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(54) **HIGH-TEMPERATURE LOW-HYDRATION
ION EXCHANGE MEMBRANE
ELECTROCHEMICAL CELL**

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(57) **ABSTRACT**

This invention relates to an electrochemical cell having a high-temperature low-hydration (HTLH) ion exchange membrane serving as an electrolyte layer. The membrane may be a non-fluorinated ionomer membrane, such as an acid-doped polybenzimidazole (PBI) membrane. The HTLH membrane is sandwiched by an anode having a hydrogen-carrying fluid feed chamber with an inlet for receiving a hydrogen-carrying fluid, and a cathode having a hydrogen product chamber with an outlet for discharging a hydrogen product gas. The anode and cathode are electrically coupleable to an electric current source for powering the electrochemical cell to produce hydrogen gas in a reduction reaction at the cathode. The hydrogen-carrying fluid may be water, in which case the electrochemical cell serves as an electrolyzer; or, the hydrogen carrying fluid may be a hydrogen gas, in which case the cell serves as a hydrogen pump.

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Related U.S. Application Data

(60) **Provisional application No. 60/375,200, filed on Apr. 23, 2002.**

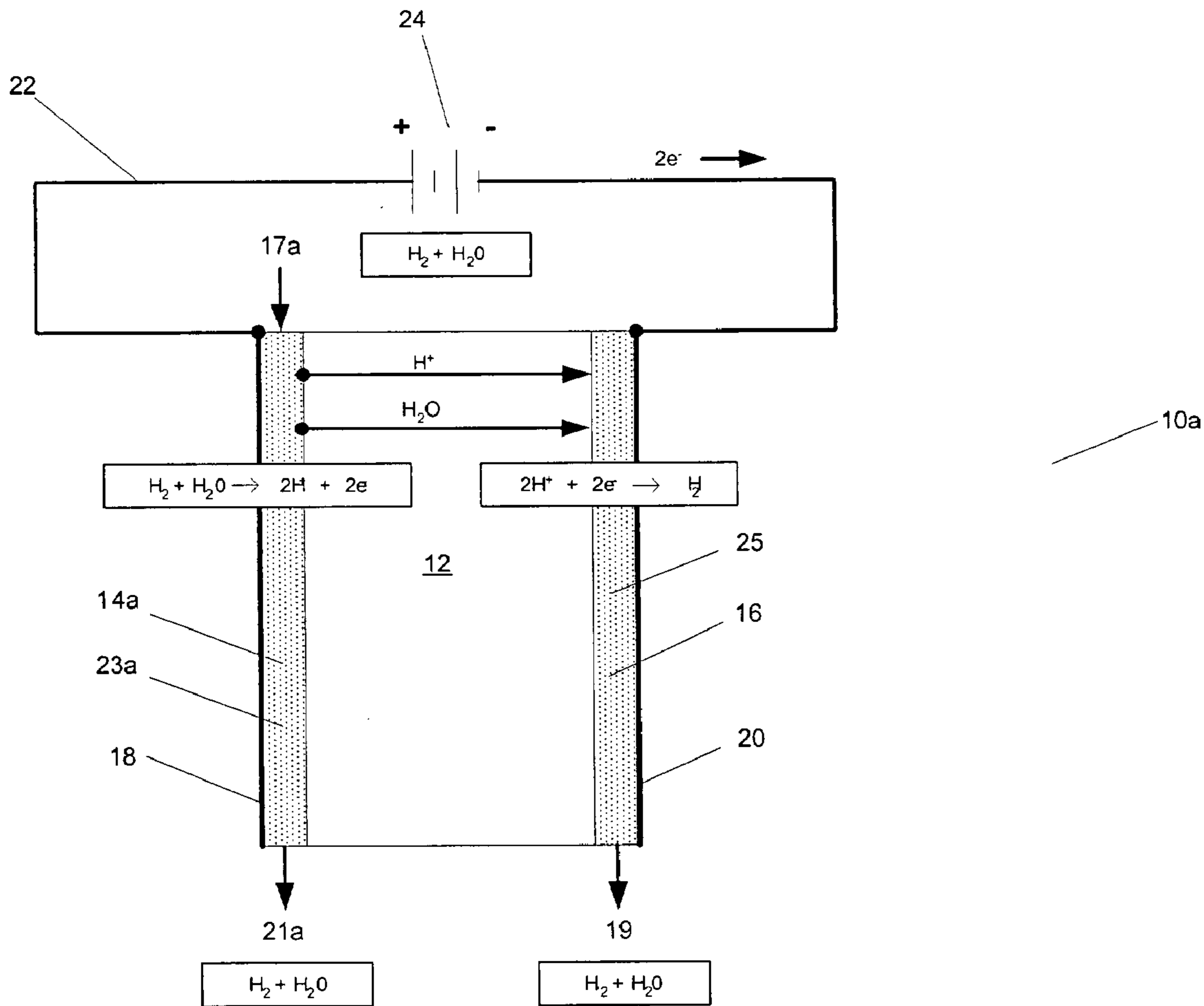


Fig. 1a

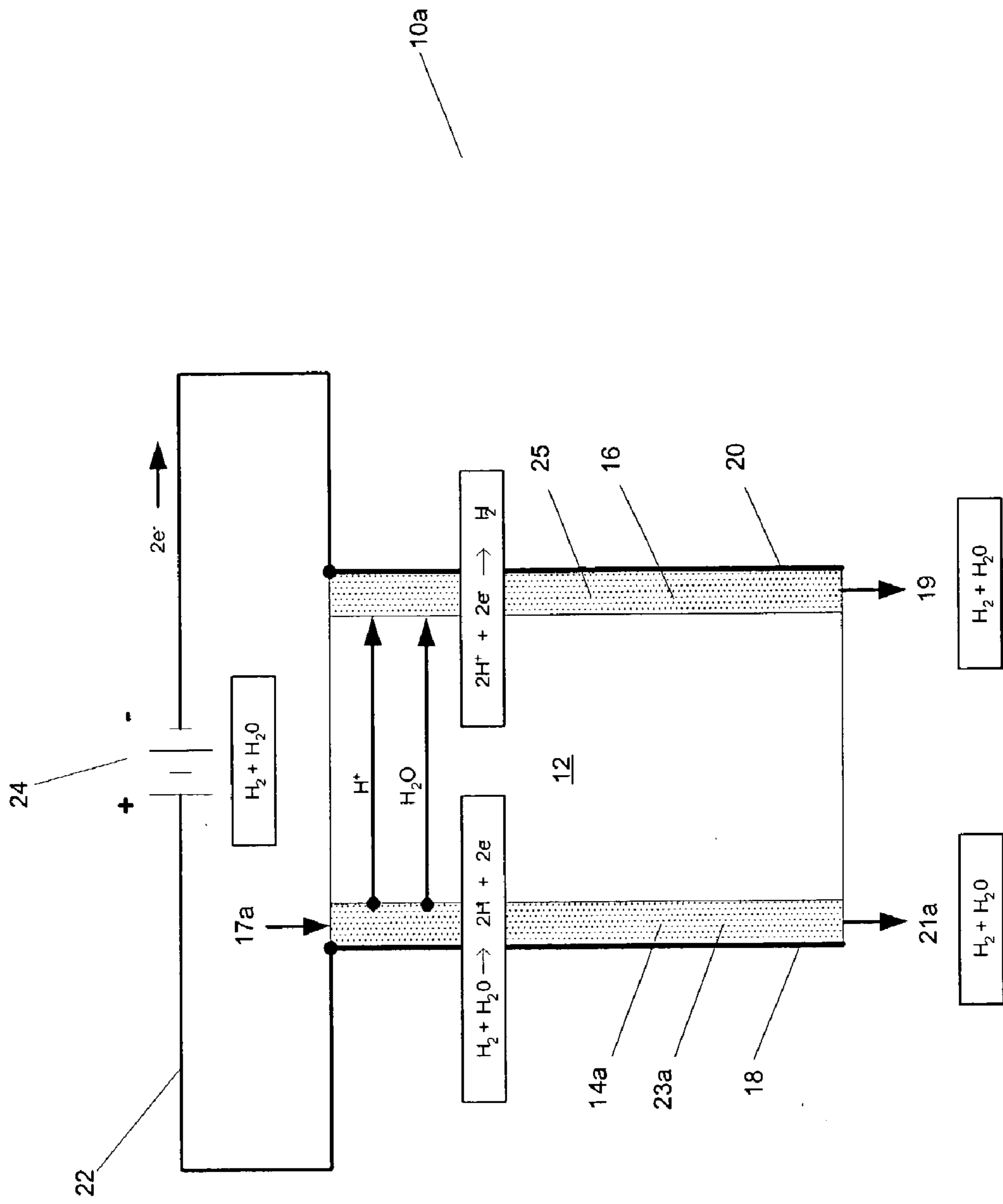


Fig. 1b

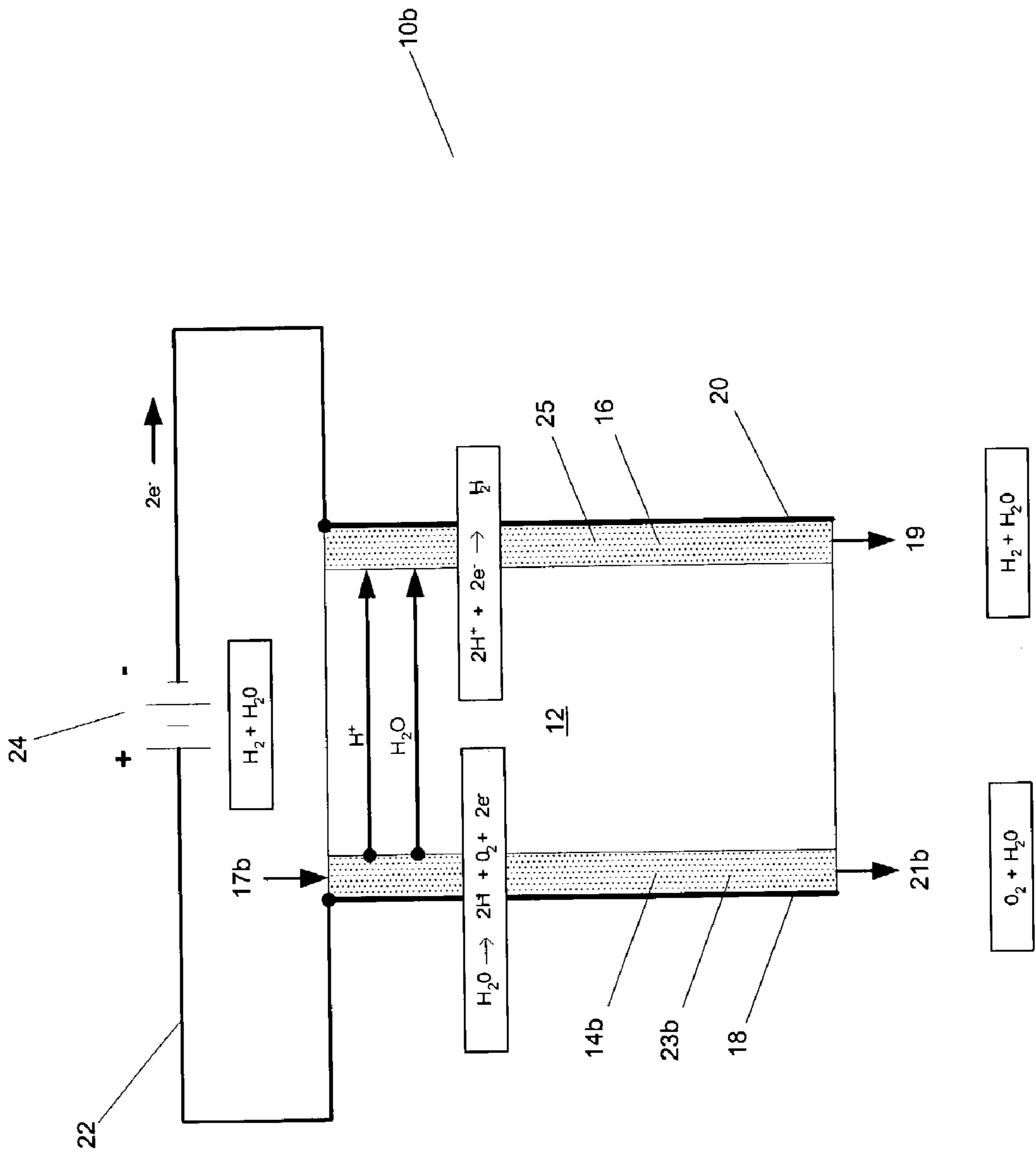


Fig. 2

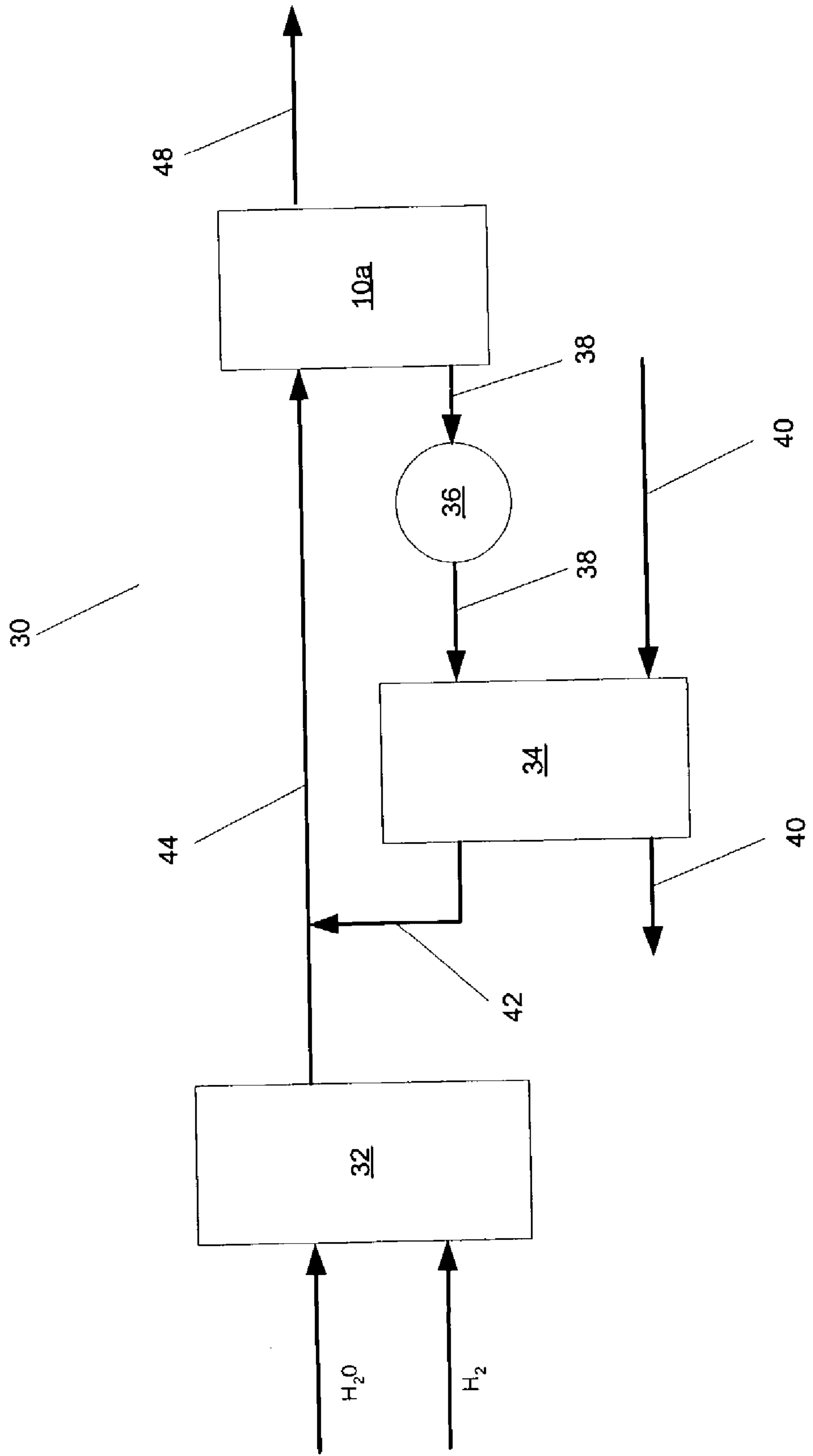


Fig. 3

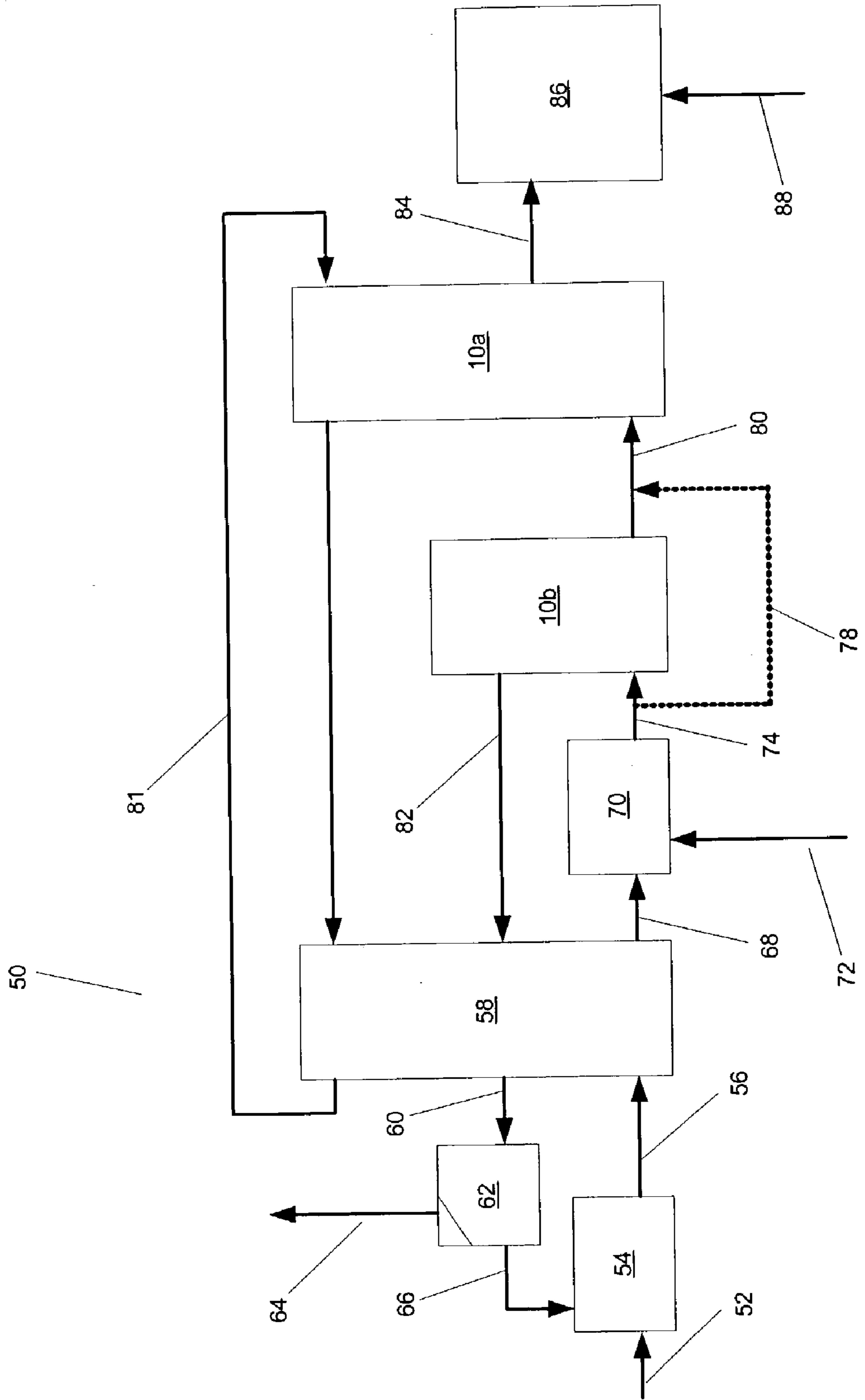


Fig. 4

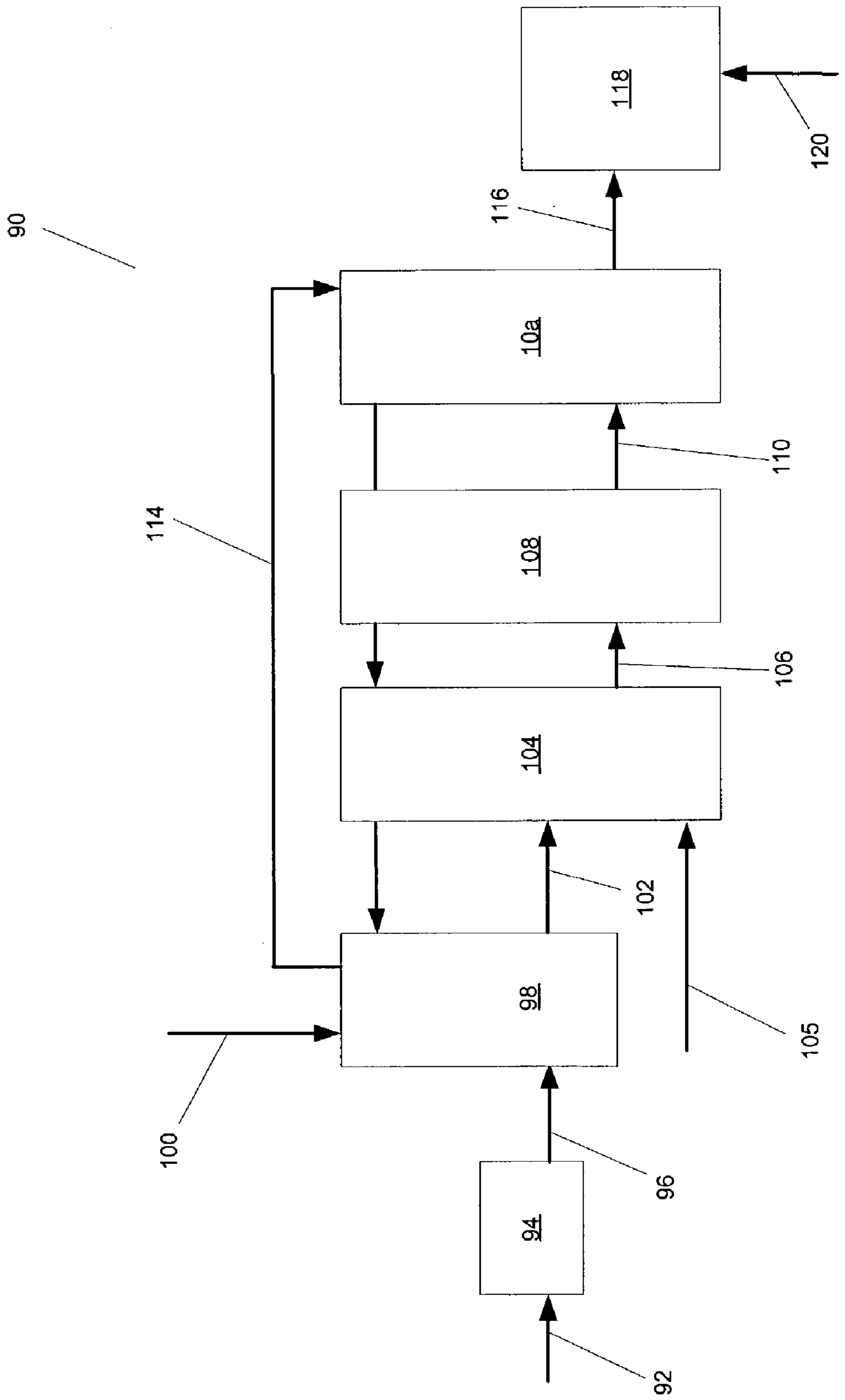


Fig. 5

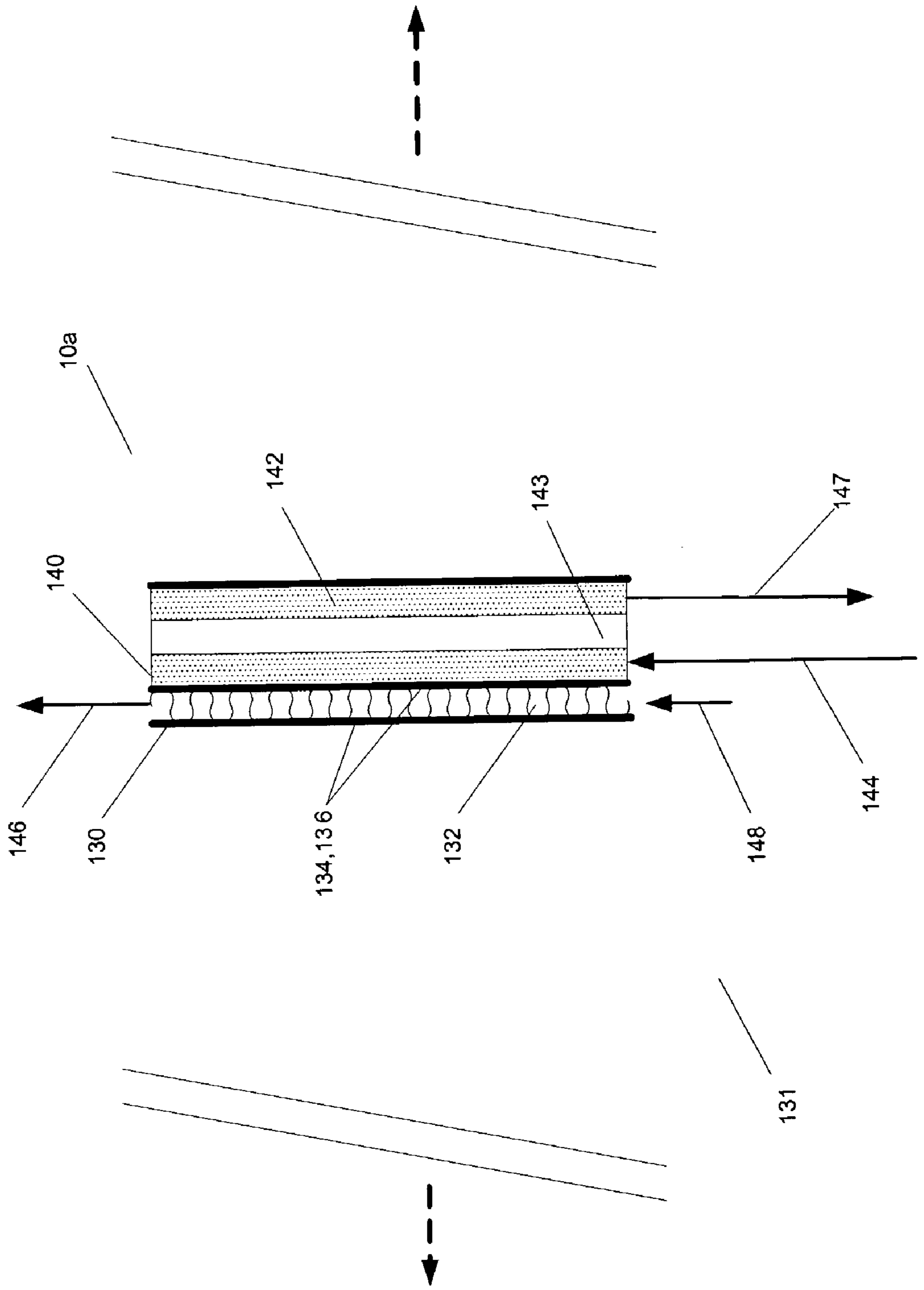


Fig. 6

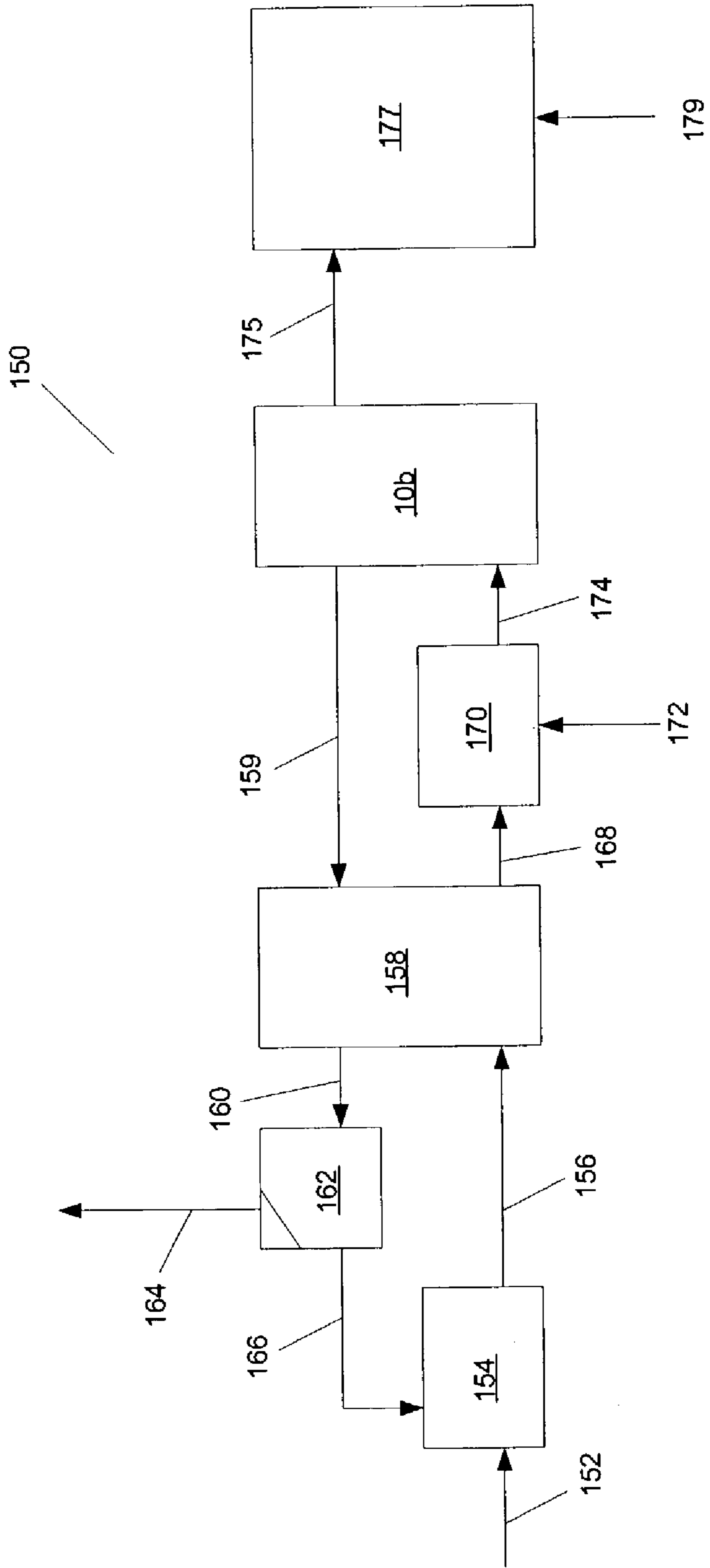
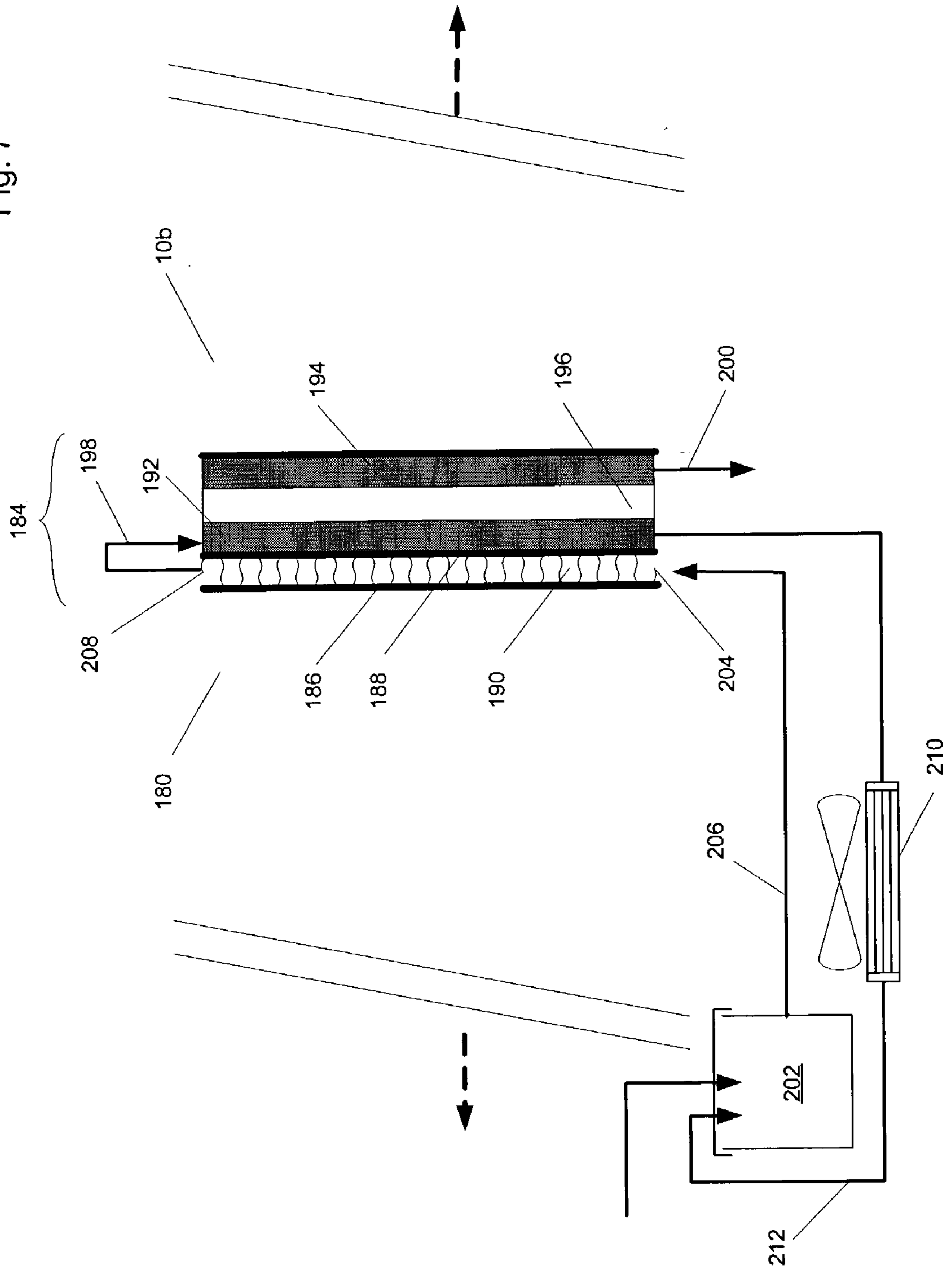


Fig. 7



HIGH-TEMPERATURE LOW-HYDRATION ION EXCHANGE MEMBRANE ELECTROCHEMICAL CELL

RELATED APPLICATIONS

[0001] This application incorporates by reference and claims priority from U.S. provisional application No. 60/375,200 entitled High Temperature Electrochemical Pump Using Low Hydration Ion Exchange Membrane filed Apr. 23, 2002.

TECHNICAL FIELD

[0002] The present invention relates to the use of High-Temperature Low-Hydration (HTLH) ion exchange membranes as an electrolyte in an electrochemical cell.

