US 20080245672A1

(19) **United States**
(12) **Patent Application Publication**
Little et al.
(10) **Pub. No.: US 2008/0245672 A1**
(43) **Pub. Date: Oct. 9, 2008**

(54) ELECTROCHEMICAL METHODS TO GENERATE HYDROGEN AND SEQUESTER CARBON DIOXIDE

(75) Inventors: **C. Deane Little**, Boulder, CO (US); **Timothy C. Heffernan**, Indianapolis, IN (US); **Joseph V. Kosmoski**, Wildomar, CA (US); **C. Gordon Little**, Boulder, CO (US)

Correspondence Address:
FAEGRE & BENSON LLP
PATENT DOCKETING
2200 WELLS FARGO CENTER, 90 SOUTH SEVENTH STREET
MINNEAPOLIS, MN 55402-3901 (US)

(73) Assignee: **NEW SKY ENERGY, INC.**, Boulder, CO (US)

(21) Appl. No.: **12/062,269**

(22) Filed: **Apr. 3, 2008**

Related U.S. Application Data

(60) Provisional application No. 60/921,598, filed on Apr. 3, 2007.

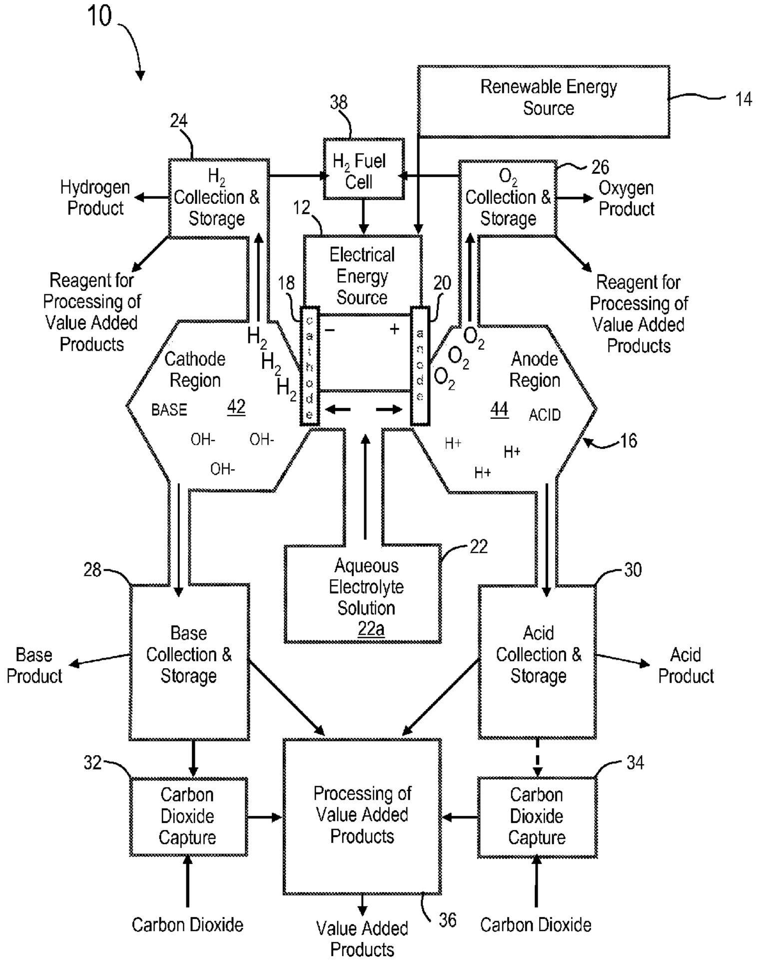
Publication Classification

(51) **Int. Cl.**
C25B 1/04 (2006.01)
C25B 1/00 (2006.01)

(52) **U.S. Cl.** **205/555**; 205/343

(57) ABSTRACT

A carbon dioxide negative method of manufacturing renewable hydrogen and trapping carbon dioxide from the air or gas streams is described. Direct current renewable electricity is provided to a water electrolysis apparatus with sufficient voltage to generate hydrogen and hydroxide ions at the cathode, and protons and oxygen at the anode. These products are separated and sequestered and the base is used to trap carbon dioxide from the air or gas streams as bicarbonate or carbonate salts. These carbonate salts, hydrogen, and trapped carbon dioxide in turn can be combined in a variety of chemical and electrochemical processes to create valuable carbon-based materials made from atmospheric carbon dioxide. The net effect of all processes is the generation of renewable hydrogen from water and a reduction of carbon dioxide in the atmosphere or in gas destined to enter the atmosphere.



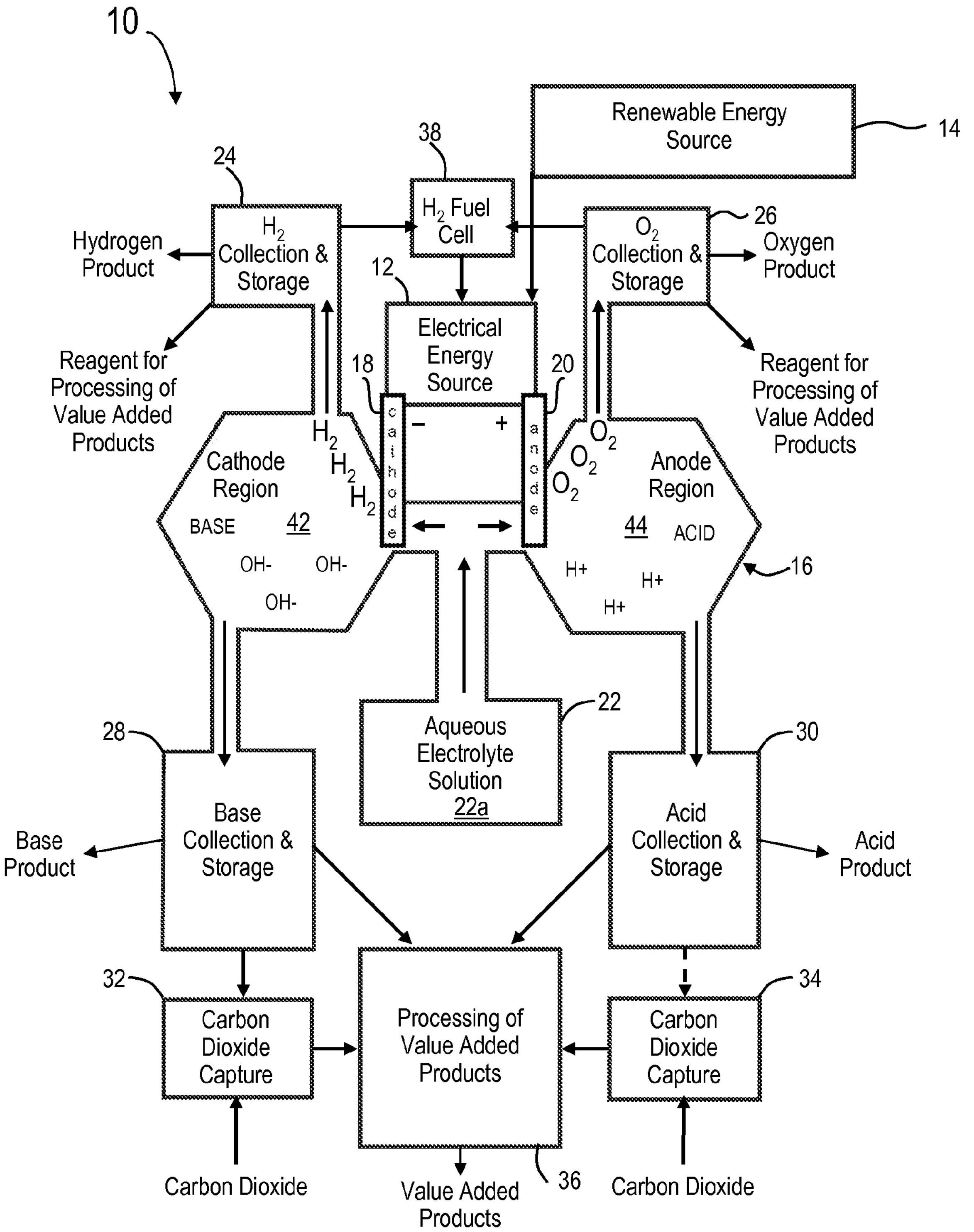

10
Renewable Energy Source
14
38
H2 Fuel Cell
24
H2 Collection & Storage
Hydrogen Product
26
O2 Collection & Storage
Oxygen Product
12
Electrical Energy Source
Reagent for Processing of Value Added Products
Reagent for Processing of Value Added Products
18
20
cathode
anode
−
+
H2
H2
H2
O2
O2
O2
Cathode Region
BASE
42
OH-
OH-
OH-
Anode Region
44
ACID
H+
H+
H+
16
22
Aqueous Electrolyte Solution
22a
28
Base Collection & Storage
Base Product
30
Acid Collection & Storage
Acid Product
32
Carbon Dioxide Capture
Carbon Dioxide
34
Carbon Dioxide Capture
Carbon Dioxide
Processing of Value Added Products
36
Value Added Products

FIG. 1

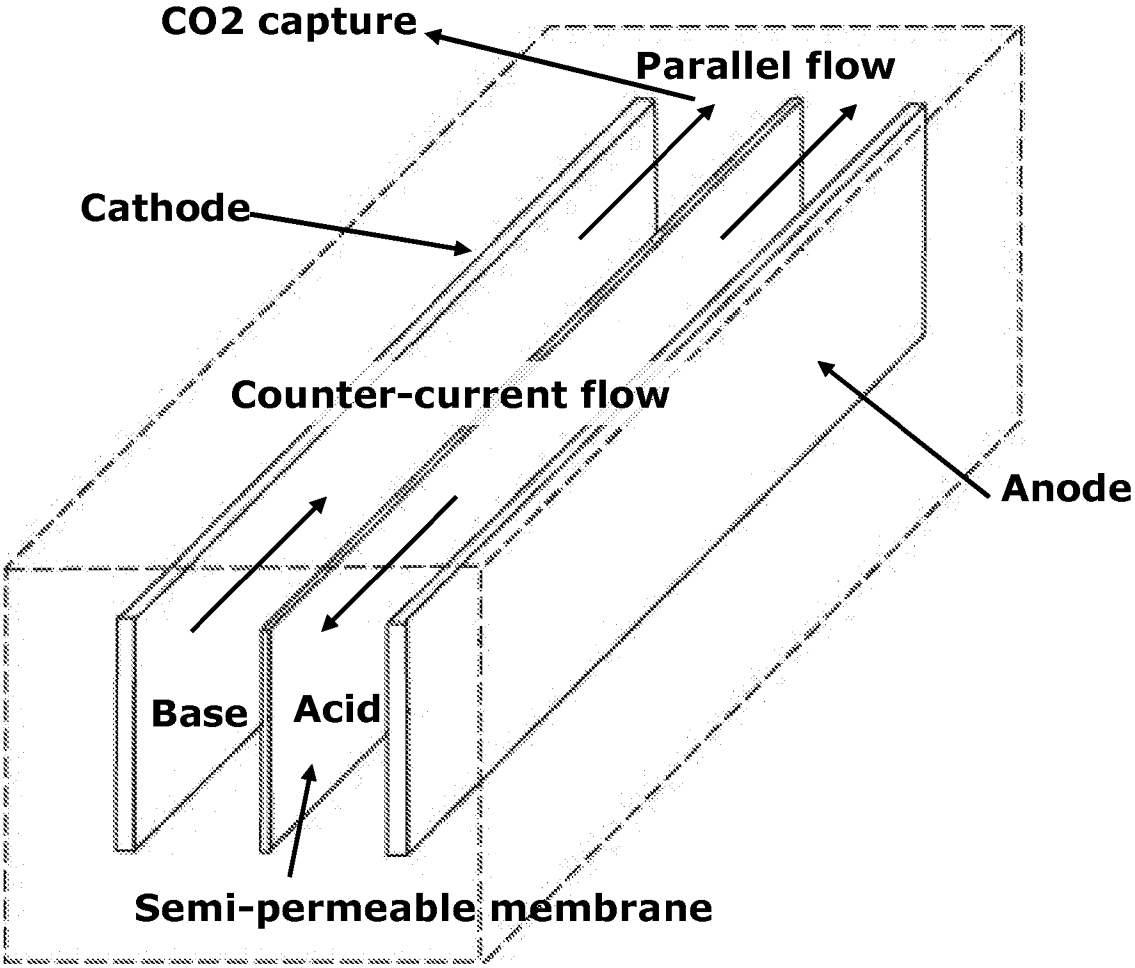

CO2 capture
Parallel flow
Cathode
Counter-current flow
Anode
Base
Acid
Semi-permeable membrane

