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(71) Applicants: **BOSHAUS PTY LTD** [AU/AU]; 123b Colin St, West Perth, Western Australia, 6005 (AU). **NATIONAL UNIVERSITY OF SINGAPORE** [SG/SG]; 21 Lower Kent Ridge Road, Singapore 119077 (SG).

(72) Inventors: **BHATTACHARJEE, Gaurav**; E5-02-09, Department of Chemical and Biomolecular Engineering, College of Design and Engineering, National University of Singapore, Singapore 117585 (SG). **LOANE, Antony William**; 123b Colin St, West Perth, Western Australia, 6005 (AU). **LINGA, Praveen**; E5-02-23, Department of Chemical and Biomolecular Engineering, College of Design and Engineering, National University of Singapore, Singapore 117585 (SG).

(74) Agent: **JONNALAGADDA, Naresh Kumar** et al.; K&S PARTNERS Intellectual Property Attorneys, 515-B, Platinum Tower, 5th Floor, Sohna Road, Sector 47, Gurgaon - 122002, National Capital Region, India, Gurgaon 122002 (IN).

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(54) Title: PROCESS FOR PRODUCTION OF HYDROGEN

(57) Abstract: The present disclosure relates to a process for production of hydrogen and metallic oxide(s). The process comprises reacting a hydrogen generating substance with water at elevated temperature and pressure conditions.



PROCESS FOR PRODUCTION OF HYDROGEN

CROSS -REFERENCE TO RELATED APPLICATIONS

This application claims the benefit of Indian Patent Application No.
5 202311021045, filed March 24, 2023, which is incorporated by reference herein in its
entirety.

TECHNICAL FIELD

The present disclosure generally relates to a process for production of hydrogen and
metallic oxide(s), by reacting a hydrogen generating substance with water at elevated
10 temperature and pressure conditions.

BACKGROUND

Hydrogen is considered the “fuel of the future” due to its ability to produce energy
without carbon emissions. It can be used in a variety of applications in addition to a green
energy carrier, including vehicle fuel, electricity production, petrochemical refining,
15 mineral processing, and chemical manufacturing. Many developed and developing
countries are investing in research on hydrogen production and storage as part of their
transition to a low-emission future. Hydrogen can be produced using different methods,
based on fossil fuel-based production, and electrolysis.

The three primary current and proposed options for large scale hydrogen production
20 are "grey H₂" (fossil fuel-based production), "blue H₂" (fossil fuel-based production
combined with carbon capture), and "green H₂" (production from renewables and zero-
emission sources). Currently, over 95% of the world's hydrogen is produced using either
steam methane reforming (SMR) or reforming of fossil fuels such as petroleum and coal,
while a small amount comes from electrolysis.

25 Reforming

To date the primary commercial pathway for hydrogen production is reforming hydrocarbons, with steam methane reforming (SMR) being the most prevalent. The feedstock requires significant pre-processing, and the reforming reaction produces a mixture of hydrogen and CO called Syngas, which requires several steps of post-processing. Close to 50% of global hydrogen demand is met via SMR, which produces direct emissions of 8 kg CO₂/kg H₂. Carbon capture and storage (CCS) methods are being implemented to decarbonize hydrogen production, but remain problematic and costly. Autothermal reforming (ATR) is an alternative to SMR, using oxygen instead of air for combustion as a heat source, and allowing for variable H₂:CO ratios.

10 Electrolysis

Electrolysis of water to produce hydrogen and oxygen represents the main focus of the proposed "green hydrogen" industry, and can be powered by renewable sources such as solar, wind, and hydro. Practical electrolysis using an electrolyser at 15 bar pressure may consume 50 kW·h/kg (180 MJ/kg), and a further 15 kW·h (54 MJ) if the hydrogen is compressed for use in hydrogen cars. The feedstock for the electrolysis process is high-purity water, requiring a significant level of processing, and the overall efficiency of electrolysis is very low, with only about 4% of H₂ produced worldwide created by electrolysis.

Therefore, there is a need in the art to provide a process for production of hydrogen which may overcome one or more or all drawbacks of the prior art.

SUMMARY

Accordingly, the disclosure herein provides a process for production of hydrogen. The disclosure herein also provides a process for production of hydrogen and metallic oxide(s). In one aspect, these processes comprise reacting a hydrogen generating substance with water at elevated temperature and pressure conditions.

In another aspect, the present disclosure provides a process for production of hydrogen; wherein the process comprises reacting a hydrogen generating substance with water in presence of a catalyst at elevated temperature and pressure conditions.

In yet another aspect, the process provided herein is a green, low-cost hydrogen
5 production process that offers several advantages over competing hydrogen production processes such as steam methane reforming (SMR) and electrolysis. In some aspects, the process provides several advantages, including a zero-carbon footprint, higher thermodynamic and mechanical efficiency, simpler process requirements, and/or smaller production plant sizes that offer both operational and economic benefits. In a further aspect,
10 the process provided herein produces pure hydrogen at high pressure suitable for both efficient storage and transport to market, and direct refueling of hydrogen fuel cell vehicles. Additionally, the metallic oxide by-product(s) (which in the case of using iron as a feedstock would be synthetic magnetite, whilst other metallic feedstocks or a combination thereof would produce related metallic oxides) represents another advantage over
15 competing hydrogen production processes as it may hold practical industrial and commercial value in its own right.

BRIEF DESCRIPTION OF THE DRAWINGS

The features of the present disclosure will become fully apparent from the following description taken in conjunction with the accompanying figures. With the understanding
20 that the figures depict only several embodiments in accordance with the disclosure and are not to be considered limiting of its scope, the disclosure will be described further through use of the accompanying figures:

Figure 1 illustrates an exemplary bench-scale plant as disclosed herein.

Figure 2 illustrates an exemplary evolution of the pressure and temperature profiles
25 within the reactor with respect to time (in hours) from points A to E for Run 1 as disclosed herein.

Figure 3 illustrates an exemplary evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) from point E onwards in Run 1 as disclosed herein.

Figure 4 illustrates (a) Grey colored finely divided pure Iron (Fe) powder used for the three experimental runs disclosed herein, and (b) Dried and crushed blackish solid phase product recovered post reaction from Run 1.

Figure 5 illustrates p-XRD analysis of the dried and crushed solid phase product recovered post reaction from Run 1 (metallic oxide, specifically synthetic magnetite in this case of iron being the metallic feedstock).

Figure 6 illustrates an exemplary evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) from points A to D for Run 2 as disclosed herein.

Figure 7 illustrates (a) Evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) from point C onwards in Run 2, and (b) The pressure and temperature evolution within the reactor for the 10 hours of the experiment immediately preceding point C.

Figure 8 illustrates standard gas chromatography peaks (signals) obtained for (a) Pure Hydrogen (H₂) gas, and (b) Pure Nitrogen (N₂) gas.

Figure 9 illustrates gas chromatograms obtained for two individual gas samples - (a) Sample 1, and (b) Sample 2 - recovered from inside the reactor post completion of Run 2, i.e., post the reaction between Iron powder and water in Run 2.

Figure 10 illustrates wet, jet black solid phase product recovered post reaction from Run 2 being metallic oxide (specifically synthetic magnetite in this case of iron being the metallic feedstock).

Figure 11 illustrates an exemplary evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) from points A and B for Run 3.

Figure 12 illustrates (a) Evolution of the pressure and temperature profiles within the reactor for the first 48 hours in Run 3 post the attainment of point A in the system, and (b) Evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) from point B onwards in Run 3.

5 Figure 13 illustrates gas chromatograms obtained for two individual gas samples - (a) Sample 1, and (b) Sample 2 - recovered from inside the reactor post completion of Run 3, i.e., post the reaction between Iron powder and water in Run 3.

 Figure 14 illustrates wet, jet black solid phase product recovered post reaction from Run 3 being metallic oxide (specifically synthetic magnetite in this case of iron being the
10 metallic feedstock).

DETAILED DESCRIPTION

 Before the present disclosure is described in greater detail, it is to be understood that the disclosure is not limited to the particular embodiments described, which may, of course, vary. It is also to be understood that the terminology used herein is for the purpose
15 of describing particular embodiments only, and is not intended to limit the present disclosure.

 Where a range of values is provided, it is understood that each intervening value, to the tenth of the unit of the lower limit unless the context clearly dictates otherwise, between the upper and lower limit of that range and any other stated or intervening value
20 in that stated range, is encompassed within the methods. The upper and lower limits of these smaller ranges may independently be included in the smaller ranges and are also encompassed within the methods, subject to any specifically excluded limit in the stated range. Where the stated range includes one or both of the limits, ranges excluding either or both of those included limits are also included in the methods.

25 Certain ranges are presented herein with numerical values being preceded by the term "about." The term "about" is used herein to provide literal support for the exact number that it precedes, as well as a number that is near to or approximately the number

that the term precedes. In determining whether a number is near to or approximately a specifically recited number, the near or approximating unrecited number may be a number which, in the context in which it is presented, provides the substantial equivalent of the specifically recited number. In an embodiment, "about" can mean within one or more
5 standard deviations, or within $\pm 30\%$, 25% , 20% , 15% , 10% or 5% of the stated value.

