Separation of Gaseous Isotopes by Diffusion

R. Sherr

pies of various molecules have been made.[6] To make these calculations, it is necessary to know the vibration frequencies of the molecules. The isomeric butenes are among the molecules which have been studied and it has sometimes been assumed that the structure of the two isomers is so similar that they may safely be assumed to have the same vibrational entropies, at least for the purpose of such calculations. Although no certain conclusions can be drawn until the spectra of these molecules have been completely analyzed, so that all the vibration frequencies are known, from the available data it would seem that this assumption is of doubtful validity. Table I shows

[6] K. S. Pitzer, J. Chem. Phys. 5, 473 (1937); L. S. Kassel, ibid. 4, 144 (1936).

that while the trans- and the cisbutenes have practically identical spectra when one considers the frequencies above 800 cm^{-1}, below that the spectra are quite different. Although this difference may be wiped out by the existence of frequencies which may not have been observed by us, such a possibility is rather remote and it would seem much more probable that there are real differences in these low frequencies. This seems not unreasonable, when one considers that in these low frequencies motions of the entire chain are involved and that these might well be different for the two compounds. It does not seem profitable, therefore, to attempt to draw definite conclusions about the existence of free rotation unless all the vibration frequencies are known.

Separation of Gaseous Isotopes by Diffusion

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The behavior of a set of mercury diffusion pumps has been investigated and the separation factors for a number of isotopic mixtures have been determined. Formulae have been derived which give the dependence of enrichment on the choice of reservoirs. In addition, Barwich's theory for the fractionation process has been extended to the case of finite reservoirs. The solution for this problem is a slowly converging series, but a qualitative picture of the dependence of equilibrium time on various factors is obtained. The separation factors which have been found for the

Hertz diffusion pumps for bromine in CH_3Br, carbon in CH_4, oxygen in O_2 and H_2O, Ne, and A are 1.005, 1.107, 1.045, 1.073, 1.198, and 1.154 per unit, respectively. Twelve pumps of the Hertz design, and seventeen modified units were used. The modifications were made in an attempt to improve the original design, but the factors obtained for these units are smaller. Auxiliary experiments indicate that the performance of the new units is seriously impaired by the presence of back diffusion and that increased factors can be obtained by eliminating the latter effect.

1. INTRODUCTION

IN recent years many problems in chemistry, biology, and physics have arisen whose solutions are facilitated by or dependent on the use of fractionated isotopes. There is consequently a growing interest in the more immediate problem of the separation of isotopes. Many fractionating devices, based on physical or chemical principles, have been applied to a number of isotopic elements with varying degrees of success.[1] One of the more promising methods has been that of separation by diffusion.

This method, although historically the first to be employed, gave no marked results until quite recently. The weakness of the earlier diffusion experiments lay in the fact that only one fractionating unit could be employed in a single operation. However, in 1932, G. Hertz[2] announced the invention of porous-walled units, so designed that a number of them could be connected in series, leading to multiple fractionation in one operation. These units have been used by a number of investigators[2,3] for the fractionation of neon, hydrogen, carbon and nitrogen.

[1] For recent reviews, see F. W. Aston, *Mass Spectra and Isotopes* (Edw. Arnold and Co., 1933). G. Champetier, Bull. Soc. Chem. 3, 1701 (1936).

[2] G. Hertz, Zeits. f. Physik 79, 108 (1932).
[3] Harmsen, Zeits. f. Physik. 82, 589 (1932); Harmsen, Hertz, and Schutze, Zeits. f. Physik 90, 703 (1934); Wooldridge and Smythe, Phys. Rev. 50, 233 (1936).

The difficulty of preventing the accumulation of impurities during operation and the possibility of obtaining higher separation factors led Hertz[4] to the development of the mercury diffusion pumps to be described below. The behavior of individual pumps under various conditions has been investigated by Barwich[5] and separation

experiments on neon, water vapor, and argon have been made by Barwich,[5] and Kopfermann and Kruger.[6]

The primary object of the present experiments has been a wider study of the potentialities of this method of separating isotopes. The usefulness of a fractionating device is determined by the degree of separation which can be obtained, by the yield,

[4] G. Hertz, Zeits. f. Physik **91**, 810 (1934).
[5] Barwich, Zeits. f. Physik **100**, 166 (1936).

