to the theory. There are several perturbing factors involved, and although an attempt has been made to evaluate their effects, one cannot be certain that they have all been accounted for with sufficient accuracy. In particular, it is possible that the electrons in equilibrium with the mesotrons affect the measured absorption to some extent. Moreover, there is some uncertainty in the number of mesotrons that are scattered out of the beam. In conclusion, we believe that the theoretical values are in approximate agreement with the experimental results and it appears probable that the discrepancies are not due to any fundamental inadequacy of the theory.

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## Performance of a Hot Wire Clusius and Dickel Column<sup>\*,\*\*</sup>

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The separation factor for argon isotopes has been measured as a function of the gas pressure in a hot wire type of Clusius and Dickel column. The variation obtained is in accord with the predictions of the Furry, Jones, and Onsager theory. It is found empirically that the explicit expressions given by Furry, Jones, and Onsager for the flat, parallel wall case can be modified to give correct results for the extreme cylindrical case if the constants of the gas are evaluated at the proper mean temperatures. The existence of turbulence in the gas causes a slight, if any, decrease in the separation factor determined by extrapolation from the experimental data obtained under conditions of lamellar flow.

# INTRODUCTION

N 1938 Clusius and Dickel,<sup>1</sup> and subsequently Brewer and Bramley,<sup>2</sup> cascaded the small effect of thermal diffusion in separating isotopes by putting the gaseous isotopic mixture into the annular space between two concentric cylindrical surfaces, mounted vertically. By maintaining the inner surface at a higher temperature than the outer, convection currents are set up which carry the isotope that concentrates toward the hot surface to the top of the column, thus greatly

increasing the transverse separation caused by the radial temperature gradient. Batteries of Clusius and Dickel columns coupled in series have since been used successfully to obtain large concentrations of important rare isotopes.<sup>3</sup>

The theory of the Clusius and Dickel method was first developed by Furry, Jones, and Onsager<sup>4</sup> in 1939 (hereinafter referred to as FJO) and also by Waldmann<sup>5</sup> and Debye.<sup>6</sup> FJO calculated explicit expressions for the vertical change in concentration from their general theory for the case of a temperature gradient between two flat,

<sup>\*</sup> Part of a dissertation presented for the degree of Doctor of Philosophy at Yale University. \*\* This paper was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war.

<sup>\*\*\*</sup> Now with the Signal Corps Engineering Labora-tories, Bradley Beach, New Jersey. <sup>1</sup>K. Clusius and G. Dickel, Naturwiss. 26, 546 (1938);

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<sup>&</sup>lt;sup>3</sup> W. W. Watson, Phys. Rev. 57, 562A (1940) and 57, 899 (1940); E. F. Shrader, Phys. Rev. 58, 475L (1940); S. B. Welles, Phys. Rev. 59, 920A (1941); R. Fleischmann, b. weiles, r hys. Rev. 39, 920A (1941); K. Fleischmann, Physik. Zeits. 41, 14 (1940); A. O. Nier and J. Bardeen, J. Chem. Phys. 9, 690 (1941).
 <sup>4</sup>W. H. Furry, R. Clark Jones, and L. Onsager, Phys. Rev. 55, 1083 (1939).

<sup>&</sup>lt;sup>5</sup> L. Waldmann, Zeits. f. Physik 114, 53 (1939).

<sup>&</sup>lt;sup>6</sup> P. Debye, Ann. d. Physik 36, 284 (1939).

parallel, vertical walls. Furry and Jones<sup>7</sup> extended the theory to the cylindrical case and published their results for two special cases.

The theoretical dependence, according to FJO, of the amount of separation of C<sup>12</sup>H<sub>4</sub> and C<sup>18</sup>H<sub>4</sub> on the gas pressure has been verified experimentally by Nier<sup>8</sup> and by Taylor and Glockler<sup>9</sup> using concentric metal cylinders. Measurements of this dependence have been reported for the extreme cylindrical case where the inner surface is an electrically heated wire.<sup>10</sup> The use of a hot wire permits the maintainance of much larger temperature gradients without excessive power inputs.