Unless defined otherwise, all technical and scientific terms used herein have the same meaning as commonly understood by those of ordinary skill in the art to which the disclosure belongs. Although any methods and materials similar or equivalent to those described herein can be used in the practice or testing of the present disclosure, preferred
10 methods and materials are described. For the purposes of the present disclosure, the following terms are defined below.

The articles "a" and "an" are used herein to refer to one or to more than one (i.e., to at least one) of the grammatical object of the article. By way of example, "an element" means one element or more than one element.

15 As used herein, the term "comprises" or "comprising" is generally used in the sense of include, that is to say permitting the presence of one or more features or components.

The term "hydrogen generating substance" refers to a substance that reacts with water at elevated temperature and pressure conditions to produce hydrogen.

In an embodiment, the present disclosure provides a process for production of
20 hydrogen. In certain embodiments, the present disclosure provides a process for production of hydrogen and metallic oxide(s). The processes comprise reacting a hydrogen generating substance with water at elevated temperature and pressure conditions.

In certain embodiments, the hydrogen generating substance is selected from a group comprising iron, an alkali metal, an alkaline earth metal, a transition metal, a post transition
25 metal, and any combination thereof. Examples of alkali metals include but are not limited to sodium, potassium, and lithium. In certain embodiments, the alkaline earth metal comprises magnesium or calcium. In certain embodiments, the transition metal comprises

zinc. In other embodiments, the post transition metal comprises aluminum or lead. In certain embodiments, the hydrogen generating substance is iron. In certain embodiments, the hydrogen generating substance is any combination of iron and the transition metal copper.

5 In certain embodiments, the hydrogen generating substance may be used as a solid reactant or a solid-state feed in the process. In other embodiments, the hydrogen generating substance may be of any size and/or shape. In some embodiments, the hydrogen generating substance is in powdered form.

10 In certain embodiments, any water may be used to react with the hydrogen generating substance to produce hydrogen. In some embodiments, the water is selected from a group comprising deionized water, distilled water, sea water, tap water, partially purified water, brackish water, and wastewater. In some embodiments, the water is non-purified water. In some instances, the water is deionized water. In further instances, the water is sea water. The water may be in liquid form, or gaseous/vapor form. In certain
15 embodiments, the water is in the liquid form.

 In certain embodiments, the present disclosure provides a process for production of hydrogen and metallic oxide(s) by reacting a hydrogen generating substance with water at an elevated temperature and elevated pressure; wherein the elevated temperature is at least about 95-100 °C, and in certain other embodiments from about 100 °C (373.2 K) to about
20 700 °C (973.2 K); and wherein the elevated pressure is at least about 80 bar, and in certain other embodiments from about 80 bar to about 1500 bar. In certain embodiments, the elevated temperature is from about 100 °C to about 700 °C, from about 100 °C to about 600 °C, from about 100 °C to about 500 °C, from about 100 °C to about 400 °C, from about 100 °C to about 300 °C, or from about 100 °C to about 200 °C. In certain embodiments,
25 the elevated pressure is about 80 bar to about 1400 bar, about 80 bar to about 1300 bar, about 80 bar to about 1200 bar, about 80 bar to about 1100 bar, about 80 bar to about 1000 bar, about 80 bar to about 900 bar, about 80 bar to about 800 bar, about 80 bar to about 700 bar, about 80 bar to about 600 bar, about 80 bar to about 500 bar, about 80 bar to about 400

bar, about 80 bar to about 300 bar, about 80 bar to about 200 bar, about 80 bar to about 100 bar, or about 80 bar to about 90 bar.

In certain embodiments, the hydrogen generating substance reacts with water at an elevated temperature and elevated pressure for a predetermined time. In certain
5 embodiments, the hydrogen generating substance reacts with water at an elevated temperature of about 100 °C and at an elevated pressure of about 80 bar to produce hydrogen and metallic oxide(s). In the case of using iron as a metallic feedstock, the metallic oxide by-product would be synthetic magnetite, whereas use of other metallic feedstocks or a combination thereof would produce related metallic oxide(s).

10 In certain embodiments, the hydrogen generating substance reacts with water at an elevated temperature and elevated pressure for a predetermined time.

In certain embodiments, one mole hydrogen generating substance is reacted with 1.3 moles of water at an elevated temperature and elevated pressure for a predetermined time.

15 In certain embodiments, the reaction of a hydrogen generating substance with water is optionally carried out in the presence of a catalyst to accelerate the reaction. The catalyst is selected from a group comprising salts and acids. Examples of salts include, but are not limited to, chlorides, hydroxides, sulfates, carbonates, phosphates, and nitrates of metals such as copper, sodium, calcium, potassium, and magnesium. The catalyst may present in
20 an amount from about 0.01 wt% (100 ppm) to about 100 wt% (1,000,000 ppm). In certain embodiments, NaCl is used as a catalyst, and in other embodiments, the catalyst is used in an amount of about 1 wt%.

In certain embodiments, the process comprises reacting iron with water at an elevated temperature and an elevated pressure to produce a product comprising metallic
25 oxide and a gas phase; condensing the water vapor in the gas phase comprising water vapor and hydrogen gas; and separating the condensed water from the gas phase; wherein the elevated temperature and pressure are same as defined above.

In certain embodiments, the reaction is carried out in a reactor. Figure 1 illustrates an exemplary bench-scale plant or system. As shown in Figure 1, the bench-scale plant or system comprises a vessel or reactor. In certain embodiments, the vessel or reactor is capable of withstanding elevated temperature and pressure conditions. In some
5 embodiments, the vessel or reactor is SS-316L pressure vessel or reactor. The reactor has one inlet pipe (used to supply nitrogen gas for a variety of purposes including to build up the pressure for the experiments, plus feedstock water) and two outlet pipes (one connects to a vent, and the other for discharge of gaseous products to the downstream plant), respectively. The reactor is further connected to a pressure relief valve, a pressure gauge /
10 transmitter, and a temperature gauge / transmitter. In certain embodiments, the pressure transmitter and temperature transmitter connect into a data acquisition system which records requisite data at regular (1 second or 20 seconds) intervals. The body of the reactor is substantially enveloped in a heater for maintaining the elevated temperature required for the experiments. Downstream of the reactor the plant also comprises at least a condenser,
15 a gas / liquid separator, a pressure control valve, and a gas receiver.

Accordingly, in certain embodiments, the present disclosure provides a process for generating hydrogen, comprising: reacting iron with water in a reactor at elevated temperature and pressure to produce a solid phase comprising metallic oxide, and a gas phase comprising hydrogen gas and water vapor; sending the gas phase to a condenser to
20 condense the water; and sending the condensed water to a gas/liquid separator to separate it from the gas phase. In some embodiments, the condenser is configured to a second predetermined temperature suitable for efficient condensation of bulk water. In some embodiments, the recovered water may undergo further treatment for reuse or disposal. The process further comprises directing the separated water into a receiver for storage. In
25 some embodiments, the process further comprises isolating or removing the metallic oxide. The metallic oxide may be isolated or removed by any method known to a person skilled in the art, such as filtration, evaporation, a combination of evaporation and filtration, and the like. In certain embodiments, the process produces pure metallic oxide.

Thus, in certain embodiments, the present disclosure provides a system for
30 production of hydrogen, comprising:

at least one reactor containing water, hydrogen generating substances, and optionally a catalyst; the reactor is defined with at least one inlet and two outlets;

a heater enclosing the reactor, wherein the heater is configured to maintain the reactor at a first predetermined temperature;

5 a condenser connected to the gas phase outlet of the reactor and configured to a second predetermined temperature for the purpose of condensing the water from the reactor gaseous products;

a gas / liquid separator connected to the outlet of the condenser, for the purpose of recovering bulk water from the gas phase product stream;

10 a pressure control valve connected to the gas phase outlet of the gas / liquid separator, and configured to maintain the first pre-determined pressure and thereby provide control of the reaction pressure; and

a receiver connected to the outlet of the pressure control valve, for the purpose of storing the product hydrogen.

15 In certain embodiments the reactor lid incorporates a vent valve (for the purpose of depressuring the reactor following completion of the reaction), and a pressure safety valve to avoid overpressure of the reactor.

In certain embodiments, the system comprises a source of nitrogen gas connected to the reactor for purposes including system purging, and pre-pressurisation of the reactor
20 (where appropriate).

In certain embodiments, the system comprises a pump connected to the reactor for the purposes of supplying feedstock water, and also pre-pressurisation of the reactor (where appropriate).

In certain embodiments of the system, the first predetermined temperature is at least
25 about 95-100 °C and the first predetermined pressure is at least about 80 bar.

In certain embodiments of the system, the second predetermined temperature is in a range from about 30 °C to 100 °C.

In certain embodiments, the present disclosure provides a system for continuous production of hydrogen, wherein the system comprises:

- 5 • at least one first reactor wherein the process of claims 1 to 16 takes place; a second reactor in re-charge mode for removal of product (e.g. hydrogen and metallic oxide) and installation of fresh reactants (e.g., water, hydrogen generating substance, and optionally a catalyst); and a third reactor in hot-standby mode ready for production upon completion of the reaction cycle
- 10 of the first reactor;
- synchronization of the reactor cycle to ensure that one reactor is in production mode, another in re-charge mode, and another in hot-standby mode at any given time; and
- 15 • computer-controlled robotic automation facilitating the continuous product flow operation, with specialized modifications to the reactors including quick opening closures, location of all process and instrument connections on the static component of the reactor, mechanical fixings on the "rotor" component suitable for robotic operation, and limit switches and interlocks to verify closure status of the reactor.

