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Experiments in Enrichment of Helium with Isotope He^3

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Equipment is described for the enrichment of helium with isotope He^3 based on the use of thermomechanical effect and rectification. An evaluation is made of the possible degree of enrichment and extraction of He^3 by means of thermal diffusion.

AS is well known natural helium consists basically of the isotope of mass 4 and contains very small amounts of the mass 3 isotope. The usual fraction of He^3 is only 10^{-8} --10^{-7} of He^4. Therefore, in order to obtain even a small amount of helium with a significant concentration of He^3 from natural helium it is necessary to enrich it by a factor of several hundred thousand. Pollard and Davidson[1] first proposed to use the thermomechanical effect for this purpose. The most effective method was developed by Esel'son and Lazarev[2]. In the method of Ref. 2 the apparatus worked cyclically and the enrichment per one cycle was about 200. The purpose of the present work, which has been conducted with considerable interruptions since 1949, was to develop equipment of sufficient yield for the extraction of He^3 from its mixture with He^4 and its further purification from He^4.

1. EQUIPMENT FOR EXTRACTION OF He^3

Several types of apparatus for the extraction of He^3 from natural helium have been devloped and tested. The first is shown in Fig. 1. The temperature in the Dewar flask *1* containing liquid helium (from which H^3 was extracted)was maintained at 2.3° K. A temperature of 1.8° K was established in the internal Dewar flask *2*. Helium entered from the external flask through the regulating valve *3* and a copper tube 0.4 diameter × 1.0 mm into the collector *4*. The level in the collector was maintained near the center of the top ball. The temperature in the collector was measured with the thermometer *8* and was somewhat lower than the λ point (2.19° K). Since the temperature in flask *1* was higher than in the collector *4*, the thermal flow was upward due to conduction of heat along the copper tupe and to the heat carried by the helium from the bottom of the collector. The thermal flow carried with it He^3 and the liquid helium depleted of He^3 was pressed into flask *2* through the copper tube *5* (0.4 × 1.0 mm). The speed of the pressure transfer was regulated by valve *6*. In this as in other apparatus the transitions from glass to metal were accomplished by welds between the copper and glass. The evaporated He^3 enriched helium in the collector *4* was passed into the rectification column *7*, the upper portion of which was cooled by contact with the helium contained in flask *2*. The column consisted

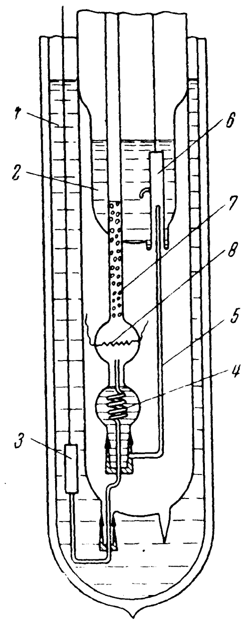

FIG. 1. Apparatus for the extraction of He^3 by thermal flow and rectification.

of a glass tube 3 mm in diameter and 100 mm long. The tube was filled with rings of 0.15 mm constantan wire cut from a spiral 1 mm diam. The more readily boiling He^3 was collected in the region of lower temperatures at the top of the column and the more readily condensable He^4 flowed back into the collector.

Enrichment up to 0.2% of He^3 was obtained with this equipment which corresponds to an increase in concentration by a factor of 10^5. It was further established that the concentration of He^3 in the output was of the order of 10^{-8}. The equipment was cyclic in operation and permitted the processing, with a single loading, of 0.75 liter of liquid helium in four hours.