FIG. 2

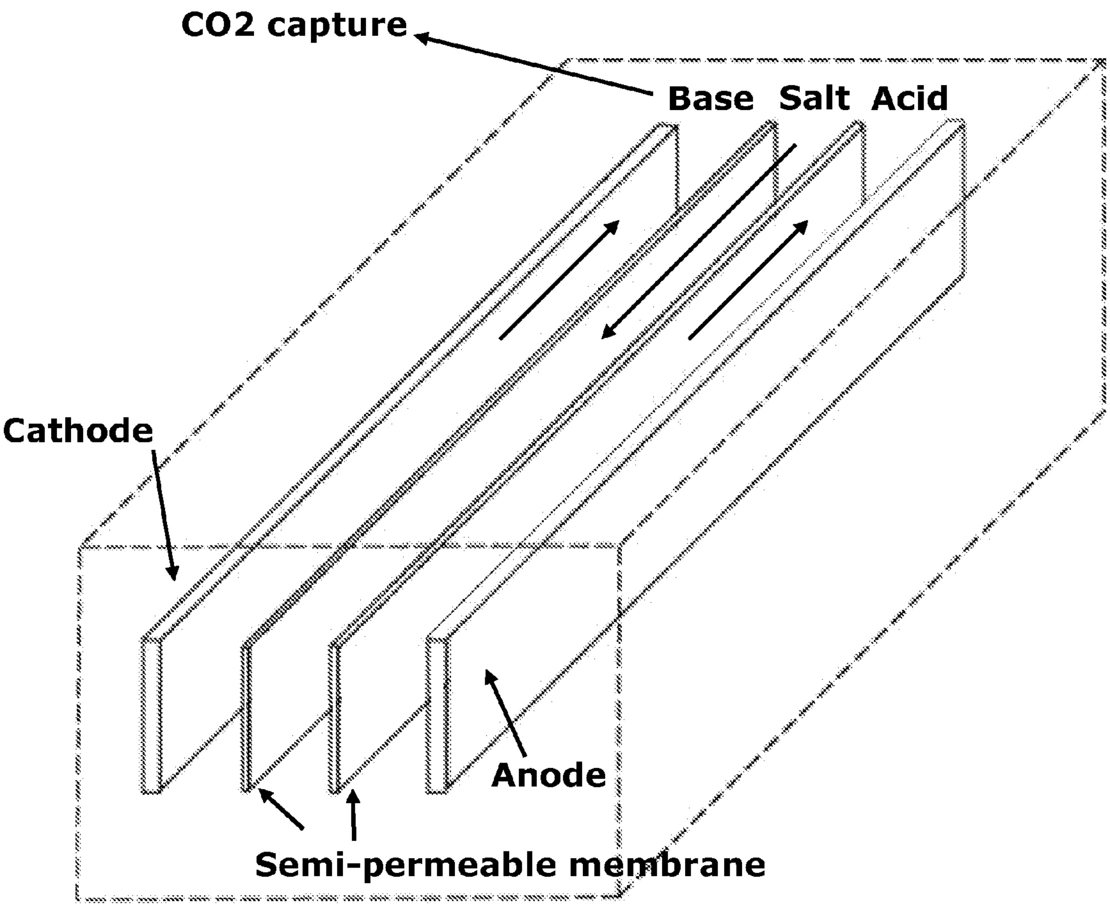

CO2 capture
Base
Salt
Acid
Cathode
Anode
Semi-permeable membrane

FIG. 3

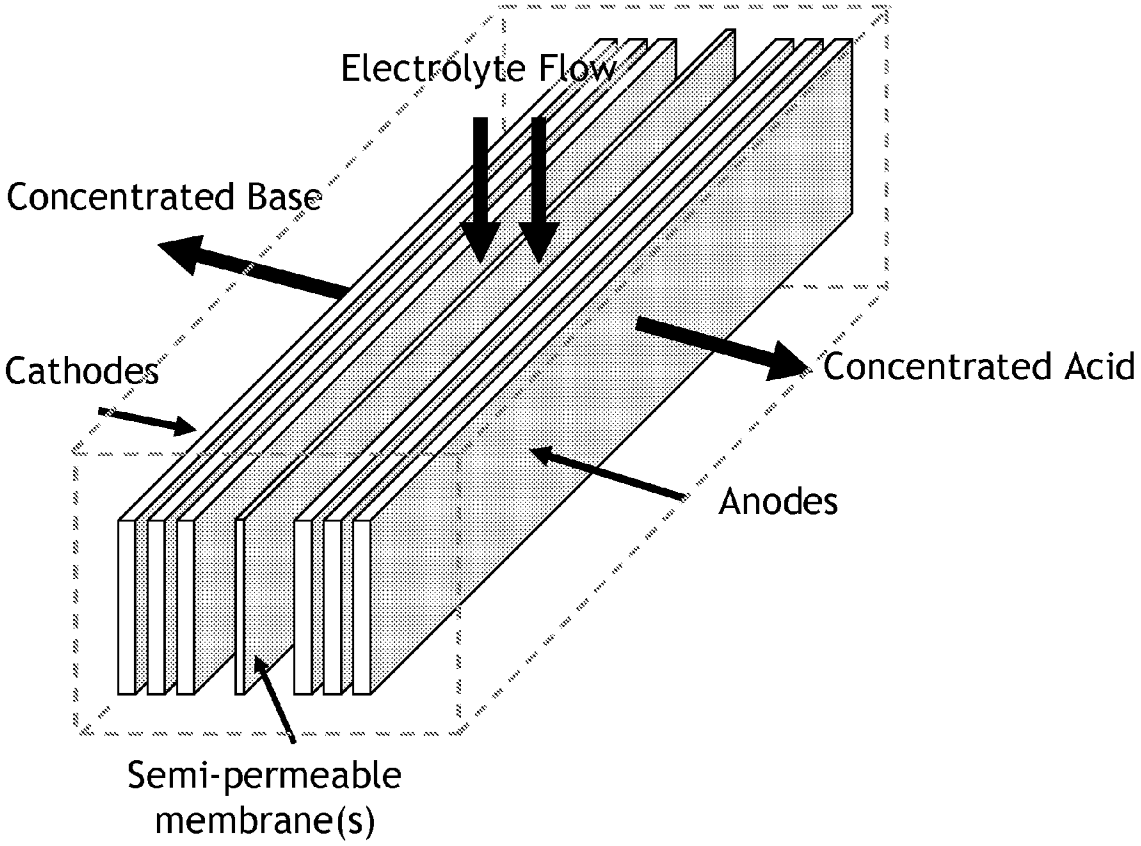

Electrolyte Flow
Concentrated Base
Cathodes
Concentrated Acid
Anodes
Semi-permeable membrane(s)

