

Separation of Helium Isotopes by Rectification and Thermo-Osmosis

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The main characteristics of a packed rectification column for separation of a $\text{He}^3 - \text{He}^4$ mixture and of a filter for enriching the solution with the light helium isotope by means of the thermomechanical effect have been determined experimentally. The results are compared with theory. A brief description of a continuous-action double ray mass spectrographic gas analyzer is presented.

IN RECENT YEARS much work has been devoted to the experimental study of the properties of He^3 and its solutions in He^4 . However the light helium isotope is interesting not only as a subject for investigation. Evaporation of liquid He^3 under evacuation makes it possible to obtain temperatures of the order of $0.3 - 0.4^\circ \text{K}$ and thus to increase significantly the scale of low-temperature experiments.

At the present time the basic source of He^3 is tritium ($\text{T}_1^3 \rightarrow \text{He}_2^3 + \beta + \nu$, with a half-life of 12 years), a very expensive hydrogen isotope which is obtained in atomic reactors. It is known that He^3 is contained also in natural helium, which, however, practically worthless for obtaining anywhere near reasonable amounts^{1,2} of He^3 , since the concentration of the light isotope in the natural gas is extremely low, about $10^{-5} - 10^{-6} \%$. In working with $\text{He}^3 - \text{He}^4$ mixtures it is often necessary to extract the He^3 from mixtures containing no less than $0.01 - 0.1 \%$ of the light isotope.

The extreme value of He^3 necessitates the fullest possible separation of the mixture even when small quantities of the original product limit the yield of He^3 . Furthermore, the separation process should be sufficiently rapid, and this is particularly important when dealing with low concentrations. He^3 and He^4 have different boiling points, and in the liquid state the two isotopes are miscible in all respects.*

Solutions of $\text{He}^3 - \text{He}^4$ have no azeotropic points⁴. Under these conditions rectification is the simplest and most convenient method for separation of the mixture, especially since the separation coefficient

$$\alpha = y(1-x)/x(1-y)$$

(y, x are the relative molar concentrations $\text{He}^3/(\text{He}^3 + \text{He}^4)$ in the vapor and the liquid, respec-

*Below a temperature of 0.8°K , He^3 and He^4 are no longer mutually soluble³.

tively), which characterizes the ideal degree of rectification, is very large: $\alpha = 3 - 7$ (Fig. 1).

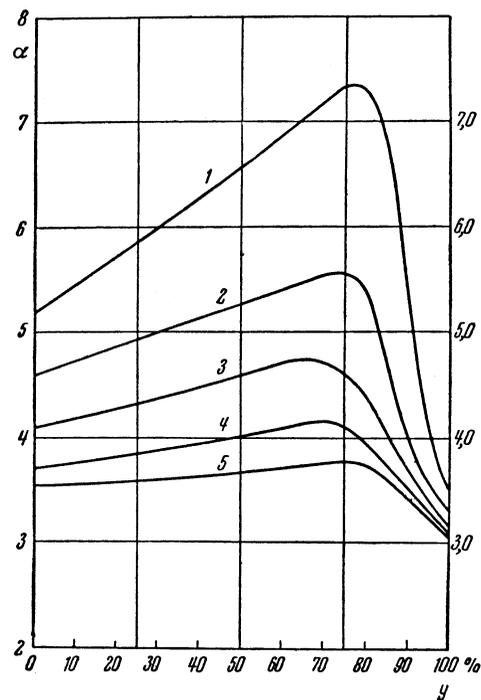


FIG. 1. The dependence of the separation coefficient α on the vapor concentration y for various pressures, calculated from experimental data^{4,30}.

There exist in addition many special methods for the separation of helium isotopes, which make use of the superfluidity of the He^4 in the solution. These involve enrichment with the aid of a thermal flux^{2,5,6}, conducting away the He^4 in a film^{7,8}, or through a thin filter^{1,2} (thermo-osmosis). As is shown by experiment, the best results are obtained by thermo-osmosis, which makes possible reliable extraction of He^3 with sufficient output^{1,2}.

In the present work we investigate the fundamen-

tal results of an experimental study of the rectification and thermo-osmosis processes which are necessary for the calculation and effective use of rectification columns and filters.

RECTIFICATION

Experiments on rectification were conducted with an adiabatic column whose design is shown in Fig. 2. The column 1 itself (a 7.5×8 tube of stainless steel) consists of two sections, 3 and 4 cm long, filled with 2.5 mm rings of constantan wire 0.25 mm in diameter. Vapor intake 2 in the central part of the column, valve 3, and tube 4 serve to draw off the mixture for analysis. Column chamber 5 is a copper cylinder externally heated by heater 6. A resistance thermometer 7 made of phosphor bronze is wound on the side surface of the chamber. The chamber is filled with the mixture through coil 8. Column condenser 9 serves also as the bottom of upper bath 10 which can be filled through valve 12 by liquid helium from Dewar flask 11 (lower bath). The temperature of the condenser is regulated by the evacuation rate of the upper bath. The column itself is surrounded by a vacuum jacket 13 which, after the apparatus is assembled, is evacuated to a pressure of 10^{-3} mm Hg. The vapor is removed from the condenser by tube 14 and from the chamber through coil 8. The glass parts of the apparatus (the upper bath and the upper part of the vacuum jacket) are joined to the metal by the copper-glass bonds 15. Wire-mesh cone 16 supports the packing and improves the conditions for the entry of vapor into the column.

