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LOW TEMPERATURE PHYSICS

L. C. JACKSON

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by

L. C. JACKSON

M.Sc. (Lond); Doct.Wis.Nat. (Leiden)

WITH 33 DIAGRAMS

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PREFACE TO SECOND EDITION

THE period between the appearance of the first edition of this monograph and the outbreak of the Second World War was one of greatly increased activity in Low Temperature Physics. New laboratories were founded in many parts of the world and the established laboratories produced an ever larger volume of publications in this most fascinating and profitable branch of physical research. It is therefore all the more impossible to give a complete account of the subject within the scope of the present monograph. It may serve as an introduction for readers whose main interest lies elsewhere, but may also stimulate some to make a closer acquaintance with physical research at the lowest temperatures. The plan of the first edition has been retained and the various sections have been brought up-to-date.

References to the original literature have been omitted and the reader is referred instead to books and collected articles from which more detailed information can be obtained.

L. C. JACKSON

BRISTOL, *April* 1946

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CHAPTER I

THE PRODUCTION OF LOW TEMPERATURES

THE LIQUEFACTION OF AIR, HYDROGEN AND HELIUM

ALL physical experiments at very low temperatures involve the use of liquefied (or solidified) gases in some way or other. The liquefaction of gases is thus an important branch of low temperature physics, and the principles underlying the methods employed will therefore be discussed briefly, with special reference to the cases of air, hydrogen and helium. With the aid of these three liquefied gases it is possible directly or indirectly to experiment over the whole range from room temperature down to 1° K. or even lower.

There are two main types of method available for the large scale liquefaction of air (and hydrogen and helium), that which employs the Joule-Thomson effect (expansion of a gas with the performance of " internal " work) to produce the necessary cooling (Linde, Hampson) and that which employs in part at least the reversible expansion of the gas with the performance of " external " work (Claude, Heylandt).

" INTERNAL WORK " METHODS OF GAS LIQUEFACTION

The essentials of a liquefier using the Joule-Thomson effect are shown diagrammatically in Fig. 1. Compressed gas enters the inner tube of the " heat interchanger " H at a pressure p_A and temperature T_A. It then expands at the valve V to a pressure p_B and the temperature falls on account of the performance by the gas of " internal work " in expanding against the forces of attraction between the molecules. The cooled expanded gas then passes up the outer tube of the interchanger, cooling the incoming compressed gas as it does so. The expanded gas leaves the liquefier at the pressure p_B and temperature T_B (T_B is nearly equal to T_A if the interchanger is efficient). The cooler compressed gas expands at V and is thereby further cooled and so on until the temperature at the lower end of the interchanger has fallen by this " regenerative " process sufficiently low for the gas to liquefy. A part of the expanded gas will then liquefy and collect at C (temperature $= T_C$).

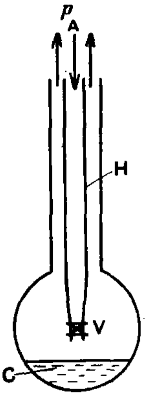

FIG. 1.

In such an expansion, provided there is no appreciable change in kinetic energy of the gas or after any change of kinetic energy has subsided, the " total heat " or " enthalpy " H of the gas before and after expansion remains constant since the process takes place adiabatically, no heat being allowed to enter the system from outside. We therefore have *

$$H_A = U_A + p_A v_A = H_B = U_B + p_B v_B \quad . (1)$$

(U = internal energy, p = pressure, v = volume per unit mass).

Theory indicates that the change of temperature dT which results when the pressure of a gas changes by an amount dp on passing a throttle or expansion valve is given by

$$\begin{aligned} \frac{dT}{dp} &= \frac{1}{C_p}\left(T\left(\frac{\partial v}{\partial T}\right)_p - v\right) = \frac{T^2}{C_p}\frac{\partial}{\partial T}\left(\frac{v}{T}\right) \\ &= -\frac{1}{C_p}\left(\frac{\partial H}{\partial p}\right)_T \end{aligned} \quad . \quad (2)$$

(C_p = specific heat of gas at constant pressure.)
The change of temperature dT is known as the " differential Joule-Thomson effect ", and as appears from (2) its sign depends on the sign of $\left(T\left(\frac{\partial v}{\partial T}\right)_p - v\right)$. There is thus a fall

