

Citation:

H. Kamerlingh Onnes, Methods and apparatus used in the cryogenic laboratory. XV. An apparatus for the purification of gaseous hydrogen by means of liquid hydrogen, in:
KNAW, Proceedings, 11, 1908-1909, Amsterdam, 1909, pp. 883-886

with a reduction valve apparatus special precautions must be taken to observe the true expansion pressure, and to ensure absence of heat conduction to the expanding jet.

2. The special apparatus here described gives a JOULE-KELVIN effect for air at 0°C. agreeing with the experimental results of JOULE and KELVIN, and with those calculated from the experimental isotherms.

It is perhaps worth noting that in the practical application of the JOULE-KELVIN effect in the LINDE-HAMPSON process for liquefying gases, in which a reduction valve which is a good conductor of heat is used, heat conduction from the valve to the expanding gas becomes of much less importance than in an accurate determination of the JOULE-KELVIN effect, for in that case the valve and the tube which conducts the gas to it are themselves cooled in the process of regeneration by the expanded gas.

In conclusion, I gratefully acknowledge my indebtedness to Prof. H. KAMERLINGH ONNES, who invited me to undertake this research and to Prof. J. P. KUENEN for their continued interest in my work and for their helpful advice, and also to the CARNEGIE Trust for a grant in aid of the expenses of the research.

Physics. — *“Methods and apparatus used in the cryogenic laboratory.*

XV. An apparatus for the purification of gaseous hydrogen by means of liquid hydrogen. By Prof. H. KAMERLINGH ONNES.

Communication 109^l from the Physical Laboratory, Leiden.

(Communicated in the meeting of March 27, 1909).

In Communication 94^f (These Proc. Sept. 1906) a liquid hydrogen cycle was described, and attention was drawn to the fact that a continuous action of that cycle was possible only when a sufficient quantity of extremely pure hydrogen was available¹⁾. In section XI of the communication referred to the method of obtaining this supply was given. The commercial gas was purified by cooling to -205° C.

¹⁾ It can easily be seen that obstruction of the regenerator-spiral must necessarily occur when a little air (or oxygen) is present in the hydrogen. The temperature at different heights of the spiral varies with changes in the velocity of the gas stream, which can scarcely be avoided, and, in any case, occur when the circulation is temporarily stopped. The result of this is that the air is alternately frozen, melted, carried lower down in the spiral and again frozen, until finally the opening of the tube is completely plugged.

and ¹⁾ by means of a liquid-air (oxygen) separator to such an extent, that, when passed through the hydrogen liquefier it gave a quantity of liquid hydrogen (although comparatively small) before the liquefier became choked, and this liquid, by vaporisation, gave pure hydrogen. When the hydrogen liquefier had again been put in a workable condition, the operations were repeated with a new quantity of gas which had undergone preliminary purification in the liquid-air separator, and the various yields of pure hydrogen thus obtained were carefully collected and united, until the required quantity was available.

This method of operating was rather troublesome, and when once I was in possession of a sufficient quantity of pure hydrogen to keep the cycle in continuous action it was an obvious advantage to avail myself of this cycle for the purification of commercial hydrogen. In Suppl. N^o. 19 the communication was already made that an apparatus was being constructed, in which the purification of the hydrogen was effected by means of its liquefaction, while another apparatus had already been constructed in which the impurities were frozen out of the gaseous hydrogen to be purified, by means of the pure liquid hydrogen of the cycle.