20 In certain embodiments, the reactors operate under high pressure conditions, and said robotic automation ensures safe operation and transition between different reactor modes.

In certain embodiments, the synchronization of the reactor cycle is achieved through precise control and timing mechanisms managed by the plant control system.

25 In certain embodiments, the specialized modifications to the reactors enable efficient and safe operation to achieve continuous product flow of hydrogen.

In certain embodiments, the robotic automation enables remote operation and monitoring of the reactors, enhancing operational safety and efficiency.

In certain embodiments of the process, a hydrogen generating substance, water, and optionally a catalyst are added to the reactor. Then, the temperature of the reactor is slowly increased to the desired (pre-determined) temperature (referred to as “first predetermined temperature”) by turning on the heater and regulating its temperature. Simultaneously, the pressure inside the reactor is slowly increased to the first pre-determined level (referred to as “first predetermined pressure”). In some instances, optionally, nitrogen gas is used to pressure the system, which is introduced slowly into the reactor via the inlet line connected to the reactor. In certain embodiments of the process, particularly those at the upper limit of the pressure range, initial reaction pressure is attained purely by hydraulic means without the need for nitrogen gas. In this embodiment process interlocks are utilized on the reactor pressure control system to provide a smooth transition into the reaction phase. This embodiment of the process represents a substantial invention that has not been referenced or foreseen in any prior art, yet is critical for achieving the full potential of the reaction for hydrogen and metallic oxide production. The gas phase produced in the reaction is first sent to a condenser to condense the water vapor, and then the condensed water is sent to a gas/liquid separator to separate it from the gas phase.

In certain embodiments, the process disclosed herein is performed in a closed reactor. A "closed reactor" refers to a system that temporarily isolates the reaction mixture from the environment and allows for gas pressure build-up by preventing materials from exiting its enclosure. Closed reactors may have openings or covers for accessing the reaction medium and are not restricted to permanently sealed or closed structures. The closed feature of the reactor may be limited to the period of operation by elements such as a cover or a port that provide reversible access to the interior. The reactor may have any shape, including cylindrical, cubical, and rectangular, and may be constructed from a variety of materials, such as metals, plastics, and ceramics, among others, or a combination thereof, each of which represents a distinct embodiment. According to certain embodiments, the reactor is equipped with a mixing mechanism that may be mechanical, magnetic, ultrasonic, or high-pressure liquid based.

In certain embodiments, the process is performed in a continuous product flow configuration requiring a minimum of three reactors whereby the first is in “production

mode” (with a reaction in progress), the second is in “re-charge mode” (involving the removal of product and installation of fresh reactants), and the third in “hot-standby mode” (prepared for production when the first reactor completes its reaction cycle). This cycle is synchronized to ensure that, at any given time, one reactor is in “production mode, another is in “re-charge mode”, and another in “hot-standby mode”. To address the safety aspects of hot reactors in the presence of high pressure hydrogen, this continuous product flow operation employs computer controlled robotic automation, requiring specialized modifications to the reactor including, but not limited to, quick opening closures; locating all process and instrument connections on the “static” component of the reactor; mechanical fixings on the “rotor” component of the reactor suitable for robotic operation; installing limit switches and interlocks to verify the closure status of the reactor. This embodiment of the process represents a substantial invention that has not been referenced or foreseen in any prior art, yet is critical for achieving the full potential of the reaction for hydrogen and metallic oxide production.

In certain embodiments, the process further comprises a step of collecting the produced H₂. In some embodiments, the process for producing hydrogen gas comprises a step of collecting the produced H₂ by delivering H₂ gas to a gas container through a gas pipe. The gas pipe can extend from the closed reactor to the container and may include a valve to seal the reactor during the reaction period while allowing the passage of H₂ to the container. Additional embodiments may include a release system with a valve, flame retardant, or bubbler attached, and a check valve with a flame arrester in the reactor or container.

In certain embodiments, the commencement of a specific experiment is dependent on the attainment of predetermined temperature and pressure levels inside the reactor system. The experiment may not be considered to have been initiated until these levels are reached. In certain embodiments, the methodology employed for increasing the temperature and pressure inside the reactor may differ slightly for each experiment.

In some embodiments, the reaction between hydrogen generating substance and liquid water to generate hydrogen gas and solid metal oxide may occur at an initial system pressure as low as about 80 bar and a system temperature as low as about 100 °C.

5 In certain embodiments, the rate of generating hydrogen is directly proportional to the initial pressure and temperature of the system. In other words, the higher the initial pressure and temperature of the system, the greater the rate of hydrogen generation achieved. Furthermore, higher initial pressure and temperature may also lead to a higher final degree of conversion of hydrogen generating substance, indicating that the reaction proceeds more completely to generate the desired product.

10 In certain embodiments, the process disclosed herein provides pure hydrogen and is substantially devoid of impurities. In some embodiments, the produced H₂ has purity of 100%.

In certain embodiments, the present disclosure provides a process for production of hydrogen and solid metallic oxide(s), for example synthetic magnetite in the case of using iron as a metallic feedstock; wherein the process comprises reacting iron with water in the presence of a catalyst at elevated temperature and pressure conditions. In some
15 embodiments, the reaction conditions are the same as described above. In certain embodiments, the process provides pure hydrogen and solid metallic oxide(s), for example synthetic magnetite in the case of using iron as a metallic feedstock which are substantially
20 devoid of impurities. In some embodiments, the produced H₂ has purity of 100%. In other embodiments, the produced magnetite is synthetic grade having purity of at least about 98%.

In certain embodiments, the process described herein has the advantage of being able to use reactants from different sources, including waste, without requiring purification,
25 pre-treatment, or pre-processing. In some embodiments, the reactants can be purified, pre-treated, or pre-processed before being used in the process.

In certain embodiments, the process provided herein is a green, low-cost hydrogen production process that offers several advantages over competing hydrogen production

processes such as steam methane reforming (SMR) and electrolysis. In some embodiments, the process provides several advantages, including a zero-carbon footprint, higher thermodynamic and mechanical efficiency, simpler process requirements, and/or smaller production plant sizes that offer both operational and economic benefits. In further
5 embodiments, the process provided herein produces pure hydrogen at high pressure suitable for both efficient storage and transport to market, and direct refueling of hydrogen fuel cell vehicles. Additionally, the metallic oxide by-product(s) (which in the case of using iron as a feedstock would be synthetic magnetite, whilst other metallic feedstocks or a combination thereof would produce related metallic oxides) represents another advantage
10 over competing hydrogen production processes as it may hold practical industrial and commercial value in its own right.

The present disclosure is further described with reference to the following examples, which are only illustrative in nature and should not be construed to limit the scope of the present disclosure in any manner.

15 **Materials**

Pure Iron (Fe; $\geq 99\%$, reduced, fine powder) and Sodium Chloride (NaCl; 99.0 % purity by mass (Titration with AgNO_3)) were purchased Sigma Aldrich (Merck) Pte. Ltd. Deionized water used for all the experiments was obtained using an Elga micromega (Merck Millipore Direct-Q® 3 UV) deionization apparatus. Pure Nitrogen (N_2) gas
20 (99.995 mol% purity) was procured from Air Liquide Singapore Pvt. Ltd.

EXAMPLES

EXPERIMENTAL-SET UP: BENCH-SCALE PLANT

A bench-scale plant was set up to establish 'proof of concept' for the process for production of hydrogen and metallic oxide(s) to which the present disclosure relates. Figure 1 shows
25 a schematic of the bench-scale plant employed which had at its heart, a SS-316L pressure vessel or reactor with an internal volume of 500 ml, a design pressure of 260 bar, and a design temperature of 350 °C. The lid of the reactor was connected through high-

temperature and high-pressure resistant valves to one inlet pipe (used to supply nitrogen gas to build up the pressure for the experiments) and two outlet pipes (one of these opened into a vent, and the other was used for gas sampling). A pressure safety valve also connected to the reactor lid was employed as a fail-safe for the experiments. Further, the reactor lid was equipped with a pressure transducer, a pressure gauge, and a thermowell incorporating a temperature gauge. While the pressure transducer and the pressure gauge directly measure the pressure inside the reactor at all times during the experiments, a thermocouple was inserted into the thermowell to constantly monitor the temperature during the experiments. The pressure transducer, pressure gauge, and thermocouple all connected into a data acquisition system which recorded the requisite data at regular (1 second or 20 second) intervals. A ceramic band heater which enveloped the entire body of the reactor (excluding the lower flange and the reactor lid) was used to maintain the elevated temperature necessary for the experiments. The plant design also consisted of a condenser, two condensate pots, a pressure control valve, and a gas accumulator (receiver) downstream of the reactor. However, in the case of the current experiments, for proof of concept experimental runs the reactor outlet was closed and there was no use of the unit components installed downstream of the reactor.

