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- (72) Inventor; and
- (71) Applicant : **NEE, Han** [US/US]; 7 Sommet, Newport Coast, CA 92657 (US).
- (74) Agent: **STREETER, Tom**; Target Technology Company, LLC, 564 Wald, Irvine, CA 92618-4637 (US).
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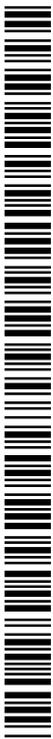
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(54) Title: METHODS OF MAKING AND USING PALLADIUM ALLOYS

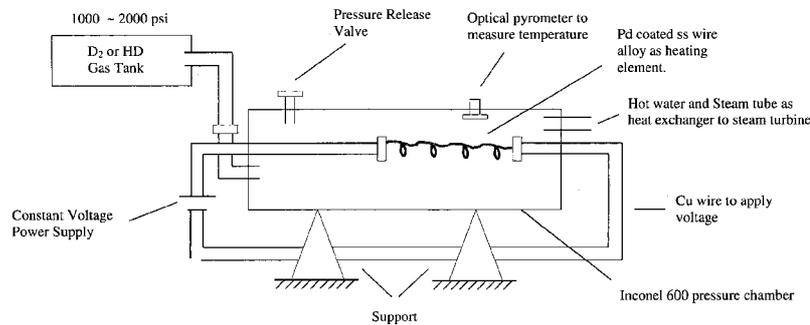


Fig 2. Deterium Gas Charging and Energy Generation Device

(57) Abstract: A gas, comprising deuterium gas (D2), hydrogen gas (H2), or a gas of their isotope (HD), singly or in combination, is dissolved into a metal comprising an alloy of palladium with boron, carbon, oxygen, or any combination thereof. The temperature is about 400°C to about 1065°C, and the gas pressure is about 0.1 atmospheres to about 1000 atmospheres. The metal consists of a single phase. An electric field, magnetic field, or ultrasonic excitation is applied to the metal.

## METHODS OF MAKING AND USING PALLADIUM ALLOYS

## BACKGROUND OF THE INVENTION

The invention relates to energy generation using various palladium alloys and  
 5 methods of making the palladium alloys. The alloys can also be used as hydrogen  
 gas purification membrane and electrodes for electrochemical processes.

In a hydrogen gas purification membrane, a palladium alloy is used due to its high  
 diffusivity and solubility of hydrogen. With impure hydrogen at the high pressure  
 (P-high) side, hydrogen gas will diffuse through the palladium alloy membrane with  
 10 area A to the low pressure (P-low) side. The following equation describes the  
 permeation phenomenon:

$$\text{Permeation rate} = D \times S [(P\text{-high})^{1/2} - (P\text{-low})^{1/2}] / t \quad \text{----- equation A -----}$$

where t is the thickness of the membrane, and D and S are, respectively, the  
 diffusion coefficient and the solubility of hydrogen atoms in the palladium alloy.

15 Hydrogen gas dissolves in the metal as atoms according to the following equation:



where a hydrogen molecule dissociates in metal into two hydrogen nuclei (each a  
 proton with a nuclear spin of quantum number 1) and two electrons, which join the  
 valance electrons of the host lattice. The same equations also apply to other  
 20 hydrogen isotopes such as deuterium and tritium.

The typical operation temperature of the Pd membrane is between 350 to 550  
 degrees C to get high permeability of hydrogen or its isotopes. Other gases either  
 dissolve very little or diffuse slowly in the membrane. They thus will be separated  
 from the hydrogen by the membrane.

The workhorse of the membrane material has been palladium with 23-25% silver by weight. This material exists as a single phase from room temperature to the highest operation temperature.

US patents 6,764,561 and 7,381,368 disclose a two-phase palladium-boron alloy for this purpose. This material will undergo phase changes if operated between room temperature and 450 degrees C. Because different Pd-B phases have different lattice spacing, cycling between room temperature and high temperature will cause lattice misfit to grow and stress between the phases which will eventually leads to cracks of the membrane. This material can operate at lower temperature, say below 400 to 410 degrees C to avoid phase transformation, but this will lower its permeation rate, because the diffusion rate of hydrogen nucleus lowers with temperature, which lowers the membrane's efficiency. Furthermore, because of the concentration gradient of the hydrogen in the membrane from the high pressure side to the low pressure side, the relative amount of the two phases will vary across the thickness of the membrane. This causes additional stresses at the interphase of the two phases which may leads to cracks. Therefore, its usefulness as a hydrogen membrane material is limited.

US patent 5,518,556 discloses a single phase boron-containing palladium or platinum alloy, but it was made by a chemical vapor deposition process and used as a hard coating for wear resistance purpose. It is not useful as a hydrogen permeation membrane.