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* For the thermodynamical theory of the Joule-Thomson effect and its " inversion ", see Porter, " Thermodynamics ", Methuen's Monographs on Physical Subjects.

in temperature on expansion when this quantity is positive and a rise when it is negative, while there will be no change in temperature on expansion when $\left(\frac{\partial v}{\partial T}\right)_p = \frac{v}{T}$. Points satisfying this condition lie on what is known as the " inversion curve " and when plotted on a $\mathfrak{p}.\mathfrak{t}$, (reduced pressure and temperature) diagram are found to give a parabola or some similar curve according to the equation of state adopted. Points lying inside the curve represent conditions which lead to cooling on expansion, points outside the curve to warming on expansion.

In any actual liquefier the pressure drop is finite and usually large (150 atm. to 1 atm.) and the drop in temperature ΔT is produced by the " integral Joule-Thomson effect ".

$$\Delta T = \int_{p_A}^{p_B} \frac{T^2}{C_p} \frac{\partial}{\partial T}\left(\frac{v}{T}\right) \cdot dp.$$

The " integral effect " also inverts for certain conditions of p and T and gives an " inversion curve " similar to that of the " differential effect ".

In applying the Joule-Thomson effect to the liquefaction of gases it is first necessary so to choose the initial pressure and temperature of the gas that expansion produces a fall in temperature throughout the range of temperature which will exist in the liquefier. This condition presents no difficulty with air as the effect is negative at room temperature for any pressure likely to be employed. On the other hand, the effect is positive at room temperature with hydrogen and helium. In actual practice these gases are cooled to 65°-90° K. and 15° K. respectively before expansion in order to bring them well inside the " inversion curve ".

The temperature at which the gas enters the actual liquefier having then been fixed, we can enquire what pressure of the gas before expansion will give a maximum yield of liquid. This can be calculated as follows :—

Suppose the liquefier has reached a steady state and is liquefying gas at a constant rate. A certain quantity of gas enters the apparatus per second at a pressure p_A and temperature T_A. The gas expands to a pressure p_B and a certain fraction ϵ is liquefied and collects at the bottom of the liquefier at temperature T_C. The remainder passes through the heat interchanger and leaves at a pressure p_B and temperature T_B. If the interchanger is perfect $T_B = T_A$. When the steady state has been reached we can equate the " total heat " of the compressed gas entering the liquefier to the sum of the " total heat " of the liquid produced and that of the expanded gas leaving the apparatus. Or if H_A, H_B, H_C are the total heats per unit mass we have

$$H_A = (1 - \epsilon)\, H_B + \epsilon H_C$$

$$\epsilon = \frac{H_B - H_A}{H_B - H_C} \qquad . \quad . \quad . \quad (3)$$

The fraction ϵ is known as the efficiency of the liquefier. We have to determine the conditions for which ϵ is a maximum. Since now p_B, T_B, p_C, T_C and T_A are all fixed by the design of the liquefier, the yield can be varied only by varying p_A, the input pressure. From the above formula ϵ will be a maximum when H_A is a minimum (H$_B$ and H$_C$ are constant). This occurs when $\left(\frac{\partial H_A}{\partial p}\right)_T = 0$. Now from equation (2) the change in temperature produced by the differential Joule-Thomson effect is given by

$$dT = -\frac{1}{C_p}\left(\frac{\partial H}{\partial p}\right)_T \cdot dp.$$

Hence the above condition for a maximum yield of liquid requires that the input pressure and temperature are situated on the inversion curve of the differential Joule-Thomson effect. A knowledge of the inversion curve therefore permits the calculation of the most favourable input pressure for any chosen input temperature and the theoretical efficiency can be deduced when H is known as a function of p and T.

Thus for air Hausen's data for the Joule-Thomson effect show that the maximum efficiency is obtained when the input pressure is 275 atm. if the air enters at room temperature (290° K.) and expands to 1 atm. pressure. This may be compared with the conditions employed in the

Hampson * liquefier, input pressure 150-200 atm., input temperature 290° K., expansion to 1 atm. This liquefier does not therefore work under the theoretically most favourable conditions. It has, however, the advantage of simplicity and ease of manipulation and is used for the small scale production of liquid air for laboratory purposes. The theoretical efficiency for the actual input conditions is such that about 8 per cent. of the air entering the liquefier should be drawn off as liquid. Actually the efficiency of the commonly used 1 litre/hour size machine is considerably less than that.