BACKGROUND OF THE INVENTION

[0003] An electrochemical cell has multiple uses, including use as a fuel cell to generate electricity by electrochemically reacting a hydrogen fuel stream and an oxidant stream, as an electrolyzer to separate water into its constituent elements, and as a pump to increase the pressure of a particular gas, e.g. hydrogen. An electrochemical cell typically comprises an electrolyte layer sandwiched by an anode layer and a cathode layer. A feed stream supplies a hydrogen-containing fluid stream to the anode, wherein it is electrochemically reacted to produce electrons, protons, and other reaction products. The protons conduct through the electrolyte, which is proton conductive, but substantially impermeable to reactant gas. The anode and the cathode are connected to an electric circuit to create an electric potential between them and allow electric current to flow from one to the other. The protons that have conducted through the electrolyte to the cathode combine with the electrons to produce hydrogen gas, which can be at a higher pressure than the feed stream. In an electrolyzer, the feed stream is water (separated at the anode into oxygen, protons and electrons), and in a pump, the feed stream may be hydrogen gas or a hydrogen-containing gas.

[0004] Among conventional electrochemical cell electrolyzers and pumps, many use solid electrolyte technology. In solid electrolyte electrochemical electrolyzers and pumps, a solid or quasi-solid material that selectively conducts anions or cations is used as an electrolyte. Typical electrolyte materials used include fluorinated polymers, of which the polymer marketed under the trademark Nafion® is a good example. This class of material incorporates acid groups into the polymer chain and requires high levels of hydration with water in order to conduct protons. It is a characteristic of these membranes that the mechanism of proton conduction also generates a tendency for water to migrate from one side of the membrane to the other. This effect is often referred to as "electro-osmotic drag" or "protonic pumping". When used in electrochemical electrolyzers or pumps, these membranes have the following drawbacks:

[0005] Hydrogen produced at the higher pressure cathode side of the cell diffuses back through the membrane to the lower pressure anode side of the cell thereby reducing the pumping efficiency. At the pressures being considered for vehicle hydrogen fueling systems, the losses due to diffusion are on the order of 10 to 20%. The mechanism for this diffusion

is generally understood to be the diffusion of gases through the water contained in the fluorinated type membrane in its hydrated state.

[0006] Water migrates across the membrane in the direction of proton flow. The amount of water that migrates to the high pressure hydrogen side of the cell is significant. Subsequent processing of the hydrogen is usually required to separate the liquid water and to dry the hydrogen before it enters a storage device.

[0007] The membrane must be kept hydrated. Typically this is achieved by introducing water or water vapor in with the hydrogen feed stream at the anode side of the cell.

[0008] It is therefore desirable to provide an electrochemical cell that does not suffer from the problems or disadvantages associated with electrochemical cells having a fluorinated polymer electrolyte membrane. In particular, it is desirable to provide an electrochemical pump and electrolyzer that are improvements over fluorinated polymer electrolyte type electrochemical pumps and electrolyzers.

SUMMARY

[0009] According to one aspect of the invention, there is provided an electrochemical cell comprising (a) an anode comprising a hydrogen-carrying fluid feed chamber with an inlet for receiving a hydrogen-carrying fluid; (b) a cathode comprising a hydrogen product chamber with an outlet for discharging a hydrogen product gas; and (c) a high-temperature low-hydration (HTLH) membrane sandwiched between the anode and cathode; the anode and cathode being electrically couplable to an electric current source for powering the electrochemical cell to produce hydrogen gas in a reduction reaction at the cathode.