In WO 98/03699, Patterson discloses an energy producing device using nickel coated on palladium on a plastic ball as a cathode in an electrolytic cell. The cell operates between room temperature and near 100 degrees C. Patterson claimed a nuclear reaction driven by a chemical process. It had at least two substantial deficiencies, namely:

- 1) The coefficient of thermal expansion between the plastic and the metal coatings are at least an order of magnitude different. This causes stresses between

the coating layers and the core of plastic ball. Repeated temperature changes of the metal coated ball will cause the coating to flake off and will stop the energy generation process.

2) The electrolytic process was done in aqueous solutions. This limits its  
5 temperature to about the boiling point of the solution, or about 100 degrees C. A chemical process normally is a function of temperature: the higher the temperature, the higher the reaction rate. Limiting the operating temperature to about 100 degrees C means a low reaction rate and low efficiency system.

10 In US 2003/0230481 A1, Miley discloses an improved version of the above mentioned WO 98/03699 by Patterson, but the essential deficiencies of WO 98/03699 were still there.

In US 2009/0086877 A1, Hagelstein et al discloses an apparatus using deuterium and/or hydrogen deuterium in material such as palladium to generate energy from nuclear reaction. No palladium alloy composition is disclosed.

15

#### SUMMARY OF THE INVENTION

The palladium alloys used in this invention are palladium added with boron, carbon, or oxygen, singly or in combination. The concentration of oxygen as an alloying element in palladium ranges from about 0.01 atomic percent to about 1.0 atomic  
20 percent, and with a more preferred range from about 0.1 atomic percent to about 0.6 atomic percent. The concentration of carbon as an alloying element in palladium ranges from 0.01 atomic percent to about 10.0 atomic percent, and with a more preferred range from about 0.1 atomic percent to about 5.0 atomic percent. The concentration of boron as an alloying element in palladium ranges from 0.01 atomic  
25 percent to about 20 atomic percent, and with a more preferred range from about 0.1 atomic percent to about 18 atomic percent.

The preferred operating pressure is from about 0.1 atmospheres to about 1000 atmospheres, with a more preferred range of about 1 atmospheres to about 100 atmospheres at the operating condition. The preferred operating temperature for Pd-B, Pd-C and Pd-O alloys are from about 400 to about 1065 degrees C, with a more preferred temperature range from about 450 to about 750 degrees C. These alloys all exist as single phase alloys at these operating temperatures and pressures.

By applying an electric potential across the palladium alloy in various forms in the presence of deuterium gas, deuterium hydrogen or other deuterium isotope gases at the above mentioned temperature and pressure, substantial heat is generated in addition to the energy generated by resistance heating caused by an electric current passing through the alloy.

These alloys can be prepared as a coating on a suitable substrate by typical physical vapor deposition processes such as sputtering, ion plating, thermal evaporation techniques, and so on. It is preferred that the substrate have an orientation of the {100} plane being parallel to the substrate surface, so that the coating will also have the same preferred orientation.

A wrought form of the alloys can be made by power metallurgy technique by first mixing powders of the various elements and going through a pressing, isostatic pressing and sintering process or a hot isostatic pressing process. This is followed by various metal-shaping processes such as hot forging or hot rolling. The alloys are then preferably annealed in a vacuum and quenched to room temperature.

For palladium alloys of a foil form, a suitable rolling schedule can be developed to produce foils with a preferred orientation of the {100} plane being parallel to the surface and the (100) direction being parallel to the rolling direction.

The above mentioned palladium materials can be further alloyed with various oxides such as alumina, silicate, sodium oxide, calcium oxide, and so on. They are mechanically alloyed in powder form, then pressed and lightly sintered in a vacuum,

or in hydrogen or deuterium gas, to achieve a porous structure of less than full density.

The above mentioned palladium alloys in coating, foil or porous form can be used as the core material to generate energy or heat in a pressurized chamber filled with  
5 deuterium (D<sub>2</sub>) gas, hydrogen (H<sub>2</sub>) gas or their isotope (HD), singly or in combination, at the above mentioned pressure and temperature. Excess energy or heat can be generated and triggered by the application of electric field, magnetic field, ultrasonic excitation etc. to the core material.

10 These palladium alloys in various forms can also be used as hydrogen permeation membranes, hydrogen isotope separation membrane, or electrodes in electrochemical cells.

Excess heat or energy can also be generated by using the palladium alloys in various forms as a cathode with a counter electrode as an anode. The electrodes are placed in an electrolyte of deuterium water or heavy water (D<sub>2</sub>O), or semi-heavy water  
15 (HDO). The electrolyte includes an ionic solution with substantial electrical conductivity. A voltage is applied across the electrodes to electrolyze the solution and generate deuterium or HD gases. These gasses dissolve into the palladium electrode and cause the electrode to generate excess heat or excess energy.