In the larger air liquefiers using the Linde process the overall efficiency is increased by two differences in technique compared with the Hampson process. The air enters the liquefier at 200 atm. pressure after being cooled to about − 40° C. by means of an ammonia refrigerating machine, whereby the fraction of air liquefied is considerably increased. Further, the air expands to about 40 atm. instead of to 1 atm., whereby the Joule-Thomson cooling is not greatly reduced but the amount of work which has to be done in compressing the air is much less. The main bulk of the air has only to be compressed from 40 to 200 atm. and only that fraction of the air which actually liquefies and so leaves the circulation has to be compressed from 1 atm. to 200 atm. Since the work done in compressing a gas isothermally from pressure p_1 to pressure p_2 is $RT \log \left(\frac{p_1}{p_2}\right)$, it will be seen that there is a considerable saving in power required by adopting Linde's procedure. Thus 2·7 kw.h. are theoretically required to produce 1 kilogram of liquid air by simple expansion from 290° K. and 200 atm. to 1 atm., while only 0·9 kw.h. are required if the air is precooled and expanded as in the Linde process. The theoretically most favourable input pressure for an input temperature of − 40° C. and expansion to about 40 atm.

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* It is assumed that the reader is familiar with the constructional details of the Hampson and Linde air liquefiers. Diagrams illustrating the processes are to be found in most advanced text-books on Heat.

is 210 atm. in agreement with the figure (200 atm.) actually used.

Similarly one can calculate that the optimum input pressure for the liquefaction of hydrogen is about 160 atm. when the input temperature is between 63° and 80° K. and the gas expands to 1 atm. Actually pressures from 150 to 165 atm. are found to give the greatest yield of liquid, and the efficiency obtained is very close to that calculated theoretically (0·285 in place of 0·295 for the Leiden liquefier with $p_A = 150$ atm., $T_A = 63°$ K.). For helium the optimum input pressure is about 30 atm., for an input temperature of 15° K. and expansion to 1 atm. Actually this pressure gives the best results in practice and a yield of about 0·13 is obtained.

The problem of the liquefaction of hydrogen is more complicated than that of air. The compressed gas has to be cooled as far as is readily possible below the inversion temperature before being allowed to expand, some means has to be adopted to prevent the narrow tubes of the liquefier being blocked by the freezing of the traces of impurity present in commercial hydrogen (mainly oxygen) and special precautions have to be taken to eliminate the risk of explosion (a 5 per cent. hydrogen-air mixture is violently explosive).

The liquefaction of hydrogen was first achieved by Dewar in 1898. Since then a number of small and large scale plants have been erected for this purpose in various parts of the world, the best known being that at the Kamerlingh Onnes Laboratory of the University of Leiden, Holland (25 litres per hr.), and that at the Physikalisch-Technische Reichsanstalt in Berlin (15 litres per hr.).*

Fig. 2 shows diagrammatically the construction of the hydrogen liquefier of the Mond Laboratory of the Royal Society at Cambridge. A limited quantity of very pure hydrogen is compressed to 160-170 atm. and enters the

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* For a description of the liquefiers and the precautions taken for safety see Leiden Comm. No. 158 and Suppl. No. 45 (Leiden liquefier) and Geiger-Scheel, "Handbuch der Physik", Vol. XI, article by Meissner, and Wein-Harms, "Handbuch der Experimental Physik", Vol. IX, Part I, article by Lenz (Berlin liquefier).

