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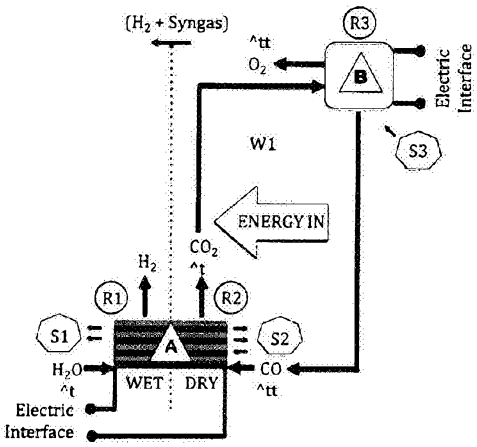
(54) **Hydrogen producing and fuel cell system.**

(57) A method for producing hydrogen, the method comprising the steps of (1) reacting metal with a controlled water supply to produce metal hydroxide, hydrogen gas and heat, the metal being preferably selected from aluminum, iron, copper and zinc or combinations thereof; (2) electrochemically reacting metal hydroxide from step (1) with an oxidizable gas, preferably carbon monoxide, to produce hydrogen, oxidized gas, preferably carbon dioxide, and metal; and (3) reducing the oxidized gas, preferably carbon dioxide, from step (2) to oxidizable gas, preferably carbon monoxide, for recycling to step (2)

Fig. 1



HYDROGEN PRODUCING AND FUEL CELL SYSTEM

Technical field

[0001] The present invention generally relates to a method and system for producing hydrogen.

Background Art

[0002] Ecological attractiveness of using hydrogen as a fuel seems a self-evidence as by burning, it produces water! So, obviously hydrogen should be the cleanest fuel on the planet.

[0003] However, the main drawback of using hydrogen as a fuel remains the problem of supply and the required high pressure storage with all the intermediate risks of flammability and explosion. Furthermore, hydrogen is very expensive to transport in comparison with all other fuels, because of its extreme low density and thus its very low calorific mass. The ecologically attractiveness is thus largely lost by the effort spent (or should we say the energy wasted) to make it safely available to the actual user.

[0004] The only way to make hydrogen both ecologically and economically attractive would be to produce it on demand when and as needed. However, while this may seem feasible for large power plants or the like, no small-sized concept seems to have been disclosed to date, let alone, a system usable in vehicles or the like.

Technical problem

[0005] It is thus an object of the present invention to provide for a method or system for producing hydrogen, electricity and heat locally/on demand, requiring no hydrogen storage. A further object of the invention is to provide an alternative method of producing and storing electrical energy and heat. The methods or systems should be holistic in the sense that it should not be limited to local thermodynamic degradation of extrinsically valuable reagents. Rather, the system should allow for a sustainable use.

General Description of the Invention

[0006] In order to overcome the above-mentioned problem, the present invention proposes, in a first aspect, a method for producing hydrogen and an apparatus arranged for effecting said method, the method comprising the steps of

(1) reacting metal with a controlled water supply to produce metal hydroxide, hydrogen gas and heat, the metal being preferably selected from aluminum, iron, copper and zinc or combinations thereof;

(2) electrochemically reacting metal hydroxide from step (1) with an oxidizable gas, such as preferably carbon monoxide to produce hydrogen, oxidized gas, such as carbon dioxide, and metal; and

(3) reducing the oxidized gas, such as carbon dioxide, from step (2) to oxidizable gas, such as carbon monoxide, for recycling to step (2).

[0007] In a second aspect, the invention proposes a method for producing electrical energy and an apparatus arranged for effecting said method, the method comprising the steps of

(1) reacting metal with a controlled water supply to produce metal hydroxide, hydrogen gas and heat, the metal being preferably selected from aluminum, iron, copper and zinc or combinations thereof;

(2) electrochemically reacting metal hydroxide from step (1) with an oxidizable gas, such as preferably carbon monoxide to produce hydrogen, oxidized gas, such as carbon dioxide, and metal;

(3) reducing the oxidized gas, such as carbon dioxide from step (2) to oxidizable gas, such as carbon monoxide, for recycling to step (2), and

(4) electrochemically reacting hydrogen gas from step (1) with oxygen to produce water and electrical energy.

[0008] Hence, this second aspect relates to a method and corresponding apparatus for using the hydrogen produced by a method or apparatus according to the first aspect, to produce electrical energy by electrochemically reacting hydrogen gas with oxygen.

[0009] In a third aspect, the present invention provides for a method or system for producing or storing electrical energy and heat, wherein the method for producing electrical energy comprises the steps of

(2') electrochemically reacting metal hydroxide with an oxidizable gas, preferably carbon monoxide, to produce electrical energy, heat, oxidized gas, preferably carbon dioxide, and metal; and

(3) reducing the oxidized gas, preferably carbon dioxide, from step (2') to oxidizable gas, preferably carbon monoxide, for recycling to step (2');

and wherein for storing electrical energy and heat, the method comprises the inverse chemical reactions of steps (2) and (3).

[0010] In the context of the present invention, the term "metal" means any appropriate chemical species, i.e. element or compound, susceptible of being oxidized (having their oxidation state increased) while reacting with water, thereby reducing (part of the) hydrogen contained in the water to dihydrogen (hydrogen gas, H_2). Appropriate metals are those able to collect oxygen from water by oxidation, in particular all kind of corrosion-able materials. Hence, the term "metal" includes so-called "base metals" which refers to metals that oxidize or corrode relatively easily, and react variably with dilute hydrochloric acid (HCl) to form hydrogen are particularly preferred. Examples include aluminum, iron, nickel, lead and zinc. In the context of the invention, copper is considered a base metal as it oxidizes relatively easily, although it does not react with HCl. The term "metal" for the purpose of the present invention does also include materials comprising said "base metals", such as minerals or combinations of minerals, e.g. calcite, zeolite, limestone, etc. Particularly preferred are metals selected among aluminum, iron, copper, zinc, etc., or combinations thereof.