FIG. 4

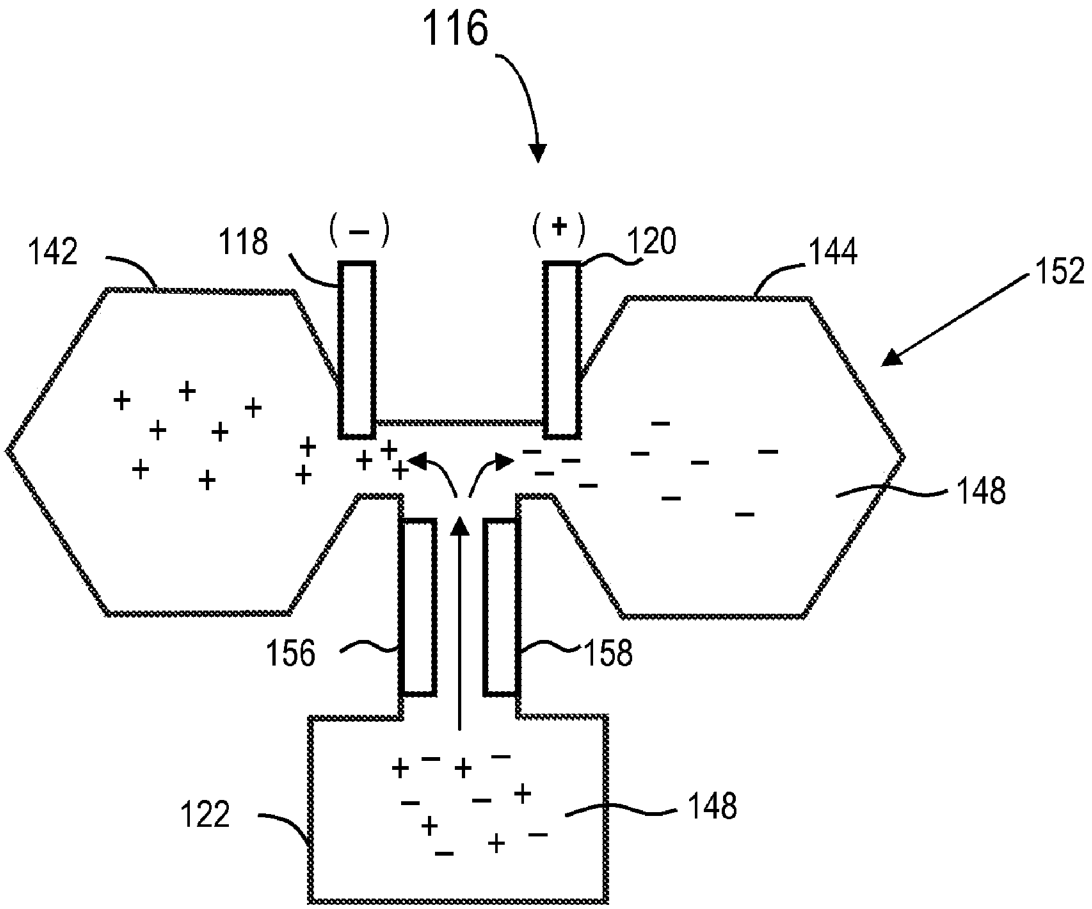


FIG. 5

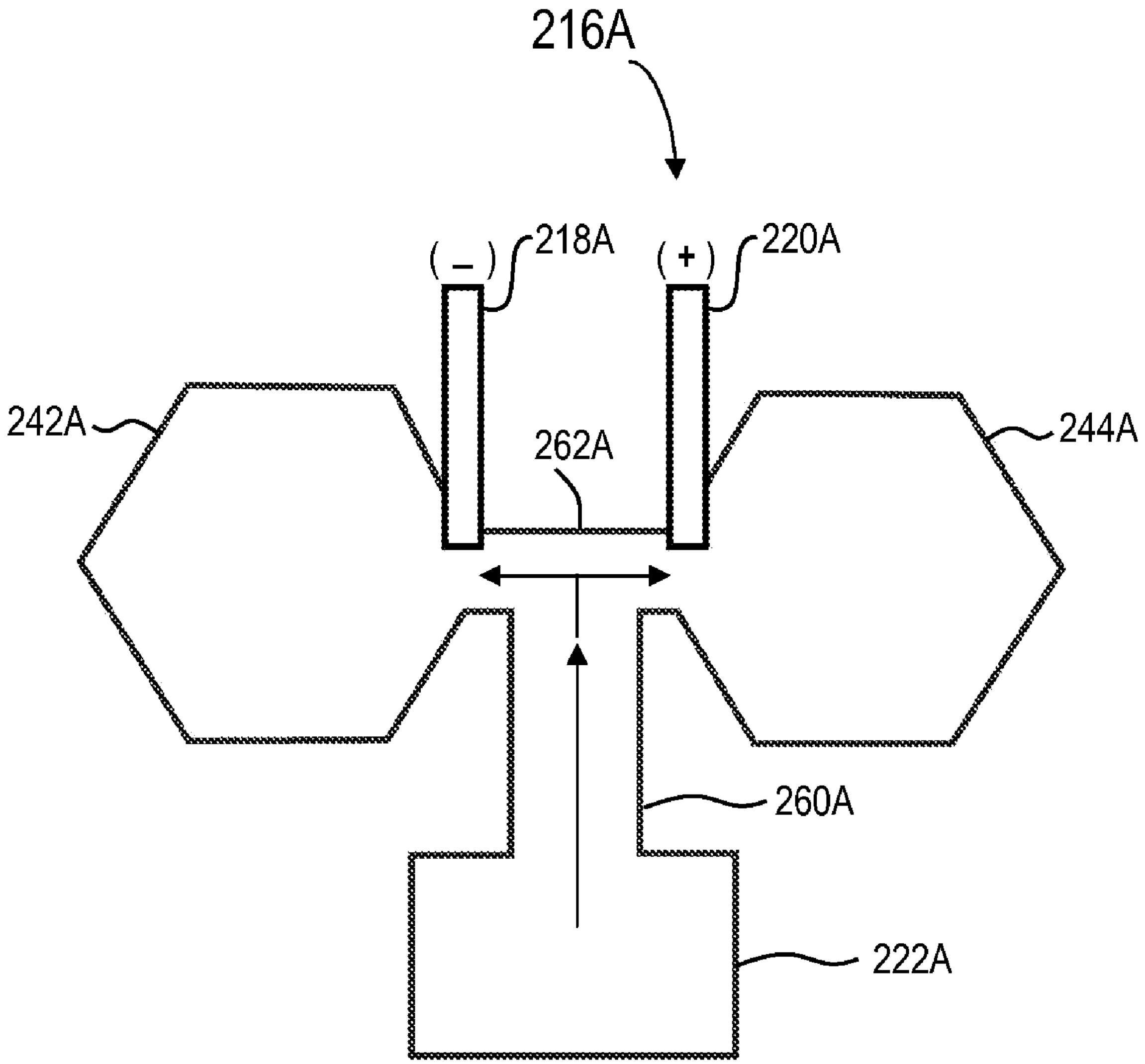


FIG. 6A

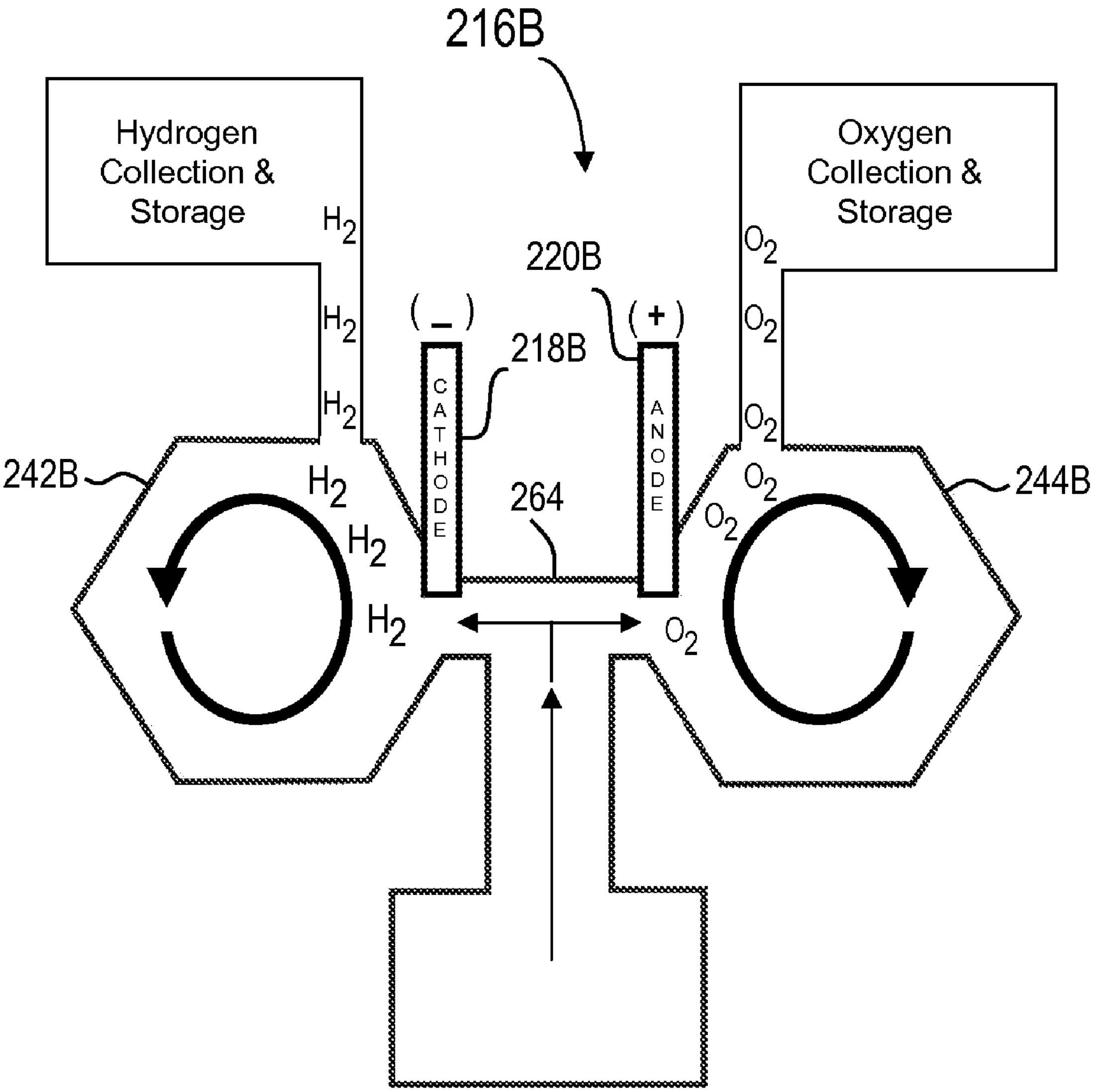

216B
Hydrogen Collection & Storage
Oxygen Collection & Storage
H_2
H_2
H_2
O_2
O_2
O_2
220B
(–)
(+)
218B
CATHODE
ANODE
242B
244B
264
H_2
H_2
H_2
O_2
O_2
O_2

FIG. 6B

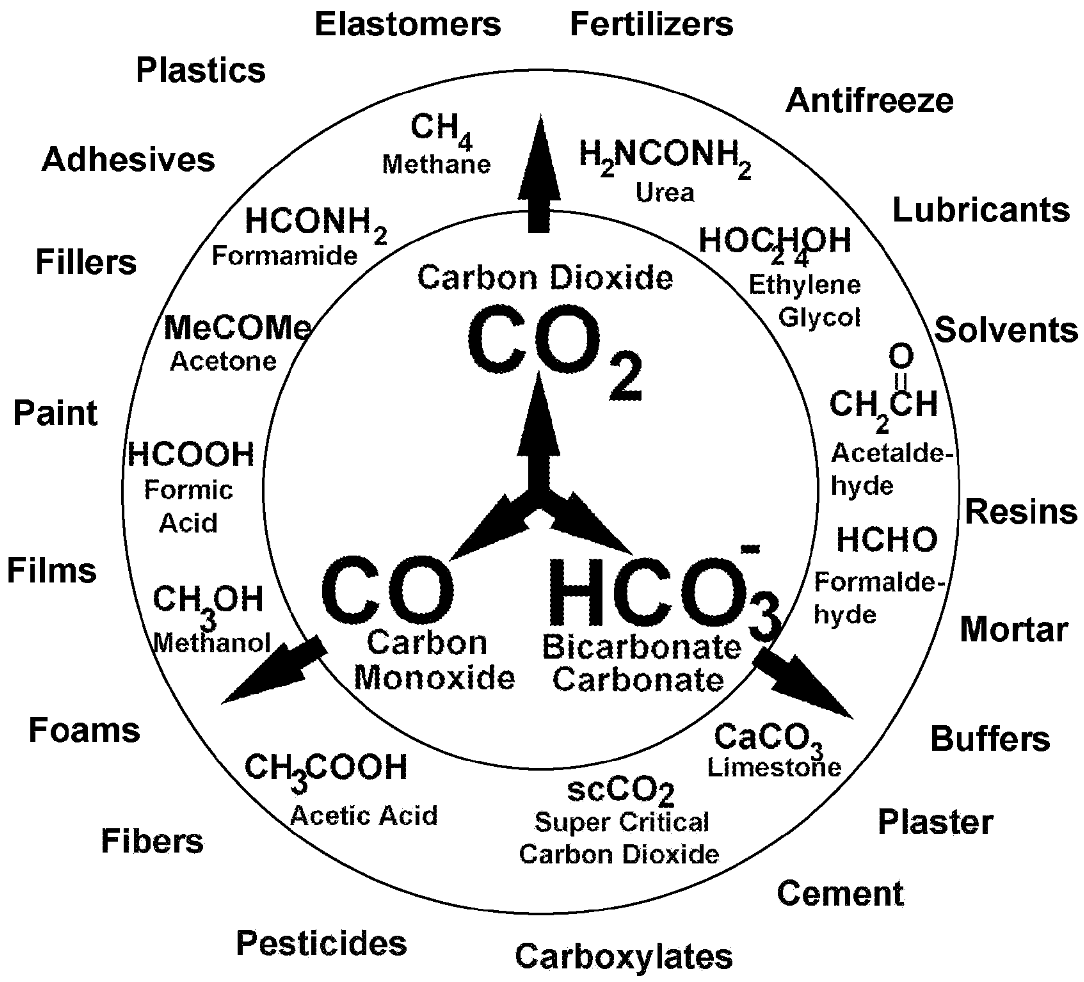

Elastomers
Fertilizers
Plastics
Antifreeze
Adhesives
Lubricants
Fillers
Solvents
Paint
Resins
Films
Mortar
Foams
Buffers
Fibers
Plaster
Cement
Pesticides
Carboxylates
CH_4
Methane
H_2NCONH_2
Urea
$HCONH_2$
Formamide
HOC_2H_4OH
Ethylene Glycol
MeCOMe
Acetone
O
CH_2CH
Acetaldehyde
HCOOH
Formic Acid
HCHO
Formaldehyde
CH_3OH
Methanol
$CaCO_3$
Limestone
CH_3COOH
Acetic Acid
$scCO_2$
Super Critical Carbon Dioxide
Carbon Dioxide
CO_2
CO
Carbon Monoxide
HCO_3^-
Bicarbonate Carbonate

FIG. 7

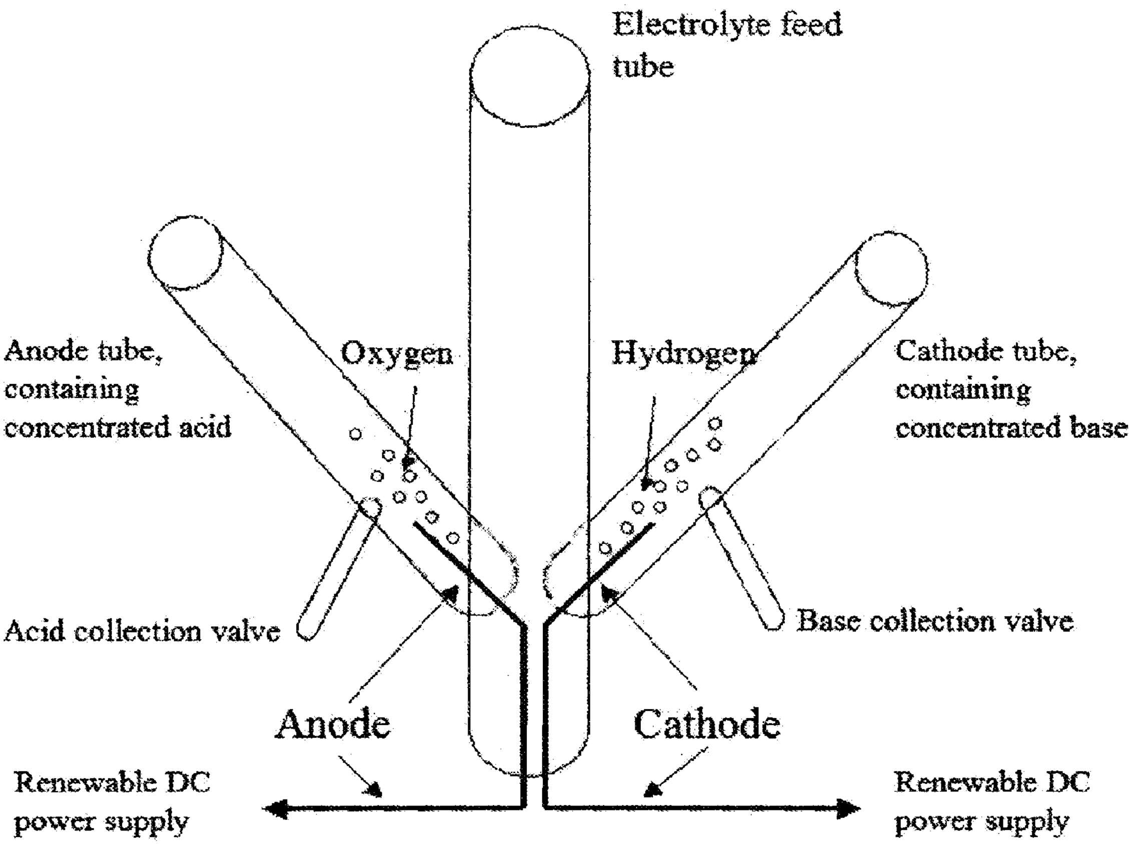

Electrolyte feed tube
Anode tube, containing concentrated acid
Oxygen
Hydrogen
Cathode tube, containing concentrated base
Acid collection valve
Base collection valve
Anode
Cathode
Renewable DC power supply
Renewable DC power supply

FIG. 8

ELECTROCHEMICAL METHODS TO GENERATE HYDROGEN AND SEQUESTER CARBON DIOXIDE

BENEFIT CLAIM

[0001] This application claims the benefit under 35 U.S.C. 119(e) of U.S. Provisional Patent Application No. 60/921,598, filed on Apr. 3, 2007, entitled "A NOVEL ELECTROCHEMICAL METHOD FOR REMOVING CARBON DIOXIDE FROM GAS STREAMS AND SIMULTANEOUSLY GENERATING HYDROGEN GAS," which is herein incorporated by reference in its entirety.