#### EXPERIMENTAL APPARATUS

An all-glass column was constructed so that the working length of the gas chamber was 200 cm. The gas chamber consisted of a 22-mm inner diameter glass tube cooled on the outside by a water jacket. The hot surface was a 20-mil tungsten wire heated by direct current to avoid vibration. Four sets of 20-mil nickel wire crosspieces were spot-welded onto the wire at 50-cm intervals to keep the wire centered in the column. The lower extremity of the wire hung freely under the tension of a 450-g cylindrical weight which consisted of a hollow copper tube filled with lead. Heating the wire in a vacuum at 1200°C for about thirty minutes served to straighten it and also to remove the oxide coating from its surface, thus greatly reducing radiant power loss needed to maintain a given temperature.

The gas chamber was terminated by two tapered ground-glass joints. The upper joint was fitted with a cap containing a sealed-in 75-mil tungsten lead. The cap was water-cooled to prevent the metal-to-glass seal and the stopcock grease used in the joint from becoming too hot. The lower ground-glass joint was fitted with a length of glass tubing surrounding the weight and terminating in a mercury cup deep enough to allow for as much as a 2.5-cm thermal extension of the wire. A copper lead from the bottom of the weight provided contact with the mercury. The lower joint was cooled sufficiently well by a flow of water through a lead pipe wrapped around the outside. Spot-welding of the wire to the leads was accomplished with the intermediate use of nickel.

## TURBULENCE MEASUREMENTS

The FJO theory was derived on the assumption that the convective flow of gas is lamellar in the annular space between the two surfaces; i.e., the convection velocity, v, is entirely in the vertical, z, direction and is a function only of the radial distance, r, from the center of the wire, except, of course, at the very ends of the column. Preliminary experiments were undertaken to determine the range of stability of lamellar flow, following the method of Onsager and Watson.<sup>11</sup>

Dissipation of the power input necessary to maintain the wire at a given temperature is caused by the following, if the flow is lamellar:

- (a) Radiation from the wire.
- (b) Transverse conduction of heat through the gas.
- $(c)\$  Conduction of heat through the spacers and leads.
- (d) Heat losses at the ends of the column due to the convective flow.

Items (a) and (c) are obviously independent of the pressure of the gas in the column. Item (b) is independent of the pressure if the mean free path of the gas molecules is much smaller than the gap space between the wire and cold wall, a condition which was strongly satisfied for the pressures used. Item (d) is pressure dependent, but its contribution to the power loss will later be experimentally demonstrated to be negligible. Consequently, with a given power input, the temperature of the wire should remain constant as the gas pressure is increased until the critical Reynolds number for this type of flow is reached, above which the lamellar flow becomes unstable and breaks up into turbulence. The turbulent swirls provide an additional, pressure-dependent mechanism for direct transfer of heat from the wire to the cold wall. Hence, above the critical turbulence pressure, the temperature of the wire should decrease with increasing gas pressure and constant power input.

The critical pressures,  $p_c$ , were determined for <u>1</u> L. Onsager and W. W. Watson, Phys. Rev. 56, 474 (1939).

<sup>&</sup>lt;sup>7</sup>W. H. Furry and R. C. Jones, Phys. Rev. **57**, 561A (1940). <sup>8</sup>A. O. Nier, Phys. Rev. **57**, 30 (1940) and **57**, 358A

<sup>(1940).</sup> <sup>9</sup> T. I. Taylor and G. Glockler, J. Chem. Phys. 8, 843 (1940).

<sup>&</sup>lt;sup>10</sup> R. Simon, Phys. Rev. **61**, 388A (1942).

wire temperatures ranging from 400° to 1700°K. With a low gas pressure in the column, the desired initial wire temperature was obtained by adjusting the power input.12 Once established, the power input was kept constant and changes in the temperature-dependent properties of the wire were recorded for increasing gas pressure. After going well beyond the critical pressure, readings were taken for decreasing pressure. The spacers and cooler spots on the wire in their vicinity introduce finite disturbances into the stream of gas which should cause general turbulence at the lowest possible Reynolds number, below which induced turbulence would be damped out. This fact was borne out experimentally as the readings for both increasing and decreasing pressure could be represented by the same curve in each case.

Figure 1 shows a typical set of curves obtained. The mean value of the pressures on each curve at which the line of zero slope intersects that with the negative slope was taken to be  $p_c$ . A slight downward trend of the "low pressure" line could be possibly detected only for the 1700°K run, which justifies the assumption that item (d) on the list of power losses is negligible. The results for nine runs are tabulated in Table I and plotted in Fig. 2.