In the experiments on rectification, we determined the dependence of the vapor concentration γ_1 in the intake 2 as well as the vapor concentration γ_u in the condenser on the pressure in the column at a constant heating rate of the chamber, and the dependence of γ_1 and γ_u on the mass velocity of the vapor at a constant pressure in the column. All the experiments were performed on a mixture with an initial concentration γ_0 of 2.6% under full reflux return (circulating system). Fig. 3 shows typical curves for γ_1 and γ_u as functions of the pressure in the column at a constant vapor mass-velocity. For the calculations which follow we used smoothed out values for γ_1 and γ_u (Tables 1 and 2). Since the liquid concentration in the chamber was no greater than $x = 0.2 - 0.3\%$ during the measurements, the vapor pressure in the column was assumed to be due to pure He^4 , and on this basis the chamber temperature

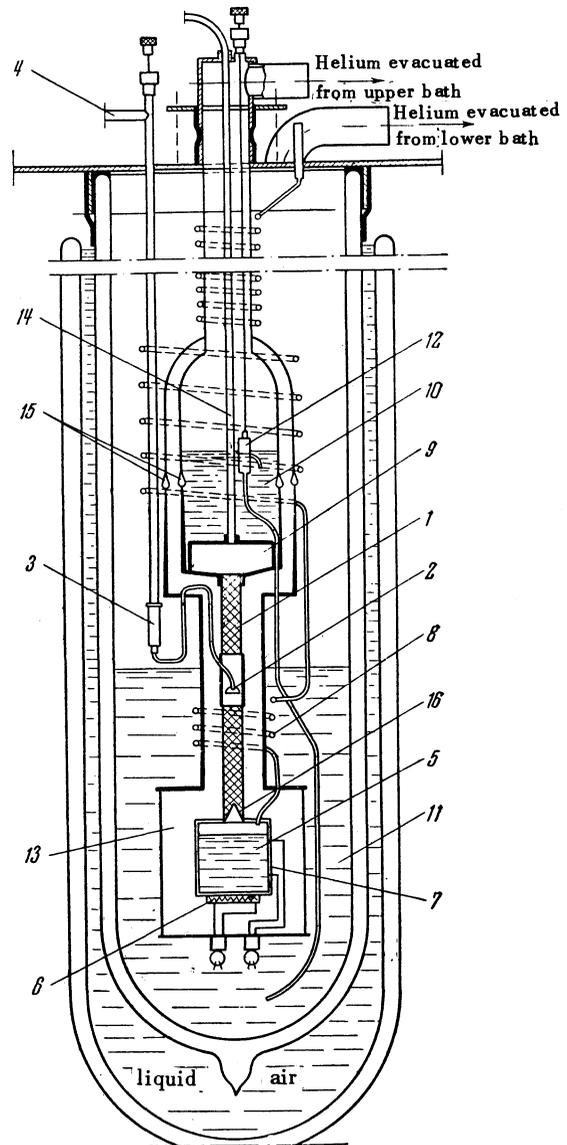


FIG. 2. Packed rectification column.

was determined.* The mass velocity U_M (measured in $\text{gm}/\text{cm}^2\text{-sec}$) of the vapor for the total input (lower) cross section of the column was determined from the power output of the electric heater and the heat of vaporization¹¹ of He^4 , and the Reynolds number Re corresponding to it was calculated from the usual relations

$$Re = U_M d_e / \psi \eta \quad (1)$$

$$d_e = 4\psi / f', \quad (2)$$

*For the determination of temperature from the He^4 vapor pressure, 1949 tables were used in all cases¹⁰.

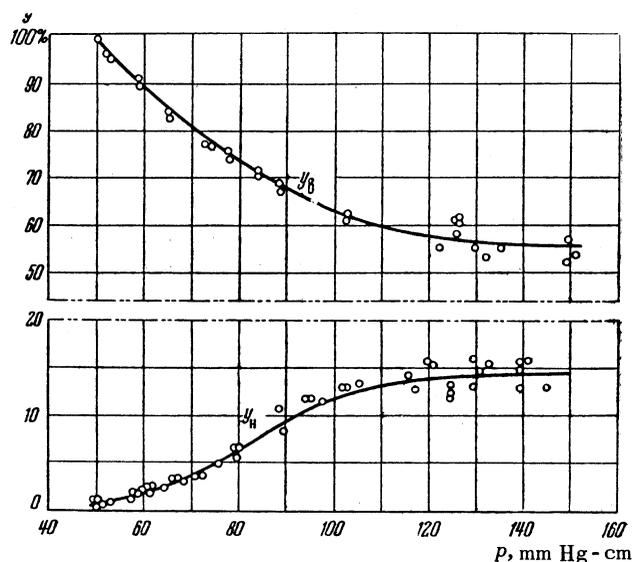


FIG. 3. The vapor concentration y_1 in the intake and y_u in the condenser as functions of the pressure in the column at constant vapor mass-velocity.

where $d_e = 0.107$ cm is the equivalent diameter, $\psi = 0.811$ is the free volume of the packing (that fraction of the volume of the column which is not occupied by packing material), and f' is the area of the packing per cm^3 of column. The magnitudes of ψ and f' are found from the weight of one cubic meter of dry, granular material and the geometry of the packing, and η is the viscosity. The measurements were performed with Re ranging from 35 to 175, which is essentially turbulent motion of the vapor¹².