liquefier at 1. It passes through the interchanger A (actually a series of fine tubes in parallel) and then through the cooling bath B where it is cooled to about 66° K. in liquid nitrogen boiling under reduced pressure. It next passes through the regenerator coil D, expands to 1 atm. at the valve E and partially liquefies into the container F. When F is about ⅓ full the liquid hydrogen passes through the tube 6 and the coil in the interchanger G. It evaporates there and passes back through the coils D and A and leaves the liquefier at 2. The hydrogen is then recompressed and returns to the apparatus again. The compressed gas is thus cooled progressively in A, B and D until the temperature falls low enough on expansion for part of the gas to liquefy in F. The heat interchange is so efficient that the expanded gas leaves at 2 at only a few degrees below room temperature. Ordinary commercial hydrogen from cylinders enters the liquefier at 3 at a pressure of 3-4 atm. It is cooled in the interchanger A and the nitrogen bath B and then passes into G. It there meets the spiral cooled with liquid hydrogen and liquefies on it at the pressure of 3 atm. The liquid hydrogen produced in the chamber G passes through the spiral in F where its temperature falls to that of liquid hydrogen boiling under 1 atm. pressure, 20° K., and is finally drawn off at the valve H into a suitable container. The impurities present in the commercial hydrogen solidify in G and not before and, as they are denser than liquid hydrogen, they collect at the bottom of G and can be removed at the end of the run. The whole

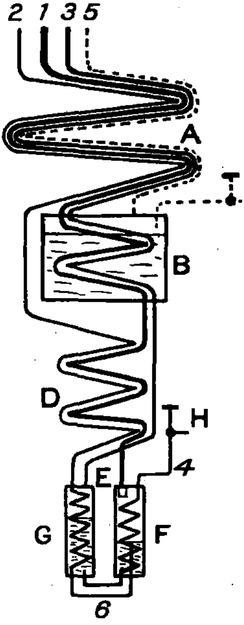

FIG. 2.

liquefier is enclosed in a metal case in which a high vacuum is maintained so as to provide good heat insulation.

The liquefier receives compressed hydrogen at the rate of 16 c. metres per hr. and produces about 4 litres of liquid per hr. The efficiency (about 0·25) is in good agreement with that calculated from expression (3) for the input conditions employed.

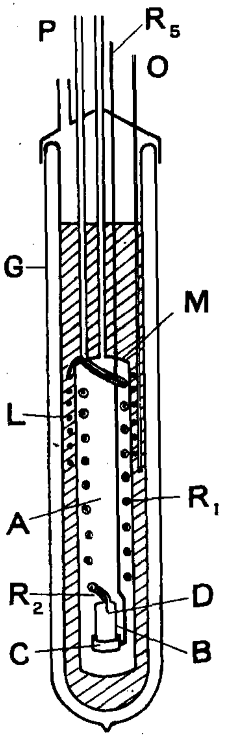

FIG. 3.

In a later model it was found possible to dispense with the two separate circulations and condense the commercial hydrogen directly into the pure liquid. The solidified impurities, as before, collected at the bottom of the container and were removed at the end of the run.

The liquefaction of helium is a matter of still greater difficulty than that of hydrogen as the gas must be cooled to hydrogen temperatures or lower* before the Joule-Thomson effect can be used to lower the temperature below the critical temperature 5·2° K. Helium was first liquefied by Kamerlingh Onnes at Leiden in 1908 and until 1923 this was the only place in the world at which physical measurements could be made in the range of temperature 1·5° K. to 4·2° K. Several relatively large scale helium liquefiers (yield 1-2 litres per hr.) are, however, now in existence. Reference may be made to the papers cited in the footnote on p. 6 for the details of the plant at Leiden and Berlin, and to p. 13 for a brief description of the

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* It is convenient to speak of "helium" or "hydrogen" temperatures when referring to the temperatures obtainable with these liquefied gases.

Kapitza helium liquefier at the Mond Laboratory, Cambridge. Fig. 3 shows the small scale helium liquefier designed by Ruhemann, which has the merit of extreme simplicity. Helium from a cylinder passes through a reducing valve, then through a tube containing charcoal immersed in liquid air to remove impurities and enters the liquefier at O at a pressure of 30-40 atm. It then traverses the coil L in the bath of liquid hydrogen in the Dewar vessel G and so is cooled well below the inversion temperature which lies below the temperatures obtainable with liquid air. The gas then passes through the inner tube R_1 of the Linde pattern interchanger A which is enclosed in the evacuated metal case M. It then expands to 1 atm. pressure at the valve D (not shown in detail) and after passing through the outer tube R_2 of the interchanger escapes into the atmosphere at P. With a rate of flow of about 1·5 litres per min. and the liquid hydrogen boiling at about 10 cm. pressure ($T = 15°$ K.), the receiver B is cooled to the temperature of the normal boiling-point of helium, 4·2° K., in 7 mins. The amount of liquid hydrogen required is quite small, about 1·5 litres. The temperature of B is indicated by the small gas thermometer C, the metal bulb of which is directly attached to the bottom of B. C is connected to a mercury manometer by means of the fine capillary R_5.