EXPERIMENTAL PROTOCOL

The experiments were conducted at elevated pressure and temperature conditions. The experiments each began with the necessary reactants (10 g iron (Fe; finely divided) and 200 ml deionized water) being loaded into the reactor and the reactor being tightly closed. In two of the reported experiments, sodium chloride (NaCl) was used as a potential catalyst. When used, NaCl was also loaded into the reactor (along with the iron particles and the water), prior to the reactor being sealed. Next, the temperature of the reactor was slowly increased to the desired (pre-determined) experimental pressure by turning on the heater and regulating its temperature. Alongside the reactor temperature, the reactor pressure was also slowly raised to the desired (pre-determined) experimental pressure. In the experiments conducted, the pressure inside the system (reactor) was created using nitrogen gas that was introduced from a cylinder slowly into the reactor, via the inlet line connected to the reactor lid. A particular experiment was only deemed to have started when the

pressure and temperature inside the system (reactor) had reached the pre-determined pressure and temperature for said experiment.

5

Results and Discussions

The Experimental Runs

Table 1 lists the various experimental parameters employed for the three experimental runs that have been disclosed in the present invention disclosure. The parameters that have been provided in Table 1 include among others, the pre-determined target experimental pressure, the pre-determined target experimental temperature, the amounts of the reactants used, the name and the amount of catalyst used (if any), and the run-time of the experiment (at the desired experimental pressure and temperature).

10

Table 1

Experiment (Amounts of Reactants Used)	Target Pressure (bar)	Target Temperature (°C)	Catalyst Used	Amount of Catalyst Used (wt% and grams)	Run Time (hours)
Run 1 (Fe: 10 grams; Deionized water: 200 ml)	80	100	Sodium Chloride (NaCl)	1 wt% (2 grams)	Total Experimental Run Time: 167.1 hours
					Actual Reaction Run Time: 47.5 hours

Run 2 (Fe: 10 grams; Deionized water: 200 ml)	80	100	Sodium Chloride (NaCl)	1 wt% (2 grams)	Total Experimental Run Time: 95.6 hours
					Actual Reaction Run Time: 40.5 hours
Run 3 (Fe: 10 grams; Deionized water: 200 ml)	100	100	Sodium Chloride (NaCl)	1 wt% (2 grams)	Total Experimental Run Time: 191.9 hours
					Actual Reaction Run Time: 182.6 hours

All three experimental runs reported were conducted at pre-determined experimental pressures and temperatures. In order to identify a successful reaction between Iron particles and water, there are three distinct markers that can be observed. Firstly, there will be an increase in pressure in the system, which is caused by the production of hydrogen gas.

5 Secondly, the presence of hydrogen gas in the gas phase product can be determined through the use of Gas Chromatography (GC). Finally, the presence of magnetite in the solid phase product can be determined through a solid-state characterization technique, such as Powder X-Ray Diffraction (p-XRD).

Experimental Run 1

- 10 The first experimental run (Run 1) was conducted in a specific manner so as to recognize the lowest pressure-temperature condition under which, within a specific period of time, markers for a successful reaction could be observed. As shown in Table 1 above, the quantities of the two reactants, finely divided Iron particles (Fe) and deionized water, used for Run 1, were 10 grams, and 200 ml, respectively.
- 15 Figure 2 illustrates the complete evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) for Run 1. As can be observed from Figure 2, for

Run 1, the temperature and pressure inside the system were first kept constant at approximately 46 °C and approximately 50 bar, respectively, for a period of about 56 hours (point A to point B in Figure 2) with no change observed in the reactor pressure. This indicates that no reaction took place for the entire duration at which the system was maintained at about 46 °C and about 50 bar, i.e., between point A and point B in the experimental run. Following this hold period, the reactor temperature and the reactor pressure were raised progressively to approximately 80 °C and approximately 76 bar, respectively. At this point the reactor temperature and pressure were held constant for a duration of about 17.6 hours (point C to point D in Figure 2). For this second hold period, once again, no pressure change was observed in the reactor whatsoever, indicating that no reaction took place between points C and D in the experimental run either. Next, the reactor temperature increased to 100 °C whereas the reactor pressure increased slightly to approximately 80.25 bar. Once the reactor temperature and pressure were attained, the reactor was left undisturbed to allow any potential reaction to initiate and continue. Interestingly, soon after the third hold period had begun (point E in Figure 2), it was observed that the pressure inside the reactor gradually started to increase while the system temperature largely remained constant. The pressure increases inside the reactor continued over approximately the next 47.5 hours, when the reaction was effectively stopped by drawing the gas phase out of the reactor to be sampled via Gas Chromatography (GC). An enlarged version of the evolution of the pressure and temperature profiles within the reactor from point E onwards in Run 1 may be seen in Figure 3. As mentioned previously, pressure increase within the system is the first marker of a successful reaction between iron and water to yield magnetite and hydrogen gas. Given that the system is closed, and the temperature of the system largely remains constant, any increase in the system pressure is a clear indicator of hydrogen gas being released inside the system due to the reaction of iron particles with water. In Run 1, the total pressure increase observed inside the system was 2.7 bar.

With regards to the solid product recovered post reaction for Run 1, Figure 4 compares the pure Fe powder that was used as the reactant (Figure 4a) and the dried and ground product obtained at the end of Run 1 (Figure 4b). Simple visual inspection reveals a striking

difference between the color of the reactant and that of the product. While the Fe powder used as the reactant is a light grey in color, the solid product obtained at the end of the experiment is significantly darker and more blackish in color, the same as the known color of Magnetite, one of the products expected in the case of a successful reaction.

5 The blackish product obtained at the end of Run 1 was then characterized with the help of the powder X-Ray Diffraction (p-XRD) technique. For this purpose, the material was first crushed into a fine powder with the help of a mortar and pestle and then p-XRD characterization was performed using a Bruker (D8 Discover model) X-Ray Diffractometer. The diffraction pattern obtained for the recovered solid sample from Run-1
10 is shown in Figure 5. As can be seen in Figure 5, the diffraction pattern indicates the presence of both synthetic grade Iron (Fe) and synthetic grade Magnetite (Fe_3O_4) in the system. The clear signal for synthetic magnetite obtained in the x-ray diffraction pattern of the solid product recovered at the end of Run-1, coupled with the pressure rise observed inside the reactor during the experiment, indicates that the reaction between Fe and water
15 indeed took place at 100 °C temperature and 80 bar pressure, yielding solid magnetite and gaseous hydrogen as products, with no impurities observed.

Experimental Run 2

Equipped with the information that the reaction between iron and water indeed takes place at 100 deg C and 80 bar, Experimental Run 2 was performed at the same target temperature
20 and pressure conditions.

The quantities of the two reactants (Iron particles and deionized water) were kept the same as in Run 1, however, in the case of Run 2, 1 wt% (2 grams) NaCl was added to the system as a potential catalyst.

Figure 6 plots the entire evolution of the temperature and pressure profiles inside the reactor
25 with respect to time (in hours) for Run 2 and shows that the reactor temperature and pressure were first slowly increased in steps up to approximately 80 °C and approximately 80 bar, respectively. Once these temperature and pressure conditions were reached, the first hold period was initiated. As it was established in Run 1 that there is no hydrogen

production at the lower temperature and pressure conditions, such as about 46 °C and about 50 bar, respectively. The methodology for Run 2 was designed such that there would be no significant hold period in the experiment prior to the system temperature and pressure reaching about 80 °C and about 80 bar, respectively. The first hold period for Run 2 thus
5 began around 8.7 hours into the experiment (point A in Figure 6) and lasted for a total of approximately 9.6 hours; the end point for this hold period is denoted by point B in Figure 6. Through the entirety of these 9.6 hours, the reactor temperature and pressure stay largely constant at about 80 °C and about 80 bar respectively, thus indicating that no reaction (production of hydrogen) takes place at these conditions. Next, the temperature of the
10 reactor was set to slowly increase to 100 °C by entering the necessary set point in the heater's controller. This regulates the temperature of the heater and by extension, that of the contents of the reactor. As can be seen in Figure 6, from point B onwards, the internal temperature of the reactor slowly starts to increase up to the desired experimental value. At point C, a sudden breakout in both the temperature and the pressure of the reactor was
15 noticed. The sudden breakout may indicate that an abrupt increase in the system temperature and pressure is a direct consequence of sudden and substantial reaction taking place between the Iron particles and water present inside the system. The pressure increase is caused by the sudden production of a substantial amount of hydrogen gas inside the system whereas the temperature increase is attributed to the reaction being exothermic in
20 nature. The temperature rise occurred for a total of about 12 hours from point C, when it stabilizes at approximately 107.7 °C. The point where the temperature of the reactor first stabilizes after the sudden breakout was marked as point D in Figure 6. The system pressure on the other hand, keeps increasing steadily from point C until the end of the reaction, about 40.5 hours later. Similar to Run 1, the reaction was essentially ended by drawing the gas
25 phase out of the reactor to be sampled via Gas Chromatography (GC). The end of the experiment is signaled by the completion of GC analysis of the gas sample drawn out of the reactor. The final pressure reading made inside the reactor for Run 2 was 89 bar. From point B to point C in Run 2, as the temperature inside the reactor increases, there is also a slight increase in the reactor pressure as a result of expansion of the nitrogen gas contained
30 inside the reactor. At point C, which is where the breakout occurred, the reactor pressure was 83.3 bar, up 2.9 bar from the value recorded at point B, which was 80.4 bar. Thus, as

a result of the reaction between Iron powder and water in Run 2, the total pressure increased by 5.7 bar. This increase in pressure was due to the production of hydrogen gas.