[0010] The fluid feed chamber may also include an outlet for discharging unreacted hydrogen-carrying fluid and/or reaction products; the discharge may be controlled by a purge valve at the outlet. The feed fluid for the fluid feed chamber may be water, and in such case, the electrochemical cell is an electrolyzer that produces hydrogen gas in a reduction reaction at the cathode, and oxygen gas in an oxidation reaction at the anode. The feed water may be in liquid or vapor form. Alternatively, the feed fluid for the fluid feed chamber may be hydrogen gas at or near ambient pressure, and the electrochemical cell in such a case is a pump that produces hydrogen gas in a reduction reaction at the cathode, at a pressure that is higher than the hydrogen feed gas.

[0011] In the electrolyzer or in the pump, the HTLH membrane may be a non-fluorinated ionomer membrane, such as an acid-doped polybenzimidazole (PBI) membrane. In particular, the PBI membrane may be doped with an acid in the group of H_2SO_4 and H_3PO_4 .

[0012] According to another aspect of the invention, there is provided an electrolyzer system comprising the electrolyzer described above, a feed water stream in fluid flow communication with the water feed chamber inlet of the electrolyzer, a hydrogen product stream in fluid flow communication with the hydrogen product chamber outlet of the electrolyzer; and a heat exchanger thermally coupled to the electrolyzer and in fluid flow communication with the water

feed stream upstream of the electrolyzer, such that heat produced by the electrolyzer is used to heat the feed water stream.

[0013] The electrolyzer system may further comprise a water vaporizer that is in fluid flow communication with the water feed stream upstream of the electrolyzer and downstream of the heat exchanger, such that feed water entering the vaporizer has been previously heated by the heat exchanger, thereby reducing the amount of energy needed to vaporize the feed water. Alternatively, the heat exchanger may be the vaporizer and if so comprises: a pair of thermally conductive separator plates, a water vaporizing channel in between the separator plates having an inlet for receiving a liquid water feed stream and an outlet for discharging a water vapor feed stream. The vaporizer is thermally coupled to the electrolyzer by at least one of the separator plates being in thermal contact with the electrolyzer.