This small liquefier permits of experiments being made at temperatures from 15° K. down to about 1·9° K. with any apparatus or material which can be enclosed in the receiver B and at relatively small cost.

THE "EXTERNAL WORK" METHODS OF GAS LIQUEFACTION

Suppose a volume of gas v_1 enters the cylinder of an engine at a pressure p_1 and then expands adiabatically and reversibly, forcing down the piston (supposed frictionless) and doing work on some external system until the volume has increased to v_2 and the pressure has fallen to p_2. The outlet valve then opens and the piston rises, forcing the gas out at pressure p_2. Then the total amount of work done by the gas will be

$$\int_2^1 vdp = H_1 - H_2,$$

and the temperature of the gas will fall from T_1 to T_2. If the equation of state of the gas can be written as $pv = (C_p - C_v)T$, (C_p, C_v specific heat at constant pressure and volume respectively), the work done will be

$$W = C_p(T_1 - T_2) = H_1 - H_2,$$

and the final temperature will be given by

$$T_2 = T_1\left(\frac{p_2}{p_1}\right)^{\frac{C_p - C_v}{C_p}} \quad . \quad . \quad . \quad (4)$$

The drop in temperature calculated from (4) is greater than that obtained by throttle expansion from pressure p_1 to pressure p_2. It would thus seem that it is preferable to cool the gas by the performance of "external" work rather than "internal" work in the liquefaction of gases. In actual practice the difficulties of the realization of the "external" work method make the two types of process of practically equal efficiency for air liquefaction.

The application of the expansion engine to the liquefaction of gases forms the basis of the Claude and Heylandt methods. Fig. 4 illustrates one of the types of liquefier employing the Claude process. Air compressed to about 40 atm. passes through the interchanger I where it is cooled to about − 80° C. by the expanded gas. It then divides, a fraction M passing through the interchanger II and 1 − M through the expansion engine E. The gas expands in the engine to 1 atm. pressure (in some Claude liquefiers in two stages 40-6·3 and 6·3-1 atm.) doing external work, such as driving a dynamo, and is cooled thereby. The cold gas then passes through the interchanger II and cools the fraction M of the high pressure gas. The degree of expansion is so chosen that the temperature of interchangers II and III is considerably lower than the critical temperature of the gas but not as low as its normal boiling-point. The fraction M still at the high pressure then condenses in interchangers II and III and after expansion to 1 atm., collects at L. A portion of the liquid evaporates in the expansion through the valve V and the vapour produced returns to the compressor via the heat interchangers.

The efficiency of the liquefier depends in a rather complicated way on the temperature and pressure of the air before expansion in the engine and the fraction 1 − M of the air passing through it. For each initial pressure there is a most favourable value of the temperature of the air before expansion in E for every value of M, and the actual conditions are best determined by experiment.

Typical figures are: pressure = 40 atm., M = 0·2 (i.e. 20 per cent. of the air goes through the throttle and 80 per cent. through the expansion engine) and $T_2 = -80°$ C.

The Heylandt process differs from the Claude process in detail rather than in principle. In this case the air is compressed to 200 atm. and a fraction 1 − M enters the expansion engine at room temperature, interchanger I of Fig. 4 being omitted. After expansion the cold air passes through an interchanger, cooling the fraction M of the compressed air which is then allowed to expand to 1 atm. at a valve and partially liquefies. The temperature of the air leaving the Heylandt expansion engine is above the critical temperature and the further cooling of the incoming

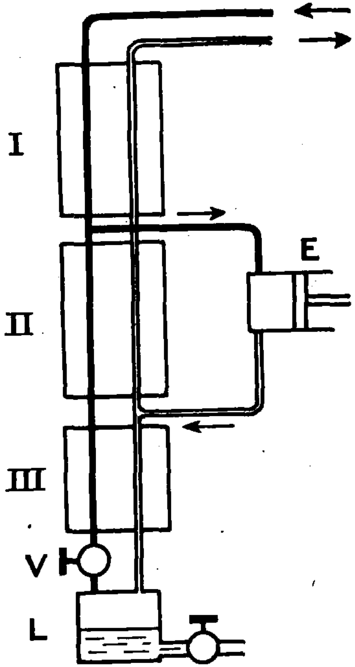

FIG. 4.

gas is obtained by the Joule-Thomson effect acting regeneratively.