[0011] In order to enhance the reaction of water with the metal and thus the production of hydrogen, the exposed surface area of the metal should be as large as possible. Any appropriate method for increasing the exposed surface area may thus be useful in the context of the present invention. In a particularly preferred embodiment, the metal in step (1) is thus in nanoparticulate form. In a still further preferred embodiment, the metal is in the form of nanoparticles comprising

aluminum. The average size of such nanoparticles is generally from 1 to 500 nm, preferably from 5 to 250 nm.

[0012] A main advantage of the present invention is that the hydrogen producing step (1) is does not require high temperatures during operation. In general, step (1) can be effected at temperatures from 5°C to 250°C, preferably from 10°C to 200°C, more preferably from 20°C to 150°C.

[0013] Appropriate reaction conditions of step (1) are known in the art.

[0014] As the metal reacts with water to produce the desired hydrogen, the metal is converted to its hydroxide(s). In the context of the present invention, it is understood that other species of the metal may be produced, such as oxides, without departing from the present invention. Without further action, the metal would be depleted rapidly. Hence, the present invention provides for a metal regeneration step (step(2)), wherein the metal hydroxide is reacted with an oxidizable gas, such as preferably carbon monoxide, in order to reduce the oxidized metal back to a lower oxidation state, preferably to its oxidation state of 0. Other oxidizable gases or mixtures thereof may be used instead of carbon monoxide as exemplified herein, examples being propane or butane.

[0015] This metal regeneration step (2) is effected in the same reactor as step (1), but are effected separately, i.e. said steps (1) and (2) are effected either separated in time or in space, or both separated in time and space. In other words, the method controls the metal regeneration reaction in order to happen in a part of the reactor where there currently is no hydrogen producing reaction (step (1), as called wet step). In the context of the invention, it is clear that step (2) need not to wait until all the metal has been oxidized. On the contrary, as this step requires electrical energy, it is preferably done when such energy is available, such as will be explained in particular below. So, while hydrogen is produced in one part of the step (1) reactor (also called reactor A below), other parts of said reactor may be submitted to the reaction of step (2) in a so-called dry step (in the absence of water) in the presence of carbon monoxide.

[0016] In step (2), the metal hydroxide reacts with carbon monoxide and electrical energy to regenerate the metal, to oxidize carbon monoxide to carbon

dioxide and to produce heat. Hence, after regeneration of the metal, it may be oxidized again in a wet step (1) to produce more hydrogen.

[0017] In a still further preferred embodiment, the reactor of steps (1) and (2) can also act (or be used) as a conventional battery wherein the metal acts as an negative pole. By providing an appropriate material as positive pole, electrical energy can be directly extracted from the so formed battery. Examples are aluminum-carbon or iron-copper batteries. In a specially preferred embodiment, the reactor is configured as an aluminum-carbon battery. Conversely, electrical energy (if available, e.g. from brake recuperation) may be used to directly reload (regenerate) the battery reactor.

[0018] The gaseous carbon dioxide produced in step (2) or (2') is then let to a further step (3) to regenerate carbon monoxide required for step (2). This step (3) may be done by any appropriate method. Preferably however, this step is effected by plasma magnetic conversion, e.g. in a magnetron.

[0019] The reaction in step (3) is strongly endothermic. However, a further advantage of the present invention is that the heat produced elsewhere in the system may be used to drive the reaction in step (3). Hence, in a particularly preferred embodiment, the reactor of step (3) is arranged close to the reactor of step (1) and (2) to reuse the heat generated there.

[0020] With a method or apparatus of the first aspect of the invention, hydrogen can thus be produced on demand (when needed).

[0021] In a second aspect, the method and apparatus is conceived to use said hydrogen to produce electrical energy in a further step (4), preferably within a conventional hydrogen fuel cell.

[0022] In a third aspect, the method and apparatus can be used in the dry pathway (in the absence of water supply) as a battery in the sense that electrical energy can be produced or stored as required using steps (2') and (3) or their reverse chemical reactions.

[0023] It might be important to stress that the methods and apparatuses presented herein are not to be considered as perpetual motion systems. Although the system is able to operate autonomously for extended times, net energy input is

necessary. However, compared to current known alternatives, the amount of energy required is reduced, different sources of energy may be utilized and thus, overall autonomy is increased.

[0024] Advantages of embodiments of the invention as described herein are the following:

[0025] The method and apparatus of the invention preferably work as a closed system in the sense that reagents are regenerated as needed or required. In particular, in a system of the first (hydrogen producing) aspect of the invention, the major constituents of the dry pathway are operated in a loop without releasing from the system. The same is also true for the wet pathway in the second (electricity producing) aspect of the invention, wherein the hydrogen is reacted back to water in step (4), which may then again be used in step (1).

[0026] Electricity may be extracted from the reactor A of steps (1) and (2) (e.g. an aluminum-carbon battery) as from a normal battery, but also by chemical transformation of aluminum with water, producing by the way hydrogen and heat.

[0027] Hydrogen will flow to the fuel cell (C) to produce electricity by reaction with air or oxygen and the heat will be used to heat up the fuel cell and the magnetic plasma diffuser (B) converting CO_2 (carbon dioxide) into CO (carbon monoxide) ready to reload the aluminum battery (A) in presence with external energy coming e.g. from brake recuperation or from the external grid system.