[0014] The fluid feed chamber outlet may discharge a water vapor and oxygen gas stream, and the system may further comprise a water recirculation circuit that comprises: a condensing heat exchanger having an inlet in fluid flow communication with the water vapor and oxygen discharge stream and an outlet for discharging a liquid water stream condensed by the heat exchanger; and a water tank in fluid flow communication with (a) the liquid water stream discharged from the heat exchanger outlet, (b) a liquid water make-up stream, and (c) the feed water stream upstream of the vaporizer, such that liquid water recovered by the condensing heat exchanger is returned to the water feed stream. Furthermore, the system may further comprise a water recirculation circuit comprising a gas/water separator with an inlet in fluid flow communication with the water vapor and oxygen gas discharge stream downstream of the heat exchanger. The separator has an oxygen gas discharge outlet, and a water discharge outlet fluidly coupled to the feed water stream upstream of the heat exchanger.

[0015] The electrolyzer system may also have a hydrogen storage chamber in fluid flow communication with the hydrogen product stream discharged from the electrolyzer.

[0016] According to another aspect of the invention, there is provided an electrochemical pump system comprising: (a) the electrochemical pump described above, (b) a hydrogen feed stream in fluid flow communication with the hydrogen feed chamber fluid inlet, (c) a hydrogen discharge stream in fluid flow communication with the hydrogen feed chamber fluid outlet and the hydrogen feed stream, (d) a heat exchanger in fluid flow communication with the hydrogen discharge stream downstream of the pump and upstream of the hydrogen feed stream, and in fluid flow communication with a coolant stream, such that the discharge stream can be cooled in the heat exchanger before joining with the feed stream; and (e) a recirculation pump in fluid flow communication with the discharge stream.

[0017] According to yet another aspect of the invention, there is provided an electrochemical pump system comprising: (a) the electrochemical pump described above, (b) a hydrogen feed stream in fluid flow communication with the hydrogen feed chamber fluid inlet, (c) a hydrogen discharge stream in fluid flow communication with the hydrogen feed chamber fluid outlet, and (d) an electrolyzer having a hydrogen discharge outlet in fluid flow communication with the hydrogen feed stream upstream of the pump, and an inlet

in fluid flow communication with a water feed stream, the electrolyzer being used to produce hydrogen from the water feed stream. The electrolyzer in this case may be an electrochemical electrolyzer comprising an HTLH membrane sandwiched between an anode and a cathode. In this pump system there may also be a heat recirculation circuit that comprises a water vaporizer in fluid flow communication with the water feed stream upstream of the electrolyzer; and a heat exchanger in fluid flow communication with the water feed stream upstream of the vaporizer and thermally coupled to at least one of the electrolyzer and the pump, such that heat generated by at least one of the electrolyzer and the pump is transferable to the water feed stream. The pump system may also include a water recirculation circuit that comprises: an oxygen and water discharge stream in fluid flow communication with the electrolyzer; and a gas/water separator having an inlet in fluid flow communication with the oxygen and water discharge stream downstream of the electrolyzer, and a water discharge outlet fluidly coupled to the feed water stream upstream of the heat exchanger, and an oxygen vent.

[0018] According to another aspect of the invention, there is provided an electrochemical pump system comprising: the electrochemical pump described above; and, a natural gas reformer comprising a hydrogen gas outlet in fluid flow communication with the hydrogen feed chamber fluid inlet of the pump and a natural gas inlet in fluid flow communication with a natural gas source. The electrochemical pump may also include a water vaporizer comprising a water feed inlet in fluid flow communication with a liquid water feed source, and a water discharge outlet in fluid flow communication with a water discharge stream that is in turn in fluid flow communication with the reformer. The electrochemical pump system may also include a thermal recirculation circuit comprising a thermal conduction conduit thermally coupling the vaporizer and at least one of the pump and reformer, such that heat generated by the reformer or the pump is transferable to the vaporizer.

[0019] According to another aspect of the invention, there is provided an electrochemical pump system comprising: the electrochemical pump described above; and, a vaporizer in fluid flow communication with a water feed stream, and being thermally coupled to the pump such that heat generated by the pump is transferable to the water feed stream in the vaporizer. The vaporizer of this system may comprise (a) a pair of thermally conductive separator plates, (b) a water vaporizing channel in between the separator plates, and (c) a water feed inlet and water vapor outlet. The vaporizer is thermally coupled to the pump by at least one of the separator plates being in thermal contact with the pump.

[0020] According to yet another aspect of the invention, there is provided an electrochemical filter comprising: (a) an anode comprising a hydrogen feed chamber with an inlet for receiving an unfiltered feed gas comprising hydrogen and contaminants, and an outlet downstream of the inlet and for discharging the contaminants; (b) a cathode comprising a hydrogen product chamber with an outlet for discharging a hydrogen product gas; and (c) a high-temperature low-hydration membrane sandwiched between the electrodes. The anode and cathode are electrically couplable to an electric current source for powering the electrochemical filter to produce the hydrogen product gas in a reduction

reaction at the cathode, thereby separating the contaminants from the hydrogen in the feed gas.

BRIEF DESCRIPTION OF THE DRAWINGS

[0021] FIGS. 1(a) and (b) are schematic illustrations of a hydrogen electrochemical pump (1(a)) and electrolyzer (1(b)) each having an HTLH electrolyte membrane.

[0022] FIG. 2 is a schematic illustration of a system for pumping hydrogen utilizing an HTLH ion exchange membrane electrochemical pump.

[0023] FIG. 3 is a schematic illustration of a system for pumping hydrogen utilizing an HTLH ion exchange membrane electrochemical pump in which the hydrogen is generated from a high temperature water electrolyzer.

[0024] FIG. 4 is a schematic illustration of a system for pumping hydrogen utilizing an HTLH ion exchange membrane electrochemical pump in which the hydrogen is generated from a natural gas steam reformer.

[0025] FIG. 5 is a schematic illustration of an integrated vaporizer cell and HTLH ion exchange membrane electrochemical pump cell.

[0026] FIG. 6 is a schematic illustration of a system for generating hydrogen utilizing an HTLH ion exchange membrane electrolyzer.

[0027] FIG. 7 is a schematic illustration of an integrated vaporizer cell and HTLH ion exchange membrane electrolyzer cell.

DETAILED DESCRIPTION

[0028] High-temperature low-hydration (HTLH) membranes are herein defined as ion exchange membranes that exhibit proton conductivity and operate at higher temperatures and require lower levels of hydration than fluorinated polymer type membranes (referred generally as hydrated membranes). In particular, HTLH membranes are operable at temperature ranges in the order of 100 to 200° C. and with a hydration level of less than or equal to 20% by weight.

[0029] One example of a suitable HTLH membrane is a non-fluorinated ionomer membrane, such as an acid-doped polybenzimidazole (PBI) membrane. Other types of proton-conducting HTLH membranes include those made with sulfonated polyphenylene oxide; polydimethylphenylene oxide phosphoric acid; polyether ketone; imidazole doped polyether ketone; a blend of sulfonated polyphenylene oxide and polyvinylidene fluoride; a blend of ortho-sulfone-sulfonated polyethersulfone and polybenzimidazole; and 1-butyl 3-methyl imidazolium trifluoromethane sulfonate doped Nafion®.

[0030] Some of the characteristics reported for HTLH membranes are: thermal stability in simulated fuel cell environments up to temperatures of 200° C., ability to operate at temperatures up to 200° C. in a fuel cell, good proton conductivity at low water activities, low crossover of methanol in direct methanol fuel cell applications, and slow oxygen reduction kinetics when used in a fuel cell membrane-electrode assembly (MEA).

[0031] An acid-doped PBI membrane was prepared by soaking a thin film of the PBI polymer in an acid solution. A typical suitable acid is H₂SO₄ or H₃PO₄. Preparation of

acid-doped PBI membranes is known in the art; for example, the article by Savadogo et al., entitled "Hydrogen/oxygen polymer electrolyte membrane fuel cell (PEMFC) based on acid-doped polybenzimidazole (PBI)" *Journal of New Materials Electrochemical Systems* 3, pp. 345-349 (2000) discloses the preparation of sulphuric-acid-doped PBI and phosphoric-acid-doped PBI for use in fuel cells. The resulting sulphuric or phosphoric acid-doped PBI membrane is comprised of acid ions bound ionically to the polymer backbone. Savadogo et al report that the phosphoric acid-doped PBI membrane is thermally stable up to the test temperature of 185° C.

[0032] In the Encyclopedia of Polymer Science and Engineering, 2nd edition, Buckley et al indicates that a phosphoric-acid doped PBI membrane retains short-term resistance to heat at temperatures as high as 500° C.

[0033] In the article entitled "Acid-Doped Polybenzimidazoles: A New Polymer Electrolyte" in the Journal of The Electrochemical Society, Volume 142, Wainright et al showed that phosphoric-acid doped PBI has high conductivity (5×10^{-2} Scm) at the test temperature of 190° C. They also found that phosphoric-acid doped PBI has low water content and low methanol crossover.

[0034] In the article entitled "Electro-osmotic Drag Coefficient of Water and Methanol in Polymer Electrolytes at Elevated Temperatures" in the Journal of The Electrochemical Society, Volume 143, No. 4, April 1996, Weng et al reported that the water electro-osmotic drag coefficient of phosphoric-acid doped PBI is essentially zero at the tested temperatures up to 200° C.

[0035] The HTLH membrane is incorporated into an electrochemical cell system that is connected to an electric current source. Upon the supply of current and a hydrogen-containing gas stream to the cell, an electrochemical process occurs: the hydrogen-containing gas is oxidized at the anode to produce protons that conduct through the membrane, and the conducted protons are reduced at the cathode to produce hydrogen product gas at higher pressure. Unlike a fuel cell, the electrochemical reduction occurs substantially free of oxygen, and therefore, the product fluid produced at the cathode is hydrogen gas. Such an HTLH electrochemical cell can be used for several purposes, such as hydrogen pumping, hydrogen filtering, and electrolysis.

[0036] The HTLH membrane has been found to have high proton conductivity at low water activity relative to hydrated membranes. This characteristic is expected to reduce the inefficiencies associated with the diffusion of gas species across the membrane. In conventional hydrated-membrane type electrochemical cell systems generating high-pressure gas on one side of the membrane, these inefficiencies can be very high. For example, in a typical electrochemical electrolyzer cell using a 0.01 inch thick Nafion® membrane and producing 6000 psig hydrogen and near ambient pressure oxygen at one (1) amp/in², the loss of current efficiency (diffusional losses) has been found to be in the order of 10 to 30%. The primary mechanism for diffusion in ion exchange membranes generally is understood in the art to be related to the diffusivity of the gas species through the water phase of the membrane, which is a function of the partial pressure differential across the membrane. Therefore, in order to maintain a reasonable current efficiency, electrolyzer cells using hydrated membranes operate at relatively

low pressure differentials. In contrast, an HTLH ion exchange membrane retains very little water relative to hydrated membranes. Thus, there is very little diffusion of gas species across the HTLH membrane via the water phase of the membrane, making it easier to operate at higher differential partial pressures, with little loss in current efficiency associated with gas diffusion. For example, an electrolyzer cell having a 0.01 inch thick HTLH membrane operating at 1 amp/in² @ 6000 psig hydrogen gas is expected to have an efficiency of around 99%.

