

Performance of a Hot-Wire Thermal Diffusion Column*†

J. W. CORBETT‡ § AND W. W. WATSON
 Sloane Physics Laboratory, Yale University, New Haven, Connecticut
 (Received August 30, 1955)

The performance of a carefully constructed, metal, hot-wire, thermal diffusion column is examined in the regions of laminar and turbulent gas flow in the column for several hot-wire temperatures using the normal argon isotopic mixture as a column gas. A new experimental method is employed which simplifies and increases the accuracy of the experiment. At the lowest temperature examined, quantitative agreement with theory is observed without the use of a parasitic remixing factor. The effect of spacers on the center wire on column performance was also examined. No significant effect was observed, this being attributed to the careful construction of the column.

INTRODUCTION

THE thermal diffusion column has been proven to be a valuable tool for the enrichment of the isotopes of gases. Furry and Jones^{1,2} extended the theory of the plane-parallel-wall column of Furry, Jones, and Onsager,³ to the extreme-cylindrical, or hot-wire column, and obtained approximate solutions by considering the molecules of the gas Maxwellian, with a thermal diffusion factor independent of temperature. Much work has been done in this laboratory^{4,5} to obtain experimental data for comparison with this theory.

Qualitative agreement with the theory has been obtained, but there were always indications of a sizable parasitic remixing factor, K_p , in the apparatus. In view of the techniques then available, this parasitic factor was not unreasonable. We therefore proposed, by employing new constructional techniques, to approach as closely as feasible to the theoretical requirements on the apparatus, and to measure the separation factor *versus* pressure dependence by examining the equilibrium behavior of the column for various temperature ratios.

APPARATUS

A column employing a metal (monel) for the outer wall of the gas chamber was carefully constructed to be as straight as possible, and hung vertically on the wall. This outer wall had internal irregularities less than two percent of the wall thickness and less than five percent eccentricity. Figures 1 and 2 indicate some of the details of the construction. The center wire (20-mil tungsten) was centered through electrically insulated

"O" ring seals and kept taut by a 4-lb weight. This avoided the use of centering cross-pieces spot-welded to the wire. The length of the gas chamber was 182 cm. The lengths of wire employed for equilibrium separation factor measurements were carefully measured so that the initial length plus the thermal expansion due to the increase to operating temperature would just equal 182 cm. Experiments using a glass column indicate that the center wire was at operating temperature to within a quarter of an inch of its ends.

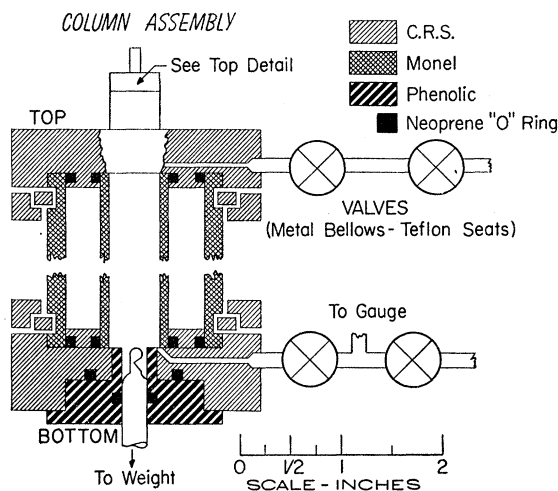


FIG. 1. Details of column construction.

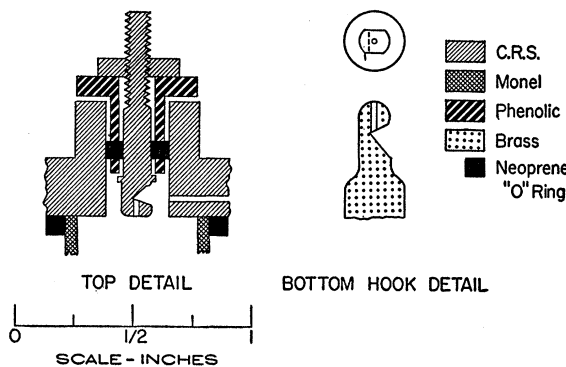


FIG. 2. Details of column construction.

