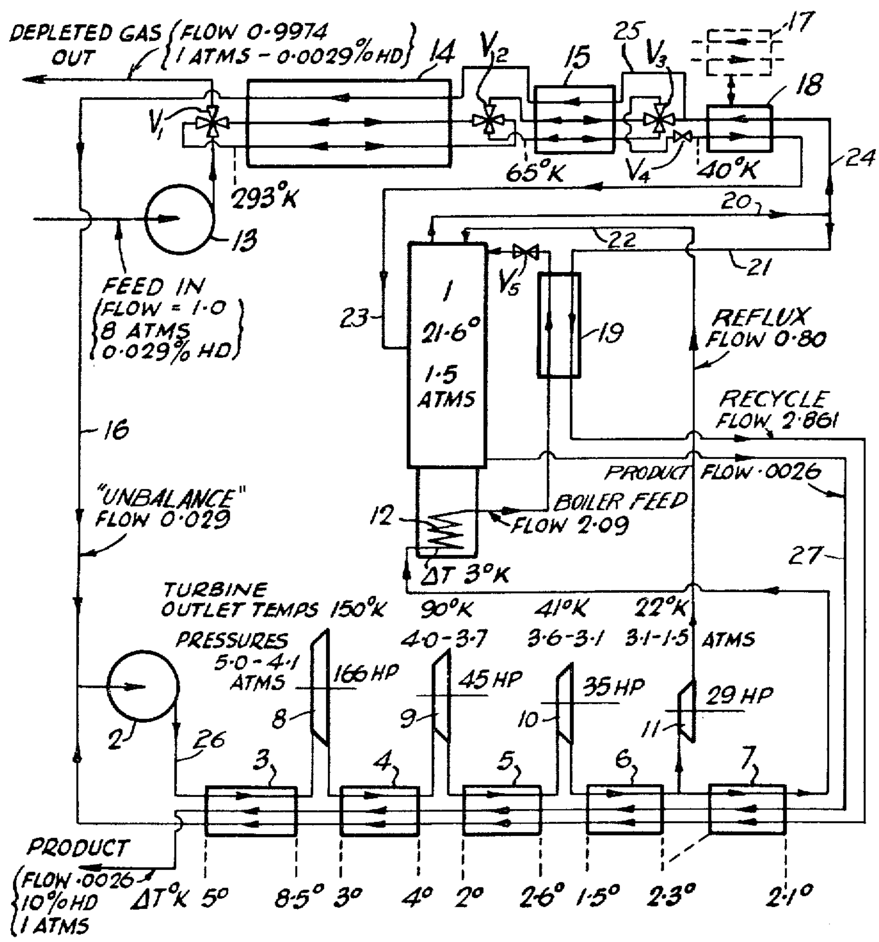
Nov. 22, 1960 W. H. DENTON 2,960,838

HYDROGEN DISTILLATION PLANT

Filed June 19, 1957



WILLIAM HAVELOCK DENTON

Inventor

Larson and Taylor

Attorneys

United States Patent Office

2,960,838

Patented Nov. 22, 1960

2,960,838

HYDROGEN DISTILLATION PLANT

William H. Denton, Harwell, England, assignor to The United Kingdom Atomic Energy Authority, London, England

Filed June 19, 1957, Ser. No. 666,509

Claims priority, application Great Britain June 19, 1956

5 Claims. (Cl. 62—28)

This invention relates to hydrogen distillation apparatus and is particularly concerned with such apparatus for the production of deuterium-enriched hydrogen.

An object of the invention is to provide apparatus having minimum power consumption and preferably capable of being fed with impure hydrogen.

According to the present invention, in a hydrogen distillation apparatus comprising a distillation column, a hydrogen feed conduit to said column, a deuterium-enriched hydrogen outlet conduit from said column, a deuterium-depleted hydrogen outlet conduit from said column, and a refrigeration circuit including an expansion turbine through which part of said depleted hydrogen is recycled to the distillation column, the refrigeration circuit comprises at least three heat exchangers and at least one additional expansion turbine connected between said heat exchangers in the hot return part of the circuit and means whereby a fraction of the depleted hydrogen not so recycled is fed into the refrigeration circuit at a point of highest temperature thereby providing increasing temperature differences at the lower temperature level of the circuit and enabling the additional turbine or turbines to be located at the highest possible temperature level without unduly small outlet temperature differences.