20 Furthermore, a palladium and/or palladium alloys of this invention can be coated by electroplating, or by an electroless plating process, onto a wire core. The core may be of copper, a copper alloy such as CDA 725, or an austenitic stainless steel such as 304S, 309S or 316S.

Before coating, the wire core is hard drawn to more than 90% reduction in diameter and then annealed to promote (100) fiber texture. After electroplating or electroless  
25 plating, the material is annealed in a vacuum at a suitable temperature to recrystallize the grain. It is then further hard drawn to gauge with more than 90% reduction. A final wire diameter of 25 to about 50 microns is preferred. The

palladium or palladium alloy should have a thickness of 1 or 2 microns, with (100) fibrous texture.

#### BRIEF DESCRIPTION OF THE DRAWINGS

5 FIG. 1 is a hydrogen permeation device.

FIG. 2 is a deuterium gas charging and energy generation device.

#### DETAILED DESCRIPTION

The palladium alloys used in this invention are palladium added with boron, carbon,  
10 or oxygen, singly or in combination. The concentration of oxygen as an alloying  
element in palladium ranges from about 0.01 atomic percent to about 1.0 atomic  
percent, with a more preferred range from about 0.1 atomic percent to about 0.6  
atomic percent. The concentration of carbon as an alloying element in palladium  
15 ranges from 0.001 atomic percent to about 5.0 atomic percent, with a more preferred  
range from about 0.001 atomic percent to about 1.5 atomic percent. The  
concentration of boron as an alloying element in palladium ranges from 0.01 atomic  
percent to about 20 atomic percent and with a more preferred range from about 0.1  
atomic percent to about 18 atomic percent.

The preferred operating pressure is from about 0.1 atmospheres to about 1000  
20 atmospheres, with a more preferred range of about 1 atmospheres to about 100  
atmospheres. The preferred operating temperature for Pd-B, Pd-C and Pd-O alloys  
are from about 400 to about 1065 degrees C, with a more preferred temperature  
range from about 450 to about 750 degrees C. The alloys all exist as a single phase  
at these operating temperatures and pressures.

25 By applying an electric potential across the palladium alloy in various forms in the  
presence of deuterium gas, deuterium hydrogen or other deuterium isotope gases at

the above mentioned temperature and pressure, substantial heat is generated in addition to the energy generated by resistance heating.

These alloys can be prepared as a coating on a suitable substrate by typical physical vapor deposition processes such as sputtering, ion plating, thermal evaporation  
5 techniques, and so on. It is preferred that the substrate have an orientation of the {100} plane being parallel to the substrate surface so that the coating will also have the same preferred orientation.

A wrought form of the alloys can be made by powder metallurgy technique by first mixing powders of the various elements and going through a pressing, isostatic  
10 pressing and sintering process or a hot isostatic pressing process. This is followed by various metal-shaping process such as hot forging or hot rolling. The alloys are then preferably annealed in a vacuum and quenched to room temperature.

For palladium alloys of a foil form, a suitable rolling schedule can be developed to produce foils with a preferred orientation of the {100} plane being parallel to the  
15 surface and the (100) direction being parallel to the rolling direction.

The above mentioned palladium materials can be further alloyed with various oxides such as alumina, silicate, sodium oxide, calcium oxide, and so on. They are mechanically alloyed in powder form, then pressed and lightly sintered in a vacuum,  
20 or in hydrogen or deuterium gas, to achieve a porous structure of less than full density.

The above mentioned palladium alloys in coating, foil or porous form can be used as the core material to generate energy or heat in a pressurized chamber filled with deuterium (D2) gas, hydrogen (H2) gas or their isotope (HD), singly or in  
25 combination at the above mentioned pressure and temperature. Excess energy or heat can be generated and triggered by the application of electric field, magnetic field, ultrasonic excitation etc. to the core material.

These palladium alloys in various forms can also be used as hydrogen permeation membranes, hydrogen isotope separation membranes, or electrodes in electrochemical cells.

5 Excess heat or energy can also be generated by using the palladium alloys in various forms as a cathode with a counter electrode as an anode. The electrodes are placed in an electrolyte of deuterium water or heavy water (D<sub>2</sub>O), or semi-heavy water (HDO). The electrolyte includes an ionic solution with substantial electrical conductivity. A voltage is applied across the electrodes to electrolyze the solution and generate deuterium or HD gases. These gasses dissolve into the palladium  
10 electrode and cause the electrode to generate excess heat or excess energy.

Furthermore, the palladium and/or palladium alloys of this invention can be coated by electroplating, or by an electroless plating process, onto a wire core. The core may be of copper, a copper alloy such as CDA 725, 99.99% purity nickel, nickel 200, or an austenitic stainless steel such as 304S, 309S or 316S.