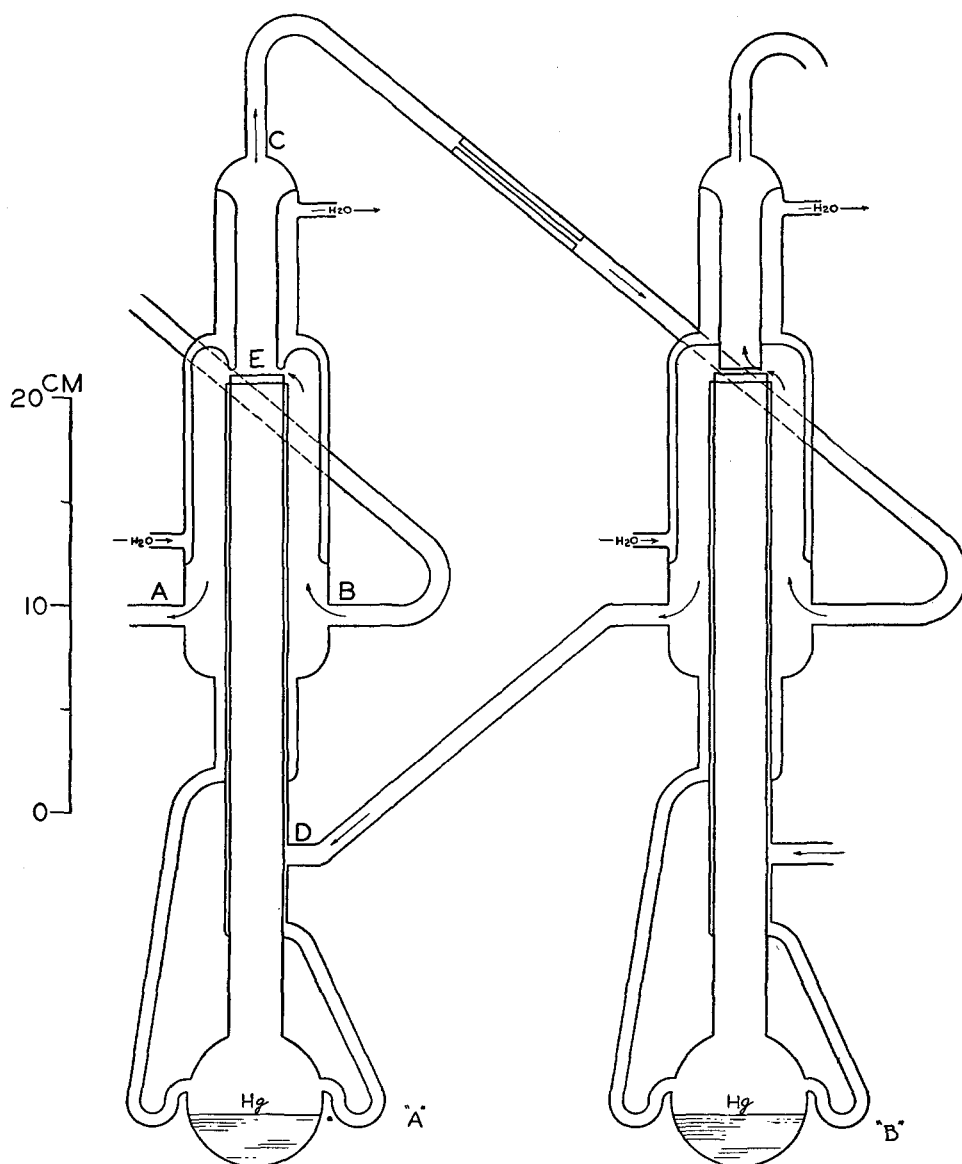[6] Kopfermann and Kruger, Zeits. f. Physik **105**, 389 (1937).