In calculating the Reynolds numbers,<sup>13</sup>  $R = v\rho L/\eta$ , the flat-wall expression,

$$R = \frac{(2w)^3 g}{18\sqrt{3}} \left(\frac{\rho}{\eta}\right)^2 \frac{T_2 - T_1}{T_2 + T_1},$$
 (1)

TABLE I. Results of the turbulence experiments.

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<i>T</i> <sub>2</sub> (°K)	pc(cm of Hg)	Reynolds number	Power input (watts)
$400 \pm 20$	$57 \pm 2$	109	9.6
$500 \pm 20$	$47 \pm 1$	76.0	15.0
$600 \pm 20$	$40\pm 2$	47.7	28.6
$700 \pm 20$	$38 \pm 1$	35.4	46.2
$900 \pm 20$	$49\pm 2$	37.1	101
$1100 \pm 20$	$61 \pm 2$	40.6	167
$1300 \pm 20$	$67 \pm 7$	34.0	270
$1500 \pm 20$	$77 \pm 7$	32.6	600
$1700 \pm 20$	$80\pm5$	26.1	1220

<sup>12</sup> The brightness temperature of the wire was measured by a Leeds and Northrup optical pyrometer. Below the visual temperature range temperatures were set by using the electrical resistance and thermal extension characteristics of tungsten. A thermocouple could not be used because it would conduct away heat rapidly enough to make the point of contact with the wire considerably cooler.





FIG. 1. Temperature dependent properties of the wire as functions of the pressure of the gas in the column for an initial wire temperature of  $1100^{\circ}$ K, which corresponds to the lines of zero slope. (a) Current through the filament of the pyrometer bulb at "matching" brightness. (b) Electrical resistance of the wire. (c) Thermal extension of the wire from its initial length at the cold wall temperature.

was used. The gap space 2w = b - a was substituted for the length dimension, L; where  $a, b \equiv$  radius of the wire, cold wall, respectively. The density of the gas (argon),  $\rho$ , was computed from the gas laws at the pressure  $p_c$  and at the temperature  $\frac{1}{2}(T_2+T_1)$  in each case, where  $T_2, T_1 \equiv$  temperature of the wire, cold wall, respectively. The viscosity,  $\eta$ , was calculated from Sutherland's formula,<sup>14</sup>

$$\eta = \eta_0 \frac{T_0 + C}{T + C} (T/T_0)^{\frac{3}{2}},$$

at  $T = \frac{1}{2}(T_2 + T_1)$  and with  $C = 140^\circ$  and  $\eta_0 = 2.10 \times 10^{-6}$  poise at  $T_0 = 273^\circ$ K. The maximum convection velocity<sup>11</sup>

$$\frac{(2w)^2 g}{18\sqrt{3}} \frac{\rho}{n} \frac{T_2 - T_1}{T_2 + T_1}$$

was used for v. g is the acceleration of gravity.

<sup>14</sup> W. Sutherland, Phil. Mag. 36, 507 (1893).



FIG. 2. The critical gas pressure,  $p_c$ , at which turbulence first appears in the column plotted vs. the hot wire temperature,  $T_2$ .  $T_1$  is the constant cold-wall temperature.

The choice of  $\frac{1}{2}(T_2+T_1)$  at which to calculate the gas constants in R is arbitrary because  $v_{\text{max}}$  does not occur in the column where  $T = \frac{1}{2}(T_2 + T_1)$ . However, calculations of R at other temperatures between  $T_1$  and  $\frac{1}{2}(T_2+T_1)$  did not result in a more consistent set of Reynolds numbers. The Reynolds numbers obtained are only about 2 percent of those for pure pressure flow through pipes.

The curve of Fig. 2 is approximately represented by Eq. (1) if we assume a constant R and  $\rho \propto p_c$ . Turbulence first occurs at lower pressures for higher wire temperatures because of the increase in convection velocity with increased temperature gradient. However, the effect of increased velocity is soon overtaken by the effect of increased viscosity and decreased density near the hot wire with increasing temperature, which makes for stability of lamellar flow. Hence a minimum occurs at  $T_2 = 650^{\circ} \pm 50^{\circ} K$  and the  $p_c vs. T_2$  curve subsequently rises.