Figure 7a provides an enlarged version of the evolution of the pressure and temperature profiles within the reactor from point C onwards in Run 2.

5 Further as can be seen from Figure 6, between points B and C in Run 2, the reactor temperature increased continually, in a slow but steady manner, towards the desired experimental temperature of 100 °C. At point C, the reactor is still on its path to attaining this desired value. Figure 6 further shows that as the reactor temperature approached the desired experimental temperature, the rate at which the temperature increased within the
10 system became increasingly inhibited. To further embellish this observation, Figure 7b has been presented which depicts the pressure and temperature evolution within the reactor for the 10 hours of the experiment immediately preceding point C. A simple quantification of Figure 7b reveals that the temperature increase in the 10 hours of the experiment immediately preceding point C is a mere 2.77 °C. This indicates that at point C, the system
15 was in fact very close to achieving equilibrium, thus making the standout observation made at point C more intriguing.

The total pressure increase achieved in Run 2 is substantially greater than that achieved in Run 1, 5.7 bar for the former as compared to 2.7 bar for the latter. In percentage terms, the pressure increase achieved in Run 2 is approximately 111% greater than that achieved in
20 Run 1. Further, the total pressure increase of 5.7 bar in Run 2 was achieved over a period of about 40.5 hours whereas the total pressure increases of 2.7 bar in Run 1 took place over a period of approximately 47 hours.

NaCl introduced to the system in Run 2 was intended to act as a catalyst for the reaction and thus the results achieved for Run 2 are along expected lines.

25 In order to confirm the production of H₂ gas resulting from the reaction between Iron (Fe) powder and water, Gas Chromatography (GC) analysis (Agilent A6980N Gas Chromatograph) was conducted on the gas phase present inside the system following the reaction for experimental run number 2 (Run 2). Standard peaks were obtained for pure

hydrogen and pure nitrogen gas samples to identify the retention times for each of these gases and the results were presented in Figures 8a and 8b, respectively. As can be seen from Figure 8, the retention time for hydrogen gas using GC apparatus and GC method (Argon used as the carrier gas) was approximately 2.6 min and that for nitrogen gas using
5 the same apparatus, the same method, and the same carrier gas was approximately 3.05 min.

Figures 9a and 9b present the gas chromatograms obtained for two individual gas samples recovered from inside the reactor post completion of Run 2, i.e., post the reaction between Iron powder and water in Run 2. The GC apparatus, method, and carrier gas employed to
10 analyze the two gas samples from Run 2 were identical to those used to obtain the standard peaks (signals) for pure hydrogen gas and pure nitrogen gas. In both Figures 9a and 9b, two distinct peaks can be observed. If the gas chromatograms are read from left to right, while the first peak appears around the 2.5 min mark for both samples analyzed and matches extremely well with the standard peak obtained for pure hydrogen gas (retention
15 time of ~2.6 min), the second peak appears around the 3.1 min mark for both samples analyzed and matches extremely well with the standard peak obtained for pure nitrogen gas (retention time of ~3.05 min). No other peaks were observed for either of the samples analyzed even after running the GC program for a minimum of 6 min per sample. For sample 1, the GC program was run for a total of 8 min and did not indicate the presence of
20 any other molecules in the gas sample. From Figure 9, it can therefore be determined that the gas phase contained in the system post reaction for Run 2 consisted only of nitrogen gas and hydrogen gas, the former present as it was used to build up the initial pressure within the system, and the latter released because of Iron powder and water reacting with each other within the system. The GC analysis of the Run 2 product gas phase yielding
25 peaks only for hydrogen gas and nitrogen gas establishes that the pressure rise observed inside the system for Runs 1 and 2 is exclusively due to the production of hydrogen gas which is a result of Iron powder and water reacting with each other at elevated temperature and pressure conditions of around 100 °C and around 80 bar, respectively.

Figure 10 presents the wet solid product obtained post reaction for Run 2. A product that is jet black in color can be observed in Figure 10, indicating that the grey Iron powder has undergone reaction with water to yield black magnetite.

5 **Experimental Run 3**

Experimental Run 3 was conducted at a target system temperature of 100 °C and a target system pressure of 100 bar, the latter represents an increase of 25% over the predetermined target system pressure for Runs 1 and 2. The quantities of the two reactants, finely divided Iron (Fe) powder and deionized water, were kept the same as in Runs 1 and 2. Maintaining consistency with Run 2, 1 wt% (2 grams) NaCl was added to the system for Run 3 as a potential catalyst.

Figure 11 shows the entire evolution of the temperature and pressure profiles inside the reactor with respect to time (in hours) for Run 3. The experimental protocol employed for Run 3 did not incorporate any significant hold period prior to achieving the target experimental temperature and pressure within the system. For Run 3, the system temperature and pressure were simply increased in a gradual stepwise manner until the respective values read approximately 100 °C and approximately 100 bar (point A in Figure 11). As may be observed from Figure 11, the pressure inside the system begins to rise as soon as the system reaches the desired experimental pressure and temperature, i.e., as soon as point A is reached in the experiment. This implies that at point A (system temperature of approximately 100 °C and system pressure of about 100.3 bar), the reaction between Iron powder and water to produce hydrogen has begun. Any rise in the system pressure prior to point A can be attributed to nitrogen gas expansion because of the increasing system temperature. From the point when the desired experimental temperature and pressure were reached in the system (point A in Figure 11), Run 3 was allowed to continue for a total of 182.6 hours or 7.6 days. Over the first 48 hours post the system, having reached point A, the pressure increase observed in the system was 3.35 bar, 24.07% greater than that observed in approximately the same time period in Run 1.

An enlarged version of the evolution of the pressure and temperature profiles within the reactor for the first 48 hours in Run 3 (post the attainment of point A in the system) is presented in Figure 12a. Over days 3 to 6 post the system having reached point A, the pressure inside the system increased at an average of $1.07 (\pm 0.20)$ bar per day, indicating steady hydrogen gas production throughout, but no substantial breakout in the reaction. On day 7 post the attainment of point A in the system, the pressure rise achieved in the system was about 2.1 bar, almost twice the average value of the pressure rise recorded over the last 4 days. A close evaluation of the pressure and temperature profiles within the system reveals a somewhat sizeable breakout event that occurred during day 7 of the experiment post the system having reached point A. This breakout event led to the pressure in the system increasing at a greater rate than that observed in the days prior. Further, the breakout event also resulted in a sudden increase in the temperature inside the system. Given that the system temperature was a regulated parameter, the sudden increase in the same leads to the inference that the reaction between Iron and water to produce hydrogen gas is an exothermic one. A similar inference was arrived at in Run 2, where the regulated system temperature exhibited a sudden, substantial increase, coincidence with the reaction breakout event. In Figure 11, the onset of the breakout event that occurred on day 7 post the attainment of point A in the system, is marked as point B. An enlarged version of the evolution of the pressure and temperature profiles within the reactor with respect to time (in hours) from point B onwards in Run 3 is provided in Figure 12b.

The total pressure increase achieved in Run 3 due to the production of hydrogen was 10.3 bar. This was the highest pressure rise observed out of the three experiments conducted which also means that the highest amount of hydrogen produced was in Run 3. The final pressure rise achieved in Run 3 was 80.7% and 281.5% higher than those achieved in Run 1 and Run 2, respectively.

No major breakout event was observed for the reaction in Run 3; this, despite the contents of the reactor being the same as in Run-2, and the employment of a higher initial pressure. Although there was a fairly modest breakout event observed in Run 3, on day 7 post the attainment of the target experimental temperature and pressure in the system, it pales in

comparison to the significantly more prominent and intense breakout event that was observed in Run 2.

Similar to Runs 1 and 2, the Run 3 reaction was ended by simply turning off the heater and drawing out the gas phase present inside the reactor for analysis via the Gas Chromatography (GC) technique. The GC apparatus, method, and carrier gas employed to analyze the Run 3 gas phase product were identical to those used to obtain the standard hydrogen and nitrogen signals (peaks) and to analyze the gaseous product from Run 2. Figures 13a and 13b present the gas chromatographs obtained for two individual gas samples recovered from inside the reactor post completion of Run 3, i.e., post the reaction between Iron powder and water in Run 3. In both Figures 13a and 13b, two distinct peaks can be observed. If the gas chromatograms are read from left to right, while the first peak appears around the 2.5 min mark for both samples analyzed and matches with the standard peak obtained for pure hydrogen gas (retention time of ~2.6 min), the second peak appears around the 3.1 min mark for both samples analyzed and matches with the standard peak obtained for pure nitrogen gas (retention time of ~3.05 min). No other peaks were observed for either of the samples analyzed even after running the GC program for a minimum of 8 min per sample. This implies that the Run 3 product gas phase comprised only two molecules, namely Hydrogen (released as a result of the reaction between Iron and Water), and Nitrogen (used to build up the initial pressure within the system).

Given the constant temperature mode of operation for Run 3, the GC result also shows that the pressure increase observed in the experiment could only have occurred due to the production of hydrogen.