CROSS-REFERENCE TO RELATED APPLICATIONS

[0002] This application is related to co-owned and co-pending applications entitled RENEWABLE ENERGY SYSTEM FOR HYDROGEN PRODUCTION AND CARBON DIOXIDE CAPTURE, filed on the same day and assigned Ser. No. ______, and to co-owned and co-pending application entitled ELECTROCHEMICAL APPARATUS TO GENERATE HYDROGEN AND SEQUESTER CARBON DIOXIDE, filed on the same day and assigned Ser. No. ______, both of which are herein incorporated by reference in their entirety.

TECHNICAL FIELD

[0003] The present invention relates to carbon dioxide capture and sequestration and generation of renewable hydrogen. More specifically, the present invention relates to the use of water electrolysis to generate renewable hydrogen and compounds used to capture and sequester carbon dioxide from the atmosphere or gas streams.

BACKGROUND

[0004] Removing carbon dioxide from the atmosphere requires a very large energy input to overcome the entropic energies associated with isolating and concentrating a diffuse gas. Current strategies for sequestering carbon dioxide from the atmosphere or for producing renewable hydrogen are either inefficient, cost prohibitive, or produce toxic by-products such as chlorine. To lower global carbon dioxide levels and reduce new carbon dioxide emissions, it is critical to develop economically feasible processes to remove vast quantities of carbon dioxide from the atmosphere or gas streams by capturing and sequestering it in a stable form, or by converting it to valuable commodity products. The production of carbon free renewable fuels is also a critical goal in the fight against global warming.

SUMMARY

[0005] According to some embodiments, the present invention is an electrochemical method of generating hydrogen and sequestering carbon dioxide from a gaseous source. A direct current is supplied from an electrical source at a predetermined voltage to a water electrolysis unit including at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions. The electrolysis cell includes an anode region separated, either passively or actively, from a cathode region. The anode and cathode regions of the electrolysis cell are electrically connected by the electrolyte. Hydrogen and base are generated at the cathode region and are individually isolated as products or reagents for down stream processes. Oxygen and acid are produced at the anode region and are individually isolated as products or reagents for down stream processes. The hydroxide ions present in the base are further reacted with a gaseous source of carbon dioxide to sequester carbon dioxide in solution as carbonate, bicarbonate, or mixtures thereof.

[0006] According to other embodiments, the present invention is an electrochemical method of producing hydrogen gas from water and bicarbonate from carbon dioxide trapped from the air or a gas stream. Direct current electricity is supplied to a water electrolysis cell having an anode region adapted to generate oxygen and protons and a cathode region adapted to generate hydrogen and hydroxide ions. The anode and cathode regions are electrically connected by the electrolyte. The hydroxide ions are removed from the cathode region and contacted with gaseous carbon dioxide from the atmosphere or gas stream to produce a solution of bicarbonate, carbonate, or mixtures thereof. The solution is further processed to isolate bicarbonate.

[0007] According to some embodiments the present invention is an electrochemical method of producing hydrogen gas from water and carbonate from the atmosphere or a gas stream. Direct current electricity is supplied to a water electrolysis cell having an anode region adapted to generate oxygen and protons and a cathode region adapted to generate hydrogen and hydroxide ions. The anode and cathode regions are electrically connected by the electrolyte. The hydroxide ions are removed from the cathode region and contacted with gaseous carbon dioxide from the atmosphere or gas stream to produce a solution of bicarbonate, carbonate, or mixtures thereof. The solution is further processed to isolate carbonate.

[0008] According to some embodiments, the present invention is an electrochemical method of producing hydrogen gas and a carbon dioxide neutral or carbon dioxide negative hydroxide base. Renewable or nuclear direct current electricity is supplied to an electrolysis cell having an anode region adapted to generate oxygen and protons and a cathode region adapted to generate hydrogen and hydroxide ions. The anode and cathode regions are electrically connected by the electrolyte. Some or all of the hydroxide ions are removed from the cathode region in the form of a base. The base solution is further processed to concentrate and purify the base, which has been manufactured without any significant carbon dioxide production.

[0009] According to still other embodiments, the present invention is an electrochemical method of producing hydrogen gas and a carbon dioxide neutral or carbon dioxide negative acid. Renewable or nuclear direct current electricity is supplied to a water electrolysis cell having an anode region adapted to generate oxygen and protons and a cathode region adapted to generate hydrogen and hydroxide ions. The anode and cathode regions are electrically connected by the electrolyte. Some or all of the protons are removed from the anode region in the form of an acid. The acid solution is further processed to concentrate and purify the acid, which has been manufactured without net carbon dioxide emissions.

[0010] According to various other embodiments, the present invention is a method of generating and maintaining separate regions of acid and base in an electrolysis chamber. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. Active and passive barriers are used to prevent recombination of the acid and base thereby maximizing the pH gradient between the two regions. The individual products are collected and isolated.

[0011] According to further embodiments, the present invention is an electrochemical method of preparing hydrogen and carbon dioxide sequestering compounds from minerals. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. The protons are present in the form of an acid and some or all of the acid is removed from the anode region. The acid is processed and then contacted with certain minerals that when activated form carbon dioxide sequestering compounds. These compounds are further reacted with a gaseous source of carbon dioxide to capture or sequester carbon dioxide from the gaseous source.

[0012] According to yet other embodiments, the present invention is a method of producing pressurized carbon dioxide. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. All products are individually collected. The hydroxide ions, processed as a base, are reacted with carbon dioxide-containing gas from the atmosphere or gas stream, resulting in a solution of carbonate or bicarbonate or a mixture thereof. The protons, processed as acid, are reacted with the carbonate or bicarbonate to release carbon dioxide in an enclosed environment to produce pressurized carbon dioxide. According to yet further embodiments, the pressurized carbon dioxide can be heated and pressurized to produce super critical carbon dioxide.

[0013] According to still other embodiments, the present invention is an electrochemical method of producing carbon dioxide neutral urea. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. Each of these products are collected and processed as reagents. Hydrogen and carbon dioxide are reacted with a source of nitrogen to produce urea.

[0014] According to still other embodiments, the present invention is an electrochemical method of producing hydrogen and carbon dioxide negative agricultural lime. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. Each of these products are collected and processed as reagents. The hydroxide ions, processed as a base, are reacted with carbon dioxide containing gas from the atmosphere or gas stream to produce a solution of carbonate. The carbonate solution is further processed with a source of calcium ions to produce $CaCO3$, agricultural lime.

[0015] According to yet other embodiments, the present invention is an electrochemical method of producing hydrogen and carbon dioxide neutral quick lime. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. Each of these products is collected and processed as reagents. The hydroxide ions, processed as a base, are reacted with carbon dioxide containing gas from the atmosphere or gas stream to produce a solution of carbonate. The carbonate solution is further processed with a source of calcium ions to produce calcium carbonate. The calcium carbonate is then heated to produce quick lime.

[0016] According to still other embodiments, the present invention is an electrochemical method of producing hydrogen and carbon monoxide from atmospheric carbon dioxide. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. These products are collected and processed as reagents. The hydroxide ions, processed as a base, are reacted with carbon dioxide containing gas from the atmosphere or gas stream to produce a solution of carbonate and bicarbonate or a mixture thereof. The solution is further reacted with protons, processed as acid, to release carbon dioxide in a controlled reaction environment. The carbon dioxide is further reacted with hydrogen produced by the water electrolysis process to form carbon monoxide.

[0017] According to yet other embodiments the present invention is an electrochemical method of producing formic acid from the air or a gas stream. Water electrolysis is used to generate protons and oxygen at the anode region while also generating hydroxide ions and hydrogen at the cathode region. These products are collected and processed as reagents. The hydroxide ions, processed as a base, are reacted with carbon dioxide containing gas from the atmosphere or gas stream to produce a solution of carbonate and bicarbonate or a mixture thereof. The solution is further reacted with the protons, processed as acid, to release carbon dioxide in an enclosed or controlled reaction environment. The carbon dioxide is further reacted with reagents produced by the electrolysis and methanol to form formic acid.

[0018] These and other aspects, processes and features of the invention will become more fully apparent when the following detailed description is read with the accompanying figures and examples. However, both the foregoing summary of the invention and the following detailed description of it represent one potential embodiment, and are not restrictive of the invention or other alternate embodiments of the invention.

BRIEF DESCRIPTION OF THE DRAWINGS

[0019] FIG. **1** is a schematic diagram of an integrated water electrolysis system according to various embodiments of the present invention.

[0020] FIGS. **2**-**5** are schematic diagrams of a water electrolysis cell according to various embodiments of the present invention.

[0021] FIGS. **6**A and **6**B are schematic diagrams of water electrolysis cells according to other embodiments of the present invention.

[0022] FIG. **7** is a schematic diagram of value-added products that may be processed from the integrated electrolysis system of FIG. **1**.

[0023] FIG. **8** is a schematic diagram of a water electrolysis unit according to an embodiment of the present invention.

[0024] While the invention is amenable to various modifications and alternative forms, some embodiments have been shown by way of example in the drawings and are described in detail below. As alluded to above, the intention, however, is not to limit the invention by those examples. On the contrary, the invention is intended to cover all modifications, equivalents, and alternatives.

DETAILED DESCRIPTION

[0025] FIG. **1** is a schematic diagram of an integrated electrolysis system **10** according to various embodiments of the present invention. As shown in FIG. **1**, the integrated electrolysis system **10** includes an electrical energy source **12**, an electrolysis unit **16** including a cathode **18** and an anode **20**, an aqueous electrolyte source **22**, a hydrogen collection and storage reservoir **24**, an oxygen collection and storage reservoir **26**, a base collection and storage reservoir **28**, and an acid collection and storage reservoir **30**. Additionally, according to various embodiments, the integrated electrolysis system includes a first carbon dioxide capture apparatus **32** connected to the base collection and storage reservoir **28**, a second carbon dioxide capture apparatus **34** connected to the acid collection and storage reservoir **30**, and a hydrogen/oxygen fuel cell **38**.

[0026] The integrated electrolysis system **10** is used to produce hydrogen, oxygen, acid, and base through water electrolysis, followed by subsequent processing of one or more of these products to capture and sequester carbon dioxide. Base produced by the integrated electrolysis system **10** is used to capture and sequester carbon dioxide. Additionally, the integrated electrolysis system **10** produces renewable hydrogen as a carbon dioxide neutral fuel. The renewable hydrogen produced by the system **10** can be used as a large-scale application for reducing global carbon dioxide pollution, a significant factor in global warming. When combined with renewable or non-carbon dioxide producing energy sources, the integrated water electrolysis system **10** creates carbon dioxide negative energy strategies for reducing the amount of carbon dioxide in the atmosphere and for producing clean, renewable hydrogen fuel. In addition, unlike traditional methods of manufacturing hydroxide base, such as the chloralkali electrolysis method, no substantial carbon dioxide or chlorine gas is produced.

[0027] Many carbon based products can be manufactured from carbon dioxide trapped by the integrated system **10**. Commercial products manufactured from carbon dioxide trapped by the integrated electrolysis system **10** are carbon dioxide negative, resulting in an overall net decrease in atmospheric carbon dioxide as gaseous carbon dioxide is converted to value-added carbon products. Sale of these products may dramatically subsidize renewable hydrogen production, making clean hydrogen an inexpensive by-product of an industrial process focused on converting atmospheric carbon dioxide into valuable carbon-based products.

[0028] As shown in FIG. **1**, the integrated water electrolysis system includes at least one water electrolysis cell **16**. Electrolysis cells are well known to those of skill in the art. According to various embodiments, an electrolysis cell includes a cathode **18** located within a cathode region **42**, an anode **20** located within anode region **44**, and an aqueous electrolyte **22***a*. Water is reduced at the cathode and oxidized at the anode. The electrolyte is responsible for charge transfer and the movement of ions within the electrolysis cell.

[0029] According to various embodiments of the present invention, the water electrolysis unit **16** includes a separate cathode region **42** and a separate anode region **44**. In some embodiments, ion selective membranes may be used to maintain separate anode and cathode regions **42** and **44**. In other embodiments, a porous glass frit, filter, or other non-selective barrier is used to maintain separate cathode and anode regions **42** and **44**.