#### SEPARATION FACTOR MEASUREMENTS

The separation factor, f, of a thermal diffusion column is defined as the ratio of the relative particle concentrations of the two isotopes at the top of the column to that at the bottom; i.e.,  $f = (c_1/c_2)_{top}/(c_1/c_2)_{bottom}$ , where the subscript 1 refers to the lighter isotope. According to FJO,<sup>4</sup>  $\ln f = 4A_d l$ , where

$$A_{d} = \frac{A}{1 + K_{d}/K_{c}} = \frac{H/2K_{c}}{1 + K_{d}/K_{c}}$$
(2)

and 2l is the length of the column.  $H, K_c, K_d$  are

the coefficients of transport, remixing by convection, and remixing by diffusion, respectively.  $A \propto \alpha D\eta / w^4 \rho \propto 1/p^2$  and  $K_d / K_c \propto (D\eta / w^3 \rho)^2 \propto 1/p^4$ . since D, the coefficient of self-diffusion of the gas, is proportional to  $\eta/\rho$ . The thermal diffusion constant,  $\alpha$ , is given by

$$\alpha = \frac{105}{118} \frac{m_2 - m_1}{m_2 + m_1} R_T, \tag{3}$$

where the m's are the respective masses of the two isotopic molecules and  $R_T(<1)$  represents a measure of the departure of the molecule from the hard-sphere model.<sup>15</sup> Hence, as a function of pressure, one may write

$$\ln f = (a'/p^2)/(1+b'/p^4). \tag{4}$$

The relation of Eq. (4) is experimentally verified if a straight line results from plotting  $p^2/\ln f vs. p^4$ . Experimental values of the constants a' and b'may be determined from the slope and intercept of this line.

The same column was used for these measurements as for the turbulence measurements except that its length was reduced to 170 cm. No endvolumes were used other than the "dead" spaces at the top and bottom of the column, estimated to be about 30 cc each. At the end of each run small samples (about 3 cc) of the gas were removed from the top and bottom of the column in break-seal tubes. These were analyzed with a Nier-type 60° mass spectrometer<sup>16</sup> built by Professor W. W. Watson, obtaining the relative particle abundances of A<sup>36</sup> to A<sup>40,17</sup> No appreciable impurities with peaks at the same places as singly ionized masses 36 or 40 were found in the

TABLE II. Results of the separation factor measurements.

Temperature range	¢(cm of Hg)	Separa	ation factor, f
$T_2 = 1500 ^{\circ}\text{K}$ $T_1 = 293 ^{\circ}\text{K}$	$\begin{array}{c} 22.0 \pm 0.2 \\ 34.0 \pm 0.2 \\ 42.2 \pm 0.2 \\ 48.0 \pm 0.2 \end{array}$	4.5 5.2 4.4 3.6	$\begin{array}{c} \pm \ 6.7\% \\ \pm \ 11.8\% \\ \pm \ 9.8\% \\ \pm \ 1.4\% \end{array}$
$T_2 = 1100^{\circ} \text{K}$ $T_1 = 293^{\circ} \text{K}$	$18.8 \pm 0.2 \\ 27.8 \pm 0.2 \\ 36.0 \pm 0.2 \\ 40.8 \pm 0.2$	3.9 2.8 2.4 1.7	$\begin{array}{c} \pm \ 2.4\% \\ \pm \ 7.8\% \\ \pm \ 4.5\% \\ \pm \ 8.2\% \end{array}$

 $^{15}$  R. Clark Jones, Phys. Rev. **58**, 111 (1940).  $^{16}$  A. O. Nier, Rev. Sci. Inst. 11, 212 (1940).  $^{17}$  Argon has three stable isotopes of natural abundances: A<sup>36</sup>-0.31 percent, A<sup>38</sup>-0.06 percent, A<sup>40</sup>-99.64 percent.



FIG. 3. Verification of the theoretical dependence of the separation factor on the gas pressure. Solid line—wire temperature of 1500°K. Broken line—wire temperature of 1100°K.

spectrometer. A<sup>38</sup> was neglected because :

a. The  $A^{38}$  peak could not be completely resolved from the large  $A^{40}$  peak.

b. The separation factor for A<sup>38</sup> from A<sup>40</sup> is only about the square root of that for A<sup>36</sup> from A<sup>40</sup>, since  $f \propto e^{\alpha}$ .

c. The presence of  $A^{38}$  does not invalidate any theoretical analysis based on a binary mixture.<sup>18</sup>

Preliminary runs were made to determine the time needed to attain equilibrium. The relaxation time was about thirty minutes for somewhere near the least favorable case. Consequently all runs were made from four to five hours long to be certain that equilibrium had been reached.