* Part of a dissertation submitted by J. W. Corbett in partial fulfillment of the requirements for the degree of Doctor of Philosophy at Yale University.

† This work was supported in part by the U. S. Atomic Energy Commission.

‡ General Electric Charles A. Coffin Fellow.

§ Now at the General Electric Research Laboratory, Schenectady, New York.

¹ W. H. Furry and R. Clark Jones, *Phys. Rev.* **69**, 459 (1946).

² R. Clark Jones and W. H. Furry, *Revs. Modern Phys.* **18**, 151 (1946).

³ Furry, Jones, and Onsager, *Phys. Rev.* **55**, 1083 (1939).

⁴ R. Simon, *Phys. Rev.* **69**, 596 (1946).

⁵ J. Donaldson and W. W. Watson, *Phys. Rev.* **82**, 909 (1951).

TABLE I. Critical pressures and Reynolds' numbers for onset of turbulence in a hot-wire thermal diffusion column. p_c is the critical pressure in cm of Hg; R is the Reynolds' number calculated by use of Eq. (120), reference 2; T_2 is the hot-wire temperature; T_1 is the cold-wall temperature.

T_2/T_1	Simon ^a		Present work	
	p_c	R	p_c	R
2	42	42	101	14
3	43	47	132	17
4	62	62	168	19
5	72	72	178	15
6	80	84	206	13

^a Interpolated from Table I, reference 4.

The effect of cross pieces on the center wire on column performance was also examined in the same manner as Donaldson and Watson⁵ did for a glass column. Then single 40-mil nickel cross pieces were spot-welded to the wire at 10-cm and 5-cm intervals.

The gas used was the ordinary cylinder isotopic mixture of argon 36, 38, and 40. The method of following the progress of the separation was to take samples of the gas, each $\frac{1}{2}$ cm³, and measure the relative concentrations with a Consolidated Engineering model 21-401 mass spectrometer. At no time did an impurity component of the gas (i.e., N₂, O₂, CO₂) exceed the amount of argon-36 in the sample. Percentages were computed solely on the basis of the three argon components. Argon-38 can be ignored so far as contributing to the column processes.⁴

The first operation with the column was to run a series of experiments in which the approach to equilibrium was investigated. Each point of this investigation consisted of from five to fifteen samples taken from the top in as quick succession as possible. As will be noticed in Fig. 1, there is a small dead volume between the top of the column proper and the valve. This volume acts as a nonconvective end volume. The succession of samples effectively measures the concentration distribution into and down along the column. This was necessary to establish which sample gave a representative measure of the performance of the column—the fourth being so chosen.

TURBULENCE MEASUREMENTS

Data were taken of the onset of turbulence in the column by measuring the temperature of the hot-wire (by electrical resistance and extension) *versus* the gas pressure. The break in the temperature *versus* pressure curve at a critical pressure, p_c , is ascribed to a change in the convective gas flow from laminar to turbulent.^{4,6} Onsager and Watson,⁶ in a concentric-tube column, found turbulence occurring for a Reynolds' number of the order of 25. Our results are compared to Simon's⁴ in Table I.

⁶ L. Onsager and W. W. Watson, Phys. Rev. **56**, 474 (1939).

SEPARATION FACTOR MEASUREMENTS

The separation factor, q , is conventionally defined as the ratio of the relative concentrations at the top of the column divided by the ratio of the relative concentrations at the bottom. This necessitates making a measurement of the concentration of the argon-36 at the bottom. Since the column was initially filled with a gas with only 0.33% A³⁶, the concentration at the bottom at equilibrium is very small. This fact has resulted in rather large experimental errors in this type of work in the past. Another procedure was followed in the present work.