When applied to apparatus in which the hydrogen feed is impure and is fed through a system of reversing heat exchangers to remove the impurities, the invention advantageously provides decreasing temperature differences at the lower temperature levels of the reversing heat exchangers, small temperature differences of the order of 1° being essential at 40° K. for removing nitrogen from the feed. The nature of the invention and other features thereof will be apparent from the accompanying flow diagram of one form of apparatus in accordance with the invention for distilling impure hydrogen.

In the diagram, a distillation column **1** is connected by means of conduits **20** and **21** and conduit **22** with a refrigeration circuit comprising a compressor **2**, heat exchangers **3–7** and expansion turbines **8–11**. Recycle gas for refrigeration leaves the column **1** by conduits **20** and **21**, while the return flow to the column is divided between (*a*) the final turbine **11**, whence it returns to the column **1** by the conduit **22**, and (*b*) the heat exchanger 7, whence it feeds the boiler **12** at the foot of the column, then passes counter-current to the recycle gas glow from the column **1** in heat exchanger **19**, and finally is expanded through valve V_5 back into the column **1**.

In this embodiment of the invention four expansion turbines are shown, but in favourable circumstances this number may be reduced to two and remain within the scope of the invention. In such circumstances, a single turbine operating, for example, with an outlet temperature of 90° K. may replace the three turbines **8**, **9** and **10** operating with outlet temperatures of 150° K., 90° K. and 41° K. respectively.

The feed of impure hydrogen is taken to a compressor **13** and thence through two stages **14** and **15** of a reversing heat exchanger provided with flow reversing valves

V_1, V_2, V_3, the counter flow being the out-flow of depleted hydrogen from the column **1**. The feed gas then passes through an expansion valve V_4, one or another of heat exchangers **17** and **18**, then through conduit **23** to the feed point of the column **1**. Depleted hydrogen leaves the column **1** by conduit **20**, passes through conduit **24** and flows countercurrent to the feed gas in heat exchanger **17** or **18**, respectively. A major part of this gas then passes countercurrent to the feed gas in the two stages **14** and **15** of the reversing heat exchanger, but a fraction is taken off by a conduit **25**, passed independently countercurrent to the feed gas through the stages **14** and **15**, and is then fed at normal temperature through conduit **16** to the inlet of the compressor **2**, which thereby feeds it by conduit **26** to the hottest point of the refrigeration circuit.

A feature of the invention is the provision of a further stage of purification below 50° K. in which the expansion is expanded to the column pressure through the expansion valve V_4 with further purification in alternating heat exchangers **17** and **18**, which are connected alternately into the apparatus. Preheat is preferably applied in known manner to the hydrogen gas stream before expansion through the valve V_4, e.g. by recirculation by a conduit through part of the last stage **15** of the reversing heat exchanger, so that the gas is warmed up slightly before expansion through the valve. By this means it is cooled in the heat exchanger to a temperature below that at which it eventually leaves the heat exchanger and therefore impurities are deposited only in the heat exchanger and not at the valve V_4, where some cooling is inevitable because of the non-ideal behaviour of hydrogen at these low temperatures (below 50° K.). Blocking of the valve V_4 is thus avoided.

By finally cooling the hydrogen at the pressure of the column, complete purification is achieved in the heat exchangers **17** and **18**, which eliminates deposition of solid impurities at an expansion valve at the column feed point. Solids deposited in the alternating heat exchangers **17** and **18** at temperatures between 50° K. and 21.6° K. are removed by warming up each of these heat exchangers alternately by means of a further gas stream, while the other is in the main circuit. Owing to the very low heat capacity of the heat exchanger metal at these temperatures, only a very small amount of energy is consumed in this process. Deuterium-enriched hydrogen product leaves the bottom of the column **1** by conduit **27** and passes separately through the recycle heat exchangers **3–7** in which it is warmed up to normal temperature.

The absolute temperature, temperature differences across heat exchangers, and pressures at various points are clearly shown in the flow sheet. The horse power of the turbines is also shown and it will be evident that most of the work is done at the higher temperature levels thus substantially reducing the power required as compared with a conventional system in which all the work is done by one turbine at the temperature level of the distillation column.

The apparatus may conveniently be described as consisting of two parts, apart from the distillation column **1**, itself: (*a*) the feed and purification section comprising the compressor **13** and the heat exchangers **14**, **15**, **17** and **18**; and (*b*) the re-cycle section which provides refrigeration in the turbines **8** to **11** and reflux gas for liquid reflux through conduit **22** to the distillation column, and comprises the compressor **2** and the heat exchangers **3** to **7**, as well as the turbines **8** to **11**. The re-cycle section (*b*) operates on pure hydrogen depleted in deuterium which reduces deuterium losses to a minimum.