[0028] Hence, a system as described herein may be delivering energy from at least four different parts of the system enhancing drastically the electricity efficiency of this cell. Electricity is extractable from following parts:

- From the fuel cell by hydrogen,
- From the battery of the reactor.

[0029] The whole system can be produced and operate in a cost-effective manner and is pollution free, because it allows recycling (almost) indefinitely the oxidized aluminum in aluminum metal.

[0030] The battery has nearly a lifelong loading capacity as compared to continuous losing of loading capacity by all other battery types.

[0031] For external energy extraction from brakes, frictions, etc., the system needs mainly current (amperes) and no tension (voltage), which means that the present battery A can extract much larger quantities of energy from the outside system and much faster (in milliseconds) without damaging the device. Current known systems can only extract at the very most about 20%. It is expected that extraction quantities with an apparatus according to the invention lies above 50 %, or even up to 80%.

[0032] The loading time from the grid system will be reduced to a few minutes. It is currently believed that a device according to the invention can be reloaded in less than 5 minutes, mainly because there is no internal resistance during loading time.

[0033] The methods and apparatuses of the invention are therefore particularly beneficial for use in vehicles, although other fields and purposes are explicitly envisaged herewith.

Brief Description of the Drawings

[0034] Preferred embodiments of the invention will now be described, by way of example, with reference to the accompanying drawings in which:

Fig. 1 is a schematic diagram of a preferred embodiment of the invention showing a method or apparatus for producing hydrogen;

Fig. 2 is a schematic diagram of a further preferred embodiment of the invention showing a method or apparatus for producing hydrogen and using said hydrogen in a fuel cell to produce electrical energy.

[0035] Further details and advantages of the present invention will be apparent from the following detailed description of several not limiting embodiments with reference to the attached drawings.

Description of Preferred Embodiments

[0036] As can be seen in Fig. 1 and 2, preferred embodiments of systems and methods of the invention as described herein comprise two pathways W1 and W2, wherein pathway W1 is also called dry pathway and pathway W2 is called wet pathway.

[0037] In a first embodiment as illustrated in Fig. 1, the method and system allows for the production of hydrogen gas in reactor A (pathway W2) by electrochemical reaction of water on an appropriate metal being oxidized to a corresponding metal hydroxide. A currently preferred metal is aluminum, but other metals, such as iron, copper, zinc, ..., or combinations thereof may also be used.

[0038] The basic principle of reactor A is to decompose water where oxygen is going to be attracted by the metal, such as aluminum, by oxidizing the metal to its hydroxide and releasing gaseous hydrogen.

[0039] The reactor A in operational mode will produce hydrogen, heat, and electricity by reaction in a plurality of connected cells, said cells being connected in a serial or a parallel connection, or a combination thereof.

[0040] The hydrogen produced by this reaction may be used in any appropriate application preferably directly, but it may be stored for later use if necessary. In another, particularly preferred embodiment, the hydrogen may be supplied directly to a hydrogen fuel cell C (see in particular the description below in relation with Fig. 2).

[0041] In fact, reactor A can be used to extract electricity as from a normal battery, e.g. an aluminum-carbon battery, but also by chemical transformation of aluminum with water, producing by the way hydrogen and heat.

[0042] The chemical reaction in reactor A during the wet pathway W2 in the case of aluminum as metal can be represented as follows:

[0043] $2\ Al + 6\ H_2O \rightarrow 2\ Al(OH)_3 + 3\ H_2 + heat$

[0044] Without any further processing step, all the metal in reactor A would be used up after a certain amount of hydrogen produced.

[0045] Hence, in a method or apparatus according to the invention, the metal is regenerated by converting (reducing) the metal hydroxide back to its metal state in the same reactor A, in the so-called dry pathway W1, i.e. in the absence of water. As this reaction takes place in the same reactor, it is either done when no hydrogen production is needed, or, preferably concomitantly but in spatially/physically separated parts of the reactor A, by electrochemical reaction of the metal hydroxide with carbon monoxide and electric energy.

[0046] The regenerating reaction in reactor A in the case of aluminum can be summarized as follows:

[0047] $Al(OH)_3 + 3\ CO + 3\ e^- \rightarrow Al + 3\ CO_2 + 3\ H^*$

[0048] The regenerated metal, e.g. aluminum, will then be available again for the production of hydrogen gas.

[0049] In a preferred embodiment of a method of the invention, the wet and dry pathways are alternated or controlled depending on the needs in terms of reactant for the hydrogen producing step (wet pathway W2) or the availability of (an excess of) electrical energy (see also below).

[0050] To complete the dry pathway, the method also comprises a further regeneration reaction in a reactor B wherein carbon dioxide is (at least partly) reduced back to carbon monoxide.

[0051] In a particularly preferred variant, the heat from the wet pathway W2 in reactor A can be used to heat up reactor B, preferably a magnetic plasma diffuser, for converting CO_2 (carbon dioxide) in CO (carbon monoxide) ready to regenerate reactor A in the dry pathway with the provision of external energy.

[0052] In a second embodiment as illustrated in Fig. 2, the method and system not only allows for the production of hydrogen gas in reactor A (pathway W2), but also the use of said hydrogen directly within a hydrogen fuel cell to produce electrical energy.

[0053] In fact, most advantageously, the hydrogen can be produced on demand, i.e. only when electrical energy is necessary. Hence, with a method and system as illustrated in Fig. 2, electrical energy can be produced from hydrogen without requiring expensive hydrogen storage, neither locally, nor for transport to the user (hydrogen fuel cell). Hence, in a particularly advantageous embodiment, the method and system may be implemented in a vehicle, the energy required e.g. for regeneration being partly produced by other components of the system itself (especially heat) or will be provided from an external source (electrical grid, plug-in battery, brake energy recuperation). Especially brake energy recuperation is of particular interest as conventional electrical energy storage devices, such as

batteries, can only recover a small fraction of the brake energy without damage and conventional fuel cell systems cannot recover any brake force energy at all.