Table II summarizes the results of runs at four different pressures each for wire temperatures of 1100°K and 1500°K. The highest pressure used in each case was well below the critical turbulence pressure for the respective temperature.  $p^2/\ln f$  is plotted against  $p^4$  in Fig. 3. The theoretical variation is evidently well satisfied in the 1500°K case. The deviation of the 1100°K points from a straight line, which appear to be random, are ascribed to difficulties occasioned with some of the analyses. The slope and intercept of each line were calculated by the method of least squares with weights on each point inversely proportional to the actual error in  $p^2/\ln f$ . By substituting the values of the constants a' and b' thus determined into Eq. (4), the experimental variation of the separation factor with pressure is

$$\ln f = \frac{3.42 \times 10^3 / p^2}{1 + 8.95 \times 10^5 / p^4} \text{ for } T_2 = 1500^{\circ} \text{K}, \quad (5)$$
  
and

$$\ln f = \frac{1.10 \times 10^3 / p^2}{1 + 1.59 \times 10^5 / p^4} \text{ for } T_2 = 1100^{\circ} \text{K}, \quad (5')$$



FIG. 4. Natural logarithm of the separation factor as a function of gas pressure with the constants a' and b' determined from FIG. 3. Solid line—wire temperature of 1500°K. Broken line—wire temperature of 1100°K.

where p is in cm of Hg. These relations are plotted in Fig. 4.

From Eq. (4) the maximum value of  $\ln f$  is  $a'/2b'^{\frac{1}{2}}$  and comes at a pressure for which  $b'/p^4=1$ . At maximum  $\ln f$ ,  $p^2/\ln f=2b'/a'$ ; hence the determination of optimum p and f can be made very simply graphically from an experimental plot such as Fig. 3. They correspond to the point on the straight line which has double the ordinate of the intercept.

## COMPARISON WITH THE THEORY

FJO give explicit expressions for A and  $K_d/K_c$ for a gas of Maxwellian molecules and for a gas of hard spheres, derived on the assumption of a temperature gradient between two flat, parallel, vertical walls. The values obtained from either of these expressions, calculating the gas constants at  $\frac{1}{2}(T_2+T_1)$  as suggested by FJO, are several times greater than the experimental ones. However, excellent agreement with experiment has been obtained by modifying the simpler expressions for the Maxwellian case<sup>19</sup> to read

and

$$A = H/2K_e = \frac{1}{4} \frac{1}{gw^4} \left( \frac{1}{\rho} \right) \Big|_{N}$$

$$K_d = \frac{1}{4} \left( \frac{Dn}{2} \right)^2 \left( \frac{r_0}{\rho} \right)$$
(6)

 $\sim$ 

 $63 \alpha / (D\eta)$ 

$$\frac{K_d}{K_c} = 1890 \frac{1}{g^2 w^6} \left\langle \left(\frac{D\eta}{\rho}\right)^2 \right\rangle_{AV} \ln\left(\frac{\gamma_0}{w}\right).$$
(7)

The  $\langle \rangle_{Av}$  denotes that the gas constants are to be suitably averaged over the gas in the column.  $r_0$  will be defined later.

One can obtain a simple expression for the temperature distribution in the column by using the inverse power repulsive model of intermolecular

<sup>&</sup>lt;sup>18</sup> R. Clark Jones, Phys. Rev. **59**, 1019 (1941). Section on "Ternary and higher order mixtures."

<sup>&</sup>lt;sup>19</sup> Equations (31) and (32) of reference 4.