15 Before coating, the wire core is hard drawn to more than 90% reduction in diameter and then annealed to promote (100) fiber texture. After electroplating or electroless plating, the material is annealed in a vacuum at a suitable temperature to recrystallize the grain. It is then further hard drawn to gauge with more than 90% reduction. The final wire diameter of 25 to about 50 microns is preferred. The  
20 palladium or palladium alloy should have a thickness of 1 or 2 microns, with (100) fibrous texture.

To make the wrought form of the palladium alloy of the current invention, one can use a powder metallurgy technique such as pressing and sintering. A more preferred method is hot isostatic pressing using powders of the alloying elements of 99.99%  
25 purity. Oxygen can be added as oxide, such as the oxides of boron, silicon, aluminum, sodium, calcium, and so on.

The powders are first thoroughly mixed in a vacuum at 100 to 200 degrees C. They are then put in a can and sealed. The can is made from 1010 plain carbon steel, extra

low carbon killed steel, or 304 stainless steel. Hot Isostatic Pressing (HIP) is done by argon at a pressure of 10,000 to 30,000 psi and a temperature of 1100 to 1200 degrees C for one hour, or until full density and homogeneous microstructure is achieved. The typical shape is either a billet form for rolling into foil, a rod form for  
5 drawing into wires, or a pancake form for a sputtering target.

For foils, the HIPed ingot goes through a series of hot rolling and cold rolling process, with more than 90% thickness reduction in the cold rolling process preferred, followed by final anneal at 900 to 1000 degrees C for an hour in a vacuum. The ingot is then quenched to room temperature, with the resulting foil  
10 having a (100) preferred orientation and a single phase. The resulting foil has high mechanical strength because of the alloying content. This offers the opportunity to produce a foil of reduced thickness for use as a hydrogen purification membrane. The hydrogen permeation rate is higher, according to equation A before.

In a typical hydrogen purification membrane as depicted in FIG. 1, impure hydrogen  
15 gas is the feed material. It has a pressure in the 3 to 7 bar range and an operating temperature of 300 to 600 degrees C. The standard membrane material is palladium with 23 to 25 % by weight of silver. The membrane has a thickness of about 25 microns. Too thick a membrane will reduce the permeation rate at any given temperature. Too thin a layer will be too fragile to withstand the pressure differential  
20 and is liable to fracture.

Palladium with boron, oxygen and carbon at low concentration is the preferred way to strengthen the membrane, because solid solution strengthening by interstitial atoms is much more effective than a substitutional solid solution hardening alloy (such as Pd-23 to 25 % Ag). Further, a single phase alloy operating at 410 degrees C  
25 or above is preferred. This avoids phase changes at the operating temperature and the resulting stress generated due to different lattice spacing for different phases.

During shut down of the hydrogen purification device, cold argon or nitrogen is used to flush the hydrogen out of the gas tubes and to quench the Pd membrane to room temperature.

The same HIP process described in the previous paragraph can also be used to make sputtering targets and to coat the palladium alloy onto suitable substrates. Suitable  
5 substrates include oxygen free high conductivity copper, 99.99% purity nickel, nickel 200, cupronickel 70/30, tin bronzes (CDA902 through 917), aluminum bronze (CDA952 through 958), stainless steel 304S, 309S and 316S, and so on, provided that the substrates are made with a (100) preferred orientation.

10 Preferably, the substrate is in a strip form with one or 2 cm in width. It is continuously fed into a sputtering machine through a series of load locks, and palladium alloys are continuously sputtered onto the substrate.

The sputtering process can be further improved by a reactive sputtering process. In this reactive sputtering process, deuterium gas and/or deuterium hydrogen gas is fed  
15 into the sputtering chamber at a partial pressure of 0.1 to  $5 \times 10^{-3}$  torr. Argon is present with a partial pressure at 1 to  $3 \times 10^{-3}$  torr. The residual gas pressure is  $5 \times 10^{-5}$  torr.

The preferred thickness of the coating is from 0.5 to about 3 microns, with a more preferred range of 1 to 2 microns. The preferred deposition rate is 0.5 nm/sec or less,  
20 with a deposition rate of 0.3 microns/sec or less more preferred. The palladium alloy coating will take on the preferred orientation (100) of the substrate.

The same substrate mentioned above can also be used as a substrate for thermal evaporation of palladium and boron simultaneously in a vacuum chamber. The chamber is equipped with a high temperature effusion cell to evaporate palladium  
25 onto the substrate. It also is equipped with an electron gun to evaporate the boron onto the substrate. A flux monitor can be used to feedback control the effusion cell and the electron gun energy for the proper ratio of boron and palladium on the substrate.

The substrate can be further heated by a boron nitride heater to a temperature of 800 to 900 degrees C. The temperature of the substrate can be monitored by an optical pyrometer. The typical vacuum pressure is between  $10^{-7}$  to  $10^{-9}$  torr. The preferred deposition rate is 0.1 nm/sec. The preferred thickness is from about 0.1 to about 5  
5 microns, with a more preferred thickness of 1 to 3 microns.

