Nov. 3, 1959 R. B. GREEN ET AL 2,911,057

HYDROGEN PURIFIER

Filed July 30, 1957

FIG. 1

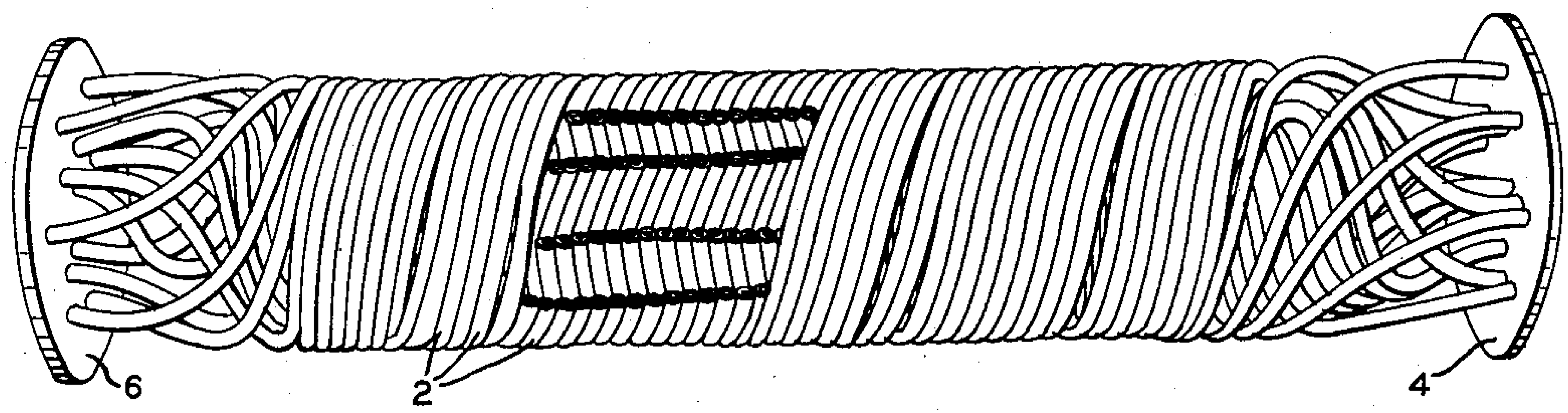


FIG. 2

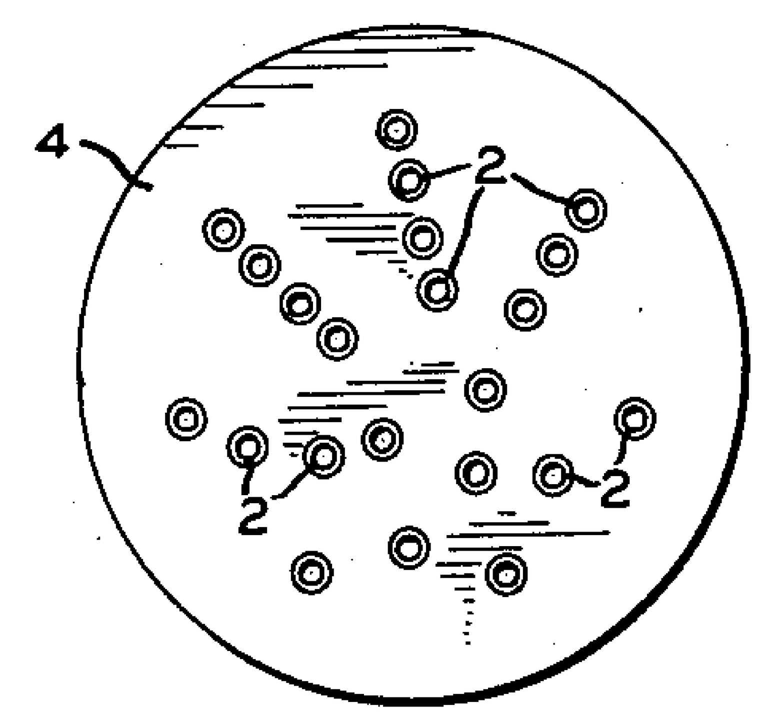
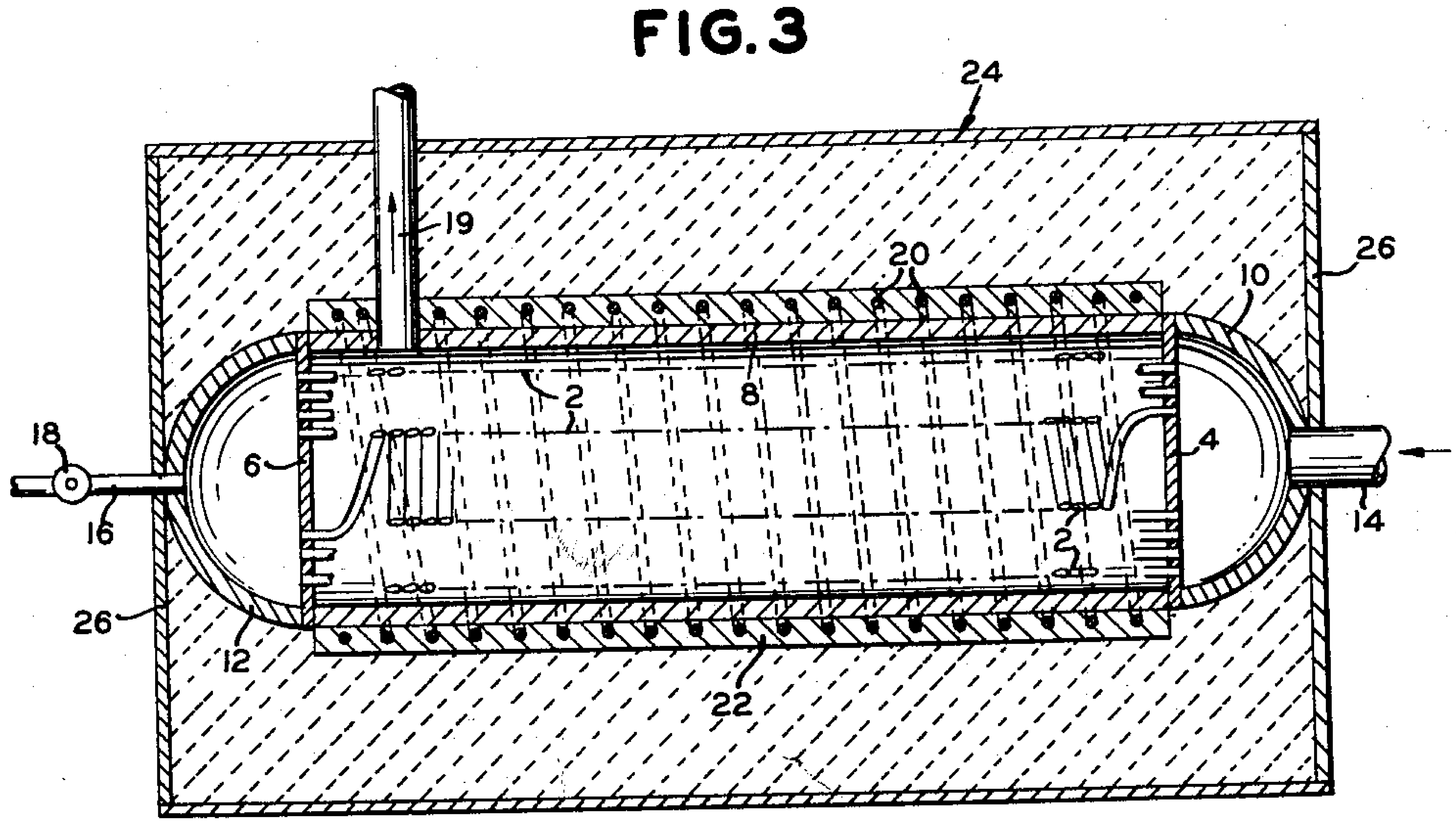


FIG. 3



INVENTORS
JAMES S. HILL
ROBERT B. GREEN

BY Karl Huber
James E. Bryan
ATTORNEYS

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HYDROGEN PURIFIER

Robert Boyce Green, Chatham, and James S. Hill, Cranford, N.J., assignors to Engelhard Industries, Inc., a corporation of New Jersey

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1 Claim. (Cl. 183—2)

This invention relates to a hydrogen purifier in which the hydrogen is purified by permeation through palladium metal.

It is known that it is possible to purify hydrogen by permeation through palladium metal, and this purification is customarily effected by the use of a palladium thimble which is inserted into a metal shell. The hydrogen is passed into the palladium thimble, and impurities are constantly withdrawn from the interior of the thimble by means of a constantly burning jet, the impurities being withdrawn with a portion of the hydrogen. These prior art devices generally are placed in a suitable furnace whereby they may be heated to the required temperature, and it is generally necessary to use a number of such devices to obtain any appreciable output of purified hydrogen.

In accordance with the present invention, a purifier is provided which is compact in size and yet which provides for a substantial output of purified hydrogen. So-called commercial hydrogen generally has a purity of about 99.5 percent, with the impurities comprising gases such as oxygen, hydrocarbons and water vapor. The purifier of the present invention operates to remove these impurities and produces a pure hydrogen, but it is also useful in purifying hydrogen having a much higher percentage of impurities than that present in commercial hydrogen, and may, for example, be used to purify a gas having only 50–60 percent hydrogen. The unit may be operated at a temperature of about 275 to 550° C., and good results have been obtained operating at a temperature of about 400° C. The pressure may be in the range of atmospheric to 100 p.s.i.g., or higher.