[0054] Indeed, with a system as described herein the huge energy output provided by brake recuperation of a vehicle (more than 65% of the total energy output) can be used for the regeneration steps. This also means that charging times on an external grid will be extremely rapid, which is actually a further advantage over conventional system, such as batteries.

[0055] The features, components and particularities of preferred variants of the embodiments illustrated in Fig. 1 and 2 will now be described more in detail by way of example.

[0056] Reactor A – aluminum-carbon regenerated battery

[0057] The basic principle of this battery is the dissociation of water in hydrogen gas and oxygen bound to oxidized Al to form in particular aluminum hydroxide ($Al(OH)_3$) at relatively low temperatures from 5°C to 250°C, preferably from 10°C to 200°C, more preferably from 20°C to 150°C.

[0058] The metal, in this case the aluminum, is provided to have a large specific surface area for contact with the reagent, i.e. water. Preferably the metal is provided as nanoparticles having sizes from 1 to 500 nm, preferably from 5 to 250 nm.

[0059] This device in operational mode produces hydrogen, heat and electricity by reaction in a plurality of connected, but physically separated cells, connected in series or in parallel.

[0060] Reactor A can be built from aluminum or similar metal frame with a plurality of cells wherein carbon nanotubes are used a positive pole of battery and aluminum nanoparticles as a negative side of the battery.

[0061] As already mentioned Al nanoparticles can be replaced with other metal or metal containing nanoparticles that are able to collect oxygen from water by oxidation, in particular all kind of corrosion able materials like Fe, Zn, Cu, or other non oxidized material metal or mineral structure.

[0062] Reactor B – magnetic plasma converter – magnetron or similar device able to easily and rapidly transfer heat energy to a gas.

[0063] Reactor C – hydrogen fuel cell

[0064] Step (4) is preferably done in a hydrogen fuel cell. Appropriate types are generally known fuel cells, such as Proton exchange membrane fuel cells (PEMFCs), Phosphoric acid fuel cell (PAFC), High-temperature fuel cells, etc. Preferably, reactor C is a stack of PEM fuel cells (PEMFCs).

[0065] Energy balance

[0066] The theoretical energy balance of a preferred system according to the invention comprising

A: Nano aluminum-graphite hydrogen energy cell

B: Plasma Magnetic CO_2/CO Converter

C: Fuel Cell

can be summarized as follows:

[0067] S1: Electro/Chemical Reaction Generated with H_2O (Heat + Electrical)

[0068] 1,2 m^3 H_2 (Specific 4 kWh), effective 3,3 kWh+ 4 kWh (heat used for system reaction, rest heat used by Peltier cells) + 4 kWh theoretical electrical energy by splitting of H_2O, effective 3,3 kWh; process temperature < 300°C / 1 kg Al

[0069] -> Total of 10,6 kWh energy produced

[0070] S4: Fuel Cell generated (Heat + Electrical):

[0071] Efficiency based on 0,75

[0072] 4 kWh specific energy, 1 kWh (heat) + 3 kWh effective / 100 g H_2 / 1 kg Al

[0073] S2: Reduction of $Al(OH)_3$ with CO and electricity from the brake force

[0074] (x) m^3 (syngas, monoatomic H) + 7 kWh (heat used for system reaction, theoretical) effective 4 kWh, the rest is coming from chemical reaction (S1),

demand of energy from plasma converter 2 kWh at a process temperature < 900°C + (X) kW brake energy

[0075] S3: Energy demand to convert CO_2 back to CO:

[0076] Total energy demand 8,9 kWh / 2,5 m^3 CO / 1 kg Al (Heat + Electrical) + demand of energy from plasma converter 2 kWh

[0077] - Heat is coming from the rest heat of the system reaction.

[0078] - Electrical (x) kWh from external grid to run the magnetron with high output; when CO is needed. Process temperature < 900°C

[0079] Demand of energy to release O_2 from Ca: 4 kWh

[0080] Total:

[0081] R1 +10,6 kWh energy produced from 1 kg of Al

[0082] R2 +4 kWh

[0083] -2 kWh

[0084] R3 -4 kWh

[0085] (R4 -2 kWh for controlling unit (Electronic, pumps, valves, etc.))

[0086] Mean:

[0087] To make 1 kg of Al available = 18,9 kWh Without brake force

[0088] From grid system 2+2+4+2 = 10 kWh Without brake force

[0089] Efficiency Theoretical 6,6 + / 10 x100% = 66 Ex. Pkt.2 heat and brake force.

Legend:

S1	Step 1 - hydrogen production
S2	Step 2 - metal regeneration
S3	Step 3 - carbon monoxide regeneration
S4	Step 4 - electricity production
A	Reactor A (hydrogen production and metal regeneration)
B	Reactor B (Oxidizable gas (carbon monoxide) regeneration)
C	Reactor C (Hydrogen fuel cell)
W1	Dry pathway
W2	Wet pathway
R1	Hydrogen producing reaction
R2	Metal regeneration reaction
R3	Oxidizable gas (carbon monoxide) regeneration reaction
R4	Electrochemical hydrogen reaction
^t, ^tt	Heat, more heat

P-LBA-001/LU

ANSPRÜCHE

1. Verfahren zur Erzeugung von Wasserstoff, elektrischer Energie und Wärme oder Speicherung von elektrischer Energie und Wärme, wobei das Verfahren zur Erzeugung elektrischer Energie folgende Schritte umfasst:
   (2) elektrochemisches Reagierenlassen eines Metallhydroxids mit einem oxidierbaren Gas, vorzugsweise Kohlenmonoxid, zur Erzeugung von elektrischer Energie, Wärme, oxidiertem Gas, vorzugsweise Kohlendioxid, und Metall; und
   (3) Reduzieren des oxidierten Gases, vorzugsweise Kohlendioxid, von Schritt (2) zu einem oxidierbaren Gas, vorzugsweise Kohlenmonoxid, zur Rezyklierung zu Schritt (2);
   und wobei zur Speicherung das Verfahren die umgekehrten chemischen Reaktionen der Schritte (2) und (3) umfasst.