[0037] In contrast to hydrated membranes, HTLH ion exchange membranes do not require the presence of significant amounts of water phase in the membrane for proton conduction. This results in reduced electro-osmotic drag of water across the membrane. Since the HTLH membrane is operable at lower levels of hydration than hydrated membranes, the product hydrogen gas is correspondingly drier. This has an important system benefit in that it reduces the requirement for dewatering the hydrogen gas produced in electrochemical cells. In typical electrochemical pump systems or electrolyzers using hydrated membranes, the dewatering equipment for drying the hydrogen gas comprises a significant portion of the system volume and weight. Using HTLH ion exchange membranes minimizes or eliminates the volume and weight of the dewatering equipment.

[0038] An electrochemical cell system using HTLH ion exchange membranes requires minimal or no water vapor feed for adequate membrane hydration. Furthermore, any water vapor required can be made using the waste heat of the electrochemical cell. The reduced hydration requirement has important system benefits in that it minimizes water handling equipment extraneous to the cell.

[0039] Electrochemical Pump

[0040] Electrochemical pumping is used to filter hydrogen from carrier gases and for compressing hydrogen to higher pressures. This process is based upon an electrochemical cell in which an anode and a cathode are connected to an electric circuit, which creates an electric potential between them and allows electrons to flow from anode to cathode. The anode and cathode are also connected by an electrolyte that allows either anions or cations to migrate from anode to cathode. An electrochemical pump based on acid-doped PBI could potentially operate in the temperature range of 100 to 200° C.

[0041] FIGS. 1 to 5 illustrate an electrochemical pump and systems that utilize HTLH ion exchange membranes for the purpose of pumping hydrogen. The characteristics of this class of membranes, as exemplified by the acid doping of PBI thin film with sulphuric or phosphoric acid, solve a number of the problems that arise from using hydrated membranes as exemplified by Nafion®.

[0042] An HTLH electrochemical pump can be integrated with a hydrocarbon reforming process to filter and purify the hydrogen rich reformat. Because of the higher operating temperature, the electrochemical pump is not particularly susceptible to carbon monoxide (CO) contamination. Carbon monoxide contamination is a typical problem with lower temperature electrochemical hydrogen oxidation half cells. For example, it is expected that an HTLH electrochemical pump operating at 160° C. can use a hydrogen feed stream having CO up to 10,000 ppm. In contrast, a Nafion-

based pump operating at below 100° C. is limited to a CO content of about 10 ppm. A high temperature, CO-tolerant electrochemical pump also reduces or eliminates the need for a selective oxidation stage in the reformer. This stage is typically used to reconvert CO to carbon dioxide (CO₂) as the reformat from the reformer stage is cooled. The water vapor in the reformat is used to hydrate the HTLH membrane in the electrochemical pump, thereby eliminating or reducing the need for a separate membrane hydration system.

[0043] An operating temperature range of 100 to 200° C. presents opportunities for thermal integration with other systems. The waste thermal energy from the electrochemical pump process could be integrated with systems using an endothermic process for hydrogen storage, such as the process disclosed by Millennium Cell, which requires heat and hydrogen to reformulate sodium borate to sodium borohydride.

[0044] The waste thermal energy from the electrochemical pump process could also be integrated with a vaporization process to vaporize liquid water for other processes. Examples of other processes are water vaporization for steam reforming and water vaporization for high temperature electrolysis.

[0045] The high temperature of an HTLH electrochemical pump allows for more compact heat rejection systems due to the higher temperature difference between the process waste thermal energy and the ambient environment used as a heat sink.

[0046] Referring now to FIG. 1(a) and according to one embodiment of the invention, an electrochemical cell 10a (with the half reactions for the oxidation and reduction of hydrogen labeled on the figure) serves as a hydrogen gas pump and has an HTLH ion exchange membrane 12 for the membrane electrolyte; a suitable membrane is PBI thin film with imbibed phosphoric acid. The electrolyte 12 is sandwiched between a pair of electrodes, namely a hydrogen feed electrode 14a, and a hydrogen product electrode 16. The hydrogen feed electrode 14a comprises a separator plate 18, and a hydrogen feed chamber 23a, which includes a fluid inlet 17a, and a fluid outlet 21a. The hydrogen product electrode 16 comprises a separator plate 20, a hydrogen product chamber 25 and a fluid outlet 19. The electrodes 14a, 16 are electrically coupled by an electric circuit 22. The circuit 22 is electrically coupled to an electric current source 24. In operation, low pressure hydrogen hydrated with at least enough water vapor to hydrate the HTLH membrane is fed into the fluid inlet 17a of the hydrogen feed chamber 23a, wherein some of the hydrogen is catalyzed and dissociated into protons and electrons. The unreacted hydrogen and water is discharged via the fluid outlet 21a, and may be recovered in a recirculation loop (not shown). The protons conduct through the HTLH membrane 12 and combine with electrons at the hydrogen product electrode 16 to produce hydrogen product gas; electrons are moved through the circuit 22 by the current source 24. Product gas is discharged through fluid outlet 19 of the hydrogen product chamber 25; a control valve (not shown) is installed at the fluid outlet 19 and operated to control the pressure of the hydrogen product gas. The hydrogen feed electrode 14a may be supplied hydrogen feed gas at near ambient pressure and the hydrogen product electrode 16 may operate to discharge product

hydrogen gas at elevated pressure. For example, the hydrogen product electrode **16** may operate at pressures up to in the order of 6000 psig or even higher depending on the mechanical strength of the cell components and the ability of the current source to supply adequate electrons.

[0047] While the hydrogen feed chamber **23a** is shown to have a fluid outlet **21a** in FIG. 1(a) such that a hydrogen feed stream passes through the hydrogen feed chamber, the pump may operate "dead-ended" at the feed chamber side and have a purge valve (not shown) attached to the feed chamber fluid outlet **21a**. In a dead-ended operation, the hydrogen feed gas is substantially completely reacted in the hydrogen feed chamber **23a**, and the purge valve remains closed during normal pump operation. The purge valve may be opened from time to time to discharge any contaminants or excess water that may have accumulated during operation.

[0048] Referring to FIG. 2, a system **30** for pumping hydrogen using an HTLH membrane electrochemical cell **10a** is shown. A humidifier **32** feeds low pressure hydrogen feed gas and water vapor to the electrochemical cell **10a** via feed line **44**. The water vapor is used to hydrate the membrane and is typically in the order of 20% by weight or less. In this system **30**, some of the low pressure hydrogen feed and water vapor is recirculated through a heat exchanger **34** to carry away waste heat from the electrochemical cell **10a**. In this connection, the low pressure hydrogen and water vapor is coupled to the electrochemical cell **10a** such that hydrogen and water vapor is fed into the hydrogen feed chamber **23a** via fluid inlet **17a** and unreacted hydrogen and water vapor is discharged from the fluid outlet **21a** into fluid conduit **38**. The discharged hydrogen and water vapor absorbs heat from the electrochemical reaction and is transmitted from the electrochemical cell **10a** to the heat exchanger **34** by a circulation pump **36** fluidly coupled to the electrochemical cell **10a** via the fluid conduit **38**. Heat from the hydrogen and water vapor stream is transferred in the heat exchanger **34** to a coolant stream **40** passing through the heat exchanger **34**, and the cooled hydrogen and water vapor stream is recirculated back into feed line **44** by fluid conduit **42**. Hydrogen is output from the electrochemical cell **10a** by way of hydrogen output line **48**.

[0049] Alternatively, a coolant other than hydrogen may be directly circulated through the electrochemical cell **10a**. Possible coolants are air, water, a water/glycol solution, etc. As a further alternative, coolant may be transmitted directly through a fluidly isolated coolant loop (not shown) in the electrochemical cell **10a** as is known in the art.

[0050] Referring to FIG. 3, a system **50** for pumping hydrogen using the HTLH membrane electrochemical cell **10a** (pump) uses hydrogen generated by an electrolyzer cell **10b**. The electrolyzer cell **10b** may use an HTLH ion exchange membrane, which is described in further detail below. In this system **50**, feed water is fed via a cold water feed conduit **52** into a water tank **54**. Feed water is then supplied from the water tank **54** via another cold water feed conduit **56** to a heat exchanger **58**. Thermal transfer conduits are used to transmit waste heat from the electrolysis and pumping processes to the heat exchanger **58**, which heats up the feed water. The heated feed water is then sent via heated water feed conduit **68** to a water vaporizer **70**, wherein thermal input means **72** (e.g. electric heater) may be applied

if needed to vaporize the water, and water vapor is transferred to the electrolyzer cell **10b** for hydrogen production via water vapor feed line **74**. If required, water vapor can be used to heat and/or hydrate the hydrogen produced by electrolyzer cell **10b** via bleed line **78** connecting water vapor feed line **74** directly to a hydrogen inlet line **80** of the electrochemical cell **10a**. Product oxygen gas and excess water vapor is discharged from the electrolyzer cell **10b** via thermal conduction conduit **82** fluidly connected to the electrolyzer cell **10b** and thermally coupled to the heat exchanger **58**, for heat transfer to feed water passing through the heat exchanger **58**. The oxygen gas and excess water is then discharged from the heat exchanger **58** to a gas/water separator **62** via discharge conduit **60** for separation into an oxygen exhaust stream **64** and water stream, which is recycled back into water tank **54** via water conduit **66**.

[0051] Low pressure hydrogen gas produced by the electrolyzer cell **10b** and water vapor is fed into the electrochemical cell **10a** via the inlet line **80**. The electrochemical pumping process can be controlled to produce high pressure hydrogen product gas; the pumping process generates heat and the heat is collected and transferred back to the heat exchanger **58** via a thermal conduction conduit **81**, which may be for example a closed water or other suitable fluid loop thermally coupled to the electrochemical cell **10a** and heat exchanger **58**. The electrochemical cell **10a** discharges hydrogen product gas through gas line **84** into a hydrogen storage system **86**. The storage system **86** may be heated by a suitable thermal input means **88**, if desired.

[0052] Alternatively, a thermal energy transfer could occur directly to the water stream being vaporized by passing the water directly through the electrolyzer cell **10b** and or the electrochemical cell **10a**.