Deuterium and or deuterium hydrogen gases can be fed into the chamber during deposition. The gas can be in molecular form or in atomic form. Atomic form is produced by first feeding molecular gas through a hot tungsten filament to dissociate the deuterium molecule. The deuterium atoms are then directed at the substrate. The  
10 partial pressure of the atomic deuterium can be between  $10^{-8}$  to  $10^{-3}$  torr.

The substrate can be rotating at between 1 to 60 rpm , with 20 to 30 rpm preferred.

After deposition, the substrate is quenched with nitrogen or argon to cool the coated part to room temperature rapidly.

It is understood that whenever the word " hydrogen" or "deuterium", it means  
15 hydrogen nucleus, atom, or molecules or deuterium nucleus, atom or molecules or their isotopes as the case may be.

## EXAMPLES

### 20 Example 1

A process to make palladium coated copper wire will be described here first.

A starting material of 99.99% purity copper is melted in a vacuum furnace and cast into a water-chilled graphite mold to make a copper bar of 9 mm in diameter. This bar is machined to 8 mm diameter and sent through a multi-pass drawing process to  
25 reduce its diameter to 0.4 mm or 400 microns, followed by an anneal at 450 degrees

C for half an hour in a vacuum. X-ray diffraction done on the 0.4 mm diameter copper wire shows a major fiber texture of (200) with a minor texture of (111) near the middle of the wire.

This wire is further degreased, cleaned and pickled in dilute hydrochloric acid, and then rinsed in DI water and dried by filtered hot air. It then is immersed in electroplating bath of the following composition: 3.7 gm/l of PdCl<sub>2</sub>, 100 gm/l of Na<sub>2</sub>HPO<sub>4</sub> · 12H<sub>2</sub>O, 20 gm/l of (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> and 2.5 gm/l of benzoic acid with balance D<sub>2</sub>O or heavy water. After dissolving the salts, the bath is heated up to the boiling point to form the palladium complex. The pH of the bath is adjusted with ammonium hydroxide to between 6 - 7.

The copper wire is made into a cathode, with a DC power supply and a current density of 10 mA per cm<sup>2</sup>, to plate 5 microns thickness of palladium at 40 to 50 degrees C. X-ray diffraction of the Pd coated Cu wire shows (200) as the dominant peak of Pd. This indicates that the electroplated Pd has a major texture in (200).

The wire is further draw down through successive smaller diameter dies to a final gauge of 25 microns in diameter, followed by cleaning, degreasing and acid pickling. It is then annealed at 450 degrees C for half an hour, then at 850 degrees C for one hour, in a vacuum.

This Pd coated wire is put into a pressure chamber. The chamber is filled with deuterium gas at 50 bars. Electric voltage is applied at two ends of the wire to pass sufficient current to heat up the wire to 450 degrees C. Heat is generated at a rate substantially higher than the resistance heating by the current passed through the wire.

## Example 2

In the same process as in Example 1, copper wire is made to a final gage of 50 microns, then cleaned and annealed in a vacuum at 450 degrees C for half an hour. It

is then electroless palladium plated in a bath of the following composition: PdCl<sub>2</sub> 4.0gm/liter, NH<sub>4</sub>OH 0.6M, Trimethylamine borate 2.5gm/liter, Mercaptobenzothiazole 3.5 mg/liter, with the balance of deuterium oxide or heavy water with pH at 11 and temperature 45 degrees C. The bath has a plating rate of 1.6 to 1.8 microns per hour. Plating time is approximately one hour.

The Pd coated wire is then put into a chamber pressurized to 50 bar with a 50:50 mixture of deuterium gas(D<sub>2</sub>) and HD. An electric current is then passed through the wire to heat up the wire, at a heating rate not more than 15 degrees C per minute, to 900 degrees C. The wire stays at 900 degrees C for one hour. The electric current is then reduced to cool the wire to 500 degrees C. The current is feed-back controlled by a temperature sensing device to keep the temperature of the wire between 500 and 650 degrees C. Heat is generated at a rate substantially higher than the resistant heating by the electrical current passed through the wire.

### 15 Example 3

This example is similar to Examples 1 and 2, but the starting wire material is 316SVM. This is an austenitic grade stainless steel. It has a typical composition in weight % as follows: Carbon 0.03% max, Manganese 2.0%, Chromium 16 to 18%, Nickel 10 to 14%, Molybdenum 2 to 3%. A clean grade is preferred, preferably using VIM (vacuum induction melting) or ESR (electroslag remelt) to minimize sulfur and phosphorus content.

The ingot is in bar form. It goes through successive wire drawing process with cold reduction of 90% or higher to a final gauge of 50 microns. An intermediate anneal before the last cold drawing is acceptable. Both the intermediate anneal and the final anneal are done at 1150 degrees C for an hour in a vacuum, followed by a quench to room temperature to preserve the austenitic microstructure and the face centered cubic lattice with dominant (100) fibrous texture.

