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(54) **Title**: A REACTOR AND A SYSTEM FOR HYDROGEN GENERATION

(57) **Abstract**: There is provided a reactor for generating hydrogen from a solid hydrogen-generating material, the reactor comprising: a chamber defined between a first end member and a second end member the second end member being spaced from the first end member along a separation direction; an inlet for receiving a fluid; an outlet for releasing hydrogen; and at least one gas transport path defined between a peripheral part of the chamber and a central part of the chamber to induce a radial flow in the chamber, wherein a maximum separation between the first end member and the second end member is less than a largest dimension of the chamber perpendicular to said separation direction.

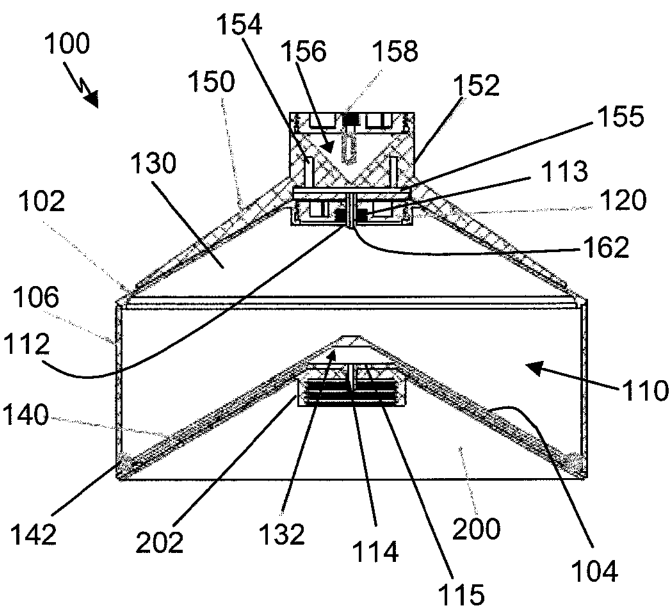

FIGURE 5

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A REACTOR AND A SYSTEM FOR HYDROGEN GENERATION

Background

The listing or discussion of a prior-published document in this specification should not necessarily be taken as an acknowledgement that the document is part of the state of the art or is common general knowledge.

The present invention relates to a reactor for hydrogen generation, and a system incorporating one or more such reactors.

In recent years there have been increased efforts to devise ways of using hydrogen as a fuel source. One such way is to inject an aqueous solution of a borohydride, particularly sodium borohydride ($NaBH_4$), as a hydrogen source, with a small quantity of a strong base as a reagent for preventing excessively fast hydrolysis, into a fixed bed reactor filled with a catalyst, to cause high-speed generation of hydrogen gas. This method suffers from many deficiencies, in particular, a low hydrogen production density – 4.6 wt% obtained on a stoichiometric basis, with borohydride being adequately reacted at a concentration of 22%.

To address these deficiencies, emphasis has shifted towards direct hydrolysis of a solid borohydride. However, use of solid fuel comes with its own challenges. In particular, it remains a challenge to design a reactor which can allow a solid borohydride, or other solid hydrogen-generating materials, to be hydrolysed safely and efficiently.

Summary

In a first aspect of the invention, there is provided a reactor for generating hydrogen from a solid hydrogen-generating material, the reactor comprising:

a chamber defined between a first end member and a second end member the second end member being spaced from the first end member along a separation direction;

an inlet for receiving a fluid;

an outlet for releasing hydrogen; and

at least one gas transport path defined between a peripheral part of the chamber and a central part of the chamber to induce a radial flow in the chamber,

wherein a maximum separation between the first end member and the second end member is less than a largest dimension of the chamber perpendicular to said separation direction.

In certain embodiments of the invention, the reactor may further comprise at least one sidewall extending between the first end member and the second end member, such that the first and second end members and the at least one sidewall form said chamber. For example:

(a) the at least one sidewall may be formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$; and/or

(b) the sidewall may be substantially cylindrical.

In further embodiments of the invention, the first end member may be a substantially conical disc. Additionally or alternatively the second end member may be a substantially conical disc.

In yet further embodiments of the invention, the maximum separation between the first end member and the second end member may be less than half the largest dimension of the chamber perpendicular to said separation direction.

In yet still further embodiments of the invention:

(a) the at least one gas transport path may be defined by a plurality of inlets on the periphery of the first or the second end member and at least one outlet at or close to the centre of the same end member; or

(b) the at least one gas transport path is defined by a plurality of outlets on the periphery of the first or the second end member and at least one inlet at or close to the centre of the same end member; or

(c) the at least one gas transport path is defined by a plurality of inlets on the periphery of the first or the second end member and at least one outlet at or close to the centre of the other end member; or

(d) the at least one gas transport path is defined by a plurality of outlets on the periphery of the first or the second end member and at least one inlet at or close to the centre of the other end member.

In further embodiments of the invention, the reactor may further comprise a separator disposed between the inlet and the outlet to divide the chamber into a first region which is in communication with the inlet and a second region which is in communication with the outlet.

In certain embodiments that may be mentioned herein, the inlet may be disposed on the first end member and the outlet may be disposed on the first or the second end member and the separator comprises a gas-permeable portion at a periphery of the second end member and a gas-impermeable portion that defines a fluid path from the gas-permeable portion to the outlet. In such embodiments, the gas-impermeable portion may be a separator plate and/or the gas-permeable portion may at least partially define the gas transport path (e.g. the separator plate may be substantially parallel to the second end member, optionally wherein:

(a) the separator plate may be located adjacent the second end member; and/or

(b)

(i) the outlet may be disposed on the second end member; or

(ii) when the outlet may be disposed on the first end member, the separator plate further comprises a gas-impermeable fluid path to said outlet).

In yet further embodiments of the invention, the reactor may further comprise a heat collector adjacent to the inlet, optionally wherein the heat collector may be formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$. Alternatively or additionally, the first end member and/or the second end member may be formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

In embodiments of the invention, the reactor may comprise a solid hydrogen-generating material disposed within the chamber. For example, the solid hydrogen-generating material may be sodium borohydride or a sodium borohydride-containing composition.

In yet further embodiments of the invention, the reactor may further comprise a heating element having a contact surface, wherein the contact surface has a shape which is complementary to a shape of an external surface of the second end member.

For the avoidance of doubt, any technically feasible combination of the embodiments of the first aspect of the invention is explicitly contemplated.

In a second aspect of the invention, there is provided a system for generating hydrogen from a solid hydrogen-generating material, comprising:

a housing formed from an insulating material, the housing having at least one recess shaped to accommodate a reactor according to the first aspect of the invention and any embodiment (or combination of embodiments) thereof; and

at least one heating element provided in each said recess that is arranged to be in thermal contact with the second end member of a reactor when the reactor occupies the recess.

In embodiments of the invention, the at least one heating element may have a contact surface which has a shape which is complementary to a shape of a contact surface of the second end member.

In further embodiments of the invention, the system may further comprise:

(a) a liquid tank for supplying liquid to the inlet of the at least one reactor. For example, the liquid tank may be connected to the inlet *via* a pump; and/or

(b) a buffer tank for receiving hydrogen from the outlet of the at least one reactor.

In yet further embodiments of the invention, the recess of the system may further comprise a heat collector integral to or removably attachable to the housing, where the heat collector is adjacent to the inlet of a reactor according to the first aspect of the invention and any embodiment (or combination of embodiments) thereof, and is in thermal contact with the first end member of said reactor when said reactor occupies the recess, optionally wherein the heat collector is formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

For the avoidance of doubt, any technically feasible combination of the embodiments of the second aspect of the invention is explicitly contemplated.

Brief Description of the Drawings

Embodiments of the invention will now be described, by way of non-limiting example only, with reference to the accompanying drawings in which:

Figure 1 is a side plan view of a first embodiment of a reactor;
Figure 2 is a cross-section through the line 2-2 of Figure 1;
Figure 3 is a block diagram of a system incorporating the reactor of Figures 1 and 2;
Figure 4 is a side plan view of a second embodiment of a reactor;
Figure 5 is a cross-section through the line 5-5 of Figure 4;
Figure 6 is a block diagram of a system incorporating the reactor of Figures 4 and 5;
Figure 7 is a plan diagram of a second end member of an alternative embodiment of the invention;
Figure 8 shows a side plan view of a further alternative embodiment of the reactor and system.
Figure 9 depicts the hydrogen output flow-rate versus time of a reactor according to Example 1.
Figure 10 depicts the hydrogen output flow-rate versus time of a reactor according to Example 2.

Detailed Description of Embodiments

Example embodiments of the invention will now be described more fully hereinafter with reference to the accompanying drawings; however, the invention may be embodied in different forms and should not be construed as limited to the embodiments set forth herein. Rather, these embodiments are provided so that this disclosure will be thorough and complete, and will fully convey exemplary implementations to those skilled in the art.