[0030] The cathode region **42** and the anode region **44** are electrically connected by an aqueous electrolyte solution **22***a* supplied from the electrolyte source **22**. The aqueous electrolyte solution **22***a* may include electrolyte solution, such as sodium, potassium, calcium, or magnesium sulfate, nitrate, or carbonate. According to various embodiments, the aqueous electrolyte includes an alkali salt. The alkali salt is substantially free of chloride and is a salt of the groups 1(IA) or 2(IIA) of the periodic table. Exemplary electrolytes suitable for use with the present invention include, but are not limited to, the following: sodium sulfate, potassium sulfate, calcium sulfate, magnesium sulfate, sodium nitrate, potassium nitrate, sodium bicarbonate, sodium carbonate, potassium bicarbonate, potassium carbonate, calcium carbonate, and magnesium carbonate. According to other embodiments, the aqueous electrolyte solution can include sea water and/or sea salt.

[0031] According to one exemplary embodiment of the present invention, the aqueous electrolyte solution **22***a* is a saturated solution of sodium sulfate prepared by adding an excess of sodium sulfate to about 1000 liters of clean distilled water placed in a 1200 liter electrolyte processing and storage reservoir. The solution is maintained at about 30 degrees Celsius (° C.) while being mechanically mixed overnight. After filtering, the resultant solution is pumped into the electrolysis unit **16** using a pump or gravity feed. In one embodiment, the aqueous electrolyte solution **22***a* contains substantially no chloride such that the electrolysis unit **16** and/or integrated electrolysis system **10** produce essentially no chlorine gas. In one embodiment, the electrolysis reaction within the water electrolysis unit **16** and/or integrated electrolysis system **10** produces less than about 100 parts per million (ppm) of chlorine, particularly less than about 10 ppm of chlorine, and more particularly less than about 1 ppm of chlorine.

[0032] The concentration of the aqueous electrolyte solution **22***a* can vary depending on the demands of the electrolysis cell and the overall system **10**. The electrolyte concentration may vary with changes in the temperature, pH, and/or the selected electrolyte salt. According to one embodiment, the concentration of the aqueous electrolyte solution **22***a* is approximately 1M. According to another embodiment a saturated aqueous electrolyte solution **22***a* is maintained within the electrolysis cell.

[0033] FIG. **2** shows a schematic diagram of a water electrolysis cell with a single permeable membrane. The water electrolysis cell includes parallel cathode and anode chambers that contain closely spaced electrodes separated by a semi-permeable membrane. This configuration maintains high electrical conductivity while minimizing loss of acid and base to recombination within the water electrolysis cell.

[0034] Fresh aqueous electrolyte solution flows in the same direction in both the cathode and anode chambers, gradually becoming more basic in the cathode chamber and more acidic in the anode chamber. Alternatively, fresh aqueous electrolyte solution may be introduced through one of the cathode chamber and anode chamber. In this case, selective ion flow across an anion or cation specific membrane would ensure production of a highly pure acid or base, respectively. This water electrolysis cell can be operated in parallel or counter-current flow modes. Counter-current flow minimizes chemical gradients formed across the semi-permeable membrane and may reduce the energy required to create such gradients and produce highly concentrated acid and base. In a counter-current system, the highest concentrations of hydronium and hydroxide ions and their counter-ions are never located directly across the semi-permeable membrane from one another, but instead reach maximum strength opposite incoming fresh aqueous electrolyte solution in the counter-cell. This design avoids the need to create a 13-14-unit pH gradients across the semi-permeable membrane, instead producing no higher than a 7-unit pH gradient between either strong acid and neutral electrolyte, or strong base and neutral electrolyte.

[0035] FIG. **3** shows a schematic diagram of a water electrolysis cell with two semi-permeable membranes. The water electrolysis cell is a parallel or counter-current flow three-chamber water electrolysis cell. A narrow central feed reservoir (such as electrolyte source) of fresh aqueous electrolyte solution is introduced between a first semi-permeable membrane and a second semi-permeable membrane that separate the cathode chamber and anode chamber. As illustrated in counter current mode, concentrated aqueous electrolyte solution enters the central feed reservoir at a first end of the water electrolytic cell and concentrated base and acid exit the cathode chamber and the anode chamber, respectively. At a second end of the water electrolytic cell, dilute base and acid enter the cathode chamber and the anode chamber, and water or dilute aqueous electrolyte solution exits the central feed reservoir. This counter-current design reduces salt contamination of base and acid produced and minimizes the chemical gradients formed across the permeable membranes. In some embodiments, the design may also be used to desalinate salt water.

[0036] In practice, the cathode chamber in FIG. **3** is initially filled with dilute base, and the anode chamber is filled with dilute acid, maintaining electrical conductivity between the electrodes. Cations flow from the central feed reservoir through the first semi-permeable membrane closest to the cathode chamber, combining with hydroxide ions formed at the cathode chamber to generate concentrated hydroxide base. Anions flow from the electrolyte solution source through the second semi-permeable membrane to the anode chamber, combining with protons formed at the anode chamber to produce concentrated acid. The semi-permeable membranes may be ion-selective (anion- or cation-specific) membranes, or may be passive barriers minimizing fluid flow, allowing passage of anions or cations in either direction. Regardless of membrane selectivity, such a 3-cell system can operate with parallel flow in all cells, or with counter-current flow between the central feed reservoir and the cathode chamber and anode chamber on either side. The counter-current flow system minimizes chemical gradients across the membranes, because high concentrations of base and acid exit the cathode chamber and anode chamber opposite highly concentrated fresh electrolyte entering the central feed reservoir. In parallel flow mode certain design advantages are also realized.

[0037] FIG. **4** is a schematic diagram of a stacked water electrolysis cell according to some embodiments of the present invention. The stacked porous electrodes may be used in some embodiments to maximize acid and base production. According to one embodiment, as shown in FIG. **4**, the water electrolysis cell includes two or more porous anode-cathode pairs aligned in a closely spaced parallel configuration. Semi-permeable or ion selective membranes are optionally included between the inner pair of electrodes. The membranes function to contain a narrow electrolyte feed reservoir located between the inner pair of porous anodes or cathodes. Fresh electrolyte flows from the reservoir outward, contacting the first pair of electrodes, where water oxidation occurs at the anode and water reduction occurs at the cathode. Thus, as water passes through each pair of electrodes it becomes increasingly acidic or basic. In one embodiment, the electrodes may consist of fine mesh screens, porous micro or nanosphere materials or thin plates with numerous flow channels penetrating the electrode. Varying DC voltages in the range of about 1.2 to about 10 Volts are supplied to these electrode pairs to maximize the production of acid in the anode chamber and base in the cathode chamber.

[0038] According to some embodiments, the aqueous electrolyte solution **22***a* may undergo additional processing prior to entering the anode and cathode regions of the cell. FIG. **5** is a schematic diagram of a water electrolysis cell **116** configured to process the aqueous electrolyte solution **148** prior to its introduction into the electrolysis cell **116**. As shown in FIG. **5**, the electrolysis cell **116** includes an electrolysis chamber **152** including a cathode **118** located within a cathode region **142** and an anode **120** located within an anode region **144**. The electrolysis chambers **152** is fluidly coupled to an electrolyte source **122**. According to various embodiments, the electrolysis cell **116** also includes a cathode electrolyte preparation electrode **156** and an anode electrolyte preparation electrode **158**.

[0039] As shown in FIG. **5**, the electrolyte preparation electrodes **156** and **158** are located within the aqueous electrolyte flow path from the electrolyte source **122** to the electrolysis chamber **152**. According to various embodiments, a voltage is applied to the electrolyte preparation electrodes **156** and **158** that is less than the minimal theoretical voltage required for water electrolysis. According to some embodiments, the applied voltage is less than about 1.2V. When a potential is applied to the preparation electrodes **156** and **158** the preparation electrodes **156** and **158** act like charged poles, attracting the ions of the opposite charge. According to some embodiments, the electrolyte preparation cathodes **156** attracts cations and repel anions. Similarly, the electrolyte preparation anodes **158** attracts anions and repel cations. This process presorts the ions present in the electrolyte solution **148** prior to its introduction into the electrolysis chamber **152**.

[0040] Referring again to FIG. **1**, a direct current is supplied to the water electrolysis unit **16** from the electrical energy source **12** to electrolyze the aqueous electrolyte solution to produce hydrogen, oxygen, acid, and base. According to some embodiments, as shown in FIG. **1**, a renewable energy source can be coupled to the electrical energy source to supply energy to the integrated system. Exemplary renewable energy sources include, but are not limited to, the following: wind, solar, hydroelectric, oceanic, tidal, geothermal, and fuel cells using renewable hydrogen. These renewable energy sources do not generate carbon dioxide. Other energy sources that may generate carbon dioxide may also be used to provide energy to the electrical energy source including biofuel, biomass, coal, methane and the like. According to one embodiment, nuclear energy may also be used to provide energy to the integrated system **10**. According to yet further embodiments, a renewable energy source that generates substantially no carbon dioxide may be coupled with a conventional energy source to supplement and/or off-set the amount of energy supplied to the electrical energy source from the conventional energy source.

[0041] According to one embodiment, the direct current is supplied to the electrolysis unit **16** at a predetermined voltage sufficient to initiate water electrolysis within the electrolysis cell **16**. According to one embodiment, the predetermined voltage supplied to the electrolysis cell is at least 1.2 volts. According to other embodiments, the predetermined voltage supplied to the cell ranges from about 1.2 volts to about 10.0 volts. The result of the electrolysis reaction within the cell **16** is the formation of protons and oxygen gas at the anode region, and hydroxide ions and hydrogen gas at the cathode region. The protons combine with anions present in the electrolyte solution to form acid. Similarly, the hydroxide ions combine with cations present in the electrolyte solution to form base.

[0042] The continuous production of acid and base during water electrolysis results in a pH difference between the cathode region **42** and the anode region **44** of the electrolysis cell **16**. According to one embodiment, the difference in pH between the cathode region **42** and the anode region **44** is at least 4 pH units. According to other embodiments, the difference in pH between the cathode region **42** and the anode region **44** is at least 8 pH units. The difference in pH between the cathode regions and anode regions **42** and **44** can be maintained by preventing the catholyte formed in the cathode region **42** and the anolyte formed in the anode region **44** from combining.

[0043] FIG. **6**A is a schematic diagram of an electrolysis cell **216**A according to one embodiment of the present invention. FIG. **6**B is a schematic diagram of another electrolysis cell **216**B according to another embodiment of the present invention. Each of the cells **216**A and **216**B as shown in FIGS. **6**A and **6**B are configured to maintain a separate cathode region **242**A, **242**B and a separate anode region **244**A, **244**B within the electrolysis cell **216**A, **216**B using fluid dynamics. Additionally, according to further embodiments, the cells **216**A, **216**B are configured to maintain a pH difference between the cathode region **242**A, **242**B and the cathode region **244**A, **244**B of at least 4 pH units and more particularly, of at least 6 pH units.

[0044] In one embodiment, as shown in FIG. **6**A, the electrochemical cell **216**A has a "T" configuration. The "T" shaped cell **216**A includes an elongated vertical portion **260**A branching bi-directionally into a horizontal portion **262**. A continuous supply of fresh electrolyte flows up through the elongated portion of the "T" shaped shell, indicated by the arrows, from the electrolyte source **222**A. Once the electrolyte has reached the horizontal portion **260**A of the "T" shaped cell **216**A, the electrolyte then flows in opposite directions towards closely spaced cathode and anode regions **242**A and **242**B. According to various embodiments, the bi-directional flow rate of the electrolyte through the cell **216**A is greater than the rate of ion migration due to the applied electric field and diffusion. Thus, the contents of the cathode and the anode regions **242**A and **244**A cannot recombine, and the pH difference between the anode and the cathode regions **242**A and **244**A can be maintained.

[0045] In another embodiment, as shown in FIG. **6**B, convective currents within the cathode and anode regions **242**B and **244**B assist in maintaining a pH difference between the cathode region **242**B and the anode region **244**B of the electrolysis cell **216**B. As shown in FIG. **6**B, hydrogen gas is formed at the cathode **218**B and rises in the form of bubbles in the electrolyte solution. The rising bubbles create convective currents in the cathode region **242**B. Similarly, oxygen produced at the anode **220**B rises in the form of bubbles, creating convective currents in the anode region **244**B. Additionally, the electrolysis cell **216**B includes a constricted pathway **264** fluidly coupling the cathode and anode regions **242**B and **244**B. The convective currents in the cathode and the anode regions **242**B and **244**B in combination with the constricted fluid pathway **264** between the cathode and anode regions **242**B and **244**B assist in maintaining a pH difference between the cathode and anode regions **242**B and **244**B of at least 4 pH units and, more particularly, of at least 6 pH units. In further embodiments, an electrolysis cell combining the features of the electrolysis cell shown in FIG. **6**A and the features of the electrolysis cell shown in FIG. **6**B can be utilized.

