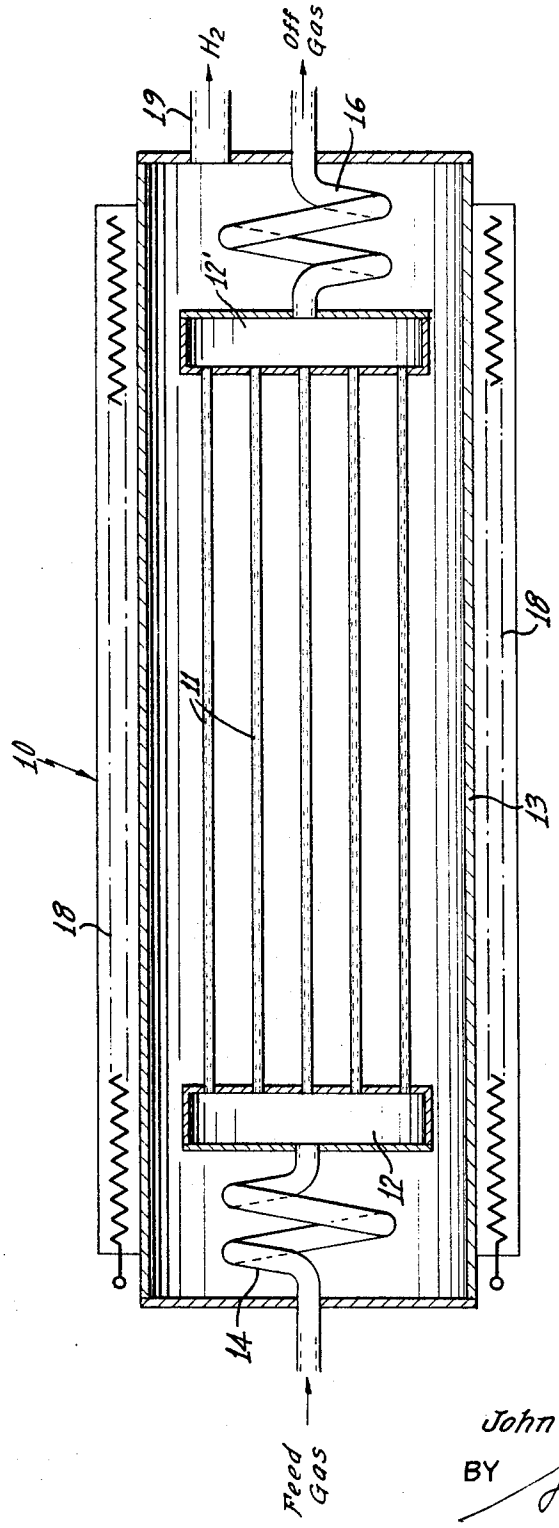
July 16, 1968 J. H. KOCH, JR 3,392,510

HYDROGEN DIFFUSION APPARATUS

Filed April 1, 1965



INVENTOR
John H. Koch, Jr.
BY Samuel Kahn
ATTORNEY

United States Patent Office 3,392,510 Patented July 16, 1968

3,392,510

HYDROGEN DIFFUSION APPARATUS

John H. Koch, Jr., Nutley, N.J., assignor to Engelhard Industries, Inc., Newark, N.J., a corporation of Delaware

Filed Apr. 1, 1965, Ser. No. 444,656
1 Claim. (Cl. 55—158)

The present invention relates to hydrogen purification apparatus and particularly to an improved unit for purification of hydrogen by diffusion through a non-porous hydrogen-permeable metal.

It is well known to purify hydrogen by diffusion through a thin wall of non-porous hydrogen-pereable metal. Impure gas containing hydrogen is brought into contact with the metal wall under heat and pressure and only pure hydrogen permeates the wall and diffuses through it.

Known hydrogen-permeable metals include palladium and palladium alloys such as palladium-silver and palladium-nickel. A suitable palladium-silver binary alloy contains 25% by weight silver.

One form of apparatus used to purify hydrogen by the diffusion process employs a bundle of thin-walled tubes of palladium alloy, fixedly supported at the ends within a collection chamber. Impure feed gas containing hydrogen is fed into the bores of the tubes, and hydrogen which diffuses through the tube walls is collected in and removed from the surrounding chamber. In such apparatus, heating means is ordinarily provided since hydrogen diffusion through palladium alloys is ordinarily effected at elevated temperature, e.g., 300 to 600° C.

For efficiency in the use of palladium or palladium alloy diffusion tubes, wall thicknesses between about 3 mils and 6 mils are employed. Because of the expansion and contraction of the thin-walled metal tubes due to hydrogen adsorption and removal as well as dimensional changes due to thermal stresses which occur on heating and cooling, there is great stress on the tubes and joints between tubes and headers, resulting in failure particularly at the joints.