In the drawing figures, the dimensions of layers and regions may be exaggerated for clarity of illustration. Like reference numerals refer to like elements throughout.

As the invention allows for various changes and numerous embodiments, particular embodiments will be illustrated in the drawings and described in detail in the written description. However, this is not intended to limit the present invention to particular modes of practice, and it will to be appreciated that all changes, equivalents, and substitutes that do not depart from the technical scope are encompassed in the present invention. In the description, certain detailed explanations of related art are omitted when it is deemed that they may unnecessarily obscure the essence of the invention. While such terms as "first," "second," etc., may be used to describe various components, such components must not be limited to the above terms. The above terms are used only to distinguish one component from another. The terms used in the present specification are merely used to describe particular embodiments, and are not intended to limit the present invention. An expression used in the singular encompasses the expression of the plural, unless it has a clearly different meaning in the context. In the present specification, it is to be understood that the terms such as "including" or "having," etc., are intended to indicate the existence of the features, numbers, steps, actions, components, parts, or combinations thereof disclosed in the specification, and are not intended to preclude the possibility that one or more other features, numbers, steps, actions, components, parts, or combinations thereof may exist or may be added. Also, expressions such as "at least one of," when preceding a list of elements, modify the entire list of elements and do not modify the individual elements of the list.

In embodiments herein, the word “comprising” may be interpreted as requiring the features mentioned, but not limiting the presence of other features. Alternatively, the word “comprising” may also relate to the situation where only the components/features listed are intended to be present (e.g. the word “comprising” may be replaced by the phrases “consists of” or “consists essentially of”). It is explicitly contemplated that both the broader and narrower interpretations can be applied to all aspects and embodiments of the present invention.

Referring initially to Figure 1 and Figure 2, there is shown a reactor 100 having a first end member 102 which has an inlet 112. The first end member 102 is a substantially conical disc and the inlet 112 is located at the apex of the cone. A second end member 104, which in the depicted embodiment has a substantially conical outer portion, is disposed opposite the first end member 102, and a sidewall 106, which is substantially cylindrical, extends between the first and second end members 102, 104 such that the first and second end members 102, 104 and the sidewall 106 form an enclosure for a chamber 110 defined between the first end member 102 and the second end member 104. A cylindrical sidewall 106 is preferred as such a shape is simple to manufacture, and provides even distribution of hoop stress along the sidewall 106.

In some embodiments, the sidewall 106 may be omitted. For example, the first end member 102 may be a first conical disc having a first apex angle, and the second end member 104 may be a second conical disc having a second apex angle which is different (larger) than the first apex angle.

The chamber 110 is suitable for containing a solid hydrogen-generating material in particulate form, such as sodium borohydride or a sodium borohydride-containing composition, for reaction with a gaseous reagent received via the inlet 112 (e.g., water vapour). The reagent may be received directly through the inlet 112 in the gas phase, or may be received in the liquid phase and then heated within the chamber 110 to transform it to the gas phase. The gaseous reagent then reacts with the particulate composition to produce hydrogen, which is released from an outlet 114 of the second end member 104.

The reactor 100 is in thermal contact with a heating element in the form of a heater base 200 which has a contact surface (not shown) having a shape which is complementary to the shape of the external surface of the second end member 104. In particular, the heater base 200 has a conical portion which matches the shape of the conical outer portion of the second end member 104 such that the second end member 104 can sit snugly on the heater base 200. The heater base 200 is formed from a highly thermally conductive material such as an aluminium alloy. Preferably, an additional thermally conductive layer, such as a thermally conductive paste, is disposed between the heater base 200 and the second end member 104 to improve contact and to substantially eliminate air gaps.

When used herein, the term "thermal contact" is intended to relate to any type of contact that enables the transfer of thermal energy. This transfer of thermal energy may be accomplished by direct physical contact between the components, but it may also be accomplished without any physical contact, such as by any known heat transfer method, such as by convection and/or radiation heating etc.

Heat is applied to the reactor 100 by the heating element 200 to ensure that the internal temperature of the chamber is high enough to maintain the gaseous reagent in the gas phase, thereby avoiding choking of the reaction due to condensation and chemical aggregation.

The first end member 102 is spaced from the second end member 104 along a separation direction (for example, a central axis joining the inlet 112 and the outlet 114) such that the maximum separation between the first and second end members 102, 104 is less than a largest dimension of the chamber 110 perpendicular to the separation direction (e.g., less than the diameter when the sidewall 106 is cylindrical), and preferably less than half the largest dimension. By arranging the maximum separation to be less than the largest lateral dimension of the chamber 110, such that the reactor 100 is relatively "flat", conduction of heat between the second end member 104 and the first end member 102 is facilitated. This is important because it allows relatively homogeneous heating inside the reactor to maintain it above the boiling point of the gaseous reagent.

Disposed in the chamber 110 between the inlet 112 and the outlet 114 is a separator which divides the chamber 110 into a first region 130 which is in communication with the inlet 112, and a second region 132 which is in communication with the outlet 114. In this embodiment, the first region 130 is much smaller than the second region 132, and the particulate composition is placed in the second region 132. By making the first region 130 smaller than the second region 132, the space available for receiving the particulate composition is increased. The first region 130 may be very narrow since it only needs to receive a small volume of liquid reagent at any given time for conversion to gas to react with the particulate composition in the second region 132.

The separator has a gas-permeable portion, and functions to prevent movement of particulate matter from the first region 130 to the second region 132 or vice versa (which may block the inlet, if the particulate composition is stored in the second region, or the outlet if it is stored in the first region), whilst allowing the gaseous reagent to traverse the gas-permeable portion to react with the particulate composition.

For example, as shown in Figure 1, the separator may comprise a separator plate 140, preferably having a shape which is complementary to that of the first end member 102 (in this case, a conical shape). By having a shape complementary to that of the first end member 102, the volume of the second region 132 of the chamber 110 is maximised. The gas-permeable portion 142 may be located at the periphery of the separator plate 140, such that it forms a seal between the perimeter of the separator plate 140 and the sidewall 106. The gas-permeable portion 142 may be a carbon fibre filter, for example. Advantageously, by providing the gas-permeable portion at the perimeter of the separator plate 140, the gaseous reagent is directed to the sidewall 106 to define a gas transport path between the peripheral part of the chamber 110 and the centre of the chamber 110, and a radial flow field of gaseous reagent into the particulate composition is created, thereby resulting in better fuel utilisation and higher gas conversion efficiency since the gas reagent has a longer path length through the solid hydrogen-generating material in the chamber.

In some embodiments, the separator need not comprise a separator plate 140. For example, the separator may simply be a gas-permeable filter which does not allow the solid material to reach the outlet 114, thereby preventing blocking the outlet 114. In such embodiments, to define a gas transport path and create a radial flow of gaseous reagent towards the chamber 110 centre, the sidewall 106 may have a plurality of apertures formed therein (each of which is in communication with inlet 112), or a central inlet port may be provided such that gaseous reagent flows from the chamber 110 centre to the sidewall 106. Such arrangements are discussed in more detail in respect of Figures 7 and 8.

The filter components used in embodiments of the present invention may be of any type which can withstand high temperature (e.g., 100°C and above), is formed from a material that does not react in the reaction environment in the chamber 110, and has a pore size smaller than the smallest expected particle size.

The separator plate 140 may be made relatively thick, in which case it may serve as a heat collector. The heat collector may absorb heat due to the reaction enthalpy, which can then be used to heat liquid reagent received through the inlet 112 into the first region to evaporate it. Preferably, the separator plate 140 has a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

An insulating member 144, made of foamed silica gel for example, may be provided at the apex of the separator plate 140. Since the inlet 112 is at the apex of the first end member 102, and the apex of the separator plate 140 is adjacent the apex of the first end member 102, if a liquid reagent is received at the inlet 112, the region surrounding the apex of the separator plate 140 will tend to have the lowest temperature, since in general the second region 132 will be at a temperature higher than the boiling point of the liquid reagent. Accordingly, the insulating member 144 prevents the particulate composition in the chamber 110 from contacting the coldest part of the separator plate 140, thereby improving homogeneity of heating of the chamber 110 contents.

The reactor 100 may be provided as a sealed, single-use unit. Alternatively, it may comprise a resealable opening which may be used to fill and re-fill the chamber 110 with a solid reactant. For example, as shown in Figure 1, the second end member 104 may comprise a cap 120 which is received in a neck 122 extending from the conical portion of the second end member 104 via a screw-threaded connection. The outlet 114 of the second end member 104 may form part of the cap 120. When the reactor 100 is seated in the heater base 200, the neck 122 of the second end member 104 may nest in a corresponding sleeve 202 of the heater base 200. It is not essential that the cap 120 protrude from the conical portion of the second end member 104, but having it do so allows for greater space inside the chamber 110, and also provides means for positively locating the reactor 100 in the heater base 200. A gas-permeable filter, such as a layer of carbon porous paper (not shown), may be provided in the cap

120 to prevent particulate material from blocking the outlet 114 while permitting hydrogen to be released from the outlet 114.