From Figures 10 and 13, it becomes evident that the peak for hydrogen gas is significantly more pronounced in the chromatogram obtained for the Run 3 product gas phase. It can also be stated that the hydrogen gas peak obtained in the chromatogram for the Run 3 product gas phase covers an area that is substantially larger when compared to that covered by its counterpart obtained in the Run 2 chromatogram. The more prominent signal attained and the greater area under the peak discerned in the case of the Run 3 chromatogram

indicates a higher concentration of hydrogen gas in the product gas phase for Run 3, matching well with the higher overall pressure increase observed in the experiment.

Figure 14 illustrates the wet solid product obtained post reaction for Run 3. Similar to the observation made in the case of Run 2, the solid product obtained after the completion of Run 3 was jet black in color, indicating that the grey Iron powder has undergone reaction with water to yield black magnetite.

Component	Reference Numeral
Reactor	1
Condenser	2
H ₂ Receiver/Accumulator	4
Gas/liquid separator	6
Container	8
Pump/Water pump	10
Heater	12
Pressure control valve (PCV)	14
Valve VB	16
N ₂ Supply	18
First predetermined pressure	P1
Second predetermined pressure	P2
First predetermined temperature	T1
Second predetermined temperature	T2
Storage conditions	20

Although the foregoing disclosure has been described in some detail by way of illustration and examples for purposes of clarity of understanding, it is readily apparent to those of ordinary skill in the art in light of the teachings of this disclosure that certain changes and modifications may be made thereto without departing from the spirit or scope of the present disclosure.

Accordingly, the preceding merely illustrates the principles of the disclosure. It will be appreciated that those skilled in the art will be able to devise various arrangements which, although not explicitly described or shown herein, embody the principles of the disclosure and are included within its spirit and scope. Furthermore, all examples and conditional language recited herein are principally intended to aid the reader in understanding the principles of the disclosure and the concepts contributed by the inventors to furthering the art and are to be construed as being without limitation to such specifically recited examples and conditions. Moreover, all statements herein reciting principles, aspects, and embodiments of the disclosure as well as specific examples thereof, are intended to encompass both structural and functional equivalents thereof. Additionally, it is intended that such equivalents include both currently known equivalents and equivalents developed in the future, i.e., any elements developed that perform the same function, regardless of structure. The scope of the present disclosure, therefore, is not intended to be limited to the exemplary embodiments shown and described herein.

WE CLAIM:

1. A process for production of hydrogen by reacting a hydrogen generating substance with water at elevated temperature and pressure conditions.
- 5 2. The process as claimed in claim 1, wherein the elevated temperature is at least about 95-100 °C; and the elevated pressure is at least about 80 bar.
3. The process as claimed in claim 1, wherein the elevated temperature is from about 100 °C (373.2 K) to about 700 °C (973.2 K) and the elevated pressure is from about 80 bar to about 1500 bar.
- 10 4. The process as claimed in any of claims 1 to 3, wherein initial reaction pressure is generated purely by hydraulic means without the introduction of nitrogen gas.
5. The process as claimed in any of claims 1 to 4, wherein one mole hydrogen generating substance reacts with 1.3 moles of water.
- 15 6. The process as claimed in any of claims 1 to 5, wherein the process is optionally carried out in the presence of a catalyst selected from a group comprising salts and acids.
7. The process as claimed in claim 6, wherein the catalyst is present in an amount from about 0.01 wt% (100 ppm) to about 100 wt% (1,000,000 ppm).
8. The process as claimed in any one of claims 1 to 7, wherein the process
20 comprises condensing the water vapor in the resulting gas phase comprising water vapor and hydrogen gas; and separating the condensed water from the gas phase.
9. The process as claimed in any one of claims 1 to 8, wherein the produced H₂ has purity of about 100%.
- 25 10. The process as claimed in any one of claims 1 to 7, wherein the process produces pure hydrogen at high pressure suitable for both efficient storage and transport to market, and direct refueling of hydrogen fuel cell vehicles.
11. The process as claimed in any one of claims 1 to 10, wherein the process further produces pure metallic oxide.
- 30 12. A process for hydrogen production, comprising:

- adding a hydrogen generating substance, water, and optionally a catalyst to a reactor;
 - slowly increasing the temperature of the reactor to a first predetermined temperature of at least about 95-100 °C by activating a heater and regulating its temperature;
 - simultaneously, slowly increasing the pressure inside the reactor to a first predetermined pressure of at least about 80 bar;
 - optionally, introducing nitrogen gas into the reactor via an inlet line connected to the reactor to pressurize the system in certain instances;
 - attaining initial reaction pressure purely by hydraulic means without the use of nitrogen gas, particularly those at the upper limit of the pressure range;
 - utilizing process interlocks on the reactor pressure control system to ensure a smooth transition into the reaction phase;
 - sending the gas phase produced in the reaction to a condenser to condense the water; and
 - sending the condensed water to a gas/liquid separator to separate it from the gas phase.
13. The process as claimed in claim 12, wherein the hydrogen generating substance comprises an alkali metal, an alkaline earth metal, a transition metal, a post transition metal, and any combination thereof.
14. The process as claimed in claim 12, wherein the catalyst is selected from a group comprising salts and acids.
15. The process as claimed in claim 12, wherein the reactor pressure control system comprises one or more sensors, valves, and/or controllers configured to regulate pressure within the reactor during the reaction phase.
16. The process as claimed in any of claims 1 to 15, wherein the reaction takes place in a closed reactor.
17. A system for production of hydrogen, comprising:

at least one reactor containing water, one hydrogen generating substance, and optionally a catalyst; the reactor is defined with at least one inlet and two outlets;

5 a heater enclosing the at least one reactor, wherein the heater is configured to maintain the reactor at a first predetermined temperature;

a condenser connected to the gas phase outlet of the reactor, and configured to a second predetermined temperature for the purpose of condensing the water from the reactor gaseous products;

10 a gas/liquid separator connected to the outlet of the condenser, for the purpose of recovering bulk water from the gas phase product stream;

a pressure control valve connected to the gas phase outlet of the gas / liquid separator and configured to maintain the first pre-determined pressure and thereby provide control of the reaction pressure; and a receiver connected to the outlet of the pressure control valve, for the purpose of storing the product hydrogen.

18. The system as claimed in claim 17, comprises a source of nitrogen gas connected to the reactor for purposes including selective system purging, and pre-pressurisation of the reactor.

19. The system as claimed in claim 17, comprises a pump connected to the reactor for the purposes of selectively supplying feedstock water, and also pre-pressurisation of the reactor.

20. The system as claimed in claim 17, wherein the first predetermined temperature is at least about 95-100 °C and the first predetermined pressure is at least about 80 bar.

21. The system as claimed in claim 17, wherein the second predetermined temperature is in a range from about 30 °C to 100 °C.

22. A system for continuous production of hydrogen, comprising:

- at least one first reactor wherein the process of claims 1 to 16 takes place; a second reactor in re-charge mode for removal of product and installation of fresh reactants; and a third reactor in hot-standby

mode ready for production upon completion of the reaction cycle of the first reactor;

- synchronization of the reactor cycle to ensure that one reactor is in production mode, another in re-charge mode, and another in hot-standby mode at any given time; and
- computer-controlled robotic automation facilitating the continuous product flow operation, with specialized modifications to the reactors including quick opening closures, location of all process and instrument connections on the static component of the reactor, mechanical fixings on the "rotor" component suitable for robotic operation, and limit switches and interlocks to verify closure status of the reactor.

23. The system as claimed in claim 22, wherein the reactors operate under high pressure conditions, and said robotic automation ensures safe operation and transition between different reactor modes.

24. The system as claimed in claim 22, wherein said synchronization of the reactor cycle is achieved through precise control and timing mechanisms managed by the computer-controlled robotic automation.

25. The system as claimed in claim 22, wherein said specialized modifications to the reactors enable efficient and safe operation of the continuous product flow process for hydrogen and metallic oxide production.

26. The system as claimed in claim 22, wherein said robotic automation enables remote operation and monitoring of the reactors, enhancing operational safety and efficiency.