[0046] Once concentrations of base and acid reach a minimum increase of one hundred fold relative to their initial electrolyte concentration, the base and acid are removed from the cathode region and anode region of the electrolysis cell. According to some embodiments, the base and the acid are capable of achieving an increase of over about 100,000 times their initial electrolyte concentration. According to one embodiment, the base and the acid formed at the cathode and anode regions are pumped to their respective collection and storage reservoirs in the integrated system **10**. According to another embodiment, positive pressure may be applied to remove the base and acid from the cathode and anode regions. According to yet another embodiment, the base and acid may be removed from their respective cell regions via gravity feed. Fresh electrolyte is then delivered from the aqueous electrolyte source to equilibrate the volume of liquid in the cathode region and the anode region. According to one embodiment, the removal of acid and base and introduction of fresh electrolyte may be accomplished by a batch-wise process. According to another embodiment, the removal of base and acid and the introduction of fresh electrolyte may be accomplished by a continuous process, creating a continuous flow electrolysis system.

[0047] According to some embodiments, the electrolyte flow rate can be adjusted to overcome undesirable ion migration, eliminating acid-base recombination and/or mixing of the electrolyte from the cathode and anode regions. According to other embodiments, the electrolyte flow rate can be adjusted to increase, decrease and/or maintain the concentrations of the base and acid produced in their respective regions of the electrolysis cell **16**.

[0048] Referring back to FIG. **1**, after water in the aqueous electrolyte solution has been electrolyzed to produce hydrogen, oxygen, base and acid, the products are sequestered and collected. The gases are routed from the cathode **18** or anode **20** to storage or flow systems designed to collect such gases. The low density of the gases relative to the aqueous electrolyte solution causes the gases to rise. The reaction regions are designed to direct this flow up and out of the cathode **18** and anode **20** and into adjacent integrated areas. The hydrogen, base, oxygen and acid are physically diverted for collection in the hydrogen sequestration tank **24**, the base sequestration tank **28**, the oxygen sequestration tank **26** and the acid collection and storage reservoir **30**, respectively.

[0049] The hydrogen and oxygen are collected in the hydrogen collection and storage reservoir **24** and the oxygen collection and storage reservoir **26**, respectively. In some embodiments the hydrogen and oxygen are used to supplement the electrical energy source **12** when used as a fuel in a furnace, fuel cell **38**, or engine to provide direct current electricity for electrolysis. The hydrogen and/or oxygen may also be used to react with other products of the integrated electrolysis system **10** to create value-added products. Finally, the hydrogen and/or oxygen may be removed from the integrated electrolysis system **10** as a product to be sold or used locally as a fuel or chemical feedstock.

[0050] The acid produced by the electrolysis unit **16** is routed to the acid collection and storage reservoir **30**. According to one embodiment the pH of the acid ranges from about pH=0 to about pH=5. The acid can be processed and removed from the system for sale as a commodity. The acid may also be used to prepare certain mineral-based carbon dioxide sequestering compounds, which are then used to capture carbon dioxide from the atmosphere or gas streams. The acid may also be used as a chemical reagent by the integrated system to create other value added products. In one embodiment, the carbonate and bicarbonate salts are isolated after reacting the base with carbon dioxide. The acid can then release the carbon dioxide from the carbonate or bicarbonate salts in a controlled manner to further process the released carbon dioxide to produce value-added products. These products may include, but are not limited to: carbon monoxide, formic acid, urea, super-critical carbon dioxide, pressurized carbon dioxide, liquid carbon dioxide or solid carbon dioxide.

[0051] The base generated by the electrolysis unit **16** is sent to the base collection and storage reservoir **28** and is sold or used as a carbon dioxide neutral commodity or chemically reacted with carbon dioxide gas to form carbonate or bicarbonate. In one embodiment a pH of the base produced in the cathode region of the cell can range from about pH=8 to about pH=14. When used to capture carbon dioxide, the carbon dioxide is captured as carbonate, bicarbonate, or mixtures thereof. The carbon dioxide may be captured by reacting, sequestering, removing, transforming, or chemically modifying gaseous carbon dioxide in the atmosphere or a gas stream. The gas stream may be flue gas, fermenter gas effluent, air, biogas, landfill methane, or any carbon dioxide-contaminated natural gas source. The carbonate salts may subsequently be processed to generate a variety of carbon-based products.

[0052] The reaction of the base with the carbon dioxide can be passive, relying only on natural gas-water mixing. An example of a passive reaction includes an open-air treatment pond filled with aqueous base, or a lined bed of hydroxide crystals. The reaction of the base with carbon dioxide is spontaneous and can be enhanced by increased concentrations of base or carbon dioxide. The reaction can also proceed by active mechanisms involving the base or carbon dioxide. An example of an active reaction includes actively spraying, nebulizing, or dripping a basic solution into air or a gas stream containing carbon dioxide. In another example, carbon dioxide is actively removed by bubbling or forcing the gas stream through a column or reservoir of base generated by the electrolysis cell **16**. Combinations of active and passive carbon dioxide trapping systems are also envisioned.

[0053] In some embodiments of the present invention, sodium bicarbonate and sodium carbonate are be formed by the integrated water electrolysis system **10**. Sodium bicarbonate and sodium carbonate may be formed within the integrated electrolysis system **10**. Alternately, base may be removed from the integrated electrolysis system **10** and transported to another site to capture carbon dioxide from the atmosphere or a gas stream using the passive or active techniques previously described. By using the base and/or acid to capture carbon dioxide from the atmosphere or a gas stream, the overall integrated electrolysis system **10** sequesters substantially more carbon dioxide than it creates, resulting in a net negative carbon dioxide footprint.

[0054] FIG. **7** illustrates value-added products that may be processed from the carbon dioxide captured using the base and/or acid produced by the integrated electrolysis system **10**. The integrated electrolysis system **10** processes the value-added products from the center of the diagram outward. As previously mentioned, base generated from water electrolysis is reacted with carbon dioxide to produce carbonate and bicarbonate salts. The carbonate and bicarbonate salts can in turn be converted to carbon monoxide by chemical or electrochemical reduction or reaction of carbon dioxide with hydrogen. The combination of carbon monoxide and hydrogen is Syngas, a critical cornerstone of synthetic organic chemistry. Through additional processing of these central products, a number of chemical building blocks, such as methane, urea, ethylene glycol, acetaldehyde, formaldehyde, limestone, acetic acid, methanol, formic acid, acetone and formamide can be formed. These value added chemical building blocks can be sold as commodity chemicals or used to produce a second class of value-added products, including polymers, fabrics, urea and various building materials. These value-added end products are then removed from the integrated electrolysis system **10** and sold, resulting in profitable conversion of carbon dioxide into carbon dioxide negative products. Simultaneous production of renewable hydrogen is subsidized by sale of these carbon products, reducing the cost of renewable hydrogen production and creating a carbon dioxide negative energy strategy with potentially dramatic impacts on global warming.

[0055] The center circle of FIG. **7** depicts exemplary products that can be produced from the reaction of hydroxide base with carbon dioxide, or (in the case of carbon monoxide) by chemical reduction of captured carbon dioxide. These chemical compounds include carbon dioxide, carbon monoxide, carbonate and bicarbonate, all of which can be easily interconverted. They can be further processed to create a variety of carbon-based monomers that serve as building blocks for larger molecules. In many cases, the hydrogen, oxygen, acid and base generated by the electrolysis unit **16** can be used for this secondary processing. The carbon based building blocks can also be further processed within the integrated electrolysis system **10** to make many valuable carbon based products. Some examples of these are illustrated in the outer ring of FIG. **7**.

[0056] According to one embodiment of the present invention, an aqueous electrolyte solution is electrolyzed in a water electrolysis cell to produce hydroxide ions in the cathode region. The hydroxide ions are present in the form of a base such as sodium or potassium hydroxide. Next the hydroxide ions in the base are contacted with a source of gaseous carbon dioxide by any one of the methods as described above to sequester the carbon dioxide in solution as bicarbonate, carbonate, or mixtures thereof.

[0057] Bicarbonate and/or carbonate can be isolated from the solution to produce a bicarbonate salt, a carbonate salt, or a mixture there of. This can be accomplished by a variety of techniques. For example, the pH of the solution can be maintained between 8 or 9 to favor bicarbonate formation or maintained higher than pH 11 to favor carbonate formation. Double displacement reactions may be used to isolate different forms of carbonate or bicarbonate. More specifically, sodium carbonate is reacted with calcium chloride to form calcium carbonate, which easily precipitates from solution. Similarly, magnesium salt can also be used to convert sodium salts of bicarbonate or carbonate to less soluble magnesium salts. The calcium carbonate and magnesium carbonate can be purified and used or sold. Other processing methods for the isolation of bicarbonate and carbonate include concentration, precipitation, heating, cooling, solar evaporating, vacuum evaporating, wind evaporating and crystallizing.

[0058] According to various embodiments, solid bicarbonate and/or carbonate can be used in the production of a wide range of building materials. For example bicarbonate and/or carbonate can be used as fillers in the manufacture of plastics, elastomers, adhesives, and other polymer based materials. According to various other embodiments, the solid bicarbonate and/or carbonate can be used in the production of mortar, cement, plaster, tile, grout, wall board, synthetic stone, and the like. Finally, solid sodium bicarbonate can purified and sold as baking soda.

[0059] According to various embodiments of the present invention, base produced at the cathode region of the electrolysis cell can be utilized to produce carbon dioxide neutral or carbon dioxide negative agricultural lime and quick lime. For example, a sodium sulfate solution is electrolyzed in a water electrolysis cell to form sodium hydroxide in the cathode region. The base is concentrated such that it reaches a pH of at least pH=10 and then is contacted with a gaseous source of carbon dioxide to favor production of a sodium carbonate enriched feedstock. The carbonate-enriched feedstock is then mixed with a calcium chloride solution. Solid calcium carbonate precipitates from the feedstock to produce agricultural lime. According to further embodiments, heat may be applied to the solid calcium carbonate, produced according to the method described above to produce carbon dioxide neutral quick lime, or, if the carbon dioxide released is captured again, carbon dioxide negative quick lime.

[0060] According to another embodiment of the present invention, acid generated in the anode region of the electrolysis cell can be utilized to produce a carbon dioxide sequestering material. Water in an aqueous electrolyte solution is electrolyzed in an electrolysis cell to produce strong acid at the anode region of the cell. At least some or all of the acid is removed from the anode region and collected and stored in an acid collection and storage reservoir. According to one embodiment, the acid is concentrated either within the anode region of the cell or in the acid collection and storage reservoir such that the resulting pH of the acid ranges from about pH=0 to about pH=5. According to a further embodiment, the acid is concentrated such that it has a pH of about pH=1. The acid is then reacted with a material that when exposed to a strong acid is converted to a carbon dioxide sequestering material. Exemplary materials that can be converted to a carbon dioxide sequestering material by reaction with a strong acid include, but are not limited to, the following: certain mineral clays, sepiolite, serpentine, talc, asbestos, and various mining byproducts such as asbestos mining waste. According to one exemplary embodiment, serpentine can be dissolved in sulfuric acid procuring a solution of magnesium sulfate while precipitating silicon dioxide as sand. Addition of sodium hydroxide creates a mixture of magnesium sulfate and magnesium hydroxide. The process also converts toxic asbestos and asbestos waste into non-toxic carbon dioxide binding materials. Subsequent exposure of the magnesium solution to carbon dioxide from the atmosphere or a gas steam results in the formation of either magnesium carbonate or magnesite, both of which form precipitates. These precipitates are well-suited for production of construction blocks. According to further embodiments of the present invention, the carbon dioxide sequestering material may be further reacted with strong acid to release carbon dioxide gas under controlled conditions. The carbon dioxide released from the carbon dioxide sequestering materials may be captured and stored for further processing.

[0061] According to various other embodiments, base produced in the cathode region and acid produced in the anode region of the electrolysis cell can be used to produce concentrated or pressurized carbon dioxide gas in a controlled reaction. For example, water in an aqueous electrolyte solution is electrolyzed to produce base at the cathode region and acid at the anode region. Some or all of the base is removed from the cathode region and collected and stored in the base collection and storage reservoir. Some or all of the acid is removed from the anode region and is collected and stored in the acid collection and storage reservoir. The hydroxide ions present in the base are reacted with a gaseous source of carbon dioxide to produce a solution including bicarbonate, carbonate, or mixtures thereof. The acid is reacted with the carbonate containing solution in an enclosed container to produce highly concentrated and pressurized carbon dioxide. Alternately the carbon dioxide can be released into a pipe or flow system for transport to another site. In other embodiments, the carbon dioxide may be further concentrated and/or purified.