Referring to the accompanying drawing in which one embodiment of the purifier of the invention is shown,

Fig. 1 is a view in elevation of the tube bundle and the end plates, which constitute the purifying section of the apparatus of the invention,

Fig. 2 is an end view of the tube bundle showing the tubes secured to the end plate, and

Fig. 3 is a view in section through the assembled purifier.

Referring to Figs. 1 and 2 of the drawing, the tube bundle consists of a plurality of palladium tubes **2**, which may be of any desired number depending upon the capacity required, these tubes being secured to a pair of end plates **4** and **6**, respectively. The end plates may be made of a suitable material such as stainless steel, and the tubes are secured thereto by silver solder.

Referring to Fig. 3 of the drawing, the tube bundle **2**, and the end plates **4** and **6**, are placed inside of a stainless steel shell **8** having the stainless steel caps **10** and **12**, respectively, secured to each end of the shell by any suitable means, such as welding.

The cap **10** is provided with an inlet **14** by means of which impure hydrogen is passed into the unit, while the cap **12** is provided with the bleed tube **16**, having a valve **18** therein, the bleed tube serving to vent the impurities from the interior of the tube bundle **2**. An outlet **19** is

provided in the wall of the stainless steel shell **8**, whereby purified hydrogen which has permeated through the walls of the tubes **2**, may be withdrawn from the shell.

A resistance winding **20** surrounds the stainless steel shell **8**, this resistance winding being comprised of any suitable material such as Nichrome wire, and the winding is encased or imbedded in a layer of cement **22**, such as asbestos cement, for example.

The entire unit is mounted in an aluminum container or box **24**, having end plates **26** made from a suitable material such as asbestos cement, or the like. The purifying unit is insulated from the container **24** by means of a suitable insulating material **28**, such as magnesium oxide or the like.

In the fabrication of the purifier of the invention, the palladium tubes are wound on an arbor designed with a series of concentric holes in a head plate at one end thereof. The end of each tube is fixed in a hole, and the spacing of the holes provides the lead of the tube as it is wound. If, for example, the first layer contains four tubes, the second layer, wound with a reversed spiral directly onto the first layer, may contain five tubes, the third layer, 6 tubes, and the fourth layer, seven tubes.

The layers support each other and consecutive layers alternate right and left hand helixes. As the diameter increases with additional layers, more tubes are wound in each layer so that as the number of tubes in a layer increases, the number of turns decreases, and the length of the tubes remains constant. This process can continue until the tubes approach a straight line. After the tubes are coiled, the ends are removed from the arbor, and fitted into drilled steel end plates. The tubes are soldered with silver solder, and after a check for leaks, the coil is welded into a stainless steel pipe. The stainless steel pipe and end plates are just large enough in diameter so that the welding operation does not affect the palladium or the solder. Dome-shaped, stainless steel caps are welded onto the ends of the unit, and an inlet tube is welded into the center of one cap; an outlet tube is mounted in the side of the casing, and a bleeder tube of copper is welded into the center of the opposite cap.

After testing for leakage, the unit is wound with Nichrome wire to provide a resistance heater. The wire is wound on asbestos paper and cemented with aluminum oxide cement. The assembly is mounted inside a steel or aluminum shell, between two end discs of asbestos cement, and is filled with an insulator such as magnesium oxide or diatomaceous earth. The resistance winding is then connected to a control box and the unit is ready for use.

In one example of the use of the purifier of the present invention, the purifier had a tube bundle 10" long and 2½" in diameter. The tubes were ⅛" palladium tubes, 4 ft. long, and the tubes were 22 in number. When this unit was operated at a temperature of 300° C. and a pressure of 100 p.s.i.g. and the feed was hydrogen having a purity of 99.5 percent, a purified hydrogen was obtained in a volume of 20 cubic feet per hour.

Since the permeation of hydrogen through palladium is directly proportional to the area, it will be seen that the present invention provides for a high throughput in a unit which has very small exterior dimensions, by virtue of the high surface area presented by the plurality of tubes through which the hydrogen permeates. Also, since each of the tubes is of the same length, the possibility of one or more tubes bursting by reason of a pressure differential within the unit, is eliminated.

It will be obvious to those skilled in the art that many modifications may be made within the scope of the present invention without departing from the spirit thereof, and the invention includes all such modifications.

What is claimed is:

A hydrogen purifier comprising a shell having a plurality of palladium tubes of equal length coiled therein in layers of alternating right and left hand helixes, the ends of the tubes being secured in a pair of end plates, means for admitting impure hydrogen into the tubes, means for removing purified hydrogen from the shell, valve means for removing impurities from the interior of the tubes, a resistance winding around the exterior of the shell, and a layer of insulating cement over the resistance winding.

References Cited in the file of this patent

UNITED STATES PATENTS

2,536,610	King et al.	Jan. 2, 1951
2,671,337	Husberg	Mar. 9, 1954

FOREIGN PATENTS

22,340	Great Britain	1891