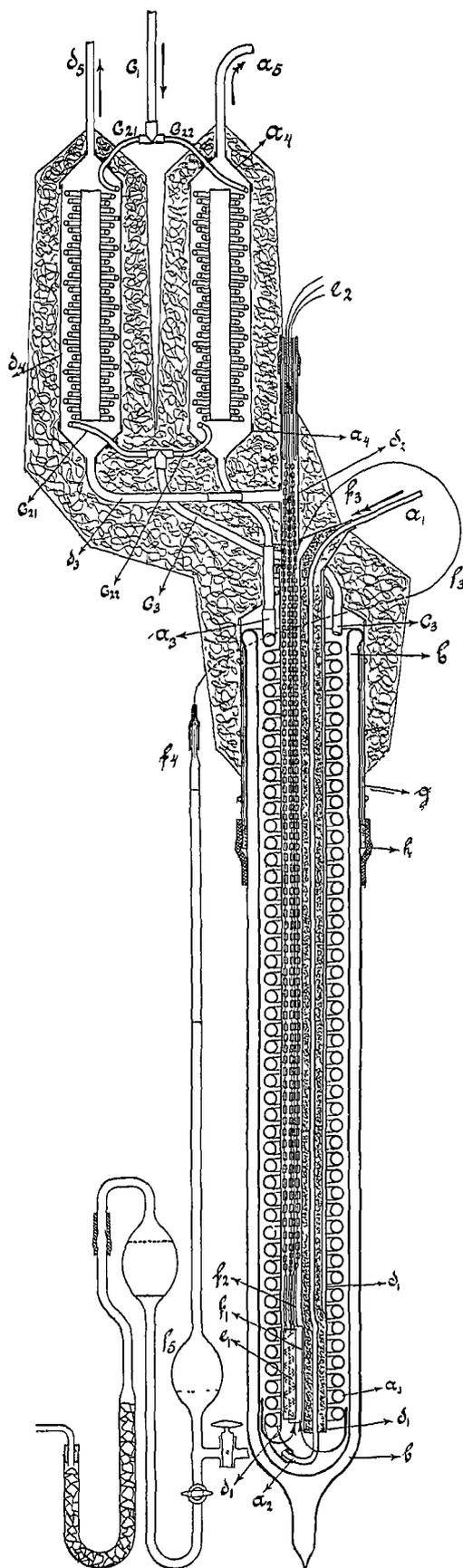
The suitability of the latter apparatus has been proved by long use; it is represented in Pl. I and a description of it is here given.

The chief portion of the apparatus is the spiral a_3 , in the lower end of which the liquid hydrogen is vaporised; it is placed in a vacuum vessel b , which is closed by means of a cap g . The hydrogen which is to be purified flows through the tube c_3 , between the vacuum glass b , and the cylinder d_1 , and along the cooled spiral in the opposite direction to that in which the gaseous hydrogen flows away, which is formed by vaporisation inside the spiral. By this means the air contained in the hydrogen is deposited on the windings of the spiral. The purified hydrogen escapes through the paper cylinder d , the copper tubes d_2 and d_3 , and the regenerator d_4 .

The liquid hydrogen is supplied through a_1 , and the insulated tube $a_1 a_2$, to the lower end of the vaporising spiral a_3 . To ascertain how much liquid hydrogen must be supplied, the temperature of the purified gas as it enters the cylinder d_1 is determined by means

¹⁾ The air separator usually works at a pressure of 60 atm. and with a velocity of 2 M.³ per hour; the impurities remaining do not then amount to more than $\frac{1}{4}\%$, and the quantity of liquid air used is about 2 litres per hour. In Comm. 94 the value $\frac{1}{20}\%$ was given: to reach this value, however, the velocity must be much smaller, in order that none of the liquid that separates out should be carried along with the current.

H. KAMERLINGH ONNES. „Methods and apparatus used in the cryogenic laboratory. XV. An apparatus for the purification of gaseous hydrogen by means of liquid hydrogen”



Proceedings Royal Acad. Amsterdam. Vol. XI,

of a hydrogen thermometer. (A resistance thermometer e , is also used; e_1 , the cylinder on which the platinum wire is wound, e_2 , the leads).

The hydrogen thermometer is arranged exactly the same as the helium-filled control thermometer in the apparatus for liquefying helium. (Cf. Comm. N^o. 108 Suppl. to the Proc. June '08) f_1 is the german-silver reservoir, $f_2 f_3$ the steel capillary, f_4 the stem, f_5 the manometer reservoir. The pressure of the hydrogen at 0° C. is chosen so that at the boiling point of hydrogen, the mercury reaches a mark at the upper end of the stem. (The pressure is then 7 cm.). The sinking of the mercury in the thermometer stem gives warning when there is not sufficient liquid hydrogen in the spiral; the supply of liquid hydrogen is so arranged that the mercury oscillates between two fixed marks.