[0062] According to other further embodiments, the carbon dioxide produced according to the method above can be converted to urea. Urea is a commonly used in the agricultural industry as a fertilizer as it is rich with nitrogen. According to various embodiments, the carbon dioxide is contacted with a source of anhydrous ammonia under pressure to produce urea. According to other embodiments, hydrogen produced at the cathode and carbon dioxide produced according to the various methods described above are reacted with nitrogen gas in an electrochemical process to produce urea.

[0063] In other embodiments, the carbon dioxide gas can be converted to useful products such as super critical carbon dioxide. Pressurized carbon dioxide gas can be adjusted to a critical temperature and a critical pressure to produce super critical carbon dioxide. Super critical carbon dioxide is widely used in the food processing and fragrance industries to extract caffeine from coffee or tea, essential oils from seeds or plant materials, or to manufacture dry ice. Recent advances have also shown super critical carbon dioxide to be a valuable reagent or solvent in the synthetic organic chemistry.

[0064] In other embodiments of the present invention, the carbon dioxide produced according to the methods described above can be converted to carbon monoxide, an essential building block in much of synthetic organic chemistry. Several well known chemical pathways are used industrially to convert carbon dioxide to carbon monoxide. In one such embodiment, the Reverse Water Gas Shift reaction utilizes hydrogen produced at the cathode to reduce carbon dioxide into carbon monoxide and water.

[0065] Carbon monoxide has many applications in bulk chemicals manufacturing. For example, aldehydes are produced by the hydroformylation reaction of alkenes, carbon monoxide, and hydrogen gas. Hydroformylation can be coupled to the Shell Higher Olefin Process to give precursors to detergents. Additionally, methanol can be produced by the hydrogenation of carbon monoxide. Finally in the Monsanto process, methanol and carbon monoxide react in the presence of a homogeneous rhodium catalyst and HI to give acetic acid. Any chemical pathway that converts carbon dioxide to carbon monoxide may be applied to carbon dioxide sequestered and released from products of the aforementioned water electrolysis/carbon dioxide capture technology described in this patent application. When manufactured from atmospheric carbon dioxide such products are carbon dioxide negative.

[0066] According to yet other embodiments of the present invention, formate and formic acid may be produced from the products of the water electrolysis reaction, described herein. For example, base produced at the cathode can be reacted with a gaseous source of carbon dioxide to produce a solution containing bicarbonate, carbonate, or mixtures thereof. The carbonate containing solution can be reacted with acid from the anode to release carbon dioxide under controlled conditions. Hydrogen gas produced by the water electrolysis reaction and methanol are added stepwise to yield formic acid. In another embodiment, a 1M cesium bicarbonate, processed from carbon dioxide sequestered in a basic solution can be electrolyzed using palladium catalysts to produce formic acid in high yield and Faradaic efficiency. In another method, the carbonate or bicarbonate is reacted with acid formed at the anode to release carbon dioxide in a controlled process. Hydrogen produced by water electrolysis and methanol are then added step wise to yield formic acid. Any chemical pathway that produces formate or formic acid using products from the aforementioned water electrolysis/carbon dioxide capture technology is contemplated by these embodiments.

[0067] According to various other embodiments of the present invention, the electrolysis products produced according to the methods described above can be used to produce methanol. Many metal oxide, zinc and zirconium catalysts are known to reduce carbon dioxide to methanol. In one such embodiment, carbon dioxide trapped from the atmosphere or a gas stream using base generated by water electrolysis is released in a controlled environment using acid also produced from the electrolysis process. The carbon dioxide and hydrogen produced by water is released in a controlled environment using acid also produced from the electrolysis process. The carbon dioxide and hydrogen produced by water electrolysis are combined and reacted over a nickel catalyst to produce methanol. In another embodiment, the Fischer-Tropsch reaction is conducted over copper or palladium to preferentially yield methanol. Any chemical pathway that produces methanol using products from the present water electrolysis and carbon dioxide capture technology is a potential pathway.

EXAMPLES

[0068] The present invention is more particularly described in the following examples that are intended as illustrations only, since numerous modifications and variations within the scope of the present invention will be apparent to those skilled in the art. Unless otherwise noted, all parts, percentages, and ratios reported in the following examples are on a weight basis, and all reagents used in the examples were commercially obtained, or may be synthesized by conventional techniques.

Example 1

[0069] A water electrolysis unit, shown in FIG. **8**, was constructed to demonstrate the feasibility of generating concentrated acid and base for carbon dioxide trapping. It consisted of a vertical central electrolyte feed tube about 2.5 centimeters (cm) in diameter, connected near its base to upward slanting anode and cathode tubes attached opposite one another. Wire, screen or flat, linear electrodes consisting of nickel, stainless steel or platinum were placed in the anode and cathode tubes near their points of attachment to the central tube. A concentrated, chloride-free electrolyte solution of aqueous sodium sulfate was introduced to the system via the central feed tube, creating an electrically conductive cell in which water was oxidized at the anode and reduced at the cathode. A small 15-watt solar panel was used to provide renewable electricity to the system.

[0070] When a DC current from the solar panel was applied to the system, hydrogen and hydroxide base were produced rapidly at the cathode while oxygen and acid formed at the anode. Hydrogen and oxygen gas flowed up the cathode and anode tubes, respectively, and were collected at the top. Acid and base accumulating in the anode and cathode tubes were collected via stopcock valves. Fresh electrolyte introduced to the central feed tube forced acid and base up the anode and cathode tubes, preventing them from recombining within the system. Within a few minutes of operation, the electrolyte in the anode cell had reached a pH of about 2, and in the cathode cell a pH of about 12, a differential of 10 pH units. Unlike the traditional chloralkali process for manufacturing hydroxide base, this renewable method of base production generated no chlorine or carbon dioxide. Sulfuric acid, a high demand commodity chemical was produced instead of chlorine.

[0071] Base produced in the cathode cell began to trap atmospheric carbon dioxide immediately, a process that was greatly enhanced by maximizing air-water exposure. This was achieved by bubbling air or gas through the basic solution or by spraying base through a column of air or carbon dioxide containing gas.

[0072] A passive trapping approach also demonstrated clear carbon dioxide capture from the air. A small amount (20 g) of crystalline NaOH was spread in a thin layer on a glass plate exposed to the air. Over the first few days the hygroscopic NaOH absorbed significant water vapor from the air, becoming a soggy mass of crystals. During the course of the next two weeks these crystals gradually dried and became opaque white in color, a visible change from the initial translucent NaOH crystals. The white crystals were a combination of sodium bicarbonate and sodium carbonate, formed from atmospheric carbon dioxide. Addition of an acid, vinegar, to these crystals resulted in vigorous bubbling as carbon dioxide was released back to the air.

Example 2

[0073] A second example used a 1-inch diameter glass tube sealed at the bottom with a porous glass frit. The frit allowed fluid and ion exchange between the inside and outside of the glass tube, creating an inner anode or cathode cell. Flat nickel or platinum electrodes were placed on opposite sides of the glass frit and attached to a 15 W DC photovoltaic panel. This system created a water electrolysis device that produced concentrated base inside the tube and concentrated acid outside the tube.

[0074] Depending on mode of operation, a pH differential of over 11 was quickly generated in this system; an acid-base concentration gradient of over 20 billion fold. The electrolyte inside the tube reached a pH of about 13, while across the frit, less than ¼ inch away, the electrolyte pH reached about 1.6. Vigorous production of hydrogen and oxygen were also observed.

[0075] A third example included a two-chamber flow-through system constructed from machined plastic. A peristaltic pump was used to circulate electrolyte solution into the anode and cathode chambers, which were physically separated by a semi-permeable membrane or filter. Variable width plastic spacers were used to vary the gaps between the electrodes and the membrane. A nickel-copper alloy was initially used as electrode material. Hydrogen and oxygen were collected at valves at the top of the device, and acid and base were continually circulated past the electrodes until sufficient concentrations were reached. A variable output DC power source was used to generate voltages sufficient to electrolyze water. Alternatively, a renewable energy source such as wind, solar, hydroelectric, geothermal or biomass energy could be used to power the device.

[0076] pH differentials of over about 10 units were quickly achieved and maintained in this system. The nickel-copper electrodes proved susceptible to corrosion at certain voltages. Corrosion-resistant electrodes such as nickel, platinum, carbon or stainless steel are best suited to the technology applications envisioned.

[0077] Overall, these experiments clearly demonstrate that water electrolysis can be used in an integrated strategy to produce renewable hydrogen and trap carbon dioxide from the air or gas streams. Given that renewable hydrogen produced by water electrolysis is already promoted as a clean alternative to fossil fuels, this combined renewable hydrogen/carbon dioxide capture technology represents a significant advance in reducing global carbon dioxide emissions.

Embodiments

[0078] Embodiment 1, is a method of generating renewable hydrogen and sequestering gaseous carbon dioxide comprising:

[0079] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one water electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0080] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0081] c) producing hydrogen gas and hydroxide ions at the cathode, wherein the hydroxide ions are present in the form of a base;

[0082] d) collecting the hydrogen gas product;

[0083] e) collecting the oxygen gas product;

[0084] f) removing some or all of the acid from the anode region;

[0085] g) removing some or all of the base from the cathode region; and

[0086] h) contacting the hydroxide ions in the base with a source of gaseous carbon dioxide to sequester carbon dioxide in solution as bicarbonate or carbonate or a mixture thereof.

[0087] A base produced according to the method of embodiment 1.

[0088] A building material comprising bicarbonate or carbonate produced according to the method of embodiment 1, wherein the building material is any one of a dry wall product, filled polyvinyl chloride, tile, grout, synthetic stone, filled resin, or an adhesive.

[0089] The method according to embodiment 1, further comprising contacting the acid produced at the anode region with a mineral compound to form a carbon dioxide sequestering material.

[0090] The method according to embodiment 1, further comprising contacting the acid produced at the anode region with a mineral compound to form a carbon dioxide sequestering material, wherein the mineral compound is any one of talc, clay mineral sepiolite, clay minerals, serpentine, asbestos, or mining byproducts.

[0091] The method according to embodiment 1, further comprising contacting the acid, hydrogen or other products produced by the processes described above with the gaseous carbon dioxide in an electrochemical cell to produce reduced carbon compounds having a general formula of $CmHxO_2n$, wherein m an integer between 1 and 6, x is an integer between 0 and 24, and n is an integer between 0 and 6.

[0092] The method according to embodiment 1, further comprising contacting the acid produced at the anode region with the gaseous carbon dioxide in an electrochemical cell to produce reduced carbon compound, wherein the reduced carbon compound is any one of formic acid, oxalic acid, formaldehyde, or methanol.

[0093] The method according to embodiment 1, further comprising chemically reducing carbon dioxide by reacting hydrogen gas produced by the system with carbon dioxide trapped by the system to produce carbon monoxide, a precursor for other synthetic processes.

[0094] The method according to embodiment 1, further comprising reacting the carbon dioxide gas and hydrogen produced at the cathode with nitrogen gas in an electrochemical process to produce urea.

[0095] The method according to embodiment 1, further comprising the steps of contacting the acid produced at the anode region with the carbonate/bicarbonate solution to produce carbon dioxide gas and contacting the carbon dioxide gas with a source of anhydrous ammonia under pressure to produce urea.

[0096] Embodiment 2 is a process of generating renewable hydrogen and producing bicarbonate comprising:

[0097] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0098] b) producing hydrogen gas and hydroxide ions at the cathode region;

[0099] c) removing some or all of the base comprising hydroxide ions from the cathode region of the electrolysis cell; and

[0100] d) contacting the hydroxide ions with gaseous carbon dioxide to produce a solution comprising bicarbonate, carbonate or a mixture thereof; and

[0101] e) isolating bicarbonate from the solution.

[0102] Embodiment 3 is a process of generating renewable hydrogen and producing carbonate comprising:

[0103] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0104] b) producing hydrogen gas and a base comprising hydroxide ions at the cathode region;

[0105] c) removing some or all of the base comprising hydroxide ions from the cathode region;

[0106] d) contacting the hydroxide ions with gaseous carbon dioxide to produce a solution comprising bicarbonate, carbonate, or a mixture thereof; and

[0107] e) isolating carbonate from the solution.

[0108] Embodiment 4 is a method of generating renewable hydrogen and producing carbon dioxide neutral or carbon dioxide negative acid comprising:

[0109] a) supplying a direct current from a renewable electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0110] b) producing oxygen gas and protons at the cathode region, wherein the protons are present in the form of an acid;

[0111] c) removing some or all of the acid from anode region;

[0112] d) collecting the acid in a reservoir; and

[0113] c) concentrating the acid.