Subsequently, the same process of Examples 1 and 2 can be followed to electroplate palladium to 2 microns thickness, with (100) preferred orientation. 0.5 micron of electroless deposition may be accomplished with a borated bath with a palladium-boron alloy, having a boron content of about 10 atomic %. An amorphous structure is produced. This is followed by an anneal at 950 degrees C for one hour in a vacuum to homogenize and recrystallize into a single phase, followed by a quench to room temperature. This produces a single phase Pd-B alloy.

The same procedure as in Example 1 and 2 is then used. The Pd coated stainless steel wire is pressurized in deuterium or deuterium hydrogen (HD) gas at 500 to 650 degrees C while a DC electric field of about 100 volts is applied across the 10 cm long wire with about 4 amperes of current flowing through the wire. Heat is generated at a substantially higher rate than the resistant heating by the applied voltage and the resultant current.

15

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## CLAIMS

I claim:

- 1) A method of dissolving a gas into a metal, wherein:
  - 5 a) the gas comprises deuterium gas (D<sub>2</sub>), hydrogen gas (H<sub>2</sub>), or a gas of their isotope (HD), singly or in combination; and wherein
  - b) the method comprises:
    - i) providing a metal comprising an alloy of palladium with boron, carbon, oxygen, or any combination thereof, wherein the alloy is characterized by consisting of a single phase at:
      - 10 (1) a temperature from about 400°C to about 1065°C; and
      - (2) a gas pressure of about 0.1 atmospheres to about 1000 atmospheres;
    - ii) exposing the metal to the gas, wherein:
      - (1) the gas or the metal has a temperature of about 400°C to about 1065°C; and
      - 15 (2) the gas has a pressure of about 0.1 atmospheres to about 1000 atmospheres; and
    - iii) applying an electric field, magnetic field, or ultrasonic excitation to the metal.
- 20 2) The method of Claim 1, wherein boron is present in the alloy in a concentration of about 0.01 atomic percent to about 20 atomic percent.
- 3) The method of Claim 1, wherein carbon is present in the alloy a concentration of about 0.01 atomic percent to about 10 atomic percent.
- 25 4) The method of Claim 1, wherein oxygen is present in the alloy in a concentration of about 0.01 atomic percent to about 1.0 atomic percent.
- 5) The method of Claim 1, wherein boron is present in the alloy in a concentration of about 0.1 atomic percent to about 18 atomic percent;
- 30 6) The method of Claim 1, wherein carbon is present in the alloy a concentration of about 0.1 atomic percent to about 5.0 atomic percent.
- 7) The method of Claim 1, wherein oxygen is present in the alloy in a concentration of about 0.1 atomic percent to about 0.6 atomic percent.
- 35

- 8) A method of dissolving a gas into a metal, wherein:
- a) the gas comprises deuterium gas (D<sub>2</sub>), hydrogen gas (H<sub>2</sub>), or a gas of their isotope (HD), singly or in combination; and wherein
  - 5 b) the method comprises:
    - i) providing a metal comprising an alloy of palladium with boron, carbon, oxygen, or any combination thereof, wherein the alloy is characterized by consisting of a single phase at:
      - (a) a temperature from about 450°C to about 750°C; and
      - 10 (b) a gas pressure of about 1 atmosphere to about 100 atmospheres;
    - ii) exposing the metal to the gas, wherein:
      - (1) the gas or the metal has a temperature of about 450°C to about 750°C; and
      - (2) the gas has a pressure of about 0.1 atmospheres to about 1000
      - 15 atmospheres; and
    - iii) applying an electric field, magnetic field, or ultrasonic excitation to the metal.
- 9) The method of Claim 8, wherein boron is present in the alloy in a concentration of about 0.01 atomic percent to about 20 atomic percent.
- 10) The method of Claim 8, wherein carbon is present in the alloy a concentration of about 0.01 atomic percent to about 10 atomic percent.
- 25 11) The method of Claim 8, wherein oxygen is present in the alloy in a concentration of about 0.01 atomic percent to about 1.0 atomic percent.
- 12) The method of Claim 8, wherein boron is present in the alloy in a concentration of about 0.1 atomic percent to about 18 atomic percent;
- 30 13) The method of Claim 8, wherein carbon is present in the alloy a concentration of about 0.1 atomic percent to about 5.0 atomic percent.
- 14) The method of Claim 8, wherein oxygen is present in the alloy in a concentration of about 0.1 atomic percent to about 0.6 atomic percent.
- 35