Since the equipment described above was of insufficient output and did not permit the uninterrupted processing of helium, another apparatus was constructed and set up which could be operated continuously with the helium added at atmospheric pressure without interrupting the operation of the equipment. In this apparatus, as in the method of Esel'son and Lazarev[2], the enrichment of He^4 was obtained by making use of the diverted He^4 in the form of a superfluid flow through a filter due to the action of thermal diffusion and besides this, by the simultaneous diversion of the He^4 enriched gas by rectification in the tube. The apparatus is shown in Fig. 2. The helium in the external flask *1* was at atmospheric pressure being replensihed as it was used up from the liquefying machine without interrupting the operation of the equipment. In the internal flask *2* the temperature was held between the limits 1.9° and 2° K by pumping and the use of heater *3*. Helium from the outer flask *1* entered collector *4* through the regulating valve *5* and the 6 mm in diameter 1.4 × 20 mm copper tubing. The tube in the inner flask was bent in the form of a spiral of an overall length of 35 cm in order to effect cooling of the helium carried by this tube. In addition, this tube was soldered to a similar tube *7* for a length of 2 cms. Due to the heat received by tube *7* at the soldered surface and the heat emitted by the heater *8*, the helium flowed in the manner of a superfluid from collector *4* through filter *9* into the inner flask. The filter consisted of a brass cartridge 7 mm in diameter, 35 mm long, densely packed with rouge. The He^3 remaining in the collector passed into the gaseous state and was enriched by rectification in tube *10* and removed from the apparatus through the same tube. According to computation the small difference in temperature formed between the collector and flask *2* was quite sufficient for the normal operation of the tubular column. The evaluation of the operation of tubular columns is presented in Ref. 3. The rate of helium processing was limited by the pumping of the helium vapor from the inner flask *2*. All the processed helium was to be pumped out by pumps at pressures from 20 to 30 mm of mercury. The equipment was capable of processing helium at the rate of 3 m^3/hr (at normal temperature and pressure) with the capactity of the vacuum pumps about 100 m^3/hr, power of the heater 82 watts and of the evaporator 31 watts. The gas collected in the first experiments was enriched to 0.02% of He^3. To determine the extraction coefficient the once-processed helium was worked over for the second time. The result showed that, within the limits of measurements errors (which amounted to 5-10%), the extraction was complete. On the basis of balance relationships the concentration of the output gas should be taken as 10^{-8} and enrichment coefficient as 2×20^4. Since the concentration of the processed helium could not be measured with sufficient accuracy it can only be stated that the common division coefficient was greater than 2×10^5. The described apparatus could be operated continuously as long as desired. The duration of its operation was determined by the operation of the liquefying equipment.

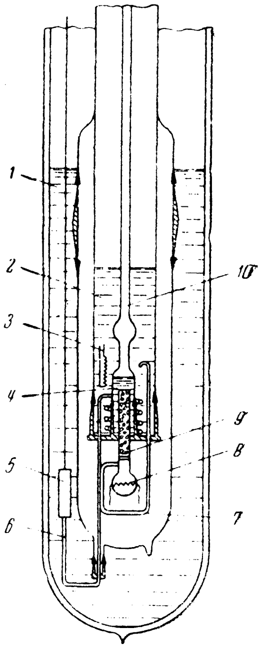

FIG. 2. Apparatus for extraction of He^3 by the thermomechanical effect and rectification.

1. EVALUATION OF THE EFFECTIVENESS OF SEPARATION METHODS

Theoretically, the coefficient of separation of He^3 by the superfluid filtration can be evaluated from the following considerations. If we assume that superflow and diffusion take place through the same effective cross section of the filter and that the velocity of the superflow is limited by the critical velocity $v_s = 20$ cm/sec, then the value of the superflow through a unit of cross section is

$$\omega_1 = \rho_s v_s,$$

and the flow of He^3 determined by diffusion will be

$$\omega_2 = D\rho m_3 X_0 / m_4 l,$$

where D nenoted the diffusion coefficient of He^3, l the length of the filter, ρ_s, the density of the superflowing helium, X_0, the molar concentration before the filter, m_3 and m_4, atomic masses of He_3 and He_4, respectively.

The molar concentration of the passing helium will be determined by:

$$X = \omega_2 m_4 / m_3 \omega_1 = D\rho X_0 / l\rho_s v_s,$$

i.e., the limit of the attainable separation coefficient is equal to

$$A = X_0 / X = l\rho_s v_s / D\rho.$$

The diffusion coefficient of He^3 in liquid helium II was determined experimentally by Beenakker et al.[4]. Their results show that the coefficient decreases rapidly from 10^2 cm^2/sec at 1.3° K to 10^{-3} cm/sec at 1.6° K and 4×10^{-5} cm^2/sec at 2.1° K. Thus, for the filter length $l = 5$ cm, we will obtain for the separation coefficient at 1.3° K $A = 10^4$, at 1.6° K $A = 8 \times 10^4$, at 2° K $A = 6 \times 10^5$ and at 2.1° K $A = 5 \times 10^5$, i.e., the separation coefficient reaches a maximum at about 2° K. In practice, the obtainable coefficient is less due to interruptions in the process when the diffusion continues and there is no superflow.