Turning now to Figure 3, there is shown an example of a system for hydrogen generation 300, comprising a reactor 100 and heater base 200, which may be of the form shown in Figures 1 and 2. The system 300 comprises a housing 305 formed from an insulating material, such as a foamed plastics material, the housing 305 having a recess 310 shaped to accommodate the reactor 100. In particular, the recess 310 may have a diameter which is slightly larger than that of the sidewall 106, such that the reactor 100 and heater base 200 fit snugly within the recess 310. The housing 305 may comprise a plurality of such recesses such that multiple reactors 100 may be accommodated.

The inlet 112 of the reactor 100 is connected to a peristaltic pump 304 (although other types of pump may of course be used), which is in turn connected to a water supply 303. An outlet of the pump 304 is connected via tubing to the inlet port 112 such that the pump 304 can supply water at a desired rate to the inlet 112 and in turn to the first region 130 to generate water vapour.

The outlet 114 of the reactor 100 is connected to a buffer tank 307 via a heat sink 306. The buffer tank 307 receives hydrogen released from the outlet 114 and helps to stabilise the hydrogen pressure prior to outputting to a hydrogen-consuming load 309 (such as a fuel cell, for example). Typically, the gas output from the reactor outlet 114 will contain water vapour as well as hydrogen. The water vapour condenses in the buffer tank 307, and the condensate can be recycled, via an additional output port of the buffer tank 307 (not shown), to the water source 303. Control of the water recycling may be exercised by means of a solenoid valve 308 placed along an output line connected to the additional output port.

An alternative embodiment of a reactor 100 is shown in Figures 4 and 5. The reactor 100 comprises a first end member 102 which is spaced from a second end member 104. The first end member 102 has an inlet 112 and the second end member 104 has an outlet 114. The first and second end members 102, 104 are spaced along a separation direction (for example, a central axis extending between inlet 112 and outlet

114) and a sidewall 106 extends between them. The sidewall 106 may, as for the previous embodiment, be substantially cylindrical. The first and second end members 102, 104 and the sidewall 106 form an enclosure for a chamber 110. Each of the first and second end members 102, 104 has a substantially conical outer portion, and the respective conical portions are substantially parallel to each other.

As for the embodiment of Figures 1 and 2, the chamber 110 is suitable for containing a solid hydrogen-generating material in particulate form, such as sodium borohydride or a sodium borohydride-containing composition, for reaction with a gaseous reagent received via the inlet 112 (e.g., water vapour) which reacts with the particulate composition to produce hydrogen, which is released from the outlet 114 of the second end member 104.

The first end member 102 is spaced from the second end member 104 along the separation direction such that the maximum separation between the first and second end members 102, 104 is less than a largest dimension of the chamber 110 perpendicular to the separation direction, and preferably less than half the largest dimension.

Disposed in the chamber 110 between the inlet 112 and the outlet 114 is a separator which divides the chamber 110 into a first region 130 which is in communication with the inlet 112, and a second region 132 which is in communication with the outlet 114. In this embodiment, the first region 130 is much larger than the second region 132, and the particulate composition is placed in the first region 130.

The separator may comprise a separator plate 140, preferably having a shape which is complementary to that of the second end member 104. In particular, the separator plate 140 is substantially conical, such that it matches the shape of the substantially conical outer portion of the second end member 104. The gas-permeable portion 142 may be located at the periphery of the separator plate 140, such that it forms a seal between the perimeter of the separator plate 140 and the sidewall 106. The gas-permeable portion 142 may be a carbon fibre filter, for example. The separator plate 140 is spaced very closely from the second end member 104, such that the volume of the first region 130 of the chamber 110 is maximised while still allowing hydrogen gas generated in the first region 130 to migrate through the filter 142 to the outlet 114.

In some embodiments, the separator need not comprise a separator plate 140. For example, as for the embodiment described above, the separator may simply be a gas-permeable filter which does not allow the solid material to reach the outlet 114, thereby preventing blocking the outlet 114. In such embodiments, to define a gas transport path and create a radial flow of gaseous reagent towards the chamber 110 centre, the sidewall 106 may have a plurality of apertures formed therein (each of which is in communication with inlet 112), or a central inlet port may be provided such that gaseous reagent flows from the chamber 110 centre to the sidewall 106.

In the embodiment of Figures 4 and 5, the heat collector is located externally of the chamber 110. In this way, if the component of the reactor 100 which contains the solid fuel is to be made disposable, that component can be made lighter and be manufactured more cheaply than if the heat collector was internal to the chamber as in Figure 2. Further, by locating the heat collector externally of the chamber 110, a greater volume is available for storage of solid fuel whilst maintaining the same external dimensions.

In particular, heat collector 150 is seated on an external surface of the first end member 102. To this end, a contact surface of the heat collector 150 has a shape which is complementary to that of the external surface of the first end member 102. For example, in the depicted embodiment, the heat collector 150 has a conical outer portion which matches the conical outer portion of the first end member 102. The heat collector may be permanently affixed to the first end member 102, but is preferably removable. Preferably the heat collector 150 has a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

The heat collector 150 has a conical outer portion and a raised central portion 152. The raised central portion 152 has an inlet 158 for receiving liquid reagent, which flows into an evaporation chamber 156 of the raised portion 152. The heat collector 150 has a nozzle 162 which is seated within an O-ring or other seal 113 located adjacent the inlet 112 of the first end member. Accordingly, a gas-tight seal is formed between the nozzle

162 and the O-ring 113, such that gas can flow from the heat collector 150 into the chamber 110 via the inlet 112. Water vapour generated in the evaporation chamber 156 flows through channels 154 formed in the heat collector into gap 155 between the heat collector 150 and the first end member 102, whereupon it flows into the nozzle 162 and thus through inlet 112 of the first end member 102.

In some embodiments, the raised central portion 152 may comprise one or more heat pipes. This improves the heat collection performance, whilst also reducing the weight of the heat collector 150.

If the reactor 100 is intended to be reusable, then as in Figure 2, it may comprise a resealable opening which may be used to fill and re-fill the chamber 110 with a solid reactant. For example, as shown in Figure 5, the first end member 102 may comprise a cap 120 which is received by screw-threaded engagement in a neck at the central portion of the first end member 102. The cap 120 and neck may be made relatively thin so as to not take up an excessive portion of the volume of the chamber 110. The inlet 112 extends through the cap 120.

The outlet 114 of the second end member 104 extends from a substantially flat central portion 115 of the second end member 104. When the reactor 100 is seated on a heater base 200, the flat portion 115 sits on top of a central portion 202 of the heater base 200, with the conical outer portion of the second end member 104 sitting on a correspondingly shaped conical outer portion of the heater base 200. Preferably, an additional thermally conductive layer, such as a thermally conductive paste, is disposed between the heater base 200 and the second end member 104 to improve contact and to substantially eliminate air gaps (e.g. the heating element may be thermally coupled to the second end member by a layer of a thermally conductive material). The thermally conductive layer can be provided on the second end member 104 or on the heater base 200.

The heater base 200 may have a screw-threaded connector 202 for attaching an external acid filter (not shown) for purifying the hydrogen gas output from outlet 114.

In Figure 6, the reactor 100 of Figures 4 and 5 is shown in use as part of a hydrogen generation system 300. Reference numerals used in Figure 6 which are the same as those in Figure 3 are used to refer to like parts. As shown in Figure 6, a reactor 100 (including heat collector 150) may be received in the recess 310 of the housing 305, and connected to water source 303 at input 112 and to heat sink 306 and buffer tank 307 at output 114 substantially as described in relation to the reactor depicted in Figure 3.

Figure 7 shows a plan view of an alternative second end member 104 of a reactor 100. This second end member may be flat or conical and may be used with a flat or conical first end member 102, which may optionally be spaced apart by a sidewall 106. In the depicted second end member, there is no separation plate, instead there is a gas permeable portion 142 (which is otherwise identical to that described in connection to Figures 4 and 5) in fluid connection *via* a first fluid connection means or apparatus 172 to one or more gas impermeable tubes 170 (four are depicted), which are then in turn fluidly connected by a second fluid connection means or apparatus 174 to the outlet 114 (not depicted), which may be situated in the second end member 104, the first end member 102, or even a sidewall 106 (if such is present) of the reactor 100. In this embodiment, the gas permeable portion 142 and gas impermeable tubes 170, along with the various connection means or apparatus 172, 174, (or, alternatively, the tubes and connection means or apparatus) may be considered to act as the separator, such that the volume of the first region of the chamber is maximised, while still allowing hydrogen gas generated in the first region to migrate through the filter 142 to the outlet 114.