- 15) The method of any of Claims 1 to 14, wherein the metal comprises:
- a) a foil; or
  - b) a porous structure of less than full density.
- 5
- 16) The method of any of Claims 1 to 14, wherein the metal comprises a coating on a substrate.
- 17) The method of Claim 16, wherein:
- 10 a) the substrate comprises a wire having a diameter of about 25 microns to about 50 microns;
  - b) the coating has a thickness of about 1 micron to about 2 microns; and
  - c) the coating has (100) fibrous texture.
- 15 18) A method of electrolyzing an electrolyte, wherein:
- a) the electrolyte comprises:
    - i) heavy water (D<sub>2</sub>O), water (H<sub>2</sub>O), or semi-heavy water (HDO), singly or in combination; and
    - ii) an ionic solution with substantial electrical conductivity; and wherein
  - 20 b) the method comprises:
    - i) placing the metal of any of Claims 1 to 4, as a cathode, in the electrolyte;
    - ii) providing a counter electrode as an anode; and
    - iii) applying a voltage across the electrodes.

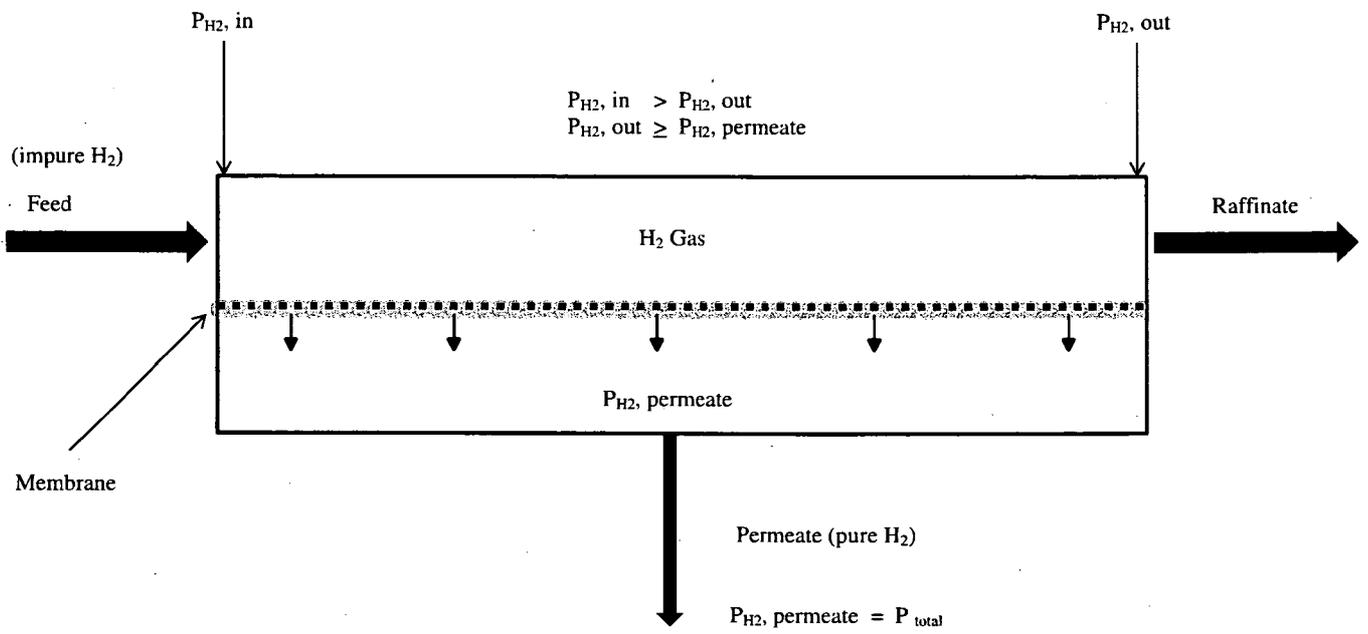


Fig 1. Hydrogen permeation device

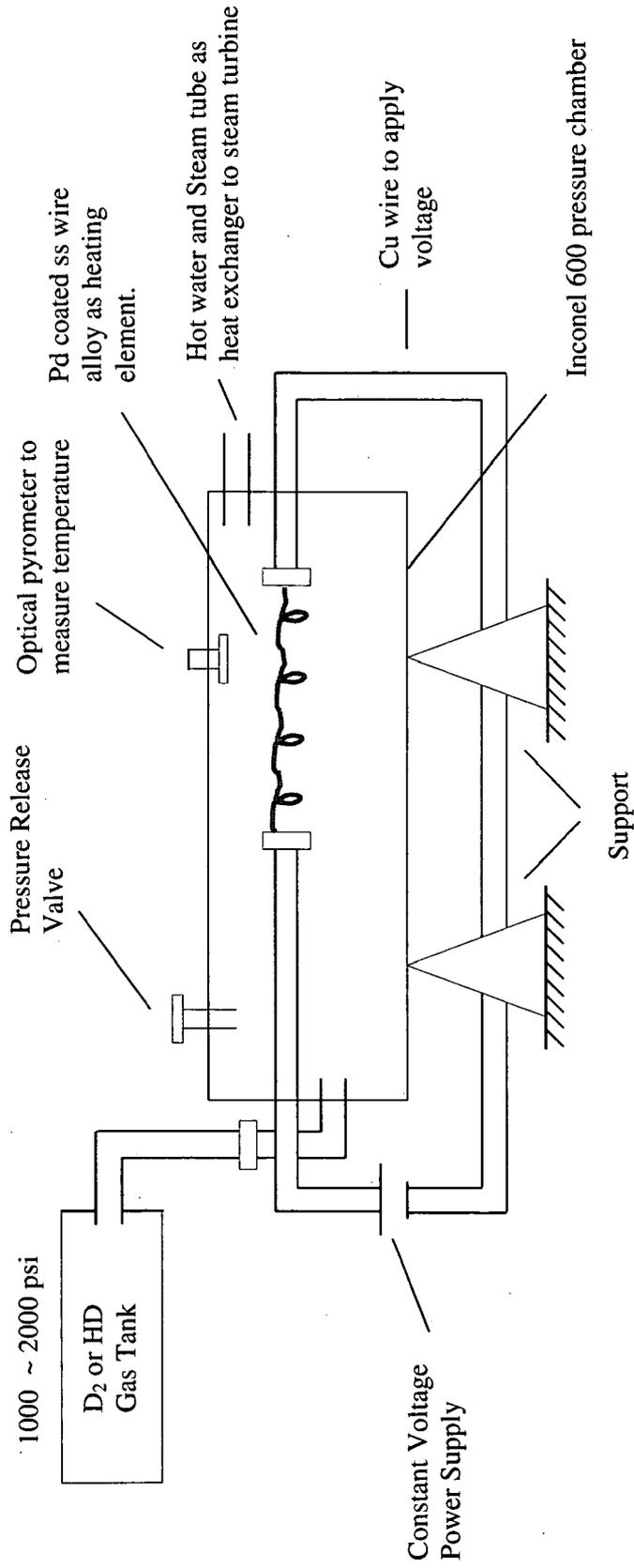


Fig 2. Deterium Gas Charging and Energy Generation Device

INTERNATIONAL SEARCH REPORT

10127000000 30.05.2012  
International application No.  
PCT/US 12/00088

A. CLASSIFICATION OF SUBJECT MATTER  
IPC(8) - B01D 53/22 (2012.01)  
USPC - 95/56; 96/4; 204/240  
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
USPC - 95/56; 96/4; 204/240  
IPC(8) - B01D 53/22 (2012.01 )

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
USPC - 95/56; 96/4; 204/240  
IPC(8) - B01 D 53/22 (2012.01) (keyword delimited)

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
PubWEST (USPT,PGPB,EPAB,JPAB); Google, Google Patent  
Search terms used: palladium alloy hydrogen deuterium absorption temperature pressure boron boride oxide carbide carbon PdO PdC  
PdB energy storage fusion electrode cathode anode wire substrate diameter thickness