The separation capacity of a column is usually characterized by the height of a transfer unit^{13,14} Φ_0 , given by

$$\Phi_0 = \frac{H}{\zeta} = H \int_{y_1}^{y_u} \frac{dy}{y^* - y}, \quad \int_{y_1}^{y_u} \frac{dy}{y^* - y} = \zeta, \quad (3)$$

where ζ is the number of transfer units, H is the length of the section of the filled column at whose ends the He^3 concentration in the vapor is y_1 and y_u , respectively, y is the vapor concentration at the core of the flow, and y^* is the vapor concentration which is equilibrium for the core of the liquid flow. The quantity $y^* - y$ is a measure of the diffusion pressure, so that Φ_0 is the height of a section of the column which gives an enrichment equal to the mean diffusion pressure in this section.

If the resistance to mass transfer in the liquid may be neglected, $y^* - y = y_i - y$ (where y_i is the

vapor concentration in equilibrium with the liquid at the phase boundary), and

$$\Phi_0 = \Phi = H \int_{y_1}^{y_u} \frac{dy}{y_i - y} = \frac{V}{k_g f},$$

where V is the volumetric rate of vapor production (in cm^3/sec), k_g is the mass-transfer coefficient in the vapor (in cm/sec), and f is the packing surface per centimeter of column height. Using the mass-transfer equation in the form¹⁵

$$Nu' = A Re^m Pr'^k \quad (4)$$

(where A is a constant, $Pr' = \eta/\rho D$ is the Prandtl diffusion number for the vapor, D is the molecular diffusion coefficient, ρ is the vapor density, and

$$Nu' = k_g d_e / D \quad (5)$$

is the Nusselt diffusion number), we arrive at¹⁶

$$\Phi = (\psi / f' A) Re^{1-m} Pr'^{1-k}. \quad (6)$$

An evaluation of the resistance to mass transfer in the vapor and liquid by Peshkov's method¹⁷ shows that in the first approximation one may neglect the resistance in the liquid. This makes it possible to use Eq. (6). Bearing in mind, however, that Re depends only weakly on the pressure, Pr' is practically constant, and*

$$\zeta = \frac{1}{\alpha - 1} \ln \frac{y_u(1 - y_1)}{y_1(1 - y_u)} + \ln \frac{1 - y_1}{1 - y_u}, \quad (7)$$

(which means that a very small change of α with pressure may cause a large change in $\alpha - 1$ and therefore in ζ), Eq. (6) should be written in the form

$$\Phi = (\psi / f' A) Re^{1-m} Pr'^{1-k} \varphi(p), \quad (8)$$

where $\varphi(p)$ is a function which accounts for the pressure-dependence of α .

The quantity $\Phi = \Phi_0$ was calculated by graphical integration of Eq. (3), with the aid of a graph of y vs. x (Fig. 4). The calculated height of the column

*Equation (7) is valid only for a circulating system with constant α .¹⁴

It was determined with the rectifying action of the condenser taken into account. As was shown by

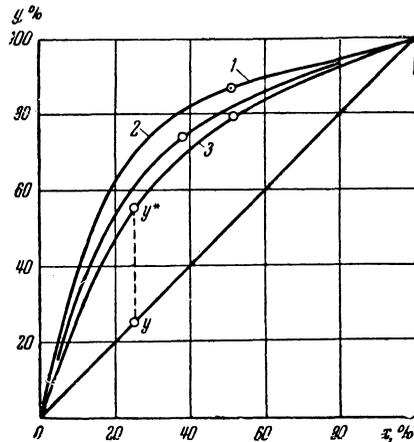


FIG. 4. Graph of y vs. x . Smoothed-out data is used^{4,30}.
1) $p = 50$, 2) $p = 100$, 3) $p = 150$ mm Hg.

measurements of the vapor concentration in the chamber, in the center of the column, and in the condenser, all at the same state of operation, about 25% of the total number of transfer units (ζ) may be assigned to the condenser. The values of Φ obtained in this way for the packed part of the column satisfy the empirical equation

$$\Phi = 353 \text{Re}^{-1.03} \varphi(p). \quad (9)$$

The values of $\varphi(p)$ are shown in Table 1. The accuracy of this equation, taking account of the spread in y_1 and y_u , is $\pm 20\%$. From Eq. (4) and (9) we obtain $\text{Nu}' \sim \text{Re}^{2.03}$. The exponent on Re usually¹⁶ lies between 0.75 and 1.18. The stronger dependence is probably explained by the poorer distribution of the liquid through the packing at low thermal loads on the column¹⁸.