(8)

force,  $F = K/d^{\nu}$ . For this model the thermal conductivity,  $\lambda \propto T^n$ , where  $n = \frac{1}{2}(\nu+3)/(\nu-1)$ . If Q is the heat flow per second per cm of length through any concentric cylindrical section of radius r in the gas chamber,  $Q/2\pi r = -\lambda(dT/dr)$ . Using  $\lambda = \lambda_0 (T/T_0)^n$  and integrating from a to r ( $T_2$  to T), we obtain

$$T_{2^{n+1}} - T^{n+1} = \frac{(n+1)QT_{0^n}}{2\pi\lambda_0} \ln\left(\frac{r}{a}\right).$$

Eliminating  $QT_0^n/\lambda_0$  by using

$$T_{2^{n+1}} - T_{1^{n+1}} = \frac{(n+1)QT_{0^{n}}}{2\pi\lambda_{0}} \ln\left(\frac{b}{a}\right),$$

we find the radial temperature distribution may be described by the relation

 $r = r_0 \equiv e^{-\gamma x}$ 

where and

 $x \equiv T^{n+1}, \quad \gamma \equiv \ln (b/a)/(x_2 - x_1)$  $r_0 \equiv \exp \left[ (x_2 \ln b - x_1 \ln a)/(x_2 - x_1) \right].$ 

T as a function of r, as given by Eq. (8), is plotted in Fig. 5. The mean qth power of the temperature, averaged over the gas in the column, is given by

$$\langle T^{q} \rangle_{Av} = \frac{1}{\pi (b^{2} - a^{2})} \int_{a}^{b} T^{q} \cdot 2\pi r dr$$

$$= \frac{2\gamma r_{0}^{2}}{b^{2} - a^{2}} \int_{x_{1}}^{x_{2}} x^{q/(n+1)} e^{-2\gamma x} dx.$$
(9)

The inverse power model is adequate only if n is considered as a slowly varying function of temperature. However, since n has been assumed to be constant in the above derivation, a suitable mean value of n must be found for given hot-wire and cold-wall temperatures. This may be done very simply experimentally as follows:

The column was filled with a given amount of gas at the cold-wall temperature,  $T_1 = 293^{\circ}$ K. The wire temperature,  $T_2$ , was then increased in steps to 1700°K and the gas pressure recorded. Within the limits of experimental error, the pressure was found to be a linear function of  $T_2$  of the form  $p = p_0 [1 + (T_2 - T_1)/7.2T_1]^{20}$ 

From the gas laws one can calculate a temperature T' which would produce the same experimental change in pressure as a given  $T_2$  if all of the gas were at T'. For  $T_2=1100^{\circ}$ K, T'=404 $\pm 1^{\circ}$ K; and for  $T_2=1500^{\circ}$ K,  $T'=460\pm 1^{\circ}$ K. The total mass of gas in the column equals

$$\int \int \rho \cdot 2\pi r dr dz = \int \int \frac{p}{R'T} \cdot 2\pi r dr dz = \text{constant},$$

where R' is the gas constant per gram. Since the maximum convection velocity is of the order of 10 cm/sec., p is not a function of r. The slight variation of p with z may be disregarded and, besides, p was measured at only one point in the column (at the bottom). Since the temperature gradient is determined by the transverse conduction of heat alone (except for comparatively small lengths at the top and bottom of the column), T is not a function of z. Consequently,

$$1/p \propto \int_a^b (1/T) \cdot 2\pi r dr.$$

Thus T' is the reciprocal of the mean reciprocal temperature.

Table III summarizes the results of numerical integration of Eq. (9) with q = -1 for  $T_2 = 1500^{\circ}$ K and  $T_1 = 293^{\circ}$ K. By interpolating in Table III it is seen that the suitable mean value of n to use



FIG. 5. Temperature of the gas in the volumn vs. the distance from the center of the wire for wire temperatures of 1500°K and 1100°K and for a cold-wall temperature of 300°K.  $a \equiv$  radius of wire;  $b \equiv$  radius of cold wall. Solid lines—for a gas of hard spheres  $(n = \frac{1}{2})$ . Broken lines—for a gas of Maxwellian molecules (n = 1). Note that the gas as a whole is "colder" as the hard sphere case is approached.

<sup>&</sup>lt;sup>20</sup> The initial pressure,  $p_0 = 30$  cm of Hg, was low enough so that this line, when plotted on Fig. 2, fell entirely below the turbulence region.

TABLE III. Calculated values of T' for  $T_2 = 1500$  °K,  $T_1 = 293$  °K.

n	<i>T'</i> (°K)
0.5	447
0.6	455
0.7	463
1.0	487

for this temperature range is  $\frac{2}{3}$ . In the same manner it was found that the value of *n* appropriate for the range 293° to 1100°K is 0.7.