The differential equation [Eq. (157), reference 2] which describes the concentration, c , in a column of length, L , and with no end-volumes, as a function of the time, t , and the distance, z , from the bottom of the column, has a solution of the form [see Eq. (160), reference 2]

$$c(z) = ke^{2Az} + f(t), \quad (1)$$

where the separation coefficient, $2A$, is defined as the ratio of H to K , H being the transport factor, K the sum of the remixing factors and $k = 2ALc_0/(e^{2AL} - 1)$, with c_0 the initial concentration in the column. The function, $f(t)$, approaches zero as $t \rightarrow \infty$. We define a z_0 such that $c(z_0) = c_0$ as $t \rightarrow \infty$; hence

$$c_0 = 2ALc_0e^{2Az_0}/(e^{2AL} - 1). \quad (2)$$

with $x \equiv 2AL$ and $\xi \equiv z_0/L$, we obtain

$$\xi = \frac{1}{x} \ln \left\{ \frac{e^x - 1}{x} \right\}. \quad (3)$$

We define a new equilibrium separation factor, q^* :

$$q^* = \left(\frac{c}{1-c} \right)_{\text{top}} / \left(\frac{c}{1-c} \right)_{\text{initial}} = \exp\{2AL(1-\xi)\}, \quad (4)$$

which involves knowing the initial and final concentrations at the top of the column, thereby simplifying the experiment and increasing the accuracy. Since ξ is a function of $2A$, the solution of Eq. 4 for the quantity $2A$ must be an iterative one. However, for small x , or $2AL$, $\xi \sim \frac{1}{2}$ (see Table II), the method is not difficult.

The values used in the theoretical calculations as well as the experimental data are given in Table III. The experimental errors are estimated as 10 percent of

TABLE II. Solutions to Eq. (3).

x	ξ	x	ξ
0.005	0.501	0.9	0.536
0.01	0.502	1.0	0.540
0.05	0.504	1.5	0.561
0.1	0.505	2.0	0.581
0.3	0.511	5.0	0.674
0.5	0.520	7.5	0.732
0.6	0.525	9.0	0.765
0.7	0.529	10.0	0.768
0.8	0.533		

TABLE III. Comparison of experimental and theoretical column performance. $r_1=0.500$ in., $r_2=0.020$ in., $r_1/r_2=25.0$. $L=182$ cm; $T_1=280^\circ\text{C}$. The following quantities are values interpolated to T_1 . $D=1.56\times 10^{-1}$ cm²/sec, $\eta=2.1\times 10^{-4}$ poise, $\rho=1.78\times 10^{-3}$ g/cc, $\alpha=1.32\times 10^{-2}$.

T_2/T_1	p inches of Hg—absolute	$2A$ ($\times 10^{-3}$ cm ⁻¹)	
		Theoretical	Experimental
2	9	3.0	3.9
	15	4.7	4.9
	20	4.20	4.25
	25	3.00	3.05
	30.5	2.20	2.20
	41	1.45	1.45
	50	0.87	0.87
3	60	0.59	0.59
	10	5.5	5.5
	15	8.4	8.2
	20	8.4	6.2
	25	6.9	4.9
	30	5.2	3.9
	41	3.1	1.95
4	51	2.0	1.40
	10	5.4	7.1
	15	10.0	11.7
	20	11.5	13.6
	25	11.0	11.6
	30	9.2	9.7
	39	6.2	6.8
	51	3.7	4.7
	71	2.05	3.15
	91	1.30	2.35
110	0.87	2.05	
5	8	4.7	4.7
	11	7.0	10.0
	16	10.5	14.3
	21	13.5	14.2
	27	13.0	9.3
	36	8.5	6.6
	43	6.3	5.3
	59	3.6	3.9
	76	2.1	3.1
	6	10	3.0
20		10.0	15.2
25		12.7	15.7
32		14.0	12.7
42		12.0	10.7
50		9.8	6.5
72		5.2	5.1
90		3.5	3.6
3	Experimental—spacer every 10 cm		
	9	3.30	
	11	4.8	
	13	8.4	
	17	9.6	
	21	7.6	
	26	6.4	
30.5	5.3		
35	4.0		
3	Experimental—spacer every 5 cm		
	8	5.0	
	12	6.1	
	14	6.6	
	18	6.5	
	22	5.4	
	30	3.7	