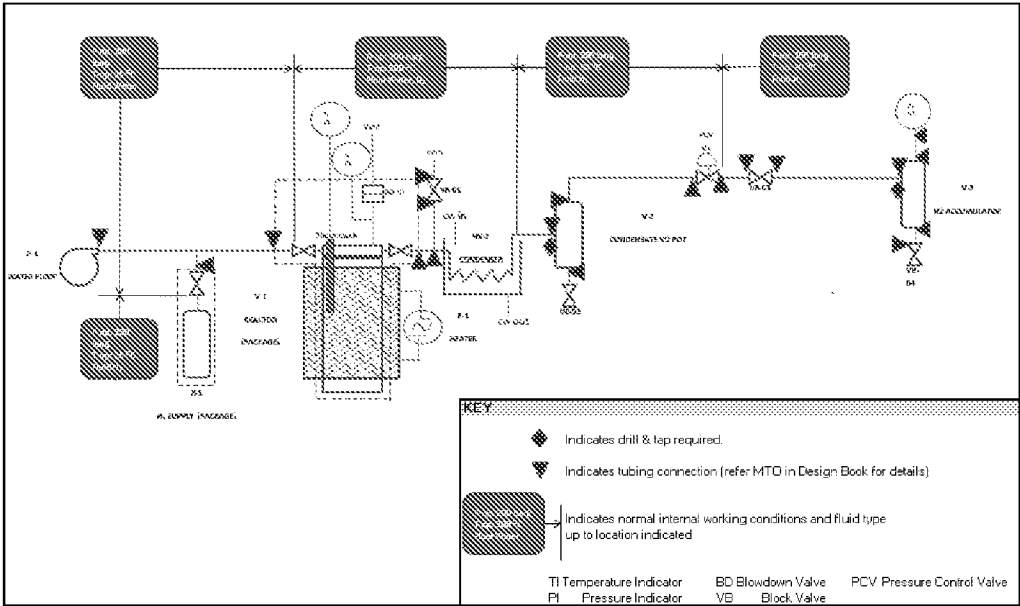


Figure 1

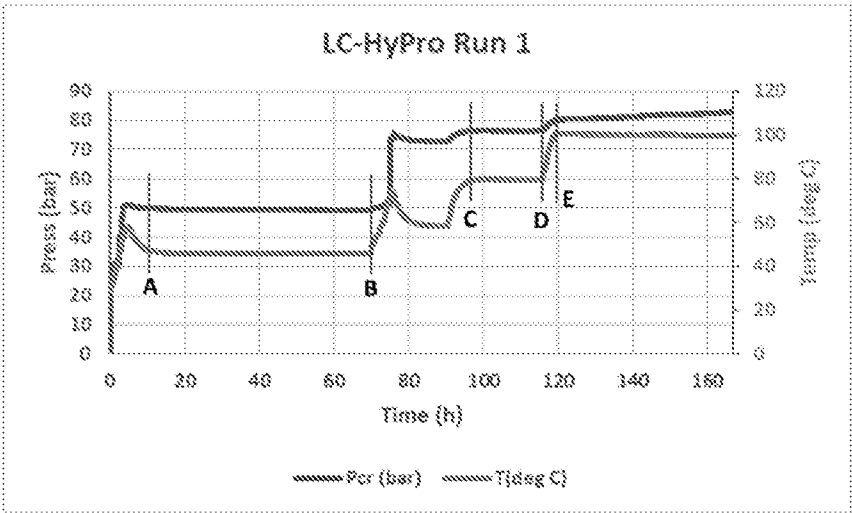


Figure 2

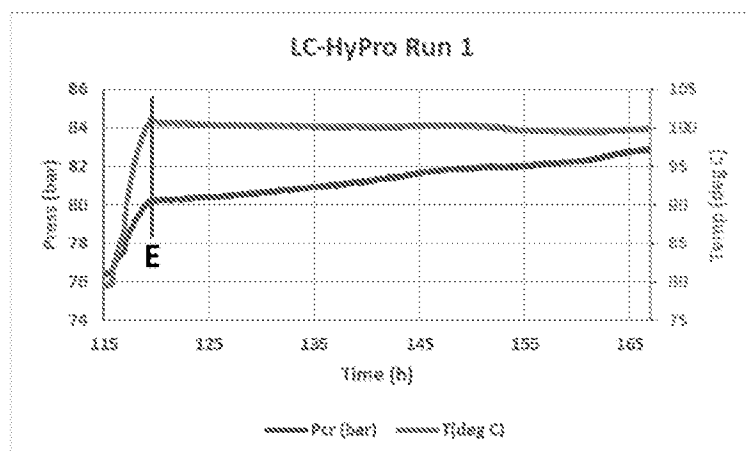


Figure 3

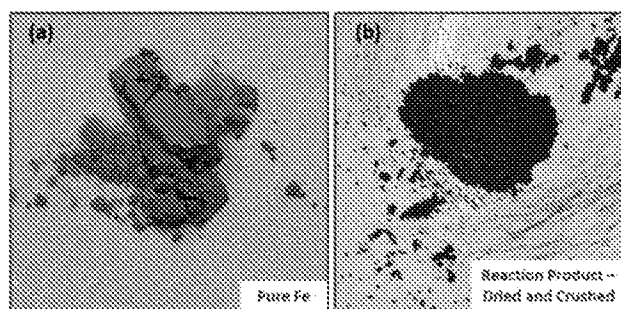


Figure 4

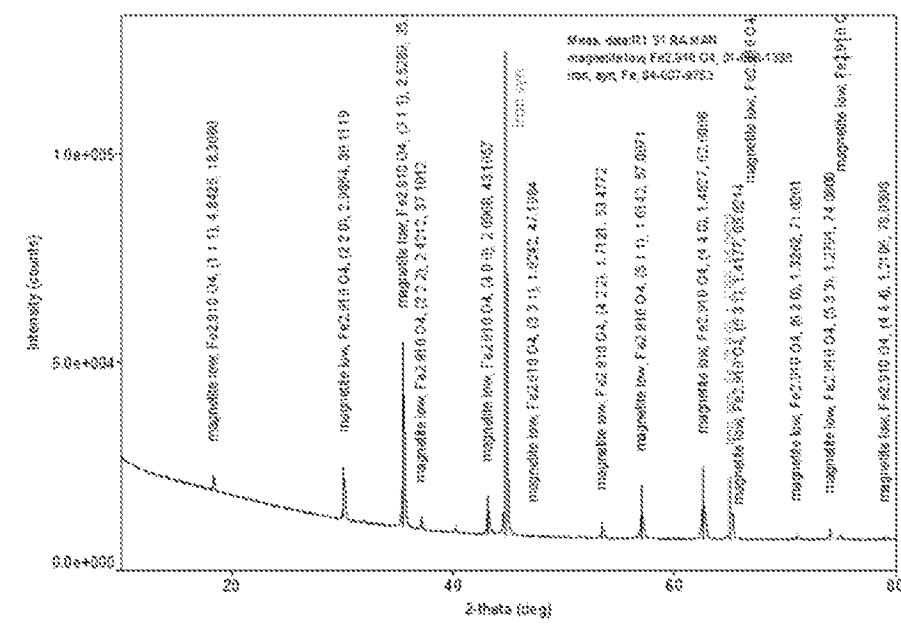


Figure 5

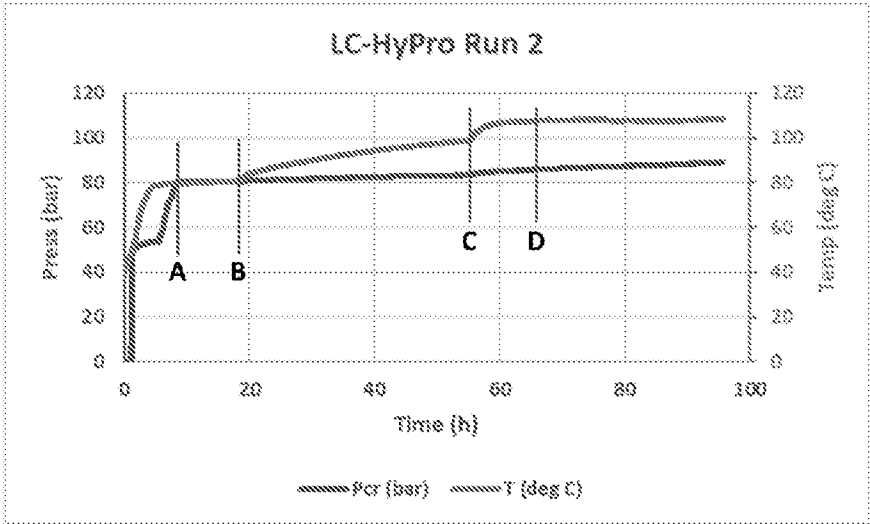


Figure 6

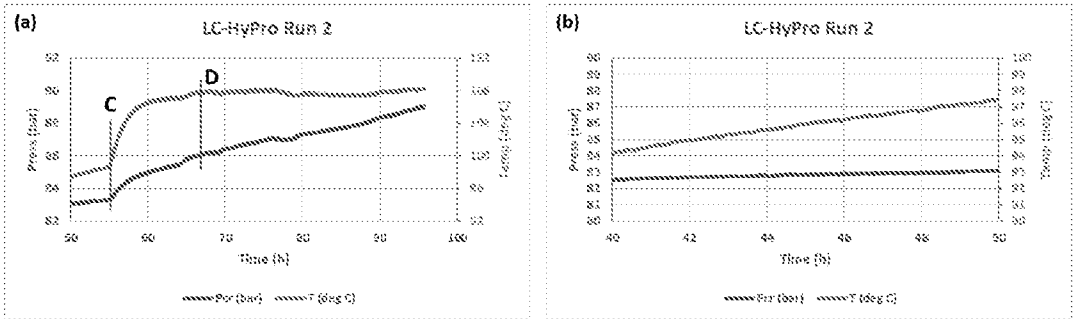


Figure 7

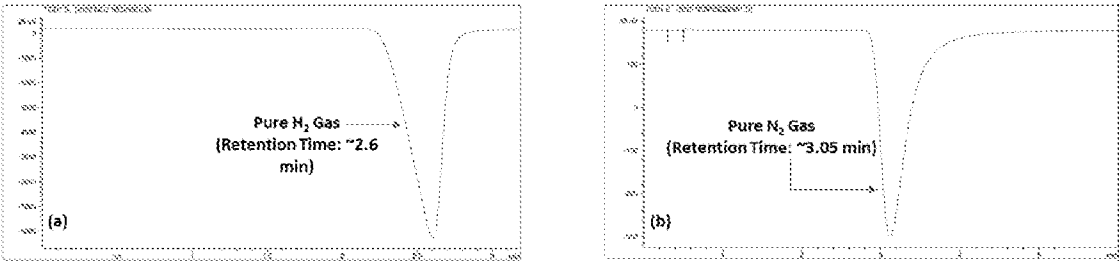


Figure 8

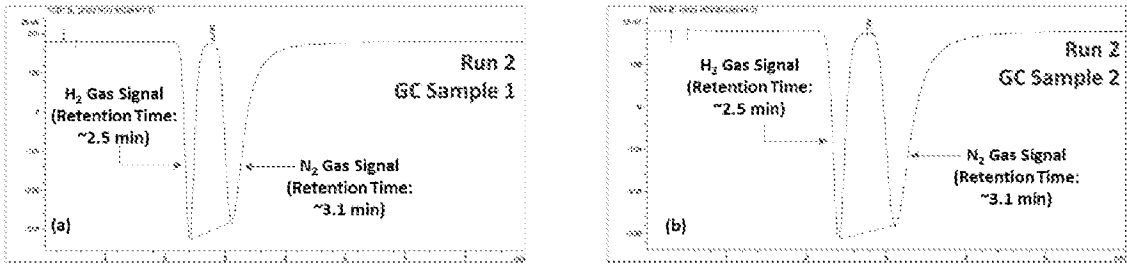


Figure 9



Figure 10

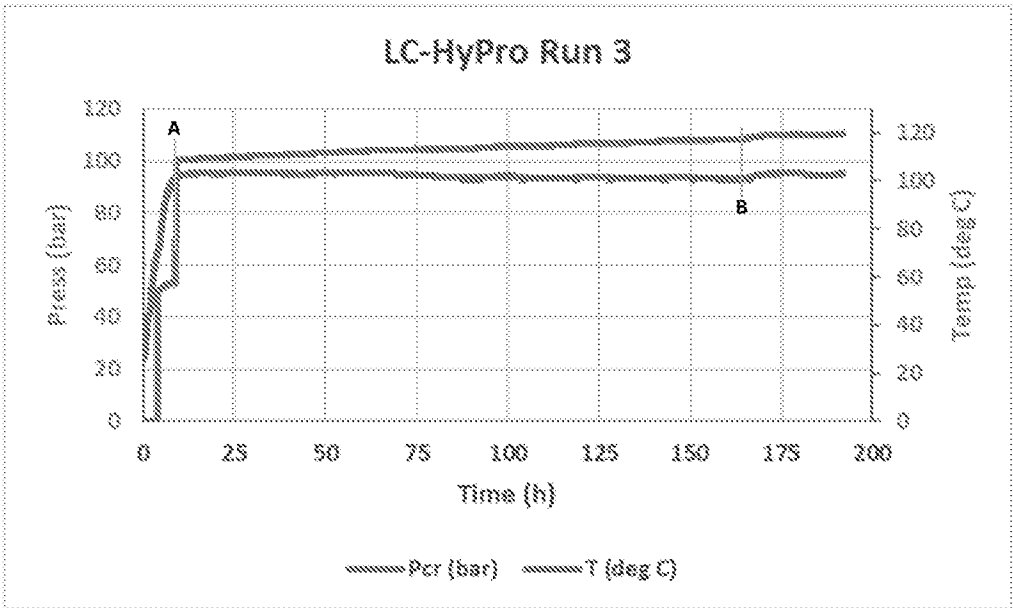


Figure 11

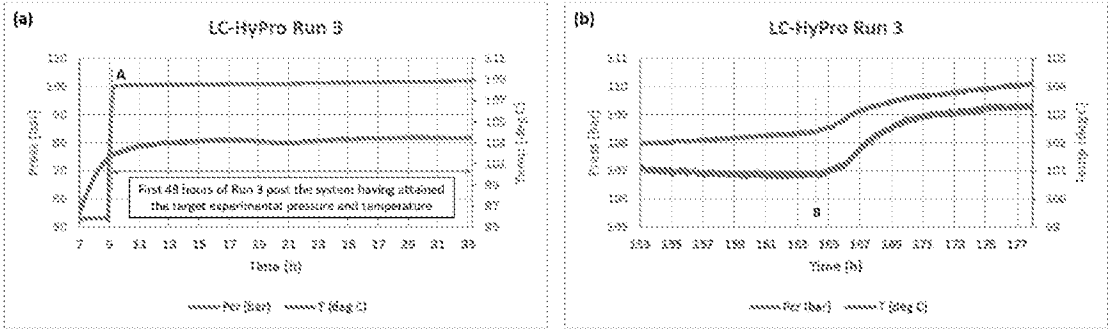


Figure 12

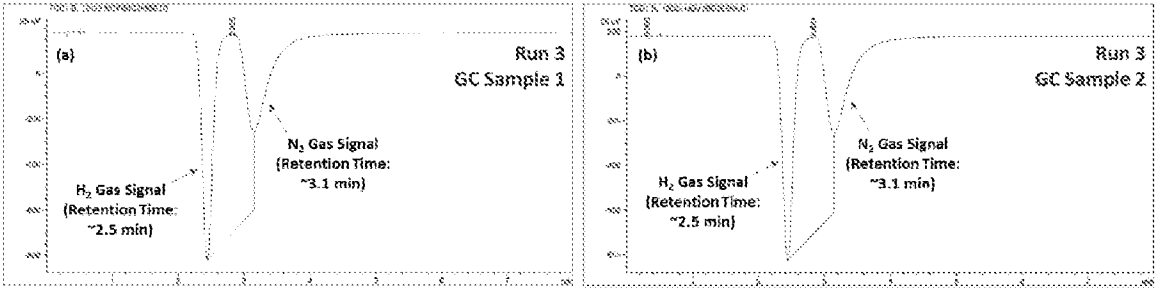


Figure 13

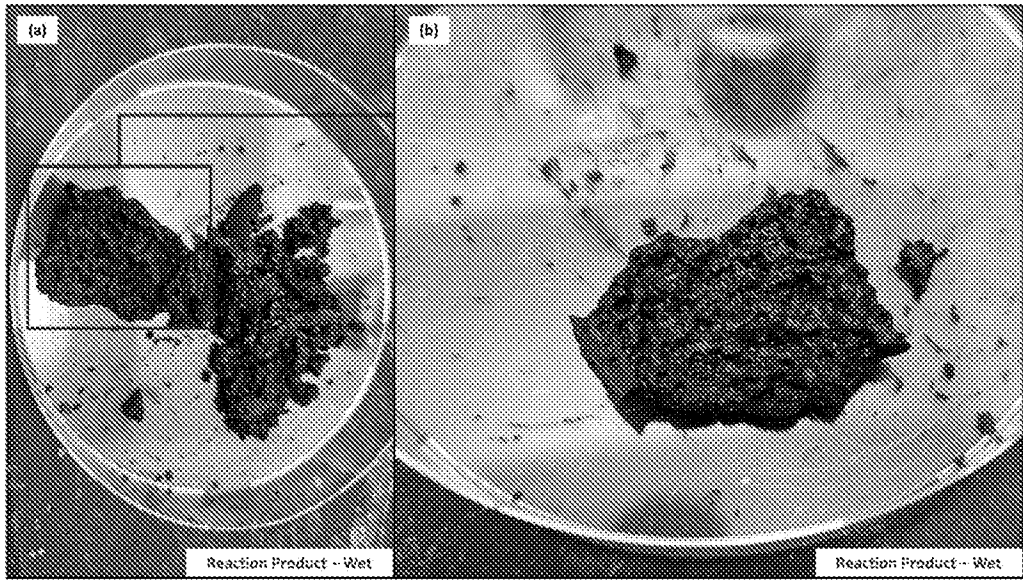


Figure 14

A. CLASSIFICATION OF SUBJECT MATTER

C01B 3/08 (2006.01) B01J 8/00 (2006.01) B01J 27/10 (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)

STNext (CASREACT, CAPLUS, REGISTRY, WPIX): Applicant/Inventor names (BOSHAUS PTY LTD; NATIONAL UNIVERSITY OF SINGAPORE; BHATTACHARJEE, Gaurav; LOANE, Antony William; LINGA, Praveen and related names), CAS numbers (hydrogen, iron, sodium chloride), keywords (hydrogen, pressure and related terms) **EPOQUE 6.30.05 (PATENW):** Applicant/Inventor names (as above), CPC/IPC symbols (C01B2203/1614, C01B2203/1628, C01B3/[08,10], C01B3, Y02E60/30), keywords (hydrogen, iron, water, sodium chloride, other metals and salt, pressure) **Online databases (Espacenet, Google Patents/Search/Scholar):** Applicant/Inventor names (as above), keywords (as above) **Internal databases:** Applicant/Inventor names (as above).

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	



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	"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
"D"	document cited by the applicant in the international application	"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
"E"	earlier application or patent but published on or after the international filing date	"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
"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)	"&" document member of the same patent family
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Date of the actual completion of the international search 30 May 2024	Date of mailing of the international search report 30 May 2024
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaaustralia.gov.au	Authorised officer Joanne Ly AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. +61 2 6283 2498

INTERNATIONAL SEARCH REPORT		International application No.
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