The first fluid connection means or apparatus 172 can be any suitable connector for connecting tubing together. For example, the connector may be a T-shaped connector made from any suitable material that can withstand the temperature of the reactor (e.g. a thermosetting polymer or metal, such as PTFE). The second fluid connecting means or apparatus 174 may be a manifold connector (made of similar materials to the first connection means or apparatus) that can accommodate all of the gas-impermeable tubes and provide at least one free connection point that can connect to the outlet 114 directly or indirectly. For example, when there are four gas impermeable tubes 170, the manifold connector may be a five-way connector, where the fifth connection means is directly attached to the outlet 114 on the second end member 104. Alternatively, the fifth connector may be provided with a gas impermeable tube that allows it to be indirectly connected to the first 102 or second 104 end members, or to the sidewall 106 (when such is present). In preferred variations of the embodiment depicted in Figure 7, there are at least four gas-impermeable tubes 170 (e.g. from four to twelve) that provide a fluid connection from the gas permeable portion 142 to the second fluid connecting means 174.

It will be appreciated that variations on the embodiment depicted in Figure 7 are possible. For example, one can dispense with the second fluid connecting means entirely and have the one or more gas impermeable tubes in direct contact with a similar number of outlets 114 instead, positioned conveniently in the reactor 100 (e.g. in the first or second end members, or in a sidewall if present). Alternatively, there may be one or more second fluid connecting means (e.g. T- or Y-shaped connectors) that enable fluid communication of two gas-impermeable tubes to a single outlet, such that a plurality of tubes are connected to a number of outlets 114 that number half the total of the plurality. Further variations on this theme are also envisaged by the current invention.

Figure 8 depicts yet a further embodiment of the current invention, one in which there is no requirement for a separator to divide the chamber 110 into a first and second region. Figure 8 depicts an alternative arrangement of the system 300 described in Figure 3, showing the housing 305 and heater base 200 attached to a reactor 100. As depicted, the reactor 100 is flat, and has a first end member 102 and second end member 104 separated by a sidewall 106 to form a chamber, where the separation of the first and second end members is less than a largest dimension of the chamber perpendicular to said separation direction, such that the required heating profile as discussed hereinbefore may be achieved.

As depicted in Figure 8, an evaporation chamber 152 is provided by a housing 154 enclosing a heat collector 150, said evaporation chamber 152 being provided with channels 156 in fluid contact with a plurality of inlets 112. Liquid water is deposited onto the heat collector 150 by way of an inlet 180 in the housing 154, following the application of heat to the reactor 100 by the heater base component 200, the liquid water is converted to steam and enters the reactor *via* the channels 156 and plurality of inlets 112 (e.g. twelve channels and inlets) on the periphery of the first end member 102. In order to induce a radial flow in the chamber, one or more outlets 114 are disposed at or around the centre of the first end member, with the outlet channel 115 being embedded in the heat collector 150.

In Figure 8, the housing 305 includes a recess 310 to accommodate the reactor, which housing makes use of clamps 350 to hold the reactor 100, heat collector 150 and heater base 200 together. In this arrangement, the clamps 350 may provide support to the reactor/heat collector/heater base arrangement to withstand internal pressure that builds up within the reactor during use, therefore maintaining the close connection of these three components.

It will be appreciated that further variations of the embodiment of Figure 8 can be envisaged. For example, the reactor and system may be arranged such that there is one or more inlets for steam 112 into the reactor 100 disposed at or around the centre of the first end member 102 and that a plurality of outlets 114 (e.g. from four to 12 or more) are disposed on the periphery of the first end member 102 to induce the required radial gas flow. For example, such further embodiments may include:

(a) a gas transport path defined by a plurality of inlets on the periphery of the first or the second end member and at least one outlet at or close to the centre of the same end member; or

(b)a gas transport path is defined by a plurality of outlets on the periphery of the first or the second end member and at least one inlet at or close to the centre of the same end member; or

(c) a gas transport path is defined by a plurality of inlets on the periphery of the first or the second end member and at least one outlet at or close to the centre of the other end member; or

(d) a gas transport path is defined by a plurality of outlets on the periphery of the first or the second end member and at least one inlet at or close to the centre of the other end member.

In any of the above embodiments, the first end member 102 and/or the second end member 104 and/or the sidewall 106 may be formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

The particulate composition used with the reactor 100 may be a sodium borohydride-containing composition, such as the composition described in PCT application no. PCT/CN2013/083551, the contents of which are incorporated herein by reference. In particular, Embodiments 1 to 10 and the composition as described hereinbelow.

The sodium borohydride-containing composition for generating hydrogen may comprise sodium borohydride and a filler. The filler is a substance having a chemical stability under an alkaline or neutral condition at a temperature in the range of 130°C to 140°C and a water solubility of less than 10 g per 100 g of water, where the chemical stability is an absence of the decomposition of the substance itself and of the occurrence of any chemical reaction of the substance with the sodium borohydride or water. The filler may have a bulk volume that is 0.02 to 16 times a bulk volume of the sodium borohydride, and may be present in a mass ratio to the sodium borohydride of equal to or less than 2:1. Further, the filler may have a bulk density of less than 16 and a mean particle size that is smaller than a mean particle size of the sodium borohydride. The alkaline condition is an alkalinity resulting from moisture-caused hydrolysis of the $NaBH_4$ or of its reaction product $NaBO_2$. The chemical reaction does not include simple hydration. For example, the filler may be $Mg(OH)_2$, which can be easily substituted by those of ordinary skill in the art for MgO, which is convertible to $Mg(OH)_2$, in part or as a whole, by simple hydration with water without significant changes in its physical and chemical properties and thus can achieve essentially the same result. For the same reason, the decomposition does not include simple dehydration. For instance, some hydroxides transform into corresponding oxides at certain temperatures by simple dehydration without significant change in its physical and chemical properties and thus can achieve essentially the same result.

This composition may address issues faced when using conventional borohydride-based hydrogen generation techniques, such as a low hydrogen production density, inadequate hydrolysis of borohydride, use of strong bases or toxic catalysts that are detrimental to the environment, and high cost.

The ratio of the bulk volume of the filler to the bulk volume of the sodium borohydride of 0.02~16 can be a ratio either before or after the two substances are mixed together. Herein, the filler functions as a physical support structure for isolating sodium borohydride particles to prevent their agglomeration, rather than a chemical reactant or a catalyst. Thus, it will be easily understood that the filler can be selected from an extensive variety of substances and each of its mass, volume, or bulk density ratios to the sodium borohydride can be selected from a wide range.

Preferably, the filler is present in a mass ratio to the sodium borohydride of equal to or less than 1:1, with equal to or less than 0.5:1 being more preferred, and with equal to or less than 0.1:1 being most preferred. A small mass ratio of the two substances is desirable for reducing the mass of the composition to result in an increased hydrogen production density. In contrast, while a larger mass ratio will not have any impact on the reaction adequacy and other technical criteria, it will increase the mass of the composition and hence leads to a lower hydrogen production density to reduce the practicality of the composition. For example, when the mass ratio is increased to 3:1, the composition will have a hydrogen production density only 1/4 of that of $NaBH_4$, causing a significant reduction in practicality.

Preferably, the filler is a substance having a bulk density of less than 2, with a substance having a bulk density of less than 0.5 (for example, $Mg(OH)_2$ which has a bulk density of 0.4) being more preferred, with a substance having a bulk density of less than 0.1 being even more preferred, and with foamed plastic pellets being most preferred (all known foamed plastic pellets have a bulk density of far less than 0.1). An extremely low bulk density of the filler is desirable for increasing the hydrogen production density of the composition. In contrast, while a larger bulk density will not have any impact on the reaction adequacy and other technical criteria, it will increase the mass of the composition and hence leads to a lower hydrogen production density to reduce the practicality of the composition. For example, if a substance with the same bulk volume but a bulk density of 16 is used in lieu of the filler in Embodiment 7 described below, the mass of the substance will be greater than 2 g, exceeding the mass of the used sodium borohydride, and accordingly, the hydrogen production density of the composition will be reduced by half and thus become less practical. Bulk density is defined as the density of a solid when packed or stacked in bulk and its unit is g/cm3. Foamed plastic pellets are generally produced from plastic pellets encapsulating thermo-expandable liquid or gas which expand tens of times its original size at a certain temperature into an extremely low bulk density. A conventional application of such plastic pellets is in the production of braille materials, in which the pellets are mixed with ink in an "unfoamed" form, and the mixture is then printed in patterns representing desired characters. When heated, the pellets expand to make the patterns embossed such that the visually impaired can read them by finger touch.

Preferably, the bulk volume of the filler is 0.2 to 8 times the bulk volume of the sodium borohydride, with 2 to 4 times being more preferred. A relatively large inter-particle distance is advantageous in preventing the agglomeration of sodium borohydride particles, and this distance can be maintained in an appropriate range when the bulk volume ratio of the filler to the sodium borohydride is set above 0.02. However, on the other hand, when the bulk volume ratio exceeds 16, even though it has no impact on the reaction adequacy and other technical criteria, the volume of the composition will become too large to meet the requirement of cost-effective storage and transportation, the mass and cost of the filler will increase to an unsuitable level, and the reactor will be required to have a larger housing which will lead to increase in weight and cost.