2. Verfahren zur Erzeugung von Wasserstoff, wobei das Verfahren folgende Schritte umfasst:
   (1) Reagierenlassen eines Metalls mit einer geregelten Wasserzufuhr zur Erzeugung von Metallhydroxid, Wasserstoffgas und Wärme, wobei das Metall vorzugsweise aus Aluminium, Eisen, Kupfer und Zink oder Kombinationen davon ausgewählt ist;
   (2) elektrochemisches Reagierenlassen des Metallhydroxids von Schritt (1) mit einem oxidierbaren Gas, vorzugsweise Kohlenmonoxid, zur Erzeugung von Wasserstoff, oxidiertem Gas, vorzugsweise Kohlendioxid, und Metall; und
   (3) Reduzieren des oxidierten Gases, vorzugsweise Kohlendioxid, von Schritt (2) zu einem oxidierbaren Gas, vorzugsweise Kohlenmonoxid, zur Rezyklierung zu Schritt (2).

3. Verfahren zur Erzeugung elektrischer Energie, wobei das Verfahren folgende Schritte umfasst:

(1) Reagierenlassen eines Metalls mit einer geregelten Wasserzufuhr zur Erzeugung von Metallhydroxid, Wasserstoffgas und Wärme, wobei das Metall vorzugsweise aus Aluminium, Eisen, Kupfer und Zink oder Kombinationen davon ausgewählt ist;

(2) elektrochemisches Reagierenlassen des Metallhydroxids von Schritt (1) mit einem oxidierbaren Gas, vorzugsweise Kohlenmonoxid, zur Erzeugung von Wasserstoff, oxidiertem Gas, vorzugsweise Kohlendioxid, und Metall;

(3) Reduzieren des oxidierten Gases, vorzugsweise Kohlendioxid, von Schritt (2) zu einem oxidierbaren Gas, vorzugsweise Kohlenmonoxid, zur Rezyklierung zu Schritt (2); und

(4) elektrochemisches Reagierenlassen des Wasserstoffgases von Schritt (1) mit Sauerstoff zur Erzeugung von Wasser und elektrischer Energie.

4. Verfahren nach Anspruch 1, 2 oder 3, wobei das Metall in Nanopartikelform, vorzugsweise Aluminium umfassenden Nanopartikel, vorliegt.

5. Verfahren nach Anspruch 4, wobei die durchschnittliche Größe der Nanopartikel 1 bis 500 nm, vorzugsweise 5 bis 250 nm, beträgt.

6. Verfahren nach irgendeinem der Ansprüche 2 bis 5, wobei Schritt (1) bei Temperaturen von 5 °C bis 250 °C, vorzugsweise von 10 °C bis 200 °C, bevorzugter von 20 °C bis 150 °C, durchgeführt wird.

7. Verfahren nach irgendeinem der Ansprüche 2 bis 6, wobei Schritt (2) in einem selben Reaktor wie Schritt (1) durchgeführt wird, wobei die Schritte (1) und (2) zeitlich und/oder räumlich getrennt durchgeführt werden.

8. Verfahren nach irgendeinem der Ansprüche 1 bis 7, wobei Schritt (3) durch plasmamagnetische Umwandlung durchgeführt wird.