For  $n = \frac{2}{3}$ :  $\nu = 13$ ,  $D = (6/5)f(\nu)\eta/\rho = 1.37\eta/\rho$ , and  $R_T = (\nu - 5)/(\nu - 1)C(\nu) = 0.597$ .<sup>4</sup> For n = 0.7:  $\nu = 11$ ,  $D = 1.40\eta/\rho$ , and  $R_T = 0.529$ .<sup>21</sup>

$$\langle D\eta/\rho \rangle_{Av} \propto \langle (\eta/\rho)^2 \rangle_{Av} \propto \langle T^{2(n+1)} \rangle_{Av}$$
. Equation (9)

can be integrated exactly for q = 2(n+1) yielding

$$\langle T^{2(n+1)} \rangle_{A_{V}} = \frac{b^{2}}{b^{2} - a^{2}} \left( x_{1}^{2} + \frac{x_{1}}{\gamma} + \frac{1}{2\gamma^{2}} \right) - \frac{a^{2}}{b^{2} - a^{2}} \left( x_{2}^{2} + \frac{x_{2}}{\gamma} + \frac{1}{2\gamma^{2}} \right)$$

Similarly,  $\langle (D\eta/\rho)^2 \rangle_{Av} \propto \langle T^{4(n+1)} \rangle_{Av}$ . From Eq. (9),

$$\langle T^{4(n+1)} \rangle_{A_{V}} = \frac{b^{2}}{b^{2} - a^{2}} \left( x_{1}^{4} + 2\frac{x_{1}^{3}}{\gamma} + 3\frac{x_{1}^{2}}{\gamma^{2}} + 3\frac{x_{1}}{\gamma^{3}} + \frac{3}{2\gamma^{4}} \right)$$
$$- \frac{a^{2}}{b^{2} - a^{2}} \left( x_{2}^{4} + 2\frac{x_{2}^{3}}{\gamma} + 3\frac{x_{2}^{2}}{\gamma^{2}} + 3\frac{x_{2}}{\gamma^{3}} + \frac{3}{2\gamma^{4}} \right)$$

Table IV lists values of these mean temperatures for the two cases in question. Using the appropriate ones at which to calculate the gas constants in Eqs. (6) and (7) ( $\rho$  by the gas laws and  $\eta$  by Sutherland's formula), and also using Eq. (3) for the thermal diffusion constant  $\alpha$ , one obtains:

$$\ln f = \frac{3.46 \times 10^3 / p^2}{1 + 9.66 \times 10^5 / p^4} \quad \text{for} \quad T_2 = 1500^\circ \text{K}, \quad (10)$$

and

$$\ln f = \frac{1.45 \times 10^3 / p^2}{1 + 1.97 \times 10^5 / p^4} \quad \text{for} \quad T_2 = 1100^{\circ} \text{K.} \quad (10')$$

When one considers the fact that any slight deviation from the cylindrical shape of the cold wall, such as might easily be expected for glass tubing, would produce appreciable changes in A and  $K_d/K_c$  which vary as  $w^{-4}$  and  $w^{-6}$ , re-

TABLE IV. Mean temperatures at which to evaluate the gas constants in Eqs. (6) and (7).

<i>T</i> <sub>2</sub> (°K)	n	$(\langle T^{10/3} \rangle_{\rm Av})^{3/10}$	$(\langle T^{20/3}  angle_{\rm Av})^{3/20}$	$(\langle T^{3.4} \rangle_{\rm Av})^{1/3.4}$	$(\langle T^{6-8} \rangle_{AV})^{1/6-8}$
1500 1100	2/3 7/10	613°K	736°K	490°K	564°K

spectively, besides introducing additional remixing due to temperature asymmetry, one finds the agreement between this semi-empirical theory and experiment is excellent. That agreement is markedly better between (10) and (5) than between (10') and (5') may be ascribed to the fact that the experimental relation (5') is nowhere near as reliable as (5). (See Fig. 3.)

