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INVESTIGATION OF THE RECTIFICATION OF A HELIUM ISOTOPE MIXTURE IN PACKLESS COLUMNS

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A study was made of the rectification of He^3—He^4 mixtures in miniature packless rectification columns at temperatures of 2.16 and 1.5°K. It is shown that such columns can be used to separate He^3 from solutions with a low content of the light isotope. It is suggested that packless columns be used to obtain He^3 of very high purity.

THE problem of the separation of helium isotopes, which had to be faced some years ago because of the need to remove He^3 from a natural mixture of isotopes, was solved successfully by the use of a combination of low-temperature methods. Because of the very low content of He^3 in natural helium, apparatus developed for this purpose[1-3] is comparatively complex. Since the mixtures currently used in physical investigations are considerably richer in He^3, simpler apparatus can be used to separate the isotopes.

The extension of the investigations of the physical properties of helium isotope solutions and the limited stocks of the rare isotope He^3 make the development of such apparatus a pressing problem.

We investigated the rectification of mixtures of helium isotopes in packless columns. In such columns, the simplicity of the apparatus is combined with a high separation power at low vapor velocities. A very important advantage of this method is the short duration of the operation, which makes it possible to remove almost all He^3 from a mixture in one cycle.

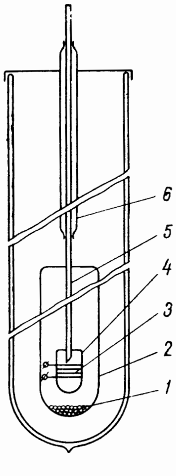

FIG. 1. Rectification column.

Figure 1 shows schematically a section through one of the investigated columns. The main part of the column is a thin-walled cylindrical tube 5 in which the rectification takes place. We used German silver or nickel tubes with an internal diameter from 1.2 to 2.0 mm and 70—200 mm in height. A glass boiler 4, having a volume of 0.3—4.0 cm^3, was soldered with kovar to the lower part of the tube and a constantan-wire heater 3 was wound on the boiler. The boiler and the lower part of the tube were placed in a vacuum jacket 2, which ensured that the rectification was adiabatic. High vacuum was maintained in the jacket by means of activated carbon 1. Another vacuum jacket 6 was used to suppress the influence of a change of the helium level in the external Dewar flask. The part of the tube between the jackets 2 and 6 acted as a column condenser. The column was placed in a conventional glass Dewar flask and was connected by a thin capillary to a mercury manometer and an inlet and outlet system. The majority of the experiments were carried out at helium temperatures of 2.16 and 1.50°K in the surrounding bath.

A typical experiment was carried out as follows. After cooling a column to a temperature of about 1.5°K, the mixture to be treated was condensed in the boiler 4, the temperature in the Dewar was increased to 2.16°K and the heater 3 was switched off. The current through the heater was selected to ensure the necessary vapor velocity in the column. In our experiments, this velocity was 0.5—1.7 cm/sec for a heater power of $\approx 10^{-3}$ W. When the heater was switched on, countercurrents of vapor and liquid were produced in the column and these resulted in a concentration of He^3[+] at the top of the column. Simultaneously, the temperature of the boiler was increased to a value at which the saturated vapor pressure of the initial solution became equal to the pressure in the column.

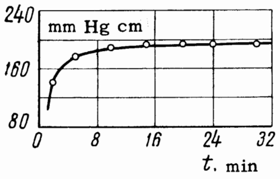

FIG. 2. Curve showing establishment of a steady state; T = 2.16°K, u = 1 cm/sec.

The variation of the pressure p in the column with time, representing the establishment of a steady state, is shown in Fig. 2. This dependence was obtained for a vapor velocity of 1 cm/sec. It is evident from Fig. 2 that approximately after 30 min the pressure in the column became equal to the saturated vapor pressure above pure He^3. After the establishment of a steady state, the vapor phase was extracted from the upper part of the column. The rate of extraction of the vapor was deduced from the rate of increase of the pressure in a calibrated volume. The investigated rates of extraction corresponded to the range of reflux ratios R from 10 to 360. The concentration of He^3 in the extracted mixture was estimated directly from the vapor pressure in the column. (To allow for the temperature drop across the tube wall, we carried out control experiments with pure He^3 and He^4 at corresponding vapor velocities.) The accuracy of the determination of the concentration was 0.3% for the extracted He^3-rich mixtures and 0.5% for He^4-rich mixtures.