FIG. 1. Mercury diffusion pumps. "*A*" is the Hertz pump; "*B*" is a unit with a modified mercury jet. The pumps are connected in series as shown.
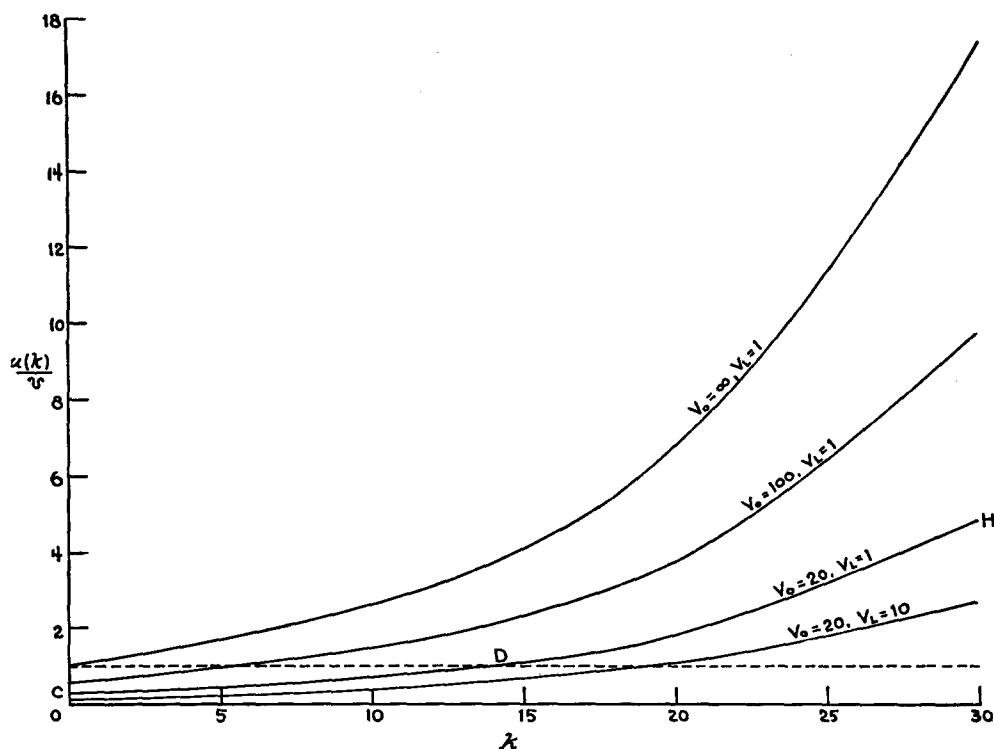
FIG. 2. Equilibrium gradient $u(k)/U$ for various choices of end volumes for small concentration of the heavier fraction. 0 corresponds to the light volume, 30 corresponds to both the Lth pump and the heavy volume. For $L=30$, $s=1.10$, and $V=0.5$. Volumes are expressed in liters.

and by the range of applicability. Since the first two of these requirements are limited by the efficiency of the individual units, the separation factors per pump for a number of gases have been determined.

The pumps are constructed and connected in series as shown in Fig. 1. Mercury vapor streams through the inner cylinder and forms a jet at E. At this point it is intercepted by a parallel cylinder of smaller radius which divides the stream so that only the inner part of it enters the cylinder. The outer portion of the stream acts as the separating agent while the central region serves to pump the gas which has diffused through the separating layer into the neighboring unit on the right. Since the lighter fraction of the gas has a greater diffusion coefficient, the gas pumped through C is enriched in this fraction. There is therefore a transport of the lighter fraction from left to right along the system of pumps through the upper connecting tubes. This is compensated by a flow of the heavier fraction

in the opposite direction through the lower connecting tubes.

The action of each pump is to increase the relative concentration of its lighter fraction by a quantity s, which defines the separation factor per unit. When equilibrium is reached, the initially uniform distribution has been transformed into an exponential concentration gradient. The light gas (the gas enriched in the lighter fraction) collects in a reservoir at the right end of the system, while the heavy gas flows into a reservoir at the left end.

2. THEORY

We confine our attention to the heavier fraction and assume for simplicity that its concentration is always much smaller than that of the lighter fraction. Then at equilibrium, by our definition of the separation factor,

$$u(k) = su(k-1), \qquad (1)$$

where $u(k)$ is the concentration of the heavier fraction in the kth pump; $k=0$ corresponds to the

reservoir at the "light" end of the system, and $k = L$ corresponds to both the Lth pump and the "heavy" reservoir. Since the total number of molecules is conserved,

$$V_0 u(0) + \sum_1^L V u(k) + V_L u(L)$$

$$= U(V_0 + V_L + LV), \quad (2)$$

where V_0 and V_L are the volumes of the "light" and "heavy" reservoirs, respectively, V is the volume of each pump, and U is the initial concentration. Transforming (2) by means of (1),

$$u(k)/U = As^k, \quad (3)$$

where A is given by

$$A = (V_0 + V_L + LV)/(V_0 + V\sum_1^L s^k + V_L s^L). \quad (4)$$

Equation (3) is represented graphically in Fig. 2. Since A is less than unity, there is an impoverishment of the heavier fraction in pumps near the light end, as is shown by the portion CD of the curve CDH. At D, enrichment begins, reaching a maximum value at $k = L$. The enrichment factor E for the entire system, defined as the ratio of the concentration at the heavy end to the initial concentration, is given by the ordinate of H. The separation factor S, the ratio of the concentrations at the heavy and light ends is given by H/C. By Eq. (3) it is seen that $S = s^L$ and is therefore independent of the choice of end volumes.