It should be noted that enrichment by thermal diffusion cannot take place at any desired high concentration. As the measurements in Refs. 5 and 6 have shown, the λ-transition is displaced with increase of concentrations into the region of lower temperatures. Thus in a 50% solution of He^3 and He^4 the superflow is already lost at 1° K. Besides, the following relation for thermal diffusion given in the work of Pomeranchuk[7] holds for not too high concentrations of He^3:

$$-SdT + \frac{dp}{\rho} - \frac{k}{m_4} d(XT) = 0,$$

where S denotes the entropy 1 gm He^4, ρ density, p pressure, m_4 the atomic mass of He^4, k Boltzman's constant and x the molar concentration of He^3. The entropy of He II in the temperature region above 1° K can be expressed by the formula $S = 0.405\,(T/2.19)^{5.5}$ cal/gm, deg, i.e., $SdT = (1/6.5)d(ST)$. Since the density of He^4 changes very little, the entire expression can be written in the form

$$d\left\{\left(\frac{kX}{m_4} + \frac{S}{6.5}\right)T - \frac{p}{\rho}\right\} = 0$$

or, along the thermal diffusion path,

$$\left(\frac{kX}{m_4} + \frac{S}{6.5}\right)T - \frac{p}{\rho} = \text{const}.$$

Since with the extraction of He^3 by thermal diffusion the concentration on one side is practically equal to zero, we obtain

$$\left(\frac{kX}{m_4} + \frac{S}{6.5}\right)T - \frac{p}{\rho} = \frac{S_0 T_0}{6.5} - \frac{p_0}{\rho}$$

or

$$X = \frac{m_4}{kT}\left(\frac{S_0 T_0 - ST}{6.5} - \frac{p_0 - p}{\rho}\right).$$

The line of λ-transitions and also the values of X computed for different values of T by the given formula are shown graphically in Fig. 3. As seen from Fig. 3 the process of thermal diffusion cannot proceed to any desired high concentrations. Therefore, the enrichment of mixtures with concentrations of He^3 higher than 10% and the purification of He^4 are more appropriately carried out by rectification. Evaluation of the operation of the rectification column was presented in Ref. 3. It was possible on the basis of these evaluations to reduce considerably the volume of experimental work in the development of the concentration equipment.

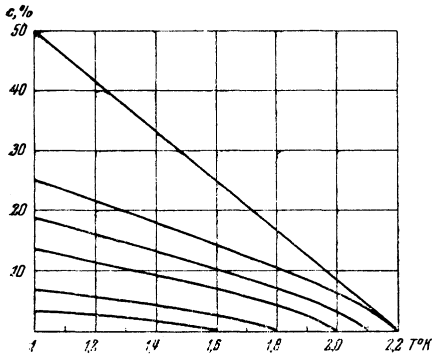

FIG. 3. The line of λ-transitions (upper) and lines $(k\epsilon/m+S_0/6.5)T-p/\rho = \text{const}$ for various values of T_0.

3. CONCENTRATION EQUIPMENT

A series of apparatus has been developed for the concentration of He^3 from mixtures of 0.1% and higher. The most effective was the apparatus shown in Fig. 4. It operates on the same principle as the one previously described. However, since it is impossible to obtain high concentration of He^3 by the thermomechanical effect, the rectification column was noticeably increased and it was in this column that the basic enrichment took place. In addition, the resulting mixture was condensed directly in the apparatus and after reprocessing collected in a special collector. The apparatus was placed in a Dewar flask *1* containing liquid helium, the resulting mixture was passed through a German silver coiled tube 1.5 × 2 mm of a total length of 800 cms. The helium condensed in the coiled tube was collected in the evaporator *3*. From the evaporator He^4 entered the glass collector *4* by superfluid flow through the filter *5* with the aid of the warmer *6*. In order to prevent diffusion of He^3 through the filter during the absence of superfluid motion, the exit opening of the filter was closed by valve *7*. The filter consisted of copper tube 2 × 4 mm, 45 mm long, densely packed with rouge. The processed helium was pumped out from the collector *4* through tube *8*.