In accordance with the present invention, apparatus is provided in which problem of failure of palladium and palladium alloy hydrogen tube diffusers due to stresses from heating and cooling in the presence of hydrogen is minimized. The hydrogen diffuser consists of a bundle of straight tubes open on each end to headers or tube sheets to which the tubes are welded or brazed securely. Each header consists of a chamber, which may be cylindrical in cross section, one wall of which is the sheet containing multiple open tube ends, and the other wall containing a single attached tube opening, preferably in central position, to which coiled inlet and outlet tubes are attached. Such coiled tubes or "pigtails" provide the sole fixed support for the tube bundle within an outer collection chamber and, in order to relieve stresses, each coiled tube is spirally coiled in opposite direction. The tube assembly moves freely in a cylindrical pressure vessel with closed ends, the coils passing through the center of the respective ends of the pressure vessel. Feed gas enters the tube bundle at one end, diffusion takes place through the straight diffusion tubes and undiffused gas passes off through the coil at the other end of the tube bundle. Diffused pure hydrogen is removed from the pressure vessel chamber.

The apparatus of the present invention is more fully described below with reference to the attached drawing which shows the apparatus in section.

Referring to the drawing, the apparatus of the invention consists of a diffusion cell **10** comprising at least one diffusion tube **11** of non-porous hydrogen-permeable metal such as palladium alloyed with silver. The diffusion

tubes **11** are attached between and open into cylindrical headers **12** and **12'**, and the tubes and headers are enclosed in a cylindrical vessel **13**. At one end of the cell, an inlet tube **14** has one end connected to and opening into one of the headers **12** and its other end opening outside the vessel **13**. Inlet tube **14** is of coiled, or "pigtail" configuration coiled either in a clockwise or counter clockwise direction. Similarly, an exhaust tube **16** at the other end of the tube bundle is connected to the other header **12'** and opens outside the tank. Exhaust tube **16** is also of coiled configuration, but coiled in a direction opposite to that of inlet tube **14**.

The diffusion cell **10** is heated by electric heating elements **18** adjacent the outside of vessel **13**. Outlet tube **19** in vessel **13** is provided for removal of diffused pure hydrogen.

In the operation of the diffusion cell, impure gas containing hydrogen is fed into the inlet tube **14** at one end and flows through the adjacent header **12**, through the tubes **11** to the other header **12'** and out of the cell through the exhaust tube **16** which may be provided with a valve to control rate of the gas flow. As the gas mixture passes through the tubes **11**, hydrogen permeates and diffuses through the walls of the tubes **11** into vessel **13** from which it is removed by outlet tube **19**.

It will be understood from the above that the sole points of fixed connection between the tube bundle and header, and the collection vessel **13**, are at the points where the inlet tube **14** and the outlet tube **16** enter the vessel **13**. Thus, any strain due to twisting or expansion of the diffusion tubes **11** is taken up by the coiled inlet and outlet tubes.

While the diffusion tubes are of palladium or palladium alloy, the remaining cell parts are suitably of stainless steel. The diffusion tubes with the header chambers and the two pigtails are first assembled with the pigtails in stretched position. The pigtails are usually of equal diameter and length, although the inlet pigtail may be of larger bore. This sub-assembly is then sealed to the jacketing chamber. Assembly with the pigtails in stretched position assists in countering the tendency to lengthening of the coils on providing internal gas pressure due to Bourdon tube effect and the tendency to stretching of the pigtails due to greater expansion of the stainless steel than the palladium alloys on heating to 300 to 600° C. in operation of the unit.

The apparatus of the present invention may be employed in horizontal or vertical position, but vertical positioning of the assembly may be preferable because movement by sagging is minimized and also because less floor space is then required.

While the apparatus of the invention has been described above in connection with a particular embodiment thereof, it will be appreciated that some modifications may be made in the structure and arrangement without departing from the spirit and scope of the invention as defined by the following claim.

I claim:

1. A hydrogen purification unit comprising an outer vessel, a plurality of thin-walled palladium-containing diffusion tubes supported at each end by a tube sheet, each of said tube sheets comprising one wall of a header composed of spaced walls defining a chamber, the opposite wall of each tube header having an opening and attached thereto a single coiled tube, one of said coiled tubes being coiled in a clockwise direction, and the other in a counter clockwise direction, the said diffusion tubes and attached headers being enclosed within said outer vessel and fixedly supported therein solely by attachment to each of said coiled tubes.

(References on following page)

3

References Cited

UNITED STATES PATENTS

2,911,057	11/1959	Green et al.	55—158
3,022,858	2/1962	Tillyer et al.	55—16
3,210,162	10/1965	Rudd	55—158 X
3,251,173	5/1966	Ehlers et al.	55—158
3,274,754	9/1966	Rubin	55—158

4

FOREIGN PATENTS

667,016	7/1963	Canada.
1,178,480	12/1958	France.

REUBIN FRIEDMAN, *Primary Examiner.*

J. W. ADEE, *Assistant Examiner.*