Preferably, the mean particle size of the filler is smaller than 0.5 times the mean particle size of the sodium borohydride, in order to facilitate the formation of microcosmic structures in which sodium borohydride particles are surrounded by particles of the filler and thus will not agglomerate together.

Preferably, the mean particle size of the sodium borohydride is larger than or equal to 0.1 mm and smaller than or equal to 2 mm, with larger than or equal to 0.2 mm and smaller than or equal to 2 mm being more preferred, and with larger than or equal to 0.2 mm and smaller than or equal to 1 mm being most preferred. When the mean particle size of the sodium borohydride is too large (larger than 2 mm), surface particles will be spaced by a larger distance from underlying particles, which is unfavorable to mass transfer and exchange and will lead to inadequate reaction. Moreover, when the mean particle size of the sodium borohydride is too small (smaller than 0.1 mm), with same bulk volume of the composition, the sodium borohydride particles will be in a very close neighborhood and hence prone to agglomeration.

The filler preferably has a water solubility of 1g/100g, and more preferably 0.1g/100g, under said condition. In general terms, the lower the water solubility, the better the filler will be kept in the solid state to function to separate sodium borohydride particles to prevent their fluidization and agglomeration. In a broader sense, $NaBO_2 \cdot nH_2O$, generated during the reaction, can also be envisioned as a filler, which is, however, an “unqualified” filler incapable of preventing the fluidization and agglomeration of sodium borohydride particles due to a relatively high solubility.

The composition may be in a form of a combination of the sodium borohydride and the filler before they are mixed together, or in a form of a mixture of the sodium borohydride and the filler. Further, the composition may also include water in a small quantity, or a product resulting from a reaction between water and the sodium borohydride, without departing from the broader scope of the invention. For instance, in certain conditions, part of the sodium borohydride may absorb a small amount of moisture and react therewith, and the composition will thereby include the remainder of the sodium borohydride, the filler and a product from the reaction.

Thus the reactor may contain a sodium-borohydride-containing composition comprising a filler. For example, the filler may be a substance having a chemical stability and a water solubility of less than 10g per 100g of water under an alkaline or neutral condition at a temperature of 130°C to 140°C, where the chemical stability is an absence of decomposition of the substance and an absence of chemical reaction with the sodium borohydride or water, and wherein the filler has a bulk volume of 0.02~16 times a bulk volume of the sodium borohydride, a mass ratio between the filler and the sodium borohydride being no greater than 2:1, the filler having a bulk density of less than 16, the filler having a mean particle size smaller than a mean particle size of the sodium borohydride, and wherein the alkaline condition is an alkalinity resulting from moisture-caused hydrolysis of the sodium borohydride or a reaction product $NaBO_2$ thereof, and wherein the chemical reaction does not include simple hydration and the decomposition does not include simple dehydration.

A method for generation of hydrogen using the above-described sodium borohydride-containing composition includes subjecting it to a temperature in the range of 110~160°C and bringing it in contact with water vapor. For example, using the reactors 100 and systems 300 described hereinbefore. The method is capable of a molar ratio of the consumed water to produced hydrogen gas of less than 0.9:1.

Preferably, in the method for generation of hydrogen, the temperature to which the composition is subjected is in the range of 120~150°C. A too high temperature can lead to over-dryness of the particles, which is unfavorable for obtaining a high reaction rate and a high reaction adequacy, whilst a too low temperature will lead to over-wetness of the particles, which may lead to their fluidization and agglomeration.

The method of the present invention achieves a high hydrogen production density in a borohydride-adequately-hydrolyzable, environmentally-friendly, low-cost, cost-efficient, easily-stoppable-and-restartable way. The present invention further allows a very low level of water consumption, which, in particular in its use in combination with a fuel cell equipped with means for recycling generated water, can result in a further increase in the hydrogen production density, and eliminate the need for water supplementation from an external source to significantly improve the maintainability and environmental independence of the fuel cell.

EXAMPLES

Example 1

The reactor and heater base of Figure 1 are applied, along with the system of Figure 3, except that a back pressure valve was used instead of a fuel cell, and that the backpressure valve was set at 2 Bar. The reactor was filled with 71.25 g of $NaBH_4$ mixed with 18.75 g of $Mg(OH)_2$.

The heater base was heated to 180°C and the temperature was controlled to maintain this temperature. When the temperature at the water input port of the reactor reached 160°C, the peristaltic pump starts up and provides a water flow rate of 0.375 mL/min into the water input port of the reactor.

Figure 9 shows the hydrogen output flow-rate of the reactor over time. As shown, in Figure 9, the temperature within the reactor may takes a period of time to reach 160 °C at the water input port of the reactor, representing a preheating period where no hydrogen gas is generated. Subsequently, hydrogen is produced as water is added to the reactor system through the water input port of the reactor until heat is no longer provided by the heating base. This results in the cooling of the reactor, such that the temperature at the water input port falls below 160°C which cuts off the addition of water to the reactor, thereby stopping the output of hydrogen. Thus, the periods of zero or low output represent periods where the reactor system was stopped, or a preheating period following the restart of the reactor system to generate hydrogen.

As can be seen from Figure 9, the reactor provides a consistent supply of hydrogen over an extended period of time, with a gradual reduction as the $NaBH_4$ is consumed.

Example 2

The reactor, heater base and heat collector of Figures 4 and 5 was used along with the system of Figure 6, except that a back pressure valve was used instead of a fuel cell, and that the backpressure valve was set at 2 Bar. The reactor was filled with 360 g of $NaBH_4$ mixed with 90 g of $Mg(OH)_2$.

The heater base was heated to 190°C and the temperature was controlled to maintain this temperature. When the temperature at the water input port of the reactor reached 160°C, the peristaltic pump starts up and provides a water flow rate of 0.38 mL/min into the water input port of the reactor and then the water flow rate is gradually increased to 0.65 mL/min.

Figure 10 shows the hydrogen output flow-rate of the reactor over time. As with Example 1, the temperature within the reactor of Example 2 takes a period of time to reach 160°C at the water input port of the reactor, representing a preheating period where no hydrogen gas is generated. Additionally, the periods of low or zero hydrogen output correspond to periods where the reactor system was stopped, or to preheating periods following subsequent re-starting of the system.

As can be seen from Figure 10, the supply of hydrogen gas can be sustained over a long period of time, and can be readily started and stopped to suit the needs of the user, enabling on-demand use from a single reactor system filled with a hydrogen generating material over an extended period of time.

Claims

1. A reactor for generating hydrogen from a solid hydrogen-generating material, the reactor comprising:

   a chamber defined between a first end member and a second end member the second end member being spaced from the first end member along a separation direction;

   an inlet for receiving a fluid;

   an outlet for releasing hydrogen; and

   at least one gas transport path defined between a peripheral part of the chamber and a central part of the chamber to induce a radial flow in the chamber,

   wherein a maximum separation between the first end member and the second end member is less than a largest dimension of the chamber perpendicular to said separation direction.

2. A reactor according to claim 1, comprising at least one sidewall extending between the first end member and the second end member, such that the first and second end members and the at least one sidewall form said chamber, optionally wherein the at least one sidewall is or are formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

3. A reactor according to claim 2, wherein the sidewall is substantially cylindrical.

4. A reactor according to any one of the preceding claims, wherein the first end member is a substantially conical disc and/or the second end member is a substantially conical disc.

5. A reactor according to any one of the preceding claims, wherein the maximum separation between the first end member and the second end member is less than half the largest dimension of the chamber perpendicular to said separation direction.

6. A reactor according to any one of the preceding claims, wherein:

(a) the at least one gas transport path is defined by a plurality of inlets on the periphery of the first or the second end member and at least one outlet at or close to the centre of the same end member; or

(b)the at least one gas transport path is defined by a plurality of outlets on the periphery of the first or the second end member and at least one inlet at or close to the centre of the same end member; or

(c) the at least one gas transport path is defined by a plurality of inlets on the periphery of the first or the second end member and at least one outlet at or close to the centre of the other end member; or

(d) the at least one gas transport path is defined by a plurality of outlets on the periphery of the first or the second end member and at least one inlet at or close to the centre of the other end member.

7. A reactor according to any one of claims 1 to 5, wherein the reactor further comprises a separator disposed between the inlet and the outlet to divide the chamber into a first region which is in communication with the inlet and a second region which is in communication with the outlet.

8. A reactor according to claim 7, wherein the inlet is disposed on the first end member and the outlet is disposed on the first or the second end member and the separator comprises a gas-permeable portion at a periphery of the second end member and a gas-impermeable portion that defines a fluid path from the gas-permeable portion to the outlet, optionally wherein said gas-permeable portion at least partially defines the gas transport path.