In passing, it is to be noted that experiment yields only the relative magnitudes of the three constants  $H, K_c$ , and  $K_d$ . One is also interested in the value of the coefficient of transport, H, in order to be able to estimate the relaxation time and also to "match" the mass transports of the units of a multi-stage apparatus.<sup>4</sup> Knowing the appropriate mean value of n, one may calculate easily the coefficient of remixing by longitudinal diffusion since it is merely

$$K_d = \int_a^b \rho D \cdot 2\pi r dr.$$

H can then be found from the values of  $A = H/2K_a$ and  $K_d/K_c$ .

### EFFECT OF TURBULENCE ON SEPARATION

Additional measurements of the separation factor were made for  $T_2=1500^{\circ}$ K at pressures above the critical pressure for turbulence. The results are:  $f=1.63\pm0.07$  for p=80.6 cm of Hg, and  $f=1.39\pm0.07$  for p=96.2 cm of Hg. Substituting these pressures into Eq. (5), one obtains f=1.67 and 1.44, respectively. Thus turbulence produces a slight, if any, decrease in the separation factor calculated by extrapolation from the experimental data obtained under conditions of lamellar flow.

To the author's knowledge, there exists no evidence that turbulence is definitely harmful to the separation process. It has been conjectured that turbulence is harmful since the FJO theory is based on the assumption of lamellar flow. The experiments of Groth<sup>22</sup> on xenon, with a similar

<sup>&</sup>lt;sup>21</sup> From a table of values of  $(6/5)f(\nu)$  and  $C(\nu)$  in reference 15.

<sup>&</sup>lt;sup>22</sup> W. Groth, Naturwiss. 27, 260 (1939).

hot tungsten wire apparatus, has been used as evidence that turbulence decreases separation.<sup>11</sup> For a column of 12-mm diameter and at atmospheric pressure Groth found that isotope separation increased with wire temperature but reached a maximum at 1000°. On the other hand, for a tube diameter of 5 mm, separation continued to increase up to the highest temperature used, 1750°. This behavior is to be expected if the 1100°K curve of Fig. 4 is correct. Since the ln f vs. p curves for various values of  $T_2$  overlap and the maxima shift to higher pressures with increasing  $T_2$ , there is an optimum wire temperature for maximum separation for a given fixed pressure and gap space.

Although the geometry of the convective flow of the gas is not quite the same and hence the Reynolds number may have a different magnitude, one can, perhaps, approximately apply the author's experimental turbulence curve, Fig. 2, to Groth's case by adjusting for the differences in w and  $\eta/\rho$  according to Eq. (1). This procedure multiplies the ordinates of Fig. 2 by about 0.9 and shifts the minimum to the left, since the viscosity increases more rapidly with temperature for xenon than for argon, n being closer to unity for xenon.<sup>15</sup> Since the wire temperatures used by Groth are to the right of the minimum point, an increase in temperature at constant pressure would not produce turbulence. The gas in Groth's larger diameter column may well have been turbulent at all the temperatures he used, becoming less turbulent for higher wire temperatures.

Experiments performed by Bramley and Brewer<sup>23</sup> seem to show that turbulence is definitely advantageous to the separation process. However, they used systems of corrugations or baffles that produced well-defined swirls of gas in definite paths and not isotropic turbulence. This procedure may conceivably be a favorable mechanism for the thermal diffusion process.

The failure of turbulence to produce a marked decrease in separation may be caused by the fact that the pressures used were only slightly above the critical pressure for  $1500^{\circ}$ K, and hence the turbulent swirls were not violent enough to compete with the thermal diffusion process, which maintains the transverse concentration gradient.<sup>24</sup> Measurements made at still higher pressures would be indecisive, because the extrapolated separation factor would approach too close to unity to be able to distinguish with any accuracy a possible decrease in ln *f* due to turbulence.

Another possible explanation is that turbulence does introduce a new separating mechanism as borne out by the experiments of Bramley and Brewer. A theoretical investigation of this point would be desirable.

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<sup>&</sup>lt;sup>23</sup> A. Bramley and A. K. Brewer, Science **90**, 165 (1939); J. Chem. Phys. **7**, 972L (1939).

<sup>&</sup>lt;sup>24</sup> There is no doubt that some turbulence existed in the gas for these runs because of the 1.3 percent and 4.8 percent greater power inputs at pressures of 80.6 cm and 96.2 cm of Hg, respectively, needed to maintain the wire at 1500°K than was required for the previous runs under lamellar flow conditions.