In order to obtain the maximum enrichment, A should approach unity. This is accomplished by making the light end volume as large as possible and the heavy volume small. The effect of the variation of volumes on the concentration gradient is indicated by the other curves of Fig. 2. When $V_0 = \infty$, it is seen that E and S are identical. The maximum enrichment is therefore limited by s^L.

It is to be expected that an increased enrichment is obtained at the expense of the time required to reach equilibrium, since a greater amount of gas must be circulated through the pumps. Consequently it is of interest to see how the equilibrium time depends on the size of the end volumes, the number of pumps and the

separation factor s. Barwich[5] has developed a theory for the time dependence of the separation process for the case of a small concentration of the lighter fraction of gas, with the assumption that the heavy end volume is infinite. It is possible, using Barwich's method, to investigate the case of finite end volumes by introducing more general boundary conditions.

If one assumes that the total volume V of each pump can be considered as concentrated in the immediate neighborhood of the mercury jet, the variation of the concentration $u(k)$ is due to three factors. Heavy gas from the $(k-1)$ pump is transported through D (Fig. 1) into the neighborhood of the jet (E) between the coaxial cylinders. Light gas leaves E through C, and flows into the $(k-1)$ pump. There is in addition a diffusion flow between E and the region $A - B$. If it is assumed that the gas in the latter region undergoes a complete mixing and that the gradient between E and $A - B$ is linear, one obtains the following equation: (cf. reference 5)

$$\partial u(k, \tau)/\partial \tau = (1/s)u(k+1, \tau)$$

$$- (1 + (1/s))u(k, \tau) + u(k-1, \tau), \quad (5)$$

where s is the separation factor, and τ is connected to the time t by the equation

$$t = \tau V(1/av + b/AD_0). \quad (6)$$

D_0 is the self-diffusion constant of the gas, A and b are the cross section and length of the path between E and $A - B$; a is the cross section of the connecting arms, and v is the velocity of flow through these tubes. In general $AD_0/b \gg av$, so that the second term of (6) may be neglected.

Equation (5) must be solved with the appropriate boundary conditions. For the Lth pump, it is assumed that V_L, the heavy end volume is added to V, so that the variation of $u(L)$ is due to the transport of heavy gas from the $(L-1)$ pump through D and the flow of light gas to the $(L-1)$ pump through C. Then

$$\partial u(L, \tau)/\partial \tau = \lambda_L \{u(L-1, \tau) - (1/s)u(L, \tau)\}, \quad (7)$$

where $\lambda_L = V/(V + V_L)$. This equation must be satisfied at all values of τ.

Similarly for $k = 0$, that is, for the light volume V_0, the variation of $u(0)$ is due to the flow of

light gas from the first pump and the flow of gas to the first pump. Assuming that the outflowing gas has the same composition as the gas in V_0 and that $AD_0/b \gg av$, one obtains

$$\partial u(0, \tau)/\partial \tau = \lambda_0 \{(1/s)u(1, \tau) - u(0, \tau)\}, \quad (8)$$

where $\lambda_0 = V/V_0$.

A third condition which must be satisfied by the solution of (5) is

$$u(k, 0) = U, \quad (9)$$

where U is the initial concentration of the heavier component.

When equilibrium has been reached, $\partial u(k)/\partial \tau$ vanishes. It is seen that $u(k) = su(k-1)$ satisfies Eqs. (5), (7) and (8). This steady state solution agrees with Eq. (1).

If we look for time dependent solutions of the form

$$u(k, \tau) = e^{-\mu \tau} s^k v(k) \quad (10)$$

we obtain from (5), (7), and (8) a set of three equations for $v(k)$ which constitute a boundary value problem which will have solutions for certain values of μ. Since there are $L+1$ points in the system, there should be $L+1$ distinct values of μ to each of which corresponds a function $v(k)$.