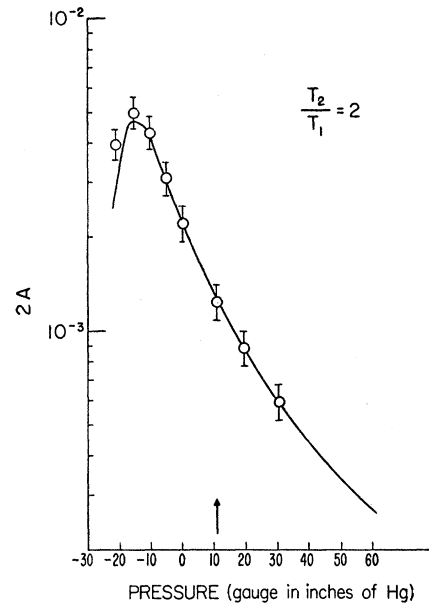


FIG. 3. Plot of separation coefficient, $2A$, versus gas pressure in the column for $T_2/T_1=2$. Solid line is the theoretical curve, with experimental points and experimental errors superimposed. Arrow indicates the onset of turbulence.

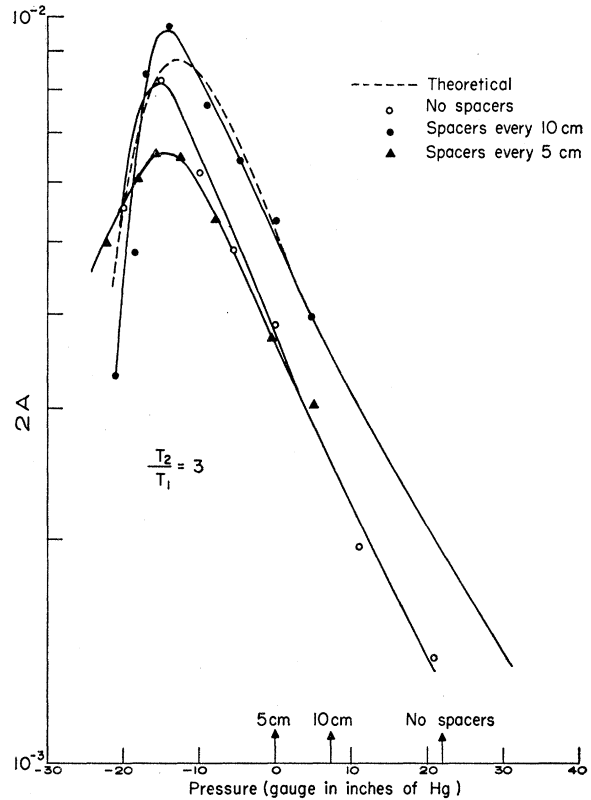


FIG. 4. Plot of separation coefficient, $2A$, versus gas pressure in the column for $T_2/T_1=3$, including the theoretical prediction for the performance with no spacers, and experimental results for a spacer every 10 cm and every 5 cm. The arrows indicate the onset of turbulence for the condition as labeled.

the values quoted. The theoretical values employ only quantities directly calculable from the theory. That is, no parasitic remixing factor is used.

CONCLUSIONS

The results presented in Table III show markedly better agreement with theory than similar results from glass columns. Indeed, the $T_2/T_1=2$ data are in excellent quantitative agreement with the theory, without employing a parasitic remixing factor (see Fig. 3). It is felt therefore that no appreciable parasitics existed in the column. Solutions of the theory which take into account the non-Maxwellian character of the gas as well as the temperature dependence of the thermal diffusion constant⁷ may very well explain the discrepancies at higher temperatures. The temperature

⁷ R. C. Srivastava, Proc. Phys. Soc. (London) A68, 294 (1955).

dependence of the thermal diffusion constant can best be represented as a linear variation with $\ln T$. The temperature dependence alone cannot account for the whole of the discrepancy, since this will increase the theoretical values of $2A$, whereas some of the experimental deviations are in the other direction.