The helium remaining in the evaporator entered the rectification column *9*, where the more readily boiling He^3 was collecting in the cold upper portion. The temperature in the upper portion of the column was maintained at the expense of the evaporation of helium from the collector *10*. Liquid helium was supplied from flask *1* through tube *11* and valve *12* by the pressure difference. Helium was added as required to make up for loss due to boiling.

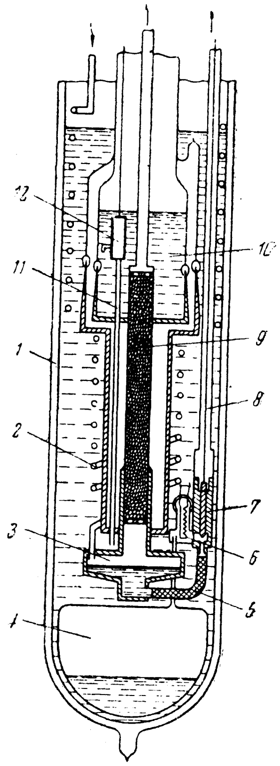

FIG. 4. Apparatus for the concentration of He^3.

The rectification column consisted of a stainless steel tube, inside diameter 9.6 mm, 0.2 mm wall thickness, 200 mm long. The entire column was filled with rings 1.5 mm diameter of 0.2 mm constantan wire.

Inasmuch as the rectification process is considerably slower than the enrichment by superflow, it was appropriate to carry out the concentration process in two cycles. The first cycle (excitation cycle) was carried out with full loading of the filter at a temperature of 1.95° K and heater power of 0.01 watt. The rate of processing of the initial gas mixture was thereby equal to 100 *l*/hr (at 0° C and 760 mm of mercury). At this processing rate, especially since the initial concentration was about 1 to 2% of He^3, the rectification column could not operate effectively and only slightly improved the singular enrichment as a result of the difference in concentrations between the liquid and gaseous state. At the top of the column the temperature was maintained at 1.5° K with the removal of the gas taking place at a pressure inside the column of 20 mm of mercury. The concentration of the removed gas was of the order of 30 to 50%. To begin the superfluid flow through the filter it was necessary to fill the collector *4* with gaseous He^4 up to equilibrium pressure, because otherwise the helium would pass through the filter by diffusion, i.e., would bring with it noticeable quantities of He^3 which decreases the degree of extraction. By following this procedure the obtained degree of extraction was 99.98% and the general division coefficient was 10^4, which is only several times smaller than the attainable limit.

When small quantities of the initial mixture were processed valve *7* was closed immediately after the cessation of the superfluid flow, all the removed gas was again condensed and the second cycle (enrichment) was carried out.

In the second cycle the temperature of the bottom of the column was set at the beginning of the process at 2.0° K, and at the top to 1.46° K. It was not possible to attain a lower temperature at the top of the column with the available pump which had a capacity of 50 m^3/hr. The pressure inside the column was thereby of the order of 45 mm of mercury. After equilibrium was reached, removal of the product began at a rate of 0.6 *l*/hr (at 0° and 760 mm of mercury). As the He^3 was being extracted, it was necessary to raise the temperature of the bath in order to maintain a pressure of 45 mm of mercury at the top of the column. The extraction process ended when the temperature of the bath reached 2.5° K. The concentration of He^3 in the removed product was 99.95 to 99.97%. After removal of the 99.95% gas and with 2.5 *l* still remaining in the apparatus, the concentration of the remaining mixture proved to be 16%.

With 2 liters of liquid helium poured initially into the outer flask, the apparatus could opperate for about 4 to 5 hours.

I consider it my pleasant duty to express my sincere thanks to Mr. A. I. Filimonov, who had constructed most of the equipment and with whom I conducted most of the experiments; also, to V.M. Kuznetsov and A. I. Uriutov who helped with the experiments; to Professor N. E. Alekseevskii in whose laboratory the mass spectroscopic analyses were carried out; and to T. K. Shusalova who performed the analyses with the mass spectroscope.

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177