9. A reactor according to claim 8, wherein the gas-impermeable portion is a separator plate.

10. A reactor according to claim 9, wherein the separator plate is substantially parallel to the second end member, optionally wherein:

(a) the separator plate is located adjacent the second end member; and/or

(b)

(i) the outlet is disposed on the second end member; or

(ii) when the outlet is disposed on the first end member, the separator plate further comprises a gas-impermeable fluid path to said outlet.

11. A reactor according to any one of the preceding claims, further comprising a heat collector adjacent to the inlet, optionally wherein the heat collector is formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

12. A reactor according to any one of the preceding claims, wherein the first end member and/or the second end member is or are formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

13. A reactor according to any one of the preceding claims, comprising a solid hydrogen-generating material disposed within the chamber.

14. A reactor according to claim 13, wherein the solid hydrogen-generating material is sodium borohydride or a sodium borohydride-containing composition.

15. A reactor according to any one of the preceding claims, further comprising a heating element having a contact surface, wherein the contact surface has a shape which is complementary to a shape of an external surface of the second end member.

16. A system for generating hydrogen from a solid hydrogen-generating material, comprising:

a housing formed from an insulating material, the housing having at least one recess shaped to accommodate a reactor according to any one of claims 1 to 15; and

at least one heating element provided in each said recess that is arranged to be in thermal contact with the second end member of a reactor when the reactor occupies the recess.

17. A system according to claim 16, comprising a liquid tank for supplying liquid to the inlet of the at least one reactor, optionally wherein the liquid tank is connected to the inlet *via* a pump.

18. A system according to claim 16 or claim 17, comprising a buffer tank for receiving hydrogen from the outlet of the at least one reactor.

19. A system according to any one of claims 16 to 18, wherein the recess further comprises a heat collector integral to or removably attachable to the housing, where the heat collector is adjacent to the inlet of a reactor according to any one of claims 1 to 15 and is in thermal contact with the first end member of said reactor when said reactor occupies the recess, optionally wherein the heat collector is formed from a material having a thermal conductivity greater than 100 $W.m^{-1}.K^{-1}$.