TABLE I

Chamber temperature, °K	Chamber pressure, mm Hg	y_1 , %	y_u , %	ζ	Φ	Re	$\varphi(p)$
2.3	50	0.75	98.5	6.52	0.865	75.2	0.246
2.49	75	5.1	76.5	2.45	2.30	71.5	0.618
2.64	100	11.8	63.1	1.63	3.47	69.0	0.903
2.77	125	14.0	57.2	1.39	4.07	66.8	1.01
2.88	150	14.5	55.2	1.38	4.10	65.0	0.995

Note. The mass velocity was 0.00492 gm/cm²-sec.

In addition to the experimental determination of the separating capacity of the column, there exist several works^{17,19,20} in which an analytic expression for the column efficiency is obtained by integration of the differential equations of mass transfer. Of particular interest are the results obtained by Peshkov¹⁷, since they include many practically important cases.

For non-extracting operation, constant-velocity vapor motion throughout the column,* and no resistance in the liquid, y_1 and y_u are related by¹⁷:

$$(1 - y_1) y_1^{-1/\alpha} = (1 - y_u) y_u^{-1/\alpha} e^{\beta H}, \quad (10)$$

$$1/\beta = (|\omega| d_e^2 / 4 D_{\text{eff}} \alpha \kappa_y) / (\alpha - 1). \quad (11)$$

*At a condenser temperature of about 1.5°K (almost pure He³) and a chamber temperature of about 2.3°K (almost pure He⁴) the vapor velocity at the condenser is about 25% greater than at the bottom of the column, so that in the first approximation the constant-velocity condition is fulfilled.

Equation (10) is derived for an empty column. On going over to a packed column, the radius of the tube in Eq. (11) is replaced by $d_e/2$. In addition, Eq. (10) is strictly speaking valid for concentrations which have been averaged in a particular way over the cross section and velocities¹⁷, although when the vapor motion is turbulent this averaging gives the values which are actually measured in experiment; here w is the velocity of the vapor averaged over the cross section (in cm/sec), $D_{\text{eff}} = D + D_t$, where D_t is the coefficient of turbulent diffusion (in cm²/sec), the coefficient κ_y is defined by the equation¹⁷

$$\left| \left(\frac{\partial y}{\partial n} \right)_i \right| = 2 \kappa_y \frac{y_i - y}{d_e} \cdot 2, \quad (12)$$

and n is the normal to the surface of phase separation. Since

$$k_g(y_i - y) = D_{\text{eff}} \left| \left(\frac{\partial y}{\partial n} \right)_i \right|,$$

Eq. (5) and (12) lead to

$$D_{\text{eff}} = \text{Nu}' D / 4 \alpha \gamma. \quad (13)$$

Inserting (13) into (11) and making use of (4), (2), and (6), we obtain

$$1/\beta = \Phi \alpha / (\alpha - 1). \quad (14)$$

The values of $1/\beta$ as calculated according to Eq. (10) and (14) are sufficiently close (Fig. 5). The coefficients α necessary for the calculation were averaged according to Eq. (7).

In the case of extraction and when the molar vapor production rate remains constant along the column, we use the same initial equations and assumptions as in deriving Eq. (10) and (14), and obtain the following expression for the number of unit transfers:

$$\zeta = \frac{H}{\Phi} = \frac{\alpha}{\alpha - 1} \beta H = \frac{1}{2} \ln \frac{-(1 - y_1)^2 + F(1 - y_1) + M}{-(1 - y_u)^2 + F(1 - y_u) + M} + \frac{2G - F}{2\sqrt{4M + F^2}} \ln \left[\frac{-2(1 - y_1) + F - \sqrt{4M + F^2}}{-2(1 - y_1) + F + \sqrt{4M + F^2}} \cdot \frac{-2(1 - y_u) + F + \sqrt{4M + F^2}}{-2(1 - y_u) + F - \sqrt{4M + F^2}} \right], \quad (15)$$

where

$$F = \frac{(\alpha - 1)(1 - y_u \gamma) - \gamma}{\alpha - 1}; \quad G = \frac{\alpha(1 - \gamma)}{\alpha - 1} + \gamma(1 - y_u);$$

$$M = (1 - y_u) \frac{\gamma}{\alpha - 1};$$

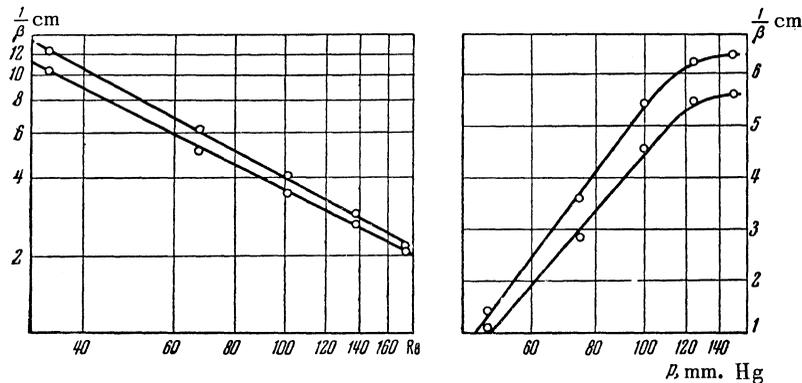


FIG. 5. Values of $1/\beta$ as calculated according to Eq. (10) (lower curve) and Eq. (14) (upper curve) for various pressures p in the column ($\text{Re} = \text{const}$) and various Re ($\rho = \text{const}$).