It can be shown that

$$v(k) = (1/s)^{k/2}\{Ce^{ik\theta} + De^{-ik\theta}\} \quad (11)$$

is a solution if θ is connected to μ by the relationship

$$\mu = 1 + 1/s - 2/(s)^{\frac{1}{2}} \cos \theta. \quad (12)$$

C and D are constants which must be chosen to satisfy the boundary conditions expressed by (7) and (8). On substituting (10) in these equations, one obtains two homogeneous linear equations whose determinant must vanish in order to give values of C and D other than 0.

L solutions of the determinantal equation are given by the values of θ which satisfy the equation

$$A + B \cos \theta = (\sin (L-1)\theta)/(\sin L\theta), \quad (13)$$

$$\text{where} \quad A = \frac{\lambda_L/(s)^{\frac{1}{2}} + \lambda_0(s)^{\frac{1}{2}} - (1+s)/(s)^{\frac{1}{2}}}{\lambda_0 + \lambda_L - \lambda_0\lambda_L}$$

$$\text{and} \quad B = 2/(\lambda_0 + \lambda_L - \lambda_0\lambda_L). \quad (14)$$

The additional solution of the secular determinant is the one corresponding to $\mu = 0$.

The complete solution is the sum of the functions corresponding to the different values of μ:

$$u(k, \tau) = \sum_n F_n e^{-\mu_n \tau} s^k v_n(k). \quad (10')$$

The functions $v_n(k)$ can be normalized by means of the orthogonality theorem appropriate to the problem,

$$\sum_{k=0}^{L} X_k s^k v_n(k) v_m(k) = \delta_{nm}, \quad (15)$$

$$\text{where} \quad X_k = \begin{cases} 1/\lambda_0 & \text{for } k=0 \\ 1 & \text{for } k=1, 2\cdots, L-1. \\ 1/\lambda_L & \text{for } k=L \end{cases} \quad (16)$$

The coefficients F_n are to be determined with reference to Eq. (9). Application of (15) to (10') yields

$$F_n = U\sum_k X_k v_n(k). \quad (17)$$

On normalizing the functions $v_n(k)$ and determining the corresponding F_n, one obtains as the complete solution

$$u(\tau, k) = Us^k(\sum_k X_k/\sum_k X_k s^k)$$
$$+ U\sum_n A_n s^{k/2} e^{-\mu_n \tau}\Theta_n(k), \quad (18)$$

where

$$\Theta_n(k) = (1 - \mu_n/\lambda_0) \sin k\theta_n - 1/(s)^{\frac{1}{2}} \sin (k-1)\theta_n$$

$$\text{and} \quad A_n = \sum_k X_k s^{-k/2}\Theta_n(k)/\sum_k X_k \Theta_n(k)^2.$$

It is observed that when $\tau = \infty$

$$u(\infty, k) = Us^k(\sum_k X_k/\sum_k X_k s^k)$$

$$= Us^k((V_0 + V_L + LV)/(V_0 + V\sum_1^L s^k + V_L s^k)).$$

This expression is identical with the steady state solution given by Eq. (3).

The determination of the coefficients A_n indicates that in general the series does not converge rapidly, so that a number of them must be calculated. It is difficult to estimate in a simple fashion the time required to reach, for example, 90 percent of the final equilibrium value for various choices of V_0, V_L, s, and L. An indication of the

TABLE I. *Variation of $1/\mu$ with V_0, with $V_L=1$, $V=0.5$, $s=1.235$, $L=30$. (The volumes are expressed in liters.)*

V_0	1	10	20	100	1000	∞
$1/\mu$	55.5	122.	215	666	2175	$\sim\infty$

TABLE II. *Variation of $1/\mu$ with V_L, with $V_0=20$, $V=0.5$, $s=1.235$, $L=30$.*

V_L	0	0.5	1	2	5	10	100
$1/\mu$	205	214	215	219	225	228	231

TABLE III. *Variation of $1/\mu$ with L, with $V_0=20$, $V_L=1$, $V=0.5$, $s=1.235$.*

L	10	20	30	50	100
$1/\mu$	92.5	185	215	223	223

TABLE IV. *Variation of $1/\mu$ with s, with $V_0=20$, $V_L=1$, $V=0.5$, $L=30$.*

s	1.235	1.182	1.119	1.085	1.043
$1/\mu$	215	141	100	90	80

influence of these factors is given by Tables I–IV which present the values of the smallest μ corresponding to different experimental conditions. After sufficient time has elapsed, the progress of equilibration is determined by the term belonging to this μ.