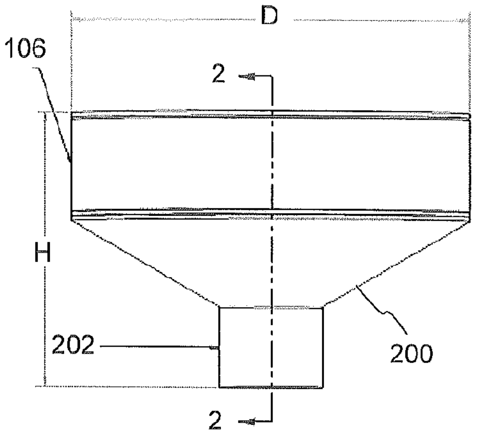

FIGURE 1

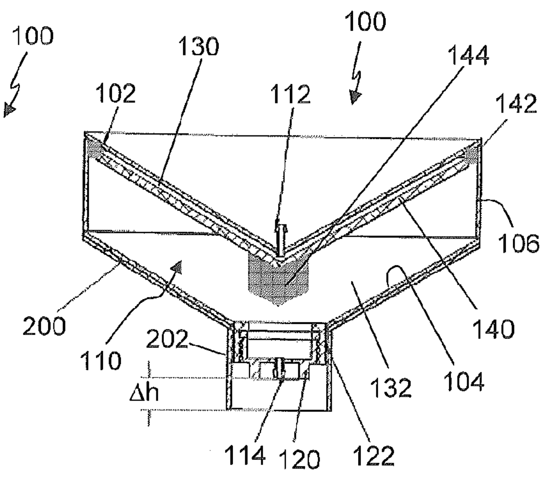

FIGURE 2

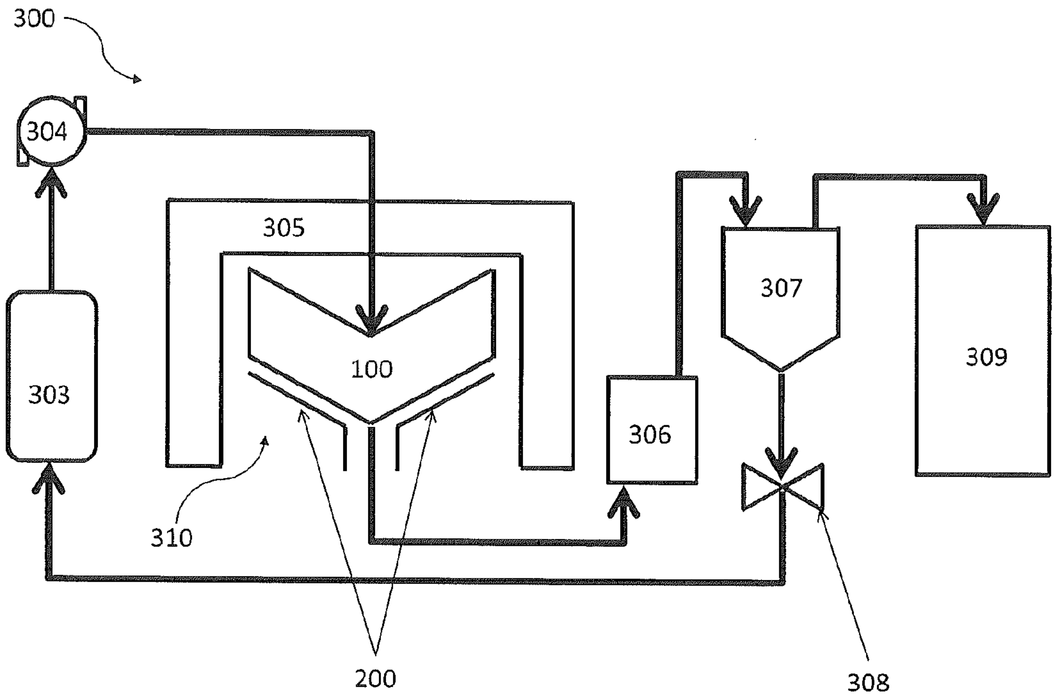

Figure 3

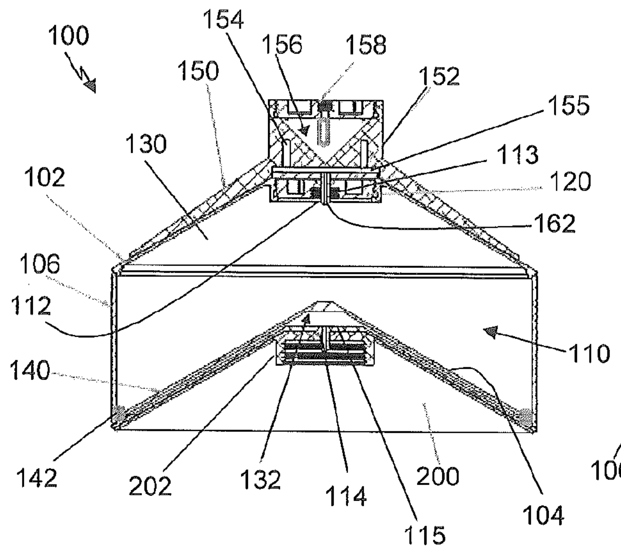

FIGURE 5

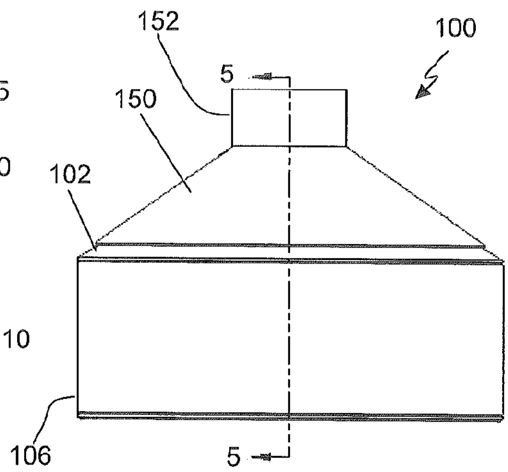

FIGURE 4

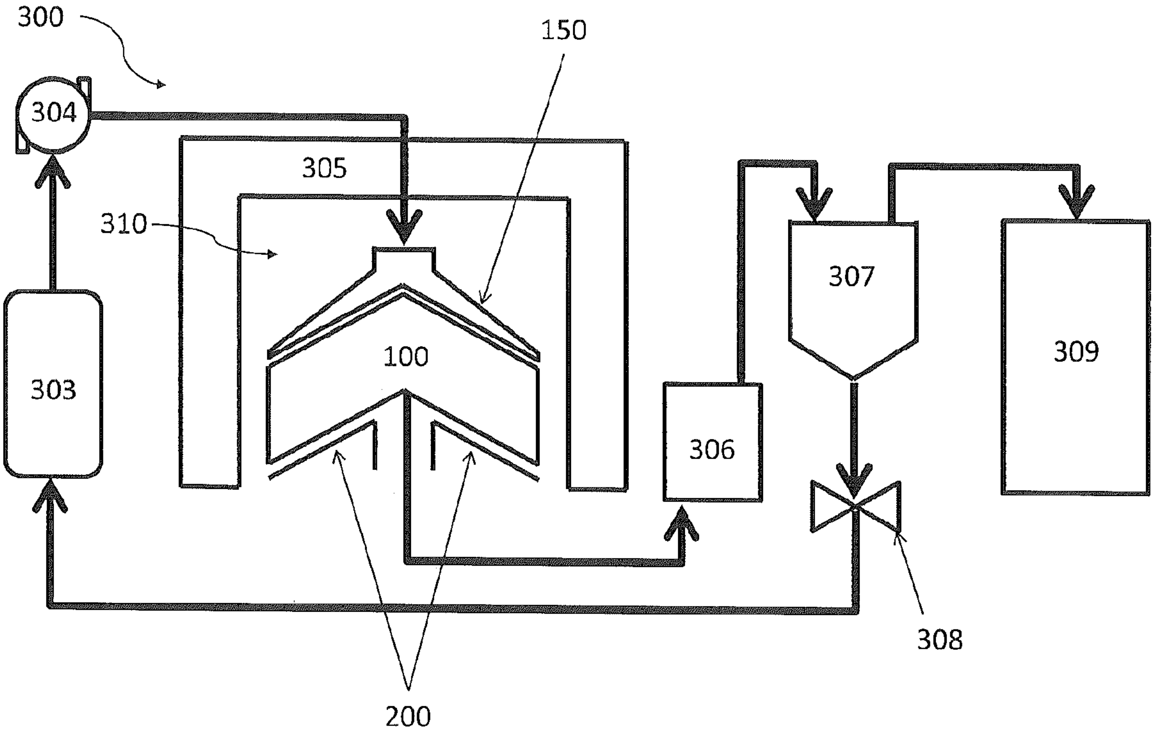

Figure 6

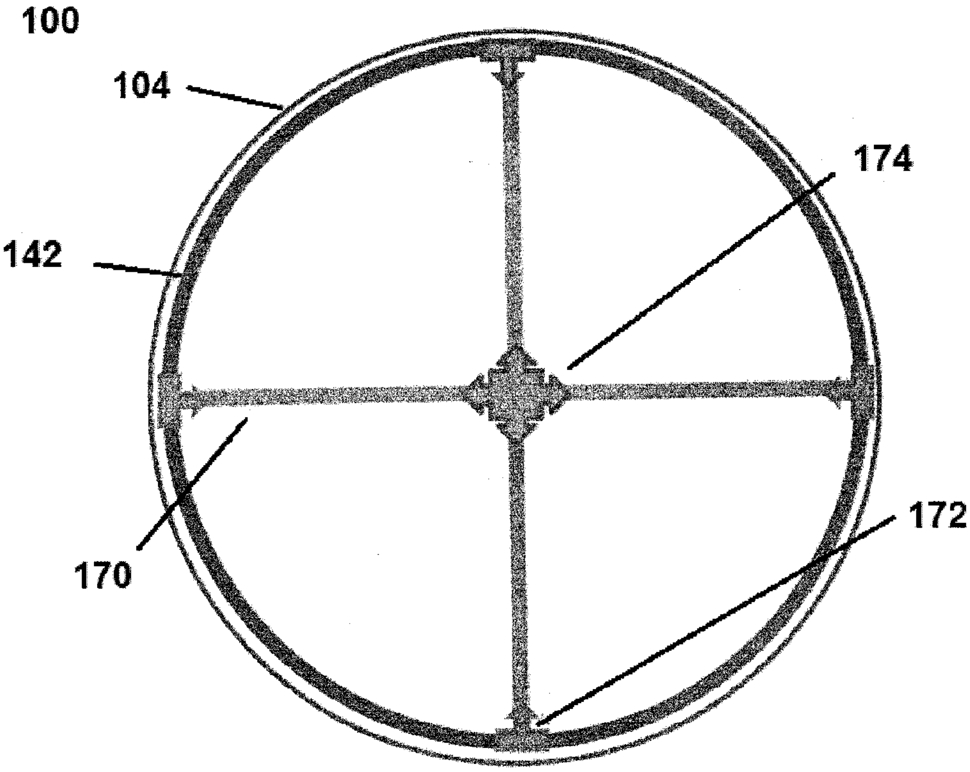

FIGURE 7

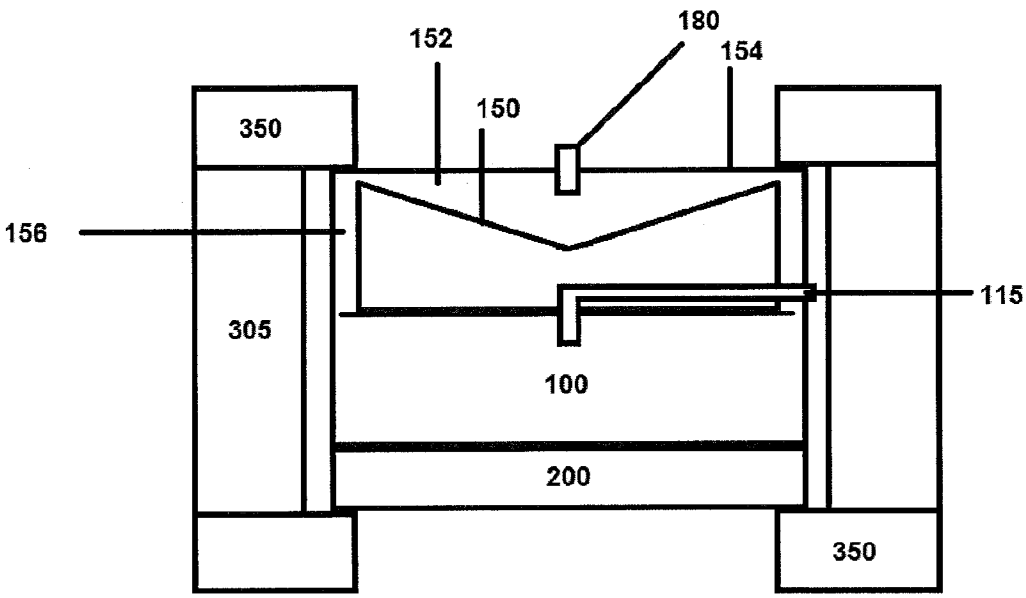

FIGURE 8

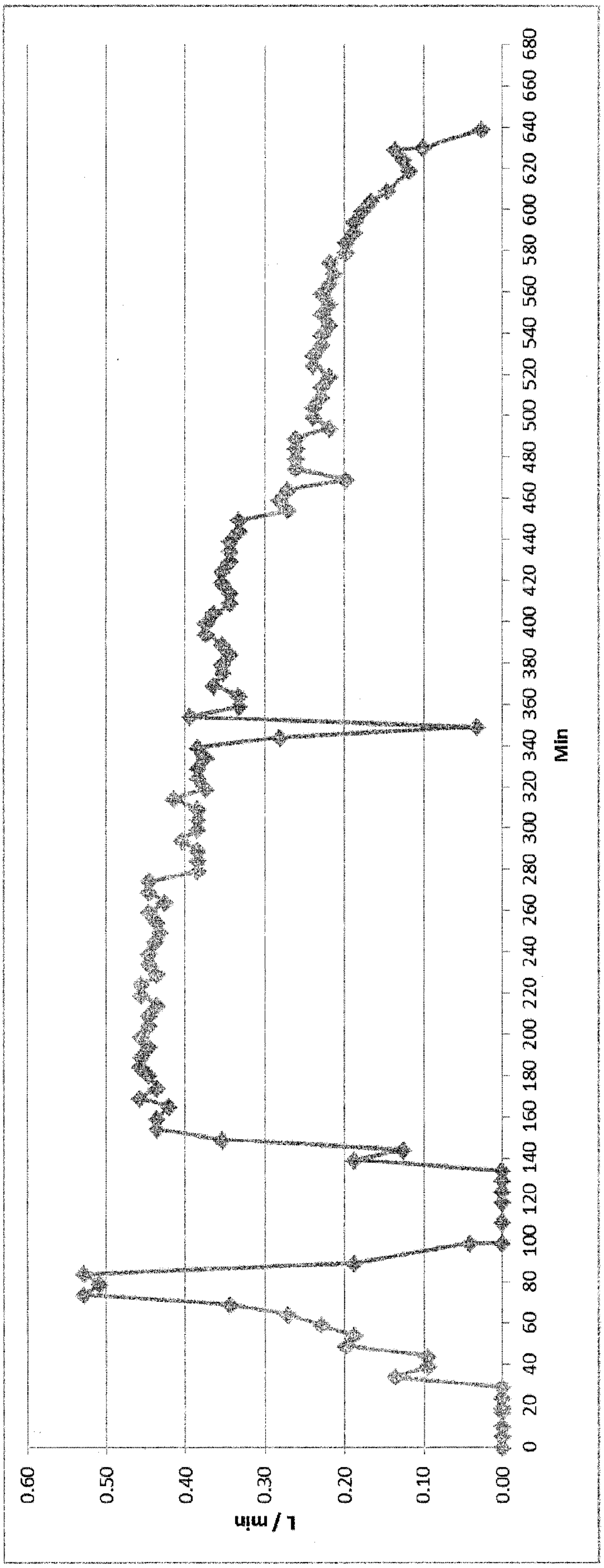
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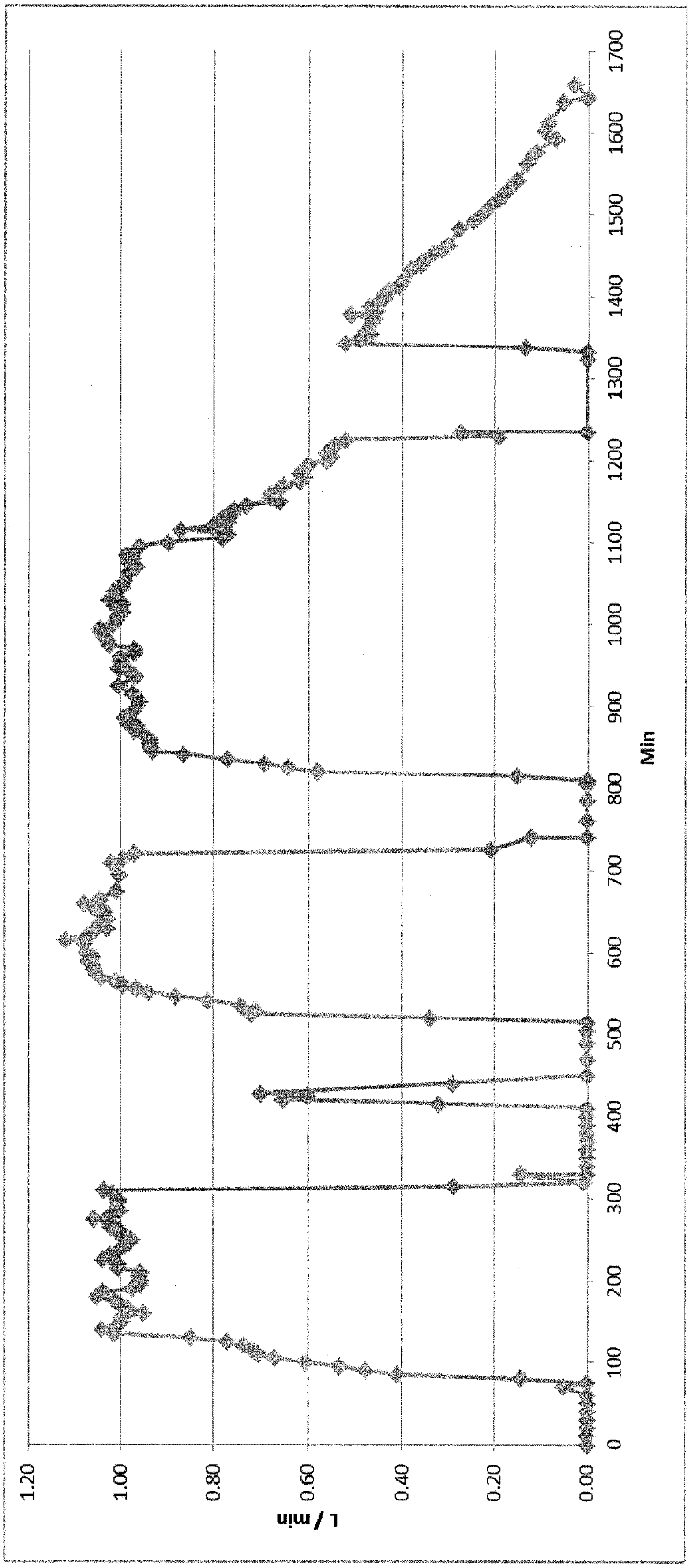
FIGURE 9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1100
1200
1300
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1500
1600
1700
Min

FIGURE 10

INTERNATIONAL SEARCH REPORT

International application No.
PCT/SG2015/050205

A. CLASSIFICATION OF SUBJECT MATTER

C01B 3/08 (2006.01) B01J 8/00 (2006.01) H01M 8/06 (2006.01) C01B 3/02 (2006.01)

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)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPIAP & EPODOC: (IPC or CC marks: C01B3/02 OR C01B3/04 OR C01B3/08 OR B01J8/00 OR B01J8/02) and (keywords: hydrogen generation reactor, chamber, shape)

Espacenet website: Applicant/inventors name search

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	Documents are listed in the continuation of Box C	

[X] Further documents are listed in the continuation of Box C [ ] See patent family annex

* 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

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"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	Date of mailing of the international search report
16 October 2015	16 October 2015

Name and mailing address of the ISA/AU	**Authorised officer**
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INTERNATIONAL SEARCH REPORT

International application No. **PCT/SG2015/050205**

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	US 2013/0115142 A1 (CHOU et al.) 09 May 2013	
X	Figures 2-8, paragraphs [0023]-[0027], [0030]-[0033]	1-3, 5-11, 13, 14
Y	As above	15
	US 2012/0115054 A1 (WALLACE et al.) 10 May 2012	
X	All figures, paragraphs [0053]-[0054], [0075]-[0080], [0085]-[0088]	1-14
Y	As above	15
	WO 2015/065289 A1 (TEMASEK POLYTECHNIC) 07 May 2015	
X	Page 9 line 10 to page 11 line 26, figures 3A-5	1, 2, 7-9, 11-14
Y	As above	15
	WO 2014/056386 A1 (JIN, K.) 17 April 2014	
Y	Figure 1	15
	US 2010/0247426 A1 (WALLACE et al.) 30 September 2010	
A	Figures 10A, 10B, 21	1-15
	RU 2553885 C1 (FEDERAL NOE G BJUDZHETNOE OBRAZOVATEL NOE) 20 June 2015	
A	Abstract and figure 1	1-15
	US 2014/0178780 A1 (REN) 26 June 2014	
A	All figures, paragraph [0032]	1-15

INTERNATIONAL SEARCH REPORT

International application No.
PCT/SG2015/050205

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
   because they relate to subject matter not required to be searched by this Authority, namely:
   the subject matter listed in Rule 39 on which, under Article 17(2)(a)(i), an international search is not required to be carried out, including

2. ☐ Claims Nos.:
   because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos:
   because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a)

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

See Supplemental Box for Details

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:
   1-15

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.

☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.

☐ No protest accompanied the payment of additional search fees.

Supplemental Box

Continuation of: **Box III**

This International Application does not comply with the requirements of unity of invention because it does not relate to one invention or to a group of inventions so linked as to form a single general inventive concept.

This Authority has found that there are different inventions based on the following features that separate the claims into two distinct groups:

Claims 1-15 are directed to a reactor for generating hydrogen from a solid hydrogen-generating material, the reactor comprising: a chamber defined between a fist end member and a second end member the second end member being spaced from the first member along a separation direction; an inlet for receiving a fluid; an outlet for releasing hydrogen; and at least one gas transport path defined between a peripheral part of the chamber and a central part of the chamber to induce a radial flow in the chamber, wherein a maximum separation between the first end member and the second end members is less than a largest dimension of the chamber perpendicular to said separation direction. The feature of the reactor having the dimensions as defined is specific to this group of claims.

Claims 16-19 are directed to a system for generating hydrogen from a solid hydrogen-generating material comprising: a housing formed from an insulating material, the housing having at least one recess shaped to accommodate a reactor, and at least one heating element provided in each said recess that is arranged to be in thermal contact with the second end member of a reactor when the reactor occupies the recess. The feature of the system comprising a housing and at least one heating element as defined is specific to this group of claim.

PCT Rule 13.2, first sentence, states that unity of invention is only fulfilled when there is a technical relationship among the claimed inventions involving one or more of the same or corresponding special technical features. PCT Rule 13.2, second sentence, defines a special technical feature as a feature which makes a contribution over the prior art.

When there is no special technical feature common to all the claimed inventions there is no unity of invention.

In the above groups of claims, the identified features may have the potential to make a contribution over the prior art but are not common to all the claimed inventions and therefore cannot provide the required technical relationship. Therefore there is no special technical feature common to all the claimed inventions and the requirements for unity of invention are consequently not satisfied *a priori*.