TABLE II

Chamber heating rate, watts	$10^2 U_M$ gm/cm ² -sec	γ_1 , %	γ_u , %	ζ	Φ	Re
0.0259	0.25	23	46	0.7	8.05	34
0.0517	0.50	11.7	57	1.45	3.98	68
0.0777	0.75	4.0	65.2	2.29	2.46	102
0.104	1.0	1.1	70.5	2.92	1.93	136
0.130	1.25	0.2	73	3.73	1.51	171

Note. The column pressure is 125 mm Hg.

and γ is the fraction of the vapor stream in the column which is extracted. If $\gamma \ll 0.1$ and $1 - \gamma_u \ll 1$, then $(\alpha - 1)/\alpha \gtrsim 0.6$, and expanding $1/F$ in a power series in γ and neglecting terms of order γ^2 and higher, Eq. (15) leads to

$$\zeta = \frac{1}{2} \ln \left[\frac{(1 - \gamma_u)^2}{1 - \gamma_u} \left(\frac{\gamma_1}{1 - \gamma_1} - \frac{\alpha}{\alpha - 1} \gamma \right) \right] + \frac{\alpha^2 - 1 + 2\alpha\gamma}{2(\alpha - 1)^2} \ln \left[\frac{\gamma_u}{1 - \gamma_u} \left(\frac{\gamma_1}{1 - \gamma_1} - \frac{\alpha}{\alpha - 1} \gamma \right)^{-1} \right] \quad (16)$$

When $\gamma = 0$, Eqs. (15) and (16) reduce to (10).

In rectification of a $\text{He}^3 - \text{He}^4$ mixture the vapor production rate at the top of the column is greater than that at the bottom by a factor of about 2, assuming that the chamber contains pure He^4 and the condenser pure He^3 . Therefore Eq. (15) and (16) can in this case be used only for a rough estimate.

If we increase the initial concentration γ_0 of the mixture, we increase the allowable (*i.e.*, for given γ_1 , γ_u , and U_M) extraction rate of the product (an increase of γ_0 by 1.5 to 4% makes it possible to increase the extraction rate by a factor of 1.5 - 2), so that the column should be supplemented with an apparatus for the preliminary enrichment of the mixture to a γ_0 value of several per cent. This is most easily done by thermo-osmosis.

To determine the maximum output of the column, one should find the critical vapor velocity, *i.e.*, the velocity at which the vapor begins to drag liquid along into the condenser (percolate).

It is known that for equal vapor velocity the pressure drop across a wet column is much greater than that across a dry one.

As has been shown by Kapitza²¹, under certain definite conditions the wave motion of a thin film of viscous liquid is more stable than laminary motion. By taking account of the disruptions in the gas current flowing across the waving surface of the liquid, one is able not only to obtain a quantitative determination of the pressure drop in a wet tube, but also to calculate the critical vapor velocity in it with good accuracy. In general, the vapor mass-velocity at the moment of percolation (U_M^{cr}) depends²¹ on the geometry of the column, the densities ρ and ρ_l of the vapor and liquid, the discharge of the liquid, its density η , and its surface tension. For the circulating state, a small temperature interval, and a given column diameter, the dependence is essentially $U_M^{\text{cr}} \sim (\rho/\rho_l)^n$, since the change of σ due to temperature is of little import ($U_M^{\text{cr}} \sim \sigma^{0.3}$), and the viscosity variation is even less ($U_M^{\text{cr}} \sim \eta^{0.2}$).

An investigation of percolation was carried out on a single-section column 200 mm long, similar design to the rectification column (Fig. 2). The moment of percolation was found from the different increases in the pressure difference between the chamber and condenser which accompanies small changes in the heating rate of the chamber. Measurements were performed both on pure He^4 and on a mixture ($\gamma_0 = 2.6\%$). All calculations on percolation were performed for the total column cross section. From the chamber pressure p_c immediately before percolation, the chamber temperature T_c was found, and the ratio of the vapor to liquid densities (ρ/ρ_l) was calculated. In performing the calculation the modulus of compressibility of the vapor $Z = 1 + Bp_c/RT_c$ was accounted for, where B is the second virial coefficient of He^4 (in $\text{n. cm}^3/\text{mole}$)²². For temperatures on the order of 2.8 - 2.9° K, the ideal gas equation gives too low a value for ρ (by 10 - 12%). Further, U_M^{cr} was calculated from the heater power and the heat of vaporization of He^4 . With a statistical error no greater than $\pm (4 - 5\%)$, both for the mixture and for pure He^4 ,

$$U_M^{\text{cr}} = 0,153 (\rho/\rho_l)^{0,46}, \\ 0,013 \leq \rho/\rho_l \leq 0,033; T_c = 2,4 \div 3,0^\circ \text{K.} \quad (17)$$