A typical dependence of the concentration x of He^3 in the extracted mixture on the amount extracted is shown by curve 1 in Fig. 3. This curve was obtained in an experiment using a column 70 mm high, 1.4 mm in diameter, and with a boiler having a volume of 3.8 cm^3. The vapor velocity was 1.7 cm/sec and the reflux number was 116. It is evident from Fig. 3 that under these experimental conditions practically pure He^3 was obtained and the volume of the intermediate fraction did not exceed 40 cm^3 of the gas under normal conditions. Estimates showed that a considerable part of this fraction resulted from the retention of the vapor phase in the column. Therefore, in order to reduce such retention, it was very desirable to reduce the temperature at which the rectification was carried out.

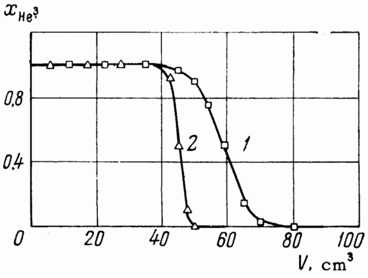

FIG. 3. Dependence of the concentration of He^{3+} at the column outlet on the yield of the substance at various temperatures in the Dewar flask: 1) T = 2.16°K; 2) T = 1.5°K.

It follows from Kuhn's formula[4]

$$n = 4LD/r^2u \tag{1}$$

(n is the number of theoretical trays when the outlet is closed, L is the height of the column, d is the diffusion coefficient, r is the radius of the column, and u is the velocity of the vapor) that the number of theoretical trays in a column decreases considerably when the diffusion coefficient is reduced; however, this reduction is compensated by an increase in the enrichment coefficient δ so that the separation, represented by the product $n\delta$, also increases considerably when the temperature is lowered. Curve 1 in Fig. 3 represents a rectification carried out at 1.5°K at the same power supplied to the boiler. We can see that the separation was considerably improved and the volume of the intermediate fraction became less than 15 cm^3.

The curves obtained were used to calculate the experimental values of n from the formula[4]

$$n_{\text{exper}} = \frac{1}{\delta} \ln \frac{c(1 - c_b)}{(1 - c)c_b}, \tag{2}$$

where c and c_b are the molar concentrations of He^3 at the top of the column and in the boiler. In this calculation, we used the points on curves 1 and 2, corresponding to different values of c_b. The number of theoretical trays in a column of 1.4 mm diameter and of L = 70 mm height, operating at T = 2.16°K, varied from 6 for $R \approx 40$ to 9.4 for $R \approx 120$. When the rate of extraction was small, the average values of n_{exper} agreed, to within 10—15%, with the values calculated using Eq. (1) for vapor velocities greater than 1 cm/sec. At lower values of these velocities, the experimental values were found to be much less than the calculated ones because the investigated conditions differed considerably from those obtainable when the outlet is closed.

The degree of enrichment $q = c(1 - c_b)/(1 - c)c_b$ for reflux numbers $R \approx 100$ was of the order of 10^4 at T = 2.16°K and of the order of 10^5 for T = 1.5°K; this enabled us to obtain He^3 of 99.9% purity from a solution containing about 1% He^3. These estimates were confirmed by control experiments in which the concentration of He^3 was measured with an MI-1305 mass spectrometer.

For a column 70 mm high and 1.4 mm in diameter, operating at 1.5°K, we obtained the following results: c = 99.998% for R = 364 and c_b = 13%; c = 99.99% for R = 22 and c_b = 9.5%; c = 99.97% for R = 18 and c_b = 2%.

A major disadvantage of packless columns is their relatively low yield: for a column of 1.4 mm diameter, operating at a vapor velocity of 1.7 cm/sec and R ≈ 100, the yield is less than 100 cm^3/hour. However, when it is necessary to separate considerable amounts of a mixture, we can use columns consisting of several tubes.[5] The results reported in the present paper indicate that a column of five tubes, 2 mm in diameter and 200 mm in height, can produce He^3 at a rate of 500 cm^3/hour.

Sherman[6] reported the obtaining of He^3 of 99.9997% purity by rectification in a column with n ≈ 5. Columns used in his investigation produced greater separation and could be employed also to prepare extremely pure helium.

From the technical and physical point of view, it would be interesting to investigate rectification at a lower temperature, when the bottom part of the column is kept at temperatures corresponding to He II. In this case, the transport of He^4 along a film should be intensified by the thermomechanical effect, which would tend to improve the separation. This effect can explain qualitatively the observation, in some cases, of a nonmonotonic change in the concentration at the end of a rectification carried out at 1.5°K.

The investigation of rectification reported in the present paper, shows that packless columns can be used successfully for the separation of mixtures with a low He^3 content and for the preparation of He^3 of very high purity.

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