The longest period (corresponding to the smallest μ value) increases rapidly with the magnitude of the light volume, but is relatively insensitive to changes in the other factors. Calculations show that the additional μ values are appreciably larger and fairly insensitive. It is of interest to note that the dependence of the equilibrium time on the number of pumps L is similar to the dependence on the heavy volume V_L. This suggests that the equilibrium time is determined primarily by the volume of the system and by the rate of transport.

It is apparent that the attainment of large enrichment factors is accompanied by a considerable increase in the equilibrium time, since the former demands the use of indefinitely large light volumes. The separation factor, on the other hand, is independent of the choice of end volumes, and is determined experimentally by the ratios of the concentrations in the end volumes. The choice of reservoirs in the present experiments ($V_0=10$–20 liters, $V_L=1$ liter) was therefore such as to insure sufficient gas for analysis and

sufficient change in the concentrations at both ends to facilitate the analyses. An attempt to obtain maximum enrichment was made only in the case of methane. Here a twelve-hour run indicated a separation factor for $C^{13}H_4/C^{12}H_4$ in the neighborhood of 13, while the enrichment factor was approximately 5 since the enriched sample contained 5–5½ percent $C^{13}H_4$ (normal 1 percent). A continued run lasting 2½ days was then made, refilling the light reservoir from time to time with normal methane. The final analysis indicated 12.4 percent $C^{13}H_4$, showing that the full value of the separation factor had been obtained.

3. APPARATUS

Our experiments were performed on several sets of pumps. A system of fourteen units of the Hertz design (Fig. 1, "A") was used to investigate the separation of the isotopes of neon, oxygen, and bromine. In these pumps the cylindrical tube which divides the mercury stream is approximated by the cusp-like shape of the inner wall of the water jacket. They shall be referred to as type A.

An attempt was made to increase the efficiency of operation by securing a sharper division of the mercury stream by means of a cylindrical tube, ring-sealed into the neck of the inner wall of the water jacket (Fig. 1, "B"). Nine pumps having a radial clearance of 1 mm between the cylinders (type B) and eight with a clearance of 2 mm (type C) were constructed and added to twelve units of type A to form a final system of 29 units.

The pumps were tested for "pinholes" and thoroughly cleaned before mounting. In order to obviate the necessity of two gas-handling systems, the pumps were mounted around a narrow rectangular frame so that both ends were adjacent. In addition to the usual evacuating system, an automatic Toepler pump, was installed. This was fed by a diffusion pump and made it possible to remove desirable gases from the entire system in a short time.

During operation, it was sometimes necessary to run continuously over periods of several days. The system must therefore be completely "tight" in order to prevent impurities from interfering with the separation process or contaminating the
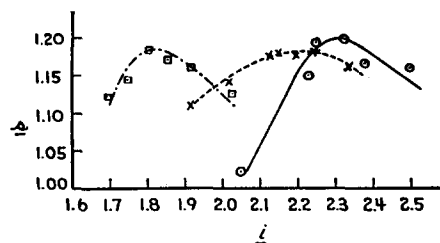
FIG. 3. Variation of separation factor with heater current for the A, B, and C units. Circle, for "A" pump; cross, for "B" pump; square, for "C" pump.

samples. While actual leaks were quickly eliminated, it took several weeks of continued pumping to get the system sufficiently hard. Several flushings with hydrogen, torching of the connectors, and heating of the inner surfaces of the pumps, (by turning off the water through the jackets for a short time), helped to remove adsorbed gases. The chief source of contamination proved to be impurities in the mercury, although this had been cleaned and distilled before using. When operations were begun, the pressure rose to 10^{-4} mm Hg in 24 hours and probably improved as time went on.

Heating was provided by Lavite electrical units whose hemispherical cavities were of the same diameter as the pump boilers. The heaters for a set of pumps were connected in parallel and the current through them was regulated by rheostats in the main leads. It was found that there was a considerable variation in the boiling rates between pumps which were receiving the same current. This variation was eliminated by adjusting the amount of asbestos packing until all of the pumps showed similar mercury streams, as indicated by a leak tester discharge.

