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A Note on the Construction and Operation of a Thermal Diffusion Column for the Separation of Isotopes

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SINCE the demonstration by Clusius and Dickel[1] that the Chapman effect coupled with a thermal syphon action could be used to separate isotopes, a number of investigators have started both theoretical and experimental studies on such columns.[2]

We have constructed a 40-foot column in which the cold surface is a two-inch standard galvanized pipe concentric with a one-inch pipe for the hot surface. This leaves an annular space of 9 mm with a mean circumference of 13.8 cm for the methane. A water jacket made of four-inch standard pipe surrounds the two-inch pipe for cooling purposes. An oxygen tank was welded on top to give a volume of 30 liters above the annular space. A one-liter glass flask fastened with a sealing wax joint to the two-inch pipe gives a total volume of 40 liters. The sampling and evacuating system along with the manometer and Pirani gage are attached to the liter flask. The one-inch pipe is closed at the lower end and is suspended from the top of the oxygen cylinder such that there is a clearance of two inches at the bottom to allow for expansion. Four sets of three steel pins each maintain the spacing in the annular space uniform. These are made from $\frac{1}{8}$-inch steel rod tapered to obtain a small point of contact with the cold surface.

The first heating element used was constructed of two loops of No. 16 nichrome wire in parallel insulated from the inside of the one-inch pipe by means of three-inch porcelain insulators ($\frac{1}{4}$ in. hole, $\frac{1}{16}$ in. wall thickness). Connection to a 220-volt line through suitable rheostats was made with heavy copper wire just below the cylinder. A second heating element was made with the same insulators using No. 14 nichrome wire covered with an asbestos sheath. These heating elements were unsatisfactory because there was not adequate provision for thermal expansion and moisture from the asbestos sheathing distilled to the top part of the column. The third heating element was constructed from one loop of No. 10 nichrome wire enclosed in double bore oval insulators (3 in.$\times\frac{1}{2}$ in.$\times\frac{1}{4}$ in.). These in turn were placed in a system of cylindrical insulators ($5\frac{1}{2}$ in.$\times\frac{9}{16}$ in. bore$\times\frac{3}{32}$ in. wall thickness). The whole was enclosed in a woven copper sheath for easy removal. Two thermocouples were imbedded in the outside of the one-inch pipe three feet from the top and bottom, respectively, and brought out through sealing wax joints.

Several experiments were performed to study the operation and behavior of the column. The abundance ratio of $C^{13}H_4$ was changed by a factor of about 2 (see Table I).

In experiments I and II using the first and second heating elements, respectively, the temperature at the two ends of the one-inch pipe were nearly the same. In Experiment III with the third heating element, however, the difference between the two ends was about 100°C. The reason for this difference in behavior of the two types of heating elements may lie in their construction in the sense that different convection currents exist within the one-inch heating tube. We have studied this point with the second heating element by filling the spaces between the insulators and the pipe with sand. However, the temperature difference between the top and bottom persisted, while the average temperature dropped from 291°C to 226°C with 0.1 mm pressure in the annular space and to 188° with 1 mm

TABLE I.

Experiment	Kilowatts	mm Hg CH_4	Cold Temp.	Ave. Hot Temp. °C	Time	Percent $C^{13}H_4$
I	3.68	180	20	236	max. conc.	2.2
II	8.40	170	20	330	$2\frac{1}{2}$ days	2.4
III	5.27	700	20	264	2 days	2.0

[1] Clusius and Dickel, Naturwiss. **26**, 546 (1938); **27**, 148 (1939).
[2] Brewer and Bramley, Phys. Rev. **55**, 590 (1939); Furry, Jones and Onsager, Phys. Rev. **55**, 1083 (1939); Gillespie, J. Chem. Phys. **7**, 530 (1939).

pressure using the same wattage. The average temperature and the temperature difference increased as the pressure in the annular space was progressively decreased. This seemed to indicate that the remaining gas inside the one-inch heating tube still carried heat to the top. We therefore evacuated the heating unit with the result that the temperature at the top and bottom reach the same value with a low pressure in the annular space. These points are being studied further because they may be of importance in the extent of separation.

It is important to use pure methane in the experiments since the heavy impurities will concentrate at the bottom and may interfere with the separation. This is shown by the relative heights of the peaks for the following masses on a mass spectrometer: Original methane for Exp. I and II: mass 16 (100); 17 (1.9); 28 (21); 30 (2.2); 32 (1.) and 44 (1.8). Final gas: mass 16 (100); 17 (3.2); 28 (128); 30 (9.6); 32 (6.5) and 44 (6.7). This sample of methane was prepared by distillation at liquid-air temperature by using a standard Podbielniak low temperature fractionating column arranged for analytical purposes and fitted with rubber connections. Evidently masses 28, 30, 32 and 44 were not removed efficiently. Owing to differences in ionizing efficiencies, the mass spectrometer does not give the correct values for the relative abundance of molecules in a gas mixture. This was further shown by the ratio of mass 16 to 28 in a gas bought as 96 percent methane, this ratio being 100 to 80 for the height of the peaks. In order to determine the usefulness of the mass spectrographic method for analyzing gases, a known mixture of nitrogen and methane containing 80 percent of the latter was analyzed in the same apparatus. The relative heights of the peaks were found to be 16 (100) and 28 (160). That is, the nitrogen peak is of the order of eight times higher than one would predict for equal ionizing efficiencies. However, the mass spectrometer does afford a satisfactory method of comparison of one sample with another. From the above figures it is also seen that mass 28 as well as the other impurities increases over fivefold which may markedly affect the separation attained in a given column.

In order to obtain very pure methane, we constructed an all glass distilling system and prepared a gas sample for Experiment III showing the following relative peaks: mass 16 (100); 17 (1.29); 28 (0.6); 30 (0.02); 32 (0.002); 44 (0.04). Final values: mass 16 (100); 17 (2.0); 28 (12.7); 30 (0.1); 32 (0.002); 44 (0.1). It is seen that this was a much purer sample of gas but the impurities concentrate several-fold in the lower sections of the column. By drawing these gases off, a further purification of the methane can be achieved.

When the results obtained so far are not as encouraging as the theory[2] would predict for this type of column, it is hoped that elimination of some of the difficulties described may give the predicted results. The optimum conditions of temperature, pressure, heavy carbon supply, surface, and space separation are yet to be determined. It is also not known whether a column with concentric tubes is as effective as a hot wire, although its capacity is greater.

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