One of the principal technical difficulties in the application of the " external work " method is that of maintaining the piston of the expansion engine gas-tight and lubricated in spite of the low temperature of the expanded air. The cylinder of the Claude engine is heat insulated and so reaches a temperature approximating to that of the expanded air, and the piston is maintained gas-tight by means of a patented impregnated leather cup which needs no further lubrication. The Heylandt engine is not insulated and remains approximately at room temperature so that ordinary lubrication can be employed. It is, however, run at high speed so that the expanded air is swept out of the cylinder before it can cool the latter appreciably.

The Claude process using a pressure of 40 atm. and with a partition ratio $M = 0{\cdot}2$ requires 0·87 kw.h. per kilogram of liquid air produced, thus nearly the same as the Linde process. The Heylandt process using a pressure of 200 atm. and $M = 0{\cdot}4$ requires about 0·7 kw.h. per kilogram of liquid air.*

It has long been realized that an expansion turbine should possess several advantages over the reciprocating engine for the production of low temperatures. It is, however, only in recent years that success has been achieved in designing such a turbine. In 1939 Kapitza produced an air liquefier which shows great promise. It works on the Claude principle, employs an expansion turbine in which the air is expanded from 5·6 to 1·5 atm. and operates with an actual overall power consumption of 1·2 kw.h. per kilogram of liquid air. It is, moreover, expected that a still better efficiency will be obtained when certain suggested improvements are incorporated and larger scale plants are made. The removal of all danger due to the use of high pressures is also a valuable feature.

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* The power consumption given above and on p. 5 are theoretical estimates based on the assumption of ideal processes, i.e. strictly isothermal compression in the compressor, no friction losses, no loss of " cold ", etc. These estimates have to be multiplied by a factor of about 2 to obtain the actual overall power requirements in practice.

Kapitza has also constructed a helium liquefier employing the Heylandt principle. Helium at a pressure of 30 atm. is cooled to 65-70° K. with liquid nitrogen, passes through a heat interchanger, expands to 2·2 atm. in an expansion engine and leaves the liquefier via the heat interchangers. A further amount of helium at 15-18 atm. is cooled by the gas from the expansion engine and then expands at a valve where part of it liquefies. The temperature of the inlet to the engine is 19° K. and that of the outlet 9° K. The final temperature drop to 4·2° K. is obtained by the Joule-Thomson effect at the valve. Ninety-two per cent. of the helium passes through the engine and 8 per cent. through the expansion valve. Fifty per cent. of the latter liquefies and the plant yields 1·7-2·0 litres of liquid helium per hr. As no liquid lubricant is possible at these temperatures, the problem was solved very ingeniously as follows: a gap of 0·05 mm. was left between the cylinder and the piston and a series of shallow grooves was cut in the latter. A small proportion of the compressed helium escapes past the piston and acts as the lubricant, the presence of the grooves ensuring that the piston moves coaxially with the cylinder. With the rapid expansion employed the loss of gas past the piston during the cooling stroke is limited to 4 per cent. of the contents of the cylinder.

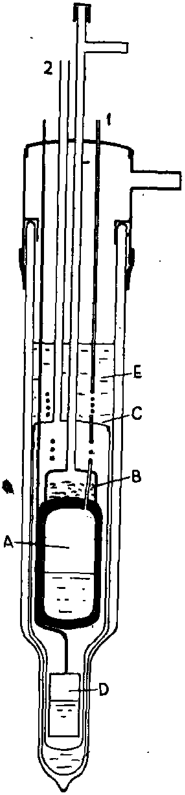

FIG. 5.

A further method for the liquefaction of helium which belongs to the "external work" class has been developed by Simon, the single expansion method. Helium is compressed via the tube 1 into the container A to a pressure of about 150 atm. and cooled