The slope of the U_M^{cr} curve as a function of ρ/ρ_l when calculated according to Kapitza's equation is close to the experimental value, so that this dependence for a packed column is the same as for a film column. A similar dependence is given by empirical equations presented in other works²³⁻²⁵. The amount of liquid trapped by the packing in the operation of the column is an important characteristic of the separation process. In periodic rectification, an increase of the ratio between the entrainment and the chamber content limits the output of the product and in many cases decreases the degree of separation²⁶. The magnitude of the entrainment makes it possible to determine the incomplete product Π , that is the minimum amount of He^3 which must be in the column during its operation. In the circulating state¹⁷ $\Pi = (S/H) \int_0^H \gamma dH$, where S is the entrainment in the whole column; using Eq. (10) and (15) we obtain

$$\Pi = \frac{S}{\zeta} \left[\frac{\alpha}{\alpha - 1} \ln \frac{1 - \gamma_1}{1 - \gamma_u} + \frac{1}{\alpha - 1} (\gamma_u - \gamma_1) \right]. \quad (18)$$

In order to determine the entrapment (Fig. 2), a known amount of mixture (2.6%) was condensed in the chamber, and then the chamber heat supply was gradually increased until all the liquid was trapped by the packing. In this case the chamber content is equal to the entrapment and the heat supply and chamber temperature determine the vapor mass velocity at the bottom end of the column. The time at which the liquid left the chamber was fixed from the readings of resistance thermometer 7 (Fig 2) which was connected into a self-balancing bridge circuit based on the EPD-17 potentiometer.

An evaluation of the entrapment for various values of U_M in calculating the thickness of the liquid film uniformly covering the packing gives a quantity on the order of $(0.7 - 1) \cdot 10^{-2}$ cm for $U_M^{cr} = 0.005 - 0.02$ gm/cm²-sec.

THERMO-OSMOSIS

The apparatus for thermo-osmosis is shown in Fig. 6. Condenser 1 is soldered to filter 2 consisting of a copper tube of diameter 2×4 mm and length 4.5 cm tightly packed with crocus. The filter and upper bath are surrounded by a vacuum jacket which is evacuated through tube 3 during assembly of the apparatus. In experiments on the efficiency of thermo-osmosis the condenser was filled through tube 6 with a known quantity of a mixture of given concentration. Evacuation of the upper and lower baths was used to establish temperatures at the end of the filter such that the He⁴ in the solution was superfluid². Calibrated collector 5 was then filled through tube 7 with gaseous He⁴ to the equilibrium pressure, thus improving² the He³ extraction from the solution. When valve 4 was opened, the superfluid helium flows into collector 5, and the He³ concentration in the condenser gradually increases. The calculated enrichment coefficient of the filter $\delta = x/x_0$ (where x and x_0 are the liquid concentrations in the condenser and collector, respectively) is on the order of $10^4 - 10^6$ (complete separation is not obtained, since He³ penetrates through the filter due to diffusion in the liquid). Measurements show that δ is in any case greater than 500. After closing valve 4 the enriched remainder was pumped into a graduated gas holder, and as a control its concentration was measured in a mass-spectrographic gas analyzer. From the known initial and final mixture concentrations and volumes, as well as from the amount of liquid in the container at a time τ , the concentration of the mixture in the condenser was

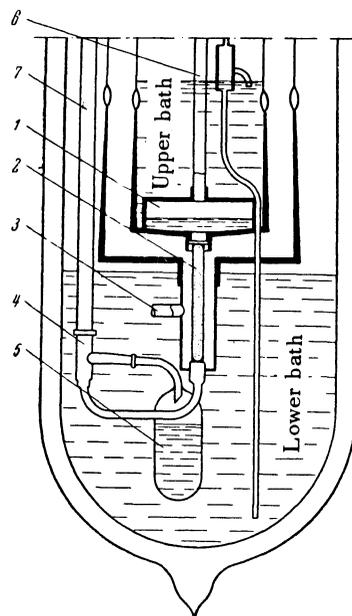


FIG. 6. Thermo-osmosis apparatus

calculated for given τ . The concentration of the liquid was calculated on the basis of the distribution coefficients γ/x as given by Esel'son and Bereznik⁴, and the vapor volume of the condenser. The output Q (in l/he) of the filter is calculated from the rate at which the collector is filled. The results obtained are shown in Fig. 7. For a given filter output and condenser temperature, the enrichment is greater for greater temperature between the ends of the filter. Qualitatively speaking, the temperature dependence of the enrichment is of the same form also for equilibrium, that is when the total discharge of liquid through the filter vanishes. The equilibrium values of x have been calculated previously². Since the output decreases rapidly as x increases, the use of thermo-osmosis at concentrations greater than 4 - 5% is not worthwhile.