[0053] Referring to FIG. 4, a system **90** for pumping hydrogen includes the HTLH membrane electrochemical cell **10a** and uses hydrogen generated by a natural gas steam reforming process. In this system **90**, feed water is fed via a cold water feed conduit **92** into a water tank **94**, and then to a water vaporizer **98** via another cold water feed conduit **96**. The water in the water vaporizer **98** is heated by a suitable thermal input means **100**. Water vapor produced by the vaporizer **98** is fed into a natural gas reformer **104** via water vapor feed line **102**, and is used in the reforming process to produce hydrogen reformat from natural gas supplied to the reformer **104** via natural gas feed line **105**. An additional reformer stage **108**, such as a shift stage, may be connected in series by fluid line **106** to operate to reform natural gas at a lower temperature stage. Reformate and water vapor then are discharged from second reformer **108** to the electrochemical cell **10a** by way of fluid line **110**. A thermal conduction conduit **114** thermally coupled to the electrochemical cell **10a**, first and second reformers **104**, **108** and vaporizer **98** is used to transmit waste heat from the reforming and pumping processes to the water vaporization process by a suitable fluid in the conduit **114**. Thermal energy means (e.g. electric heater) may be used by the vaporizer **98** if needed to vaporizer water. The hydrogen is discharged from the electrochemical cell **10a** as high pressure hydrogen to a hydrogen storage system **118** via high pressure hydrogen line **116**. The storage system **118** may be heated by a suitable thermal input means **120**, if desired.

[0054] Alternatively a thermal energy transfer could occur directly to the water stream being vaporized by passing the water directly through the reformer(s) 104, 108 and/or electrochemical cell 10a.

[0055] Referring now to FIG. 5, a vaporizer cell 130 is combined with an HTLH membrane electrochemical cell 10a to form an integrated vaporizer/pump assembly 131; multiple vaporizer/pump assemblies can be combined to form a vaporizer/pump assembly stack (as shown by repeating lines in FIG. 5). The electrochemical cell 10a is thermally coupled to the vaporizer cell 130 to allow direct transfer of thermal energy from the electrochemical pumping process to the vaporization process. The vaporizer cell 130 comprises a pair of electrically and thermally conductive separator plates 134, 136 sandwiching a current conducting heat transfer plate 132. The electrochemical cell 10a comprises a pair of flow distribution and current conducting feed and product electrode layers 140, 142 sandwiching a HTLH ion exchange membrane electrolyte 143. The electrodes 140, 142 are connected to a current source via an external circuit (not shown). Low pressure hydrogen gas is fed through an inlet 144 of the feed electrode 140, and high pressure hydrogen gas is discharged out of an outlet 147 in the product electrode 142 by the electrochemical process previously described. Heat produced by the pumping process is thermally transferred through the electrodes 140, 142 to the separator plates in adjacent vaporizer cells 130. The heat is used in the vaporization process, wherein liquid feed water is fed through an inlet 148 of the vaporizer 130 and vaporized; water vapor is discharged from an outlet 146 of the vaporizer 130.

[0056] In the electrochemical pump, the water for membrane hydration may be purified by a vaporization process and the thermal energy of the electrochemical pump process may be integrated with the vaporization process. In the present electrochemical pump system, the thermal energy of the electrochemical pump process may be integrated with any endothermic process for storing the hydrogen produced from said electrochemical pump system. For example, the thermal energy of the electrochemical pump process may be integrated with the endothermic process for converting sodium borate to sodium borohydride. 17.

[0057] Electrolyzer

[0058] According to another embodiment of the invention and referring to FIGS. 1(b), 6 and 7, an electrochemical electrolyzer and system having an HTLH ion exchange membrane are provided.

[0059] Referring to FIG. 1(b) in particular, an electrolysis cell 10b with the half reactions for the electrolysis of water (as labeled in FIG. 1(b)) serves as an electrolyzer cell and has an HTLH ion exchange membrane 12 for the membrane electrolyte; a suitable membrane is PBI thin film with imbibed phosphoric acid. The electrolyte 12 is sandwiched between a pair of electrodes, namely a water feed electrode 14b, and a hydrogen product electrode 16. The water feed electrode 14b comprises a separator plate 18, and a water feed chamber 23b, which includes a fluid inlet 17b and a fluid outlet 21b. The hydrogen product electrode 16 comprises a separator plate 20, a hydrogen product chamber 25 and a fluid outlet 19. The electrodes 14b, 16 are electrically coupled by an electric circuit 22. The circuit 22 is electrically coupled to an electric current source 24. In operation,

low pressure water vapor is fed into the inlet of the fluid inlet 17b of the water feed chamber 23b, wherein some of the water vapor is catalyzed and disassociated into oxygen molecules, protons and electrons. Product oxygen gas and unreacted water vapor are discharged via the fluid outlet 21b. The protons conduct through the HTLH membrane 12 and combine with electrons at the hydrogen product electrode 16 to produce hydrogen product gas; electrons are moved through the circuit 22 by the current source 24. Hydrogen product gas is discharged through fluid outlet 19 of the hydrogen product chamber 25; a control valve (not shown) attached to the fluid outlet 19 can be operated to control the pressure of the product gas. The water feed electrode 14b may operate at near ambient pressure and the hydrogen product electrode 16 may operate at elevated pressure. The product electrode 16 may be operated at up to around 6000 psig or higher, depending on the mechanical strength of the electrode 16 and the ability of the current source to supply adequate electrons.

[0060] This HTLH electrochemical cell 10a is suitable for use as the electrolyzer cell 10b in FIG. 3.

[0061] Preferably, the HTLH membrane electrolyzer cell 10b is fed vaporized water. This prevents the washout of acid that occurs when such membranes are washed or soaked in liquid water. The vaporization of the water feed has the added benefit of inherently separating ionic contaminants from the water vapor. In typical electrolyzers operating below the boiling point of water a system for de-ionizing the liquid water is generally used. Therefore, an advantage provided by an HTLH membrane electrolyzer cell 10b is that the need for a liquid water de-ionization system is eliminated.

[0062] Further, liquid water is typically a means for carrying contaminants into these types of electrochemical cells. Again the use of water vapor in the HTLH membrane electrolyzer cell reduces the influx of potential contaminants to the cell.

[0063] In conventional electrolyzers operating at higher pressures the separator plates and electrodes are designed to provide maximum support to the electrolyte membrane. The designs typically utilize very small flow channels for the liquid water feed that results in water flow rate and circulation requirements many times the rate of water electrolysis to ensure complete distribution of the water within the flow field. The use of water vapor feed increases the mobility of the water molecules within the electrode/reactant diffusion layer and reduces the flow circulation for distribution requirements to a minimum.

[0064] An electrolyzer cell using an acid-doped PBI membrane electrolyte is expected to be operable in the temperature range of 100 to 200° C. This presents opportunities for thermal integration with other systems. The waste thermal energy from the electrolysis process is integrable with systems using an endothermic process for hydrogen storage, such as the process disclosed by Millennium Cell, which requires heat and hydrogen to reformulate sodium borate to sodium borohydride. The waste thermal energy from the electrolysis process could also be integrated with the vaporization process to vaporize a liquid water feed. Examples of such integrated systems are shown in FIGS. 6-8 and discussed below.

[0065] Referring to FIG. 6, a system 150 for generating hydrogen uses the HTLH membrane electrolyzer cell 10b

discussed previously and has a heat exchanger **158** that uses heat produced by the electrolyzer cell **10b**. In this system **150**, feed water is fed via a cold water feed conduit **152** into a water tank **154**. Feed water is then supplied from the water tank **154** via another cold water feed conduit **156** to the heat exchanger **158**. Thermal conduction conduits **159** are used to transmit waste heat from the electrolysis process to the heat exchanger **158**, which heats up the feed water. The heated feed water is then sent via heated water feed conduit **168** to a water vaporizer **170**, wherein thermal input means **172** (e.g. electric heater) may be applied if needed to vaporize the water, and water vapor is transferred to the electrolyzer cell **10b** for hydrogen production via water vapor feed line **174**. Product oxygen gas and excess water vapor is discharged from the electrolyzer cell **10b** via thermal conduction conduit **159** fluidly connected to the electrolyzer cell **10b** and thermally coupled to the heat exchanger **158**, for heat transfer to feed water passing through the heat exchanger **158**. The oxygen gas and excess water is then discharged from the heat exchanger **158** to a gas/water separator **162** via discharge conduit **160** for separation into an oxygen exhaust stream **164** and water stream, which is recycled back into water tank **154** via water conduit **166**. The hydrogen is discharged from the electrolyzer cell **10b** as hydrogen to a hydrogen storage system **177** via high pressure hydrogen line **175**. The storage system **177** may be heated by a suitable thermal input means **179**, if desired.

[0066] Alternatively, a thermal energy transfer could occur directly to the water stream being vaporized by passing the water directly through the electrolyzer cell **10b**.

[0067] Referring now to FIG. 7, a vaporizer cell **180** is combined with an HTHL membrane electrochemical electrolyzer cell **10b** to form an integrated vaporizer/pump assembly **184**; multiple vaporizer/electrolyzer assemblies **184** can be combined to form a vaporizer/electrolyzer assembly stack (as shown by repeating lines). The electrolyzer cell **10b** is thermally coupled to the vaporizer cell **180** to allow direct transfer of thermal energy from the electrolysis process to the vaporization process. The vaporizer cell **180** comprises a pair of electrically and thermally conductive separator plates **186**, **188** sandwiching a current conducting heat transfer plate **190**. The electrolyzer cell **10b** comprises a pair of flow distribution and current conducting water feed and hydrogen product electrode layers **192**, **194** sandwiching a HTHL ion exchange membrane electrolyte **196**. The electrodes **192**, **194** are connected to a current source via an external circuit (not shown). Low pressure water vapor is fed through an inlet **198** of the water feed electrode **192**, and hydrogen gas is discharged out of an outlet **200** in the product electrode **194** by the electrolysis process previously described. Heat produced by the electrolysis is thermally transferred through the electrodes **192**, **194** to the separator plates in adjacent vaporizer cells **180**. The heat is used in the vaporization process, wherein liquid feed water is fed from a water tank **202**, through a circulating pump (not shown), through an inlet **204** of the vaporizer **180** via a water feed line **206**, and vaporized; water vapor is discharged from an outlet **208** of the vaporizer **180**. Unreacted water and oxygen gas is discharged from the electrolyzer cell **10b** and through a heat exchanger **210** to reject excess heat and return the excess water or water vapor via Water return line **212** back into the water tank **202**. Alternatively, the water return line **212** can bypass the water tank **202** and couple to the water feed line **206**, and the heat

exchanger **210** or heat exchangers can be located on either or both of the water lines **212**, **206**, as appropriate for water and heat balance, as is known in the art.