The fraction of pure depleted hydrogen, which is fed to the re-cycle section by the conduit **16**, may be termed the unbalance flow. This serves the dual purpose of

(1) transferring heat load from the feed section (e.g., superheat at the column feed point) to the warm end of the re-cycle section, and thence to the warmest turbine **8**, where it is extracted with expenditure of least power, and (2) causing the diminishing temperature difference in the feed heat exchangers **14** and **15** as one proceeds to lower temperatures, which is necessary for creating conditions for re-evaporation of solid impurities in these reversing heating exchangers **14** and **15**.

An advantage achieved by the use at least one turbine at a higher temperature in addition to the coldest turbine **11** is the considerable reduction in the pressure required of the re-cycle compressor **2** which leads to a considerable increase in the volume flow of gas through the turbines. The relatively large flow of gas in the re-cycle section achieved in the present system (e.g., 2.86 compared with 1.0 for the feed in, as shown in the diagram) also allows a large gas flow through the turbines, which is of particular advantage for the coldest turbine and is necessary to achieve high thermodynamic efficiencies. Higher gas volume flow enables the use of unduly high rotating speeds (r.p.m.) in the turbines to be avoided, since the high tip speeds necessary can be achieved by using larger turbine wheel diameter. Very high rotating speeds which would introduce serious brake and bearing problems are thus avoided. The result of this is that turbines of simpler engineering construction and more reliable operation may be used than would otherwise be possible.

These advantages are particularly important in the refrigeration of hydrogen, compared with other gases, owing to its low molecular weight imposing very high turbine tip speeds in the use of either axial flow or radial-inward flow turbines. This usually tends to require the use of multi-stage turbines, but in the present system permits the use of only single stage machines. Indeed, if the minimum temperature differences imposed by the heat exchangers in the re-cycle stream are reduced to the order of 1° at the outlet of a turbine, the number of turbines may be reduced from four to two (as hereinbefore described) and both of the latter may consist of single stage turbines.

As a further consequence of the reduction of pressure in the re-cycle section, the heat exchangers may be of cheaper construction, leading to a substantial reduction in the capital cost of the apparatus.

I claim:

1. Hydrogen distillation apparatus comprising a fractionating column having inlet means for feeding hydrogen to the column, first outlet means for withdrawing deuterium enriched hydrogen from the column and second outlet means for withdrawing deuterium depleted hydrogen from the column, a plurality of expansion turbines, a plurality of heat exchangers in a series of at least a number equal to said plurality of expansion turbines, a compressor, means communicating with said second outlet means for feeding a first portion of said deuterium depleted hydrogen in a first direction of flow successively through each of said series of heat exchangers and thence to said compressor, means communicating with said second outlet means for feeding a second portion of said deuterium depleted hydrogen to said compressor, means for feeding said first and second portions of deuterium depleted hydrogen from said compressor through successive stages each of which comprises passing the first and second portions through one of said heat exchangers in counterflow to said first direction of flow and thence through one of said expansion turbines, and means for returning said first and second portions to said fractionating column from the expansion turbine in the last of said successive stages.

2. Hydrogen distillation apparatus according to claim 1 comprising a plurality n of expansion turbines and at least a plurality $n+1$ of said heat exchangers, and further comprising means for feeding a part of said first and second portions emanating from the heat exchanger in the last of said successive stages through the extra of said heat exchangers in counterflow to said first direction of flow and thence to the boiler of said fractionating column for return ultimately to said fractionating column.

3. Hydrogen distillation apparatus according to claim 1 wherein said inlet means includes means for purifying impure hydrogen being fed to said fractionating column, said purifying means including a series of reversing heat exchangers.

4. Hydrogen distillation apparatus according to claim 3 wherein said means for feeding said second portion of deuterium depleted hydrogen to said compressor feeds at least a part of said second portion through said series of reversing heat exchangers in counterflow to said impure hydrogen.

5. Hydrogen distillation apparatus according to claim 4 wherein said purifying means further comprises means for purifying said impure hydrogen at a temperature below 50° K. and at the pressure of the fractionating column, said means for feeding said second portion passing said second portion through said last-named means in counterflow to said impure hydrogen.

References Cited in the file of this patent

UNITED STATES PATENTS

2,095,809	Gomonet	Oct. 12, 1937
2,496,380	Crawford	Feb. 7, 1950
2,502,282	Schlitt	Mar. 28, 1950
2,534,478	Roberts	Dec. 19, 1950
2,660,038	Pool	Nov. 24, 1953
2,667,044	Collins	Jan. 26, 1954