9. Vorrichtung, die für die Durchführung des Verfahrens nach irgendeinem der Ansprüche 1 bis 7 angeordnet ist.

Fig. 1

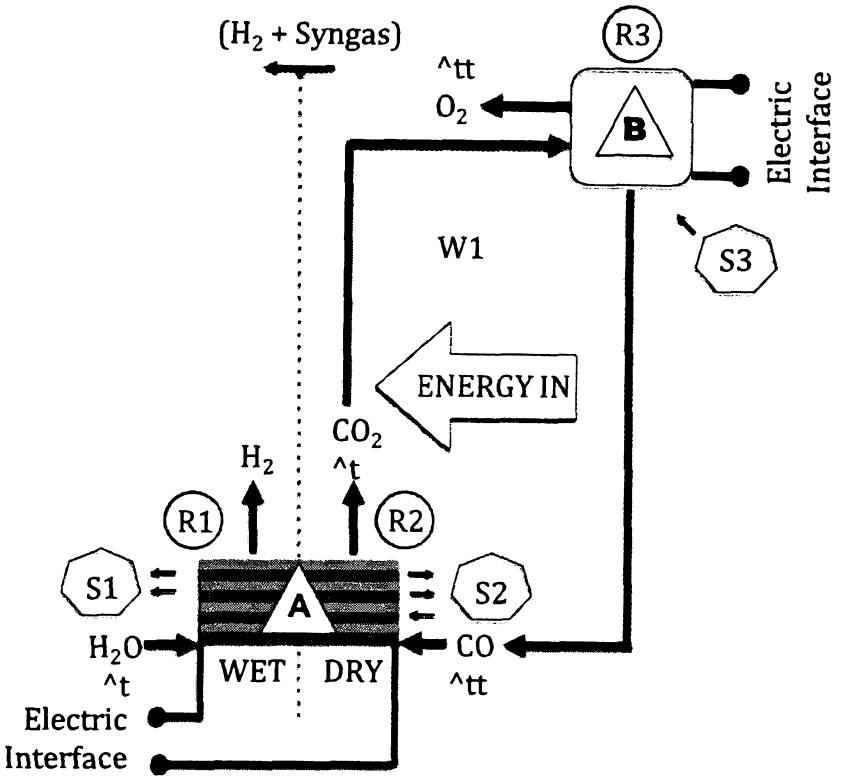
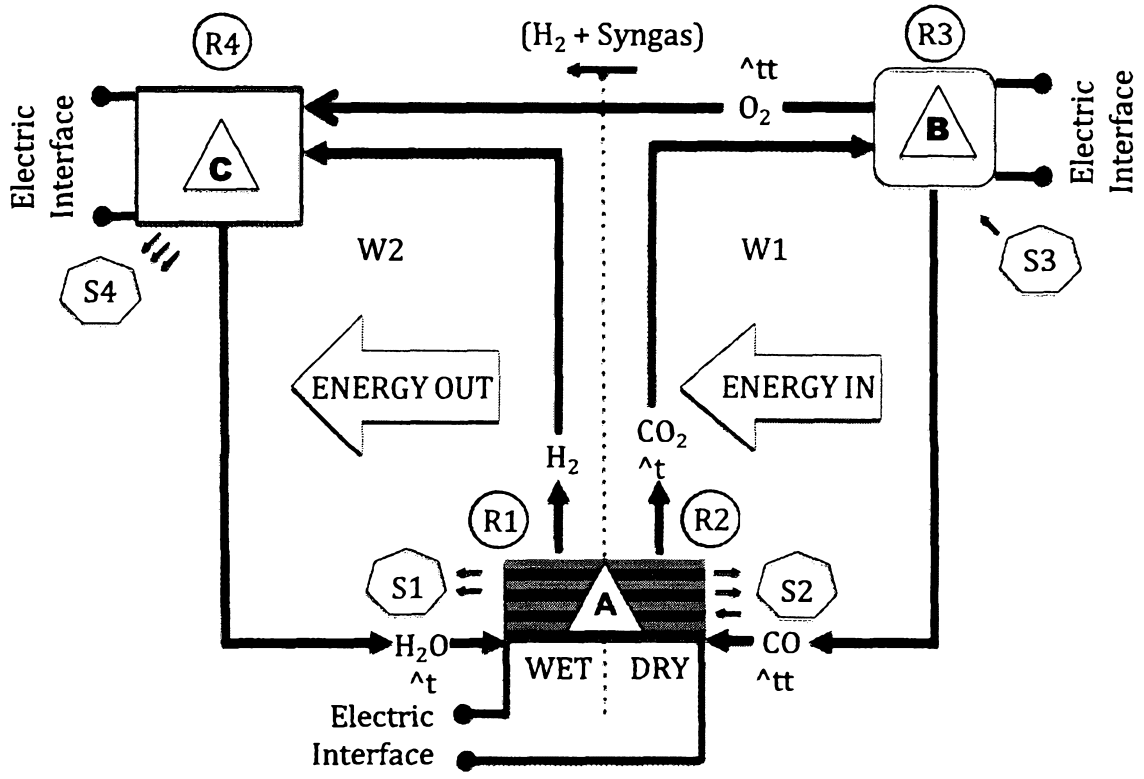

(H2 + Syngas)
^tt
O2
R3
B
Electric Interface
S3
W1
ENERGY IN
CO2
^t
H2
R1
R2
S1
A
S2
H2O
^t
WET
DRY
CO
^tt
Electric Interface

Fig. 2


R4
(H2 + Syngas)
^tt
O2
R3
Electric Interface
C
B
Electric Interface
S4
W2
W1
S3
ENERGY OUT
ENERGY IN
CO2
^t
H2
R1
R2
S1
A
S2
H2O
^t
WET
DRY
CO
^tt
Electric Interface

Abstract

A method for producing hydrogen, the method comprising the steps of (1) reacting metal with a controlled water supply to produce metal hydroxide, hydrogen gas and heat, the metal being preferably selected from aluminum, iron, copper and zinc or combinations thereof; (2) electrochemically reacting metal hydroxide from step (1) with an oxidizable gas, preferably carbon monoxide, to produce hydrogen, oxidized gas, preferably carbon dioxide, and metal; and (3) reducing the oxidized gas, preferably carbon dioxide, from step (2) to oxidizable gas, preferably carbon monoxide, for recycling to step (2).

(Fig. 1)

LE GOUVERNEMENT
DU GRAND-DUCHÉ DE LUXEMBOURG
Ministère de l'Économie
et du Commerce extérieur

National Application Number

LO 1211
LU 92860

SEARCH REPORT

in accordance with Article 35.1 a)
of the Luxembourg law on patents
dated 20 July 1992

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
	The nature of the alleged invention, insofar as it would seem to emerge from the current claims and from the description read in conjunction with the drawings, is so confusing that no meaningful search on the present subject-matter can be carried out. The reasons are as follows: 1. Independent claim 1 1.1 Claim 1 relates to a method for producing hydrogen , electrical energy and heat, wherein the method comprises the step (2) of producing electrical energy, heat, carbon dioxide, and metal; and the step (3) of reducing carbon dioxide. The method of claim 1 is totally unclear because the claimed method is directed to the production of hydrogen but none of the method steps (2) and (3) involves the production of hydrogen. Therefore, it is unclear how the hydrogen is produced. 1.2 Furthermore, the method step (2) lacks clarity. The step (2) is defined by a desired result, namely electrochemically reacting metal hydroxide with carbon monoxide to produce electrical energy, heat, carbon dioxide, and metal. It is unclear what is meant by "electrochemically reacting metal hydroxide with carbon monoxide". The description merely teaches to use a so called "aluminium-carbon battery" for carrying out the step (2) without providing support and disclosure for such method step. It is absolutely not credible that a reaction of metal hydroxide with carbon monoxide produces electrical energy. Moreover, there is no common -/--		INV. C25B1/02 H01M14/00 C01B3/08 ADD. H01M8/0656 H01M8/18 H01M12/08