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2006/0088138 A 1 (Jouanneau) 27 April 2006 (27.04.2006), entire document, especially para [0009], [001 1], [001 2], [0100]-[0105], [0042]-[0082], [0129]-[0158]	1-6, 18 -----
Y		17
Y	US 3,709,810 A (Grubb et al.) 09 January 1973 (09.01 .1973), col 1, ln 36-42	17
A	US 6,764,561 B 1 (Miles et al.) 20 July 2004 (20.07.2004), entire document	1-18
A	US 2006/0024193 A 1 (Zhao et al.) 02 February 2006 (02.02.2006), entire document	1-18
A	US 2006/0108457 A 1 (Pratt et al.) 25 May 2006 (25.05.2006), entire document	1-18
A	US 2004/0036168 A 1 (Bedinger et al.) 26 February 2004 (26.02.2004), entire document	1-18
A	US 6,426,158 B 1 (Pratt et al.) 30 July 2002 (30.07.2002), entire document	1-18
A	US 6,682,817 B 1 (della Porta et al.) 27 January 2004 (27.01 .2004), entire document	1-18

Further documents are listed in the continuation of Box C.

\* Special categories of cited documents:  
 "A" document defining the general state of the art which is not considered to be of particular relevance  
 "E" earlier application or patent but published on or after the international filing date  
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 "O" document referring to an oral disclosure, use, exhibition or other means  
 "P" document published prior to the international filing date but later than the priority date claimed  
 "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention  
 "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone  
 "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art  
 "&" document member of the same patent family

Date of the actual completion of the international search 17 May 2012 (17.05.2012)	Date of mailing of the international search report <b>30 MAY 2012</b>
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Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 2231 3-1450 Facsimile No. 571-273-3201	Authorized officer: Lee W. Young PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774
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