[0068] It is to be understood that even though various embodiments and advantages of the present invention have been set forth in the foregoing description, the above disclosure is illustrative only, and changes may be made in detail, and yet remain within the broad principles of the invention. For example, some of the components described above may be implemented using a variety of different compounds and different structures. Therefore, the present invention is to be limited only by the appended claims.

1. An electrochemical cell comprising

- (a) an anode comprising a hydrogen-carrying fluid feed chamber with an inlet for receiving a hydrogen-carrying fluid;
- (b) a cathode comprising a hydrogen product chamber with an outlet for discharging a hydrogen product gas; and
- (c) a high-temperature low-hydration membrane sandwiched between the anode and the cathode;

the anode and the cathode being electrically couplable to an electric current source for powering the electrochemical cell to produce hydrogen gas in a reduction reaction at the cathode.

2. The electrochemical cell of claim 1 wherein the high-temperature low-hydration membrane is a non-fluorinated ionomer membrane.

3. The electrochemical cell as claimed in claim 2 wherein the membrane is an acid-doped polybenzimidazole membrane.

4. The electrochemical cell as claimed in claim 3 wherein the polybenzimidazole membrane is doped with an acid selected from the group of H_2SO_4 and H_3PO_4 .

5. The electrochemical cell of claim 4 wherein the hydrogen-carrying fluid feed chamber is a water feed chamber for receiving feed water and comprises an outlet for discharging reaction product and unreacted water, and the electrochemical cell is an electrolyzer that produces hydrogen gas in a reduction reaction at the cathode, and oxygen gas in an oxidation reaction at the anode.

6. The electrochemical cell of claim 4 wherein the hydrogen-carrying fluid feed chamber is a hydrogen feed chamber for receiving hydrogen-carrying feed gas, and the electrochemical cell is a pump that produces hydrogen gas in a reduction reaction at the cathode.

7. An electrochemical electrolyzer comprising

- (a) an anode comprising a water feed chamber with an inlet for receiving feed water and an outlet for discharging unreacted water and product oxygen gas;
- (b) a cathode comprising a hydrogen product chamber with an outlet for discharging product hydrogen gas; and
- (c) a high-temperature low-hydration membrane sandwiched between the anode and the cathode;

the anode and the cathode being electrically couplable to an electric current source for powering the electrolyzer

to produce hydrogen gas in a reduction reaction at the cathode, and oxygen gas in an oxidation reaction at the anode.

8. The electrolyzer as claimed in claim 7 wherein the membrane is a non-fluorinated ionomer membrane.

9. The electrolyzer as claimed in claim 8 wherein the membrane is an acid-doped polybenzimidazole membrane.

10. The electrolyzer as claimed in claim 9 wherein the polybenzimidazole membrane is doped with an acid selected from the group of H_2SO_4 and H_3PO_4 .

11. An electrolyzer system comprising

- (a) the electrolyzer of claim 7;
- (b) a feed water stream in fluid flow communication with the water feed chamber inlet of the electrolyzer;
- (c) a hydrogen product stream in fluid flow communication with the hydrogen product chamber outlet of the electrolyzer; and
- (d) a heat exchanger thermally coupled to the electrolyzer and in fluid flow communication with the water feed stream upstream of the electrolyzer, such that heat produced by the electrolyzer is used to heat the feed water stream.

12. The electrolyzer system of claim 11 further comprising a water vaporizer in fluid flow communication with the water feed stream upstream of the electrolyzer and downstream of the heat exchanger, for vaporizing the feed water.

13. The electrolyzer system of claim 11 wherein the heat exchanger is a vaporizer and comprises a pair of thermally conductive separator plates, a water vaporizing channel in between the separator plates having an inlet for receiving a liquid water feed stream and an outlet for discharging a water vapor feed stream, the vaporizer being thermally coupled to the electrolyzer by at least one of the separator plates being in thermal contact with the electrolyzer.

14. The electrolyzer system of claim 13 wherein the feed chamber outlet discharges a water vapor and oxygen gas stream, and the system further comprises a water recirculation circuit that comprises

- (a) a condensing heat exchanger having an inlet in fluid flow communication with the water vapor and oxygen discharge stream, and an outlet for discharging a liquid water stream condensed by the heat exchanger; and
- (b) a water tank in fluid flow communication with the liquid water stream discharged from the heat exchanger outlet, a liquid water make-up stream, and the feed water stream upstream of the vaporizer, such that liquid water recovered by the condensing heat exchanger is returned to the water feed stream.

15. The electrolyzer system of claim 12 wherein the feed chamber outlet discharges a water vapor and oxygen gas stream and the system further comprises a water recirculation circuit comprising a gas/water separator in fluid flow communication with the water vapor and oxygen gas discharge stream downstream of the heat exchanger, and having an oxygen gas discharge outlet, and a water discharge outlet fluidly coupled to the feed water stream upstream of the heat exchanger.

16. The electrolyzer system of claim 15 further comprising a hydrogen storage chamber in fluid flow communication with the hydrogen product stream discharged from the electrolyzer.

17. An electrochemical pump comprising

- (a) an anode comprising a hydrogen feed chamber with an inlet for receiving hydrogen-containing feed gas;
- (b) a cathode comprising a hydrogen product chamber with an outlet for discharging product hydrogen gas;
- (c) a high-temperature low-hydration membrane sandwiched between the electrodes;

the anode and cathode being electrically couplable to an electric current source for powering the electrochemical pump to produce the product hydrogen gas in a reduction reaction at the cathode.

18. The pump as claimed in claim 17 wherein the membrane is a non-fluorinated ionomer membrane.

19. The pump as claimed in claim 18 wherein the membrane is an acid-doped polybenzimidazole membrane.

20. The pump as claimed in claim 19 wherein the polybenzimidazole membrane is doped with an acid in the group of H_2SO_4 and H_3PO_4 .

21. The pump as claimed in claim 20 wherein the hydrogen feed chamber further comprises an outlet for discharging unreacted hydrogen-containing feed gas.

22. An electrochemical pump system comprising

- (a) the electrochemical pump of claim 21;
- (b) a hydrogen feed stream in fluid flow communication with the hydrogen feed chamber inlet;
- (c) a hydrogen discharge stream in fluid flow communication with the hydrogen feed chamber outlet and the hydrogen and water feed stream;
- (d) a heat exchanger in fluid flow communication with the hydrogen discharge stream downstream of the pump and upstream of the hydrogen feed stream, and in fluid flow communication with a coolant stream, such that the discharge stream can be cooled in the heat exchanger before joining with the feed stream; and,
- (e) a recirculation pump in fluid flow communication with the discharge stream.

23. An electrochemical pump system comprising

- (a) the electrochemical pump of claim 21;
- (b) a hydrogen and water feed stream in fluid flow communication with the hydrogen feed chamber inlet;
- (c) a hydrogen discharge stream in fluid flow communication with the hydrogen feed chamber outlet;
- (d) an electrolyzer having a hydrogen discharge outlet in fluid flow communication with the hydrogen feed stream upstream of the pump, and an inlet in fluid flow communication with a water feed stream, for producing hydrogen from the water feed stream.

24. The pump system of claim 23 wherein the electrolyzer is an electrochemical electrolyzer comprising an anode, a cathode and a high-temperature low-hydration membrane sandwiched between the anode and the cathode.

25. The pump system of claim 24 further comprising a heat recirculation circuit comprising a

- (a) a water vaporizer in fluid flow communication with the water feed stream upstream of the electrolyzer; and
- (b) a heat exchanger in fluid flow communication with the water feed stream upstream of the vaporizer and ther-

mally coupled to at least one of the electrolyzer and the pump, such that heat generated by at least one of the electrolyzer and the pump is transferable to the water feed stream.

26. The pump system of claim 25 further comprising a water recirculation circuit comprising

- (a) an oxygen and water discharge stream in fluid flow communication with the electrolyzer;
- (b) a gas/water separator having an inlet in fluid flow communication with the oxygen and water discharge stream downstream of the electrolyzer, and a water discharge outlet fluidly coupled to the feed water stream upstream of the heat exchanger, and an oxygen vent.

27. An electrochemical pump system comprising:

- (a) the electrochemical pump of claim 17; and,
- (b) a natural gas reformer comprising a hydrogen gas outlet in fluid flow communication with the hydrogen feed chamber inlet of the pump and a natural gas inlet in fluid flow communication with a natural gas source.

28. The electrochemical pump system of claim 27 further comprising a water vaporizer comprising a water feed inlet in fluid flow communication with a liquid water feed source, and a water discharge outlet in fluid flow communication with a water discharge stream that is in turn in fluid flow communication with the reformer.

29. The electrochemical pump system of claim 28 further comprising a thermal recirculation circuit comprising a thermal conduction conduit thermally coupling the vaporizer and at least one of the pump and reformer, such that heat generated by the reformer or the pump is transferable to the vaporizer.

30. An electrochemical pump system comprising

- (a) the electrochemical pump of claim 17; and,
- (b) a vaporizer in fluid flow communication with a water feed stream, and being thermally coupled to the pump such that heat generated by the pump is transferable to the water feed stream in the vaporizer.

31. The pump system of claim 30 wherein the vaporizer comprises a pair of thermally conductive separator plates, a water vaporizing channel in between the separator plates, a water feed inlet and water vapor outlet, the vaporizer being thermally coupled to the pump by one of the separator plates being in thermal contact with the pump.

32. The electrochemical pump as claimed in claim 17 wherein the hydrogen feed chamber further comprises a contaminant discharge outlet downstream of the inlet, such that the contaminants in the feed gas are filtered by operation of the electrochemical pump and discharged via the contaminant discharge outlet.

33. An electrochemical filter comprising

- (a) an anode comprising a hydrogen feed chamber with an inlet for receiving an unfiltered feed gas comprising hydrogen and contaminants, and an outlet downstream of the inlet and for discharging the contaminants;
- (b) a cathode comprising a hydrogen product chamber with an outlet for discharging a hydrogen product gas;
- (c) a high-temperature low-hydration membrane sandwiched between the electrodes; and

the anode and cathode being electrically couplable to an electric current source for powering the electrochemical filter to produce the hydrogen product gas in a reduction reaction at the cathode, thereby separating the contaminants from the hydrogen in the feed gas.

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