TECHNICAL FIELDS SEARCHED (IPC)
C25B H01M C01B

The present search report has been drawn up for all claims

	Date of completion of the search	Examiner
	7 April 2016	Perednis, Dainius

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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A : technological background
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2

EPO FORM 1503 03.82 (P04C55)

LE GOUVERNEMENT
DU GRAND-DUCHÉ DE LUXEMBOURG
Ministère de l'Économie
et du Commerce extérieur

SEARCH REPORT

in accordance with Article 35.1 a)
of the Luxembourg law on patents
dated 20 July 1992

National Application Number

LO 1211
LU 92860

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
	general knowledge of "aluminium-carbon batteries" available to the person skilled in the art. 1.3 The description also does not provide support and disclosure for the method step (3). Allegedly, carbon dioxide is reduced using a so called "plasma magnetic converter". However, no common general knowledge of an apparatus called the "plasma magnetic converter" is available to the person skilled in the art. 1.4 For the above mentioned reasons the claim 1 lacks clarity to such an extent that a meaningful search of the whole claimed subject-matter of the claim 1 cannot be carried out. 2. Independent claims 2 and 3 2.1 The method step (1) in claims 2 and 3 lacks clarity. The step 1 is directed to reacting a metal selected from aluminium, iron, copper and zinc with water to produce metal hydroxide, hydrogen gas and heat. However, such reaction as described in claims and description is not possible. It is well known that although the reaction of step (1) is thermodynamically favorable, it does not proceed due to the presence of oxide layer which forms on the surface of metal (for example aluminium) particles. The oxide layer prevents water from coming into direct contact with the metal and consequently stops the reaction. Therefore, it is unclear how metal hydroxide, hydrogen gas and heat are produced in step (1). 2.2 Furthermore, the method step (2) in claims 2 and 3 lacks clarity. The step (2) is defined by a desired result, namely -/--		TECHNICAL FIELDS SEARCHED (IPC)

The present search report has been drawn up for all claims

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The present search report has been drawn up for all claims

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TECHNICAL FIELDS SEARCHED (IPC)

C25B
H01M
C01B

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The present search report has been drawn up for all claims

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EPO FORM 1503 03.82 (P04C55)

LE GOUVERNEMENT
DU GRAND-DUCHÉ DE LUXEMBOURG
Ministère de l'Économie
et du Commerce extérieur

WRITTEN OPINION

File No.	Filing date *(day/month/year)*	Priority date *(day/month/year)*	Application No.
LO1211	27.10.2015		LU92860

International Patent Classification (IPC)
INV. C25B1/02 H01M14/00 C01B3/08 ADD. H01M8/0656 H01M8/18 H01M12/08

Applicant
LB Association

This report contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☒ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☐ Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the application
- ☐ Box No. VIII Certain observations on the application

Examiner
Perednis, Dainius

LE GOUVERNEMENT
DU GRAND-DUCHÉ DE LUXEMBOURG
Ministère de l'Économie
et du Commerce extérieur

WRITTEN OPINION

File No.	Filing date *(day/month/year)*	Priority date *(day/month/year)*	Application No.
LO1211	27.10.2015		LU92860

International Patent Classification (IPC)
INV. C25B1/02 H01M14/00 C01B3/08 ADD. H01M8/0656 H01M8/18 H01M12/08

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- ☐ Box No. VII Certain defects in the application
- ☐ Box No. VIII Certain observations on the application

Examiner
Perednis, Dainius

Application No.
LU92860

WRITTEN OPINION

Box No. I Basis of the opinion

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.

2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application and necessary to the claimed invention, this opinion has been established on the basis of:

   a. type of material:

   ☐ a sequence listing

   ☐ table(s) related to the sequence listing

   b. format of material:

   ☐ on paper

   ☐ in electronic form

   c. time of filing/furnishing:

   ☐ contained in the application as filed.

   ☐ filed together with the application in electronic form.

   ☐ furnished subsequently.

3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

4. Additional comments:

WRITTEN OPINION

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step, or to be industrially applicable have not been examined in respect of

☒ the entire application

☐ claims Nos.

because:

☐ the said application, or the said claims Nos. relate to the following subject matter which does not require a search *(specify)*:

☐ the claims, the description or the drawings or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*:

☒ no search report has been established for the whole application or for said claims Nos.

☐ a meaningful opinion could not be formed as the sequence listing was either not available, or was not furnished in the international format (WIPO ST25).

☐ a meaningful opinion could not be formed without the tables related to the sequence listings; or such tables were not available in electronic form compliant with he international norm (WIPO ST.25).

☒ See Supplemental Box for further details

Application No.

WRITTEN OPINION

LU92860

Box No. I Basis of the opinion

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.

2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application and necessary to the claimed invention, this opinion has been established on the basis of:

   a. type of material:

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Application No.
LU92860

WRITTEN OPINION

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step, or to be industrially applicable have not been examined in respect of

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☐ claims Nos.

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☐ the claims, the description or the drawings or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*:

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☒ See Supplemental Box for further details

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The nature of the alleged invention, insofar as it would seem to emerge from the current claims and from the description read in conjunction with the drawings, is so confusing that no meaningful search on the present subject-matter can be carried out. The reasons are as follows:

1. Independent claim 1

1.1 Claim 1 relates to a method for producing hydrogen, electrical energy and heat, wherein the method comprises the step (2) of producing electrical energy, heat, carbon dioxide, and metal; and the step (3) of reducing carbon dioxide. The method of claim 1 is totally unclear because the claimed method is directed to the production of hydrogen but none of the method steps (2) and (3) involves the production of hydrogen. Therefore, it is unclear how the hydrogen is produced.