The improvement in performance with increasing number of spacers as noticed by Donaldson and Watson⁵ was not observed (see Fig. 4). We feel that such improvement can occur when the construction is such that the spacers improve the geometry of the column. If the construction approaches the theoretical ideal, i.e., no irregularities in the wire-to-wall spacing, column absolutely straight, etc., spacers cannot improve the performance.

Finally, we believe that the Furry, Jones, and Onsager theory is adequate, but that appropriate solutions have not been effected for all cases.

Color Centers in Alkali-Silicate Glasses Containing Alkaline Earth Ions

RYOSUKE YOKOTA

Matsuda Research Laboratory, Toyko-Shibaura Electric Company, Kawasaki-shi, Japan

(Received May 18, 1955; revised manuscript received August 26, 1955)

Samples of alkali metal oxide-silica glasses with various kinds and amounts of alkaline earth oxide calcium oxide and silica were prepared, x-irradiated, and studied by spectrophotometry. Bleaching experiments with visible light were carried out. No effect of alkaline earth content upon the absorption spectra was detected.

Room temperature bleaching of a $K_2O-SrO-SiO_2$ glass with light of wavelength corresponding to the x-ray induced absorption band at 1.95 eV, decreased absorption generally but failed to create a new absorption band such as the Z_1 band found in KCl, containing $SrCl_2$, that has been irradiated and then bleached.

Comparisons are made with alkali-halide systems containing alkaline earth halides. It is suggested that there are inadequacies in the quasi-crystalline model of glass when applied to these glasses.

I. INTRODUCTION

IN an earlier report¹ on alkali silicate and borate glasses, the author presented evidence suggesting that the visible band found in x-irradiated samples is due to electrons trapped by oxygen vacancies adjacent to alkali ions and that the ultraviolet band is due to positive holes trapped by alkali ion vacancies which are adjacent to oxygen ions.

Warren and others² have demonstrated short-range order in glasses of simple composition and the author has used this to construct an analogy between the color centers of alkali-silica glasses and those in crystalline alkali halides. The attempt seems to be successful. The present work extends the study to alkali-silica glasses containing alkaline earth oxide.

¹ R. Yokota, Phys. Rev. 95, 1145 (1954); 93, 896 (1954).

² B. E. Warren, J. Appl. Phys. 8, 645 (1937); Warren, Krutter, and Morningstar, J. Am. Ceram. Soc. 21, 259 (1938); I. Šimon and H. O. McMahon, J. Chem. Phys. 21, 23 (1953); I. Šimon and H. O. McMahon, J. Am. Ceram. Soc. 36, 160 (1953); R. Yokota, J. Phys. Soc. Japan 5, 295 (1950); 6, 489 (1951); M. L. Huggins, J. Phys. Chem. 58, 1141 (1954).

X-ray studies³ have indicated that the structure of $Na_2O-CaO-SiO_2$ glass is similar to that of Na_2O-SiO_2 glass with some of the sodium ions replaced by calcium ions.

In view of these facts, we have studied silica glasses containing alkaline earth oxide as well as alkali metal oxide to learn whether, in these glasses, the x-ray coloration can be compared with that in alkali halides containing alkaline earth halides and whether the comparison will support the "quasi-crystalline model" for glass.

Most commercial glasses are silicate and borosilicate glasses which contain alkali and alkaline earth ions. The method of preparation of the purest glass is the same as before.⁴ The alkaline earth oxides we used were Merck guaranteed reagent grade carbonates. Every sample was annealed.

³ J. Bischof, J. Am. Ceram. Soc. 24, 262 (1941).

⁴ R. Yokota, reference 2.