Since it was often desirable to run the pumps overnight, safety devices were installed which turned off the heaters in case of failure of the water supply or of sudden increase of pressure in the pumps. The water devices were U tubes filled with mercury, the electrodes being so arranged that contact was maintained as long as there was a sufficient head. The pressure device was a mercury manometer with two tungsten electrodes arranged so that a gas pressure of several centimeters caused the mercury to make contact between them. Breaking of contact in the water gauges or making of contact in the gas manometer opened a heavy duty relay circuit breaker.

4. PROCEDURE AND ANALYSIS

The study of many gases is considerably simplified by the fact that the optimum conditions of operation do not vary appreciably from one mixture to another. These optimum conditions were found by observing the degree of separation obtained for various values of heater current and gas pressure.

As the heater current is increased, the over-all separation passes through a maximum. Barwich[5] found that the separation $vs.$ current curves for a single pump passed through a maximum, fell to a minimum, and rose again. In addition different pumps exhibited different characteristics. However, if capillary tubes were inserted in the upper connecting arms, (see Fig. 1), the characteristic curves showed single broad maxima which were more or less in the same region for different pumps.

Typical calibration curves are shown in Fig. 3. The experimental data has been reduced to average current and separation factor per unit. The latter is obtained by taking the Rth root of the over-all separation factor for each set of R pumps. The differences exhibited by the three types of pumps is to be attributed largely to difference of design, and partly to natural variations which depend on the packing. These curves were obtained simultaneously from observations on the composite system of 29 units. Analyses were made of samples taken from both ends of each set of pumps. It was found that each set gave a separation factor which depended only on its heater current and was independent of the operation of its neighbors.

Similar curves obtained for different initial gas pressures usually indicated a marked decrease of separation factor with increasing pressure. (Analyses of molecular oxygen failed to disclose such a decrease.) Consequently an initial pressure of 2 mm was found to be most desirable, larger pressures decreasing the separation, and smaller pressures decreasing the yield.

The analyses of the isotopic mixtures were made with mass spectrographs. An instrument of the Dempster type was connected directly to the system of 29 units by a long narrow pipe line. This mass spectrograph required about 0.5 cc of gas for analysis and gave reasonably accurate

relative abundances when the latter were in the region from 1 to 40. Other analyses were made by Dr. J. A. Hipple, Jr., and D. L. G. Smith of this laboratory on mass spectrographs which have been described elsewhere.[7]

5. RESULTS AND DISCUSSION

The results of experiments on a number of gases is given in Table V. The first column gives the gas or gases used, while the second gives the mass numbers of the molecules involved. The third and fourth columns show the experimental separation factors s, and log s, for the three types of pumps, A, B, and C. The fifth column gives the radii of the molecules. The radii of Hg and H_2O are those used by Barwich.[5] That of CH_3Br does not appear in the literature, so that a reasonable value has been assumed. The remaining values are taken from viscosity measurements.[8]

According to Barwich,[5] the separation factor s is given by

$$\log s = a\{(r_1+r_Q)^2(m_1)^{\frac{1}{2}} - (r_2+r_Q)^2(m_2)^{\frac{1}{2}}\},$$

where r_1 and m_1 are the radius and mass of the heavier molecule, r_2 and m_2 the corresponding quantities for the lighter molecule, and r_Q is the radius of the mercury molecule. The quantity a is a constant depending on the geometry of the mercury jet and on the rate of streaming of the vapor. If we designate the bracketed expression by F then

$$\log s = aF.$$

The values of F corresponding to the various mixtures are given in the last column of Table V.

The values of the separation factors which have

TABLE V. *Separation factors for various mixtures.*

GAS	MASS NUM-BERS	SEPARATION FACTOR			LOG s			RA-DIUS ×10⁸ cm	F
		A	B	C	A	B	C		
CH_3Br	94, 96	1.005			0.005			(1.7)	(1.225)
CH_4	16, 17	1.107	1.105	1.061	.1017	0.0999	0.0592	1.6	1.427
O_2	32, 34	1.045			.0440			1.48	1.879
Ne	20, 22	1.198	1.183	1.183	.1807	.1681	.1681	1.18	1.936
H_2O	18, 20	1.073			.0705			1.4	2.253
A	36, 40	1.154	1.140	1.057	.1432	.1310	.0554	1.49	3.51
$A-CO_2$	40, 44		1.7			.531		1.49	12.61
								1.70	

[7] W. Bleakney, Phys. Rev. 40, 496 (1932); Smith, Lozier, Smith and Bleakney, Rev. Sci. Inst. 8, 51 (1937).
[8] Herzfeld, *Handbuch der Physik*, Vol. 22.