1.2 Furthermore, the method step (2) lacks clarity. The step (2) is defined by a desired result, namely electrochemically reacting metal hydroxide with carbon monoxide to produce electrical energy, heat, carbon dioxide, and metal. It is unclear what is meant by "electrochemically reacting metal hydroxide with carbon monoxide". The description merely teaches to use a so called "aluminium-carbon battery" for carrying out the step (2) without providing support and disclosure for such method step. It is absolutely not credible that a reaction of metal hydroxide with carbon monoxide produces electrical energy. Moreover, there is no common general knowledge of "aluminium-carbon batteries" available to the person skilled in the art.

1.3 The description also does not provide support and disclosure for the method step (3). Allegedly, carbon dioxide is reduced using a so called "plasma magnetic converter". However, no common general knowledge of an apparatus called the "plasma magnetic converter" is available to the person skilled in the art.

1.4 For the above mentioned reasons the claim 1 lacks clarity to such an extent that a meaningful search of the whole claimed subject-matter of the claim 1 cannot be carried out.

2. Independent claims 2 and 3

2.1 The method step (1) in claims 2 and 3 lacks clarity. The step 1 is directed to reacting a metal selected from aluminium, iron, copper and zinc with water to produce metal hydroxide, hydrogen gas and heat. However, such reaction as described in claims and description is not possible. It is well known that although the reaction of step (1) is thermodynamically favorable, it does not proceed due to the presence of

oxide layer which forms on the surface of metal (for example aluminium) particles. The oxide layer prevents water from coming into direct contact with the metal and consequently stops the reaction. Therefore, it is unclear how metal hydroxide, hydrogen gas and heat are produced in step (1).

2.2 Furthermore, the method step (2) in claims 2 and 3 lacks clarity. The step (2) is defined by a desired result, namely electrochemically reacting metal hydroxide with carbon monoxide to produce hydrogen, carbon dioxide, and metal. It is unclear what is meant by "electrochemically reacting metal hydroxide with carbon monoxide". The description merely teaches to use a so called "aluminium-carbon battery" for carrying out the step (2) without providing support and disclosure for such method step. It is absolutely unclear how a reaction of metal hydroxide with carbon monoxide can produce hydrogen, carbon dioxide, and metal. Moreover, there is no common general knowledge of "aluminium-carbon batteries" available to the person skilled in the art.

2.3 The method step (3) in claims 2 and 3 lacks clarity for reasons mentioned above in the chapter 1.3.

2.4 Consequently also claims 2 and 3 lack clarity to such an extent that a meaningful search of the whole claimed subject-matter of the claims 2 and 3 cannot be carried out.

3. Dependent claims 3-9

Moreover, none of the dependent claims 3-9 is directed to subject-matter for which a meaningful search is practicable since these claims do not contain any features which would contribute to the clarification of the independent method claims 1-3.

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The nature of the alleged invention, insofar as it would seem to emerge from the current claims and from the description read in conjunction with the drawings, is so confusing that no meaningful search on the present subject-matter can be carried out. The reasons are as follows:

1. Independent claim 1

1.1 Claim 1 relates to a method for producing hydrogen, electrical energy and heat, wherein the method comprises the step (2) of producing electrical energy, heat, carbon dioxide, and metal; and the step (3) of reducing carbon dioxide. The method of claim 1 is totally unclear because the claimed method is directed to the production of hydrogen but none of the method steps (2) and (3) involves the production of hydrogen. Therefore, it is unclear how the hydrogen is produced.

1.2 Furthermore, the method step (2) lacks clarity. The step (2) is defined by a desired result, namely electrochemically reacting metal hydroxide with carbon monoxide to produce electrical energy, heat, carbon dioxide, and metal. It is unclear what is meant by "electrochemically reacting metal hydroxide with carbon monoxide". The description merely teaches to use a so called "aluminium-carbon battery" for carrying out the step (2) without providing support and disclosure for such method step. It is absolutely not credible that a reaction of metal hydroxide with carbon monoxide produces electrical energy. Moreover, there is no common general knowledge of "aluminium-carbon batteries" available to the person skilled in the art.

1.3 The description also does not provide support and disclosure for the method step (3). Allegedly, carbon dioxide is reduced using a so called "plasma magnetic converter". However, no common general knowledge of an apparatus called the "plasma magnetic converter" is available to the person skilled in the art.

1.4 For the above mentioned reasons the claim 1 lacks clarity to such an extent that a meaningful search of the whole claimed subject-matter of the claim 1 cannot be carried out.

2. Independent claims 2 and 3

2.1 The method step (1) in claims 2 and 3 lacks clarity. The step 1 is directed to reacting a metal selected from aluminium, iron, copper and zinc with water to produce metal hydroxide, hydrogen gas and heat. However, such reaction as described in claims and description is not possible. It is well known that although the reaction of step (1) is thermodynamically favorable, it does not proceed due to the presence of

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2.3 The method step (3) in claims 2 and 3 lacks clarity for reasons mentioned above in the chapter 1.3.

2.4 Consequently also claims 2 and 3 lack clarity to such an extent that a meaningful search of the whole claimed subject-matter of the claims 2 and 3 cannot be carried out.

3. Dependent claims 3-9

Moreover, none of the dependent claims 3-9 is directed to subject-matter for which a meaningful search is practicable since these claims do not contain any features which would contribute to the clarification of the independent method claims 1-3.