MASS SPECTROGRAPHIC GAS ANALYZER (MG)

The gas analyzer is based on the PTI-4 helium leak-detector, and is a double-ray mass spectrograph with 180° deflection and separate amplification circuits for the He³ and He⁴ ion currents. The MG can be used for continuous analysis of a gas mixture in the concentration range between 0.2% for He³/(He³ + He⁴) and 0.2% for He⁴/(He³ + He⁴) with an accuracy of $\pm 5\%$ of the measured quantity. Unlike Nier's^{27,28} double-ray instrument and the domestic²⁹ MS-2M, the gas analyzer has a pulsating ion

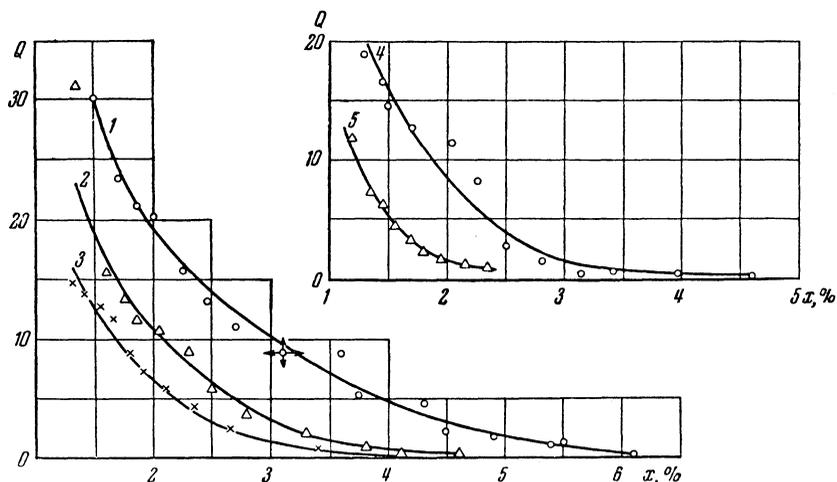


FIG. 7. The dependence of the filter output Q on the concentration of the liquid being enriched at various temperatures. Curve 1) 1.43° (solution) – 2.15° (He^4 collector); Curve 2) $1.43 - 2.02^\circ$ K; Curve 3) $1.43 - 1.86^\circ$ K; Curve 4) $1.86 - 2.15^\circ$ K; Curve 5) $1.86 - 2.02^\circ$ K.

current and ac amplification. The method for obtaining an ac voltage on the collector is the same as in the leak detector. The construction of the ion source (cathode 1 and ionizer 2, Fig. 8) is not changed. Between the ionizer and the exit slit 4 is an accelerating diaphragm 3 (with a slit of about 0.2 mm). The radii of the ion trajectories are 3.5 cm for He^{3+} and 4 cm for He^{4+} . The distance between the slit centers of the collector diaphragm 5 is about 10 mm. In the vacuum chamber of the MG are placed two acorn-tube electrometer stages 6Zh1Zh connected as triodes. The grid-leak resistors (5×10^8 ohm) are the same for both tubes. The signals from the electrometer stages are fed into ac amplifiers (the He^4 circuit uses the amplifier of the leak detector) and are then rectified. The maximum current gain, found from the ratio between the output dc current and the effective current in the input resistance of the acorn tubes, is about 5×10^8 . An electronic potentiometer EPD-17 is used to measure the ratio of the ion currents: voltages proportional to the output currents of the amplifiers are applied, with opposite polarity, to the input of the potentiometer amplifier (the output current of the He^4 circuit flows along the EPD-17 slide wire). When so connected, the apparatus gives the ion current ratio, or the molar concentration of the mixture in terms of He^3/He^4 .

Different ranges of measurement are obtained by switches which change the ratio of the gains of the amplifiers from 1 to 100. In order to test the amplifiers, an ac voltage from a PTI-4 generator can be applied to the input of the electrometer stages.

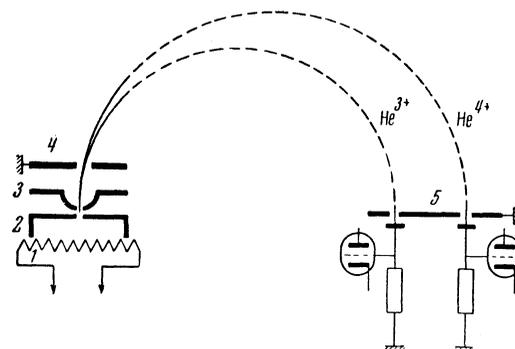


FIG. 8. Gas-analyzer vacuum chamber. The potential of cathode 1 and diaphragm 3 is $+220$ v, that of ionizer 2 is $U_{dc} + U_{ac} = 316 + 21$ v.

Under continuous analysis the gas consumption is $0.1 - 0.2$ cm^3/hr . During measurement, the gas pressure ahead of the input valve (PTI-4 throttle valve) is no greater than $20 - 30$ mm Hg, and the gas input is easily regulated; the readings of the apparatus do not depend on the gas pressure ahead of the valve within the limits of accuracy of the analysis. The background of HHH^+ ions is between 0.03 and 0.04% of the He^3 after a half hour of cathode heating. In order to obtain readings relative to zero concentration, regulation of the output current in the He^3 circuit is provided for while the ratio between the amplification factors is held constant. The resolving power of the mass spectrograph (with a maximum half-width of about 12) and the shape of the ion beams make it possible, in principle, to increase the sensitivity of the apparatus by a factor of at least 3 - 4.