The german-silver cap, g , is attached to the vacuum vessel by means of a rubber sleeve, and projects so far over the vacuum glass that the rubber does not become cold. The upper portion of the cap, g , and the regenerator are protected from external sources of heat by capoc.

As a rule the velocity, with which the hydrogen to be purified is supplied, is so regulated that 5 M³. per hour flow through the apparatus. In that case 4 litres of liquid hydrogen per hour are needed. This rate of production, however, cannot be kept up continuously on account of the fall of pressure in the cylinders containing the hydrogen to be purified, and of various preparatory and auxiliary operations, such as analyses, the coupling and changing of vessels with liquid hydrogen, etc. The time necessary for the manufacture of the liquid hydrogen and for compressing the purified hydrogen into cylinders must also be taken into account. The purification of 10 M³. as a general rule is the work of one day i. e. 8 working hours. On such a working day 25 litres of liquid air are used.

Of course, the purer the hydrogen supplied to it the longer can the apparatus remain in action. Hence, if commercial hydrogen¹⁾ is to be purified, the liquid air separator of Comm. No. 94 sect. XI is coupled to the apparatus here described. If that be done, the apparatus can be worked for hours without stopping. The gas which issues from the apparatus is practically perfectly pure.

It has now become an easy matter to obtain pure hydrogen suitable for liquefaction in the hydrogencycle, if the precaution is taken that a certain minimum store (in Leiden 10 M³.) of pure hydro-

¹⁾ The percentage impurity changes very irregularly, being sometimes quite small and sometimes very considerable in amount.

gen is always available, which with the present apparatus is now a matter of no difficulty. Formerly, when quite pure hydrogen was a costly commodity, many experiments were obstructed by the precautions necessary to properly collect the hydrogen that had evaporated; but now one need no longer be afraid of sacrificing pure hydrogen, if necessary, and since this is the case, the great objection to its being sent away is removed.

Physics. — “*On the motion of a metal wire through a piece of ice.*”

By Dr. J. H. MEERBURG. (Communicated by Prof. H. A. LORENTZ).

(Communicated in the meeting of March 27, 1909).

II.

A paper by G. QUINCKE¹⁾ which had escaped my notice at the time of my first communication on the above-mentioned subject²⁾ and which I happened to come across only some time ago, induced me take the subject up again. In this paper by QUINCKE the phenomena are dealt with, caused by occlusions of salt in the ice and it is shown that even with ice, formed from distilled water, these play a part. From this point of view the phenomena are also studied, that are observed when a metal wire sinks through ice; the turbidity where the wire has cut through, is ascribed³⁾ to occluded salt-solution with a different refractive power from the ice between which it lies. If this view is right, differences must be expected in the velocity of descent with ice of different origin. For then the slower descent — slower than theory would lead us to expect — is also a consequence of the fact that probably the salt-containing water does not re-freeze above the wire and this cause would be the more effective as the percentage of salt is greater.

So I repeated part of the experiments with ice, formed from distilled water. Boiled distilled water was frozen by means of a mixture of snow and common salt in a glass tube of about 4 cm. diameter, the freezing progressing, after BUNSEN's advice⁴⁾, from above downwards. If the freezing took place sufficiently slowly a quite clear rod of ice was formed in this way⁵⁾. Undoubtedly even this

1) G. QUINCKE Ann d. Phys. 18. p. 1.

2) These proceedings Vol. IX, p. 718.

3) G. QUINCKE l. c. p. 46.

4) G. QUINCKE l. c. p. 14.

5) Sometimes a turbidity appeared in the middle of the tube, starting as a plume in the axis, the fine ramifications extending upwards in gentle curves. It is curious