been obtained for type A for Ne, H_2O, and A are to be compared with those obtained by Barwich[5] and Kopfermann and Kruger,[6] who worked with a system of A pumps. If their data is reduced to separation factor per pump, one obtains for Ne, H_2O and A, the values 1.2, 1.085, and 1.126, respectively. The slightly higher factor for water vapor is to be attributed to the lower pressure used by Barwich, 2 mm as compared with 5 mm used in the present work. The factor for argon is smaller than that which we have obtained and may be due to the possibility that equilibrium had not been reached in the experiment of Kopfermann and Kruger. It is of interest to note the high value obtained for the $A-CO_2$ mixture. While diffusion through porous-walled tubes gives the same factor for $A-CO_2$ as for Ne,[3] the present method gives a much larger value for the former mixture.

Barwich[5] found that runs with equal mixtures of the rare gases under identical conditions gave separation factors which agree well with his theoretical formula. The present experimental values of log s do not, however, indicate the predicted linear dependence on F. This discrepancy is probably due to a number of effects.

The formula derived by Barwich is at most only approximate, due to the simplifying assumptions made, especially those concerned with the shape of the mercury stream. The latter was assumed to be cylindrical, whereas actually it is fan-shaped and changes with gas pressure and different gases. The variation of separation factor with pressure and initial concentrations (as found by Barwich by varying the relative concentrations of the mixtures mentioned above) is not unwarranted, although not contained in the theoretical formula. In the present experiments, the initial concentrations ranged from 0.02 percent H_2O^{18} to 50 percent for the bromine isotopes. It is difficult to say whether this variation of concentrations is wholly responsible for the erratic behavior of the separation factors. It is possible that the peculiarities of the gases with respect to interaction with the hot mercury stream, adsorption on the glass walls, and back diffusion may strongly influence the fractionating process.

Observation of the mercury vapor jet of the "A" pumps by means of a leak tester discharge

reveals a jet so ill defined that it seems as if its separating efficiency is almost fortuitous. Similar observations on the type B and C units show jets in which the separating and pumping layers are quite distinct and well defined. Despite the better geometry of the latter pumps, their efficiency is smaller. Furthermore, the C units are less efficient than the B, although the diffusion path in the former is larger.

Experiments with a fourth type of pump have given the reason for this failure to realize our expectations. The newest unit is essentially the same as the C pump except that it is inverted, so that the mercury streams downward. It was observed that in the other pumps large globules of condensed mercury would fall and tear holes in the mercury stream through which gas could pass through the jet without being fractionated. This effect probably accounts to a large extent for the dropping off of the separation factor with high heating current. The new pump was therefore inverted in order to eliminate this.

Tests with a mixture of $Ne - CO_2$ showed that this pump, with improved geometry, was even more inefficient. In addition to the smaller separation factor, the range of current was considerably narrower. These tests were made allowing the gas to circulate freely. The stopcocks in the system were then so adjusted that gas could enter or leave the light volume only by diffusing through the mercury jet. With a current which previously had given no separation a practically complete fractionation was obtained. Neon was run under similar conditions and gave a separation factor for $Ne^{20} : Ne^{22}$ of 1.5 or larger, as compared with the value of 1.2 obtained previously. Because of the extreme slowness with which this

separation progressed, it would not be practical despite the fact that the present pump is equivalent to more than two of the other units.

However, these tests show a direction which improvement may take. The transport of the lighter fraction in a fractionation system is insured by the pumping action of each unit. The opposite flow of the heavier fraction through the lower connectors (Fig. 1) is due to the pressure gradient which is established. This latter mass flow is offset by diffusion and unless there is a sufficient pressure gradient, the transport is nullified and the fractionating effect of the jet is not realized. While the "A" units pumped so rapidly that capillaries had to be introduced in the upper arms to reduce the transport, the "B" and "C" units gave rise to much smaller pressure differences. The resulting ill effects of back diffusion should be eliminated by using small capillaries in the lower connectors or by introducing circulating pumps.

This method of separation is applicable to a wide range of elements and gives factors which are favorable compared to those given by other methods. Since the separation factor increases exponentially with the number of pumps, any desired enrichment can be obtained by employing a sufficient number of units. The yield differs for various mixtures, but is of the order of 1 cc of enriched gas at NTP per day.