CONCLUSIONS

A combination of thermo-osmosis and rectification is a simple and effective method for extracting He³ from a mixture. The filter used in thermo-osmosis is easily connected to the column so that the whole apparatus can be placed into a single cryostat².

The results of the present work show that a packed column 8 – 10 cm long makes it possible to obtain practically pure He³ when dealing with a mixture containing only 1 – 2% of the light isotope. Using a column from 7 – 8 mm in diameter, the He³ extraction rate under these conditions may be as great as 8 – 10 l/hr. The amount of the incomplete product is then of the order of 200 – 300 cm³ of He³. However, successive rectification of the remainder at lower heating rates of the column makes it possible to decrease the amount of the incomplete product substantially.

For the column and filter together, the total coefficient of enrichment is of the order of 10⁵ – 10⁶.

Several expressions have been obtained for the calculation of the rectification process with turbulent motion of the vapor in the column, and the dependence of the filter output on the concentration of the mixture to be enriched has been determined.

In order to study the separation processes, a double-ray mass spectrographic gas analyzer has been constructed, and in spite of its relatively simple design it can be used to obtain sufficiently accurate measurements of the concentrations in various mixtures.

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¹ B. N. Esel'son and B. G. Lazarev, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 743 (1950).

² V. P. Peshkov, J. Exptl. Theoret. Phys. (U.S.S.R.) **30**, 850 (1956), Soviet Physics JETP **3**, 706 (1956).

³ W. M. Fairbank, Phys. Rev. **103**, 262 (1956).

⁴ B. N. Esel'son and N. T. Berezniak, J. Exptl. Theoret. Phys. (U.S.S.R.) **30**, 628 (1956), Soviet Physics JETP **3**, 568 (1956).

⁵ Lane, Fairbank, Aldrich, and Nier, Phys. Rev. **73**, 256 (1948).

⁶ Reynolds, Fairbank, Lane, McInteer, and Nier, Phys. Rev. **76**, 64 (1949).

⁷ Daunt, Probst, Jonston, Adrich, and Nier, Phys. Rev. **72**, 502 (1947).

⁸ Esel'son, Lazarev, and Lifshitz, J. Exptl. Theoret. Phys. (U.S.S.R.) **20**, 749 (1950).

⁹ K. Atkins and J. R. Lovejoy, Canad. Journ. of Phys. **32**, 702 (1954).

¹⁰ H. van Dijk and D. Shoenberg, Nature **164**, 151 (1949).

¹¹ W. Keesom, *Helium*, IIL, 149 (a translation of a book published by Elsevier Publishing Co., N. Y., 1942).

¹² *Processes and Apparatus in Chemical Engineering*, ed. A. G. Kasatkin, Goskhimizdat, 1953, p. 48.

¹³ T. H. Chilton and A. P. Coburn, Ind. Eng. Chem. **26**, 1183 (1934).

¹⁴ T. H. Chilton and A. P. Colburn, Ind. Eng. Chem. **27**, 255 (1935).

¹⁵ D. A. Frank-Kamenetskii, *Diffusion and Heat Transfer in Chemical Kinetics*, Publ. Acad. Sci. U.S.S.R., 1947, p. 41.

¹⁶ I. P. Usiukin and L. S. Aksel'rod, Kislород (Oxygen) **3**, 1 (1952).

¹⁷ K. P. Peshkov, J. Tech. Phys. (U.S.S.R.) **26**, 664 (1956).

¹⁸ D. W. Duncan, J. H. Koffolt, J. R. Withrow, Trans. Am. Inst. Chem. Engrs. **38**, 259 (1942).

¹⁹ W. Kuhn, Helv. Chim. Acta **25**, 252 (1942); **37**, 1407 (1954).

²⁰ J. M. Westhaver, Ind. Eng. Chem. **34**, 126 (1942).

²¹ P. L. Kapitza, J. Exptl. Theoret. Phys. (U.S.S.R.) **18**, (1948).

²² Kilpatrick, Keller, and Hammel, Phys. Rev. **94**, 1103 (1954).

²³ M. P. Malkov and K. F. Pavlov, *Very Low Temperature Handbook*, Gostekhizdat, 1947, p. 227.

²⁴ N. M. Zhavoronkov, Chemical Industry **2-3**, 12 (1944).

²⁵ C. S. Robinson and E. R. Gilliland, *Elements of Fractional Distillation*, 4th ed. McGraw-Hill Book Co. (1950), p. 440.

²⁶ *Distillation*, (Ed. American edition, A. Weisberg) IIL, 1954, pp. 126, 130.

²⁷ Nier, Ney, and Inghram, Rev. Sci. Instr. **18**, 749 (1947).

²⁸ A. O. Nier, Rev. Sci. Instr. **18**, 398 (1947).

²⁹ Trans. (Trudy) of Sci. Res. Inst. of the Radio Industry, Gosenergoizdat **4**, (24), 1955, p. 26.

³⁰ V. P. Peshkov and V. N. Kachinskii, J. Exptl. Theoret. Phys. (U.S.S.R.) **31**, 720 (1956), Soviet Physics JETP **4**, 607 (1957).