[0114] Embodiment 5 is a method of generating renewable hydrogen and producing carbon dioxide negative or carbon dioxide neutral base comprising:

[0115] a) supplying a direct current from a renewable electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0116] b) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0117] c) removing some or all of the base from the cathode region;

[0118] d) collecting the base in a reservoir; and

[0119] e) concentrating the base.

[0120] Embodiment 6 is a method of generating and maintaining separate regions of concentrated hydronium ions and concentrated hydroxide ions comprising:

[0121] a) contacting a cathode region including at least one cathode adapted to generate hydrogen gas and hydroxide ions and an anode region including at least one anode adapted to generate oxygen gas and hydronium with an aqueous electrolyte;

[0122] b) applying a DC voltage between 1.2 and 10 volts to the anode and cathode;

[0123] c) removing the hydrogen gas and hydroxide ions from the cathode region; and

[0124] d) removing the oxygen gas and hydronium ions from the anode region.

[0125] The method of embodiment 6 further comprising the step of supplying the cathode and anode regions with fresh electrolyte utilizing bi-directional flow.

[0126] The method of embodiment 6 further comprising the step of creating convection currents within the anode and the cathode regions.

[0127] Embodiment 7 is a method of generating renewable hydrogen and producing a carbon dioxide sequestering compound comprising the steps of:

[0128] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, the anode and the cathode regions electrically connected by the electrolyte;

[0129] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0130] c) removing some or all of the acid from the anode region;

[0131] d) concentrating the acid, wherein the acid has a pH ranging from about pH=0 to about pH=5; and

[0132] e) contacting the acid with a material that when exposed to a strong acid is converted to a carbon dioxide sequestering solution.

[0133] The method according to embodiment 7, wherein the material is any one of a mineral clay sepiolite, serpentine, talc, asbestos, or a mining byproduct.

[0134] The method according to embodiment 7, further comprising the step of adding base to the carbon dioxide sequestering solution.

[0135] The method according to embodiment 7, further comprising the step of contacting the carbon dioxide sequestering solution with a source of gaseous carbon dioxide.

[0136] The method according to embodiment 7, further comprising the step of precipitating and processing magnesium salts from the carbon dioxide sequestering solution.

[0137] Embodiment 8 is a method of generating renewable hydrogen and producing pressurized carbon dioxide gas comprising:

[0138] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0139] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0140] c) producing hydrogen gas and hydroxide ions at the cathode, wherein the hydroxide ions are present in the form of a base;

[0141] d) removing some or all of the acid from the anode region;

[0142] e) removing some or all of the base from the cathode region;

[0143] f) contacting the base comprising hydroxide ions with gaseous carbon dioxide to produce a solution comprising bicarbonate, carbonate, or a mixture thereof;

[0144] g) contacting the solution with acid produced at the anode region of the cell in an enclosed chamber to produce carbon dioxide gas under pressure; and

[0145] h) collecting the pressurized carbon dioxide gas.

[0146] The method according to embodiment 8, further comprising producing super critical carbon dioxide from the collected pressurized carbon dioxide gas.

[0147] Embodiment 9 is a method of generating renewable hydrogen and producing carbon dioxide negative urea fertilizer, the method comprising the steps of:

[0148] a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0149] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0150] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0151] d) removing some or all of the acid from the anode region;

[0152] e) removing some or all of the base from the cathode region;

[0153] f) contacting the base having with a gaseous source of carbon dioxide to produce a feedstock comprising bicarbonate or carbonate or a mixture thereof;

[0154] g) contacting the acid produced at the anode region with the feedstock to produce carbon dioxide gas; and

[0155] h) contacting the carbon dioxide gas with a source of anhydrous ammonia under pressure to produce urea.

[0156] Embodiment 10 is a method of generating renewable hydrogen and producing carbon dioxide negative urea fertilizer, the method comprising the steps of:

[0157] a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0158] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0159] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0160] d) removing some or all of the acid from the anode region;

[0161] e) removing some or all of the base from the cathode region;

[0162] f) contacting the base having with a gaseous source of carbon dioxide to produce a feedstock comprising bicarbonate or carbonate or a mixture thereof;

[0163] g) contacting the acid produced at the anode region with the feedstock to produce carbon dioxide gas; and

[0164] h) reacting the carbon dioxide gas and hydrogen produced at the cathode with nitrogen gas in an electrochemical process to produce urea.

[0165] Embodiment 11 is a method of generating renewable hydrogen and producing carbon dioxide negative agricultural lime comprising the steps of:

[0166] a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0167] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0168] c) producing hydrogen gas and hydroxide ions at the cathode, wherein the hydroxide ions are present in the form of a base;

[0169] d) removing some or all of the acid from the anode region;

[0170] e) removing some or all of the base from the cathode region;

[0171] f) contacting the base having a pH of greater than about pH=10 with gaseous carbon dioxide to produce a carbonate enriched feedstock;

[0172] g) contacting the feedstock with an aqueous composition comprising calcium ions; and

[0173] h) precipitating calcium carbonate from the feedstock to produce agricultural lime.

[0174] Embodiment 12 is a method of generating renewable hydrogen and producing carbon dioxide neutral quick lime comprising:

[0175] a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0176] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0177] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0178] d) removing some or all of the acid from the anode region;

[0179] e) removing some or all of the base from the cathode region;

[0180] f) contacting the base having a pH of greater than about pH=10 with gaseous carbon dioxide to produce a carbonate enriched feedstock;

[0181] g) contacting the carbonate enriched feedstock with an aqueous solution comprising calcium ions to generate calcium carbonate;

[0182] h) applying heat to the calcium carbonate to produce quick lime.

[0183] The method according to embodiment 12, wherein carbon dioxide released from applying heat to the calcium carbonate is contacted with base to regenerate the carbonate feedstock.

[0184] The method according to embodiment 12, wherein carbon dioxide released from applying heat to the calcium carbonate is sequestered and pressurized for use as supercritical carbon dioxide.

[0185] Embodiment 13 is a method of generating renewable hydrogen and producing carbon dioxide neutral or carbon dioxide negative carbon monoxide comprising:

[0186] a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0187] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0188] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0189] d) removing some or all of the acid from the anode region;

[0190] e) removing some or all of the base from the cathode region;

[0191] f) contacting the base with gaseous carbon dioxide to produce a solution comprising bicarbonate or carbonate or a mixture thereof;

[0192] g) contacting the solution with acid produced in the anode region of the cell to produce carbon dioxide gas under pressure;

[0193] h) collecting the pressurized carbon dioxide gas; and

[0194] i) reducing a portion of the carbon dioxide gas with a portion of the hydrogen gas generated at the cathode to produce carbon monoxide.

[0195] Embodiment 14 is a method of generating renewable hydrogen and producing formic acid comprising:

[0196] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0197] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0198] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0199] d) removing some or all of the acid from the anode region;

[0200] e) removing some or all of the base from the cathode region;

[0201] f) contacting the base with a gaseous carbon dioxide to produce a solution comprising bicarbonate or carbonate or a mixture thereof;

[0202] g) contacting the solution with acid produced in the anode region of the cell to produce carbon dioxide gas under pressure;

[0203] h) collecting the pressurized carbon dioxide gas;

[0204] i) reducing a portion of the carbon dioxide gas with a portion of hydrogen gas generated at the cathode to produce carbon monoxide;

[0205] j) reacting a portion of the carbon monoxide with methanol in the presence of the base produced at the cathode having a pH ranging of at least 10 to produce methyl formate; and

[0206] k) hydrolyzing the methyl formate to produce formic acid.

[0207] Embodiment 15, is a method of generating renewable hydrogen and producing formic acid comprising:

[0208] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0209] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0210] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0211] d) removing some or all of the acid from the anode region;

[0212] e) removing some or all of the base from the cathode region;

[0213] f) contacting the base having a with gaseous carbon dioxide to produce a solution comprising cesium bicarbonate; and

[0214] g) electrolyzing cesium bicarbonate to form formic acid.

[0215] Embodiment 16 is a method of generating renewable hydrogen and producing formic acid comprising:

[0216] a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

[0217] b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

[0218] c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

[0219] d) removing some or all of the acid from the anode region;

[0220] e) removing some or all of the base from the cathode region;

[0221] f) contacting the base having a with gaseous carbon dioxide to produce a solution comprising bicarbonate or carbonate or a mixture thereof; and

[0222] g) hydrogenating the bicarbonate in the presence of a catalyst to produce formic acid.

[0223] The method according to any one of embodiments 1-16, wherein the source of gaseous carbon dioxide is atmospheric carbon dioxide.

[0224] The method according to any one of embodiments 1-16, wherein the source of gaseous carbon dioxide is a gas stream.

[0225] The method according to any one of embodiments 1-16, wherein an amount of carbon dioxide sequestered is greater than an amount of carbon dioxide generated by the electrical source.

[0226] The method according to any one of embodiments 1-16, further comprising applying to the water electrolysis cell a counter-current flow adapted to introduce concentrated electrolyte into a central feed chamber separated by semi-permeable membranes from the anode and cathode regions, such that the direction of flow of the electrolyte is opposite to the direction of flow of acid and base produced at the anode and cathode regions.

[0227] The method according to any one of embodiments 1-16, further comprising maintaining a separate anode and cathode region by providing at least one ion selective membrane positioned between the anode region and the cathode region of the electrolysis cell.

[0228] The method according to any one of embodiments 1-16, further comprising maintaining a separate anode and cathode region by providing at least one non-selective, semi-permeable membrane positioned between the anode region and the cathode region of the electrolysis cell.

[0229] The method according to any one of embodiments 1-16, further comprising continuously supplying fresh electrolyte to the electrolysis cell.

[0230] The method according to any one of embodiments 1-16, further comprising supplying fresh electrolyte to the electrolysis cell in a batch-wise manner.

[0231] The method according to any one of embodiments 1-16, further comprising maintaining a pH difference between the anode region and the cathode region of at least 6 pH units.

[0232] The method according to any one of embodiments 1-16, wherein the pH of the anode region ranges from about pH=0 to about pH=5.

[0233] The method according to any one of embodiments 1-16, wherein the pH of the cathode region ranges from about pH=8 to about pH=14.

[0234] The method according to any one of embodiments 1-16, wherein the electricity supplied from the source ranges from about 1.2 to about 10.0 volts.

[0235] The method according to any one of embodiments 1-16, wherein the voltage supplied from the energy source is at least 1.2 volts.

[0236] The method according to any one of embodiments 1-16, wherein a pH of the base produced at the cathode region ranges from about pH=8 to about pH=14.

[0237] The method according to any one of embodiments 1-16, further comprising applying positive pressure to the electrolysis cell to remove the acid from the anode region and the base from the cathode region.

[0238] The method according to any one of embodiments 1-16, further comprising applying a gravity feed to the electrolysis cell to remove the acid from the anode region and the base from the cathode region.

[0239] The method according to any one of embodiments 1-16, further comprising continuously pumping acid from the anode region to an acid storage region and the base from the cathode region to a base storage region.

[0240] The method according to any one of embodiments 1-16, wherein the electrical source is a renewable energy source.

[0241] The method according to any one of embodiments 1-16, further comprising recycling the hydrogen gas to a renewable energy source to be utilized as a fuel.

[0242] The method according to any one of embodiments 1-16, further comprising recycling the hydrogen gas to a renewable energy source to be utilized as a fuel wherein the renewable energy source is a fuel cell powered by renewable hydrogen, a synthetic fuel, biomass, or biofuel.

[0243] The method according to any one of embodiments 1-16, further comprising supplying renewable energy from a renewable energy source to the electrical energy source wherein the renewable energy source is any one of a wind turbine, solar cell, hydroelectric generator, biofuel, geothermal, or oceanic.

[0244] The method according to any one of embodiments 1-16, further comprising recycling the hydrogen gas to generate renewable electricity.

[0245] The method according to any one of embodiments 1-16, further comprising recycling the hydrogen gas and the oxygen gas to regenerate water and electricity supplied to the electrolysis unit.

[0246] The method according to any one of embodiments 1-16, wherein the electrical energy source does not consume fossil based fuels.

[0247] The method according to any one of embodiments 1-16, wherein the electrical energy source is a nuclear generator.

[0248] The method according to any one of embodiments 1-16, wherein the aqueous electrolyte comprises an alkali salt substantially free of chloride ions.

[0249] The method according to any one of embodiments 1-16, wherein the aqueous electrolyte comprises a sodium salt substantially free of chloride ions.

[0250] The method according to any one of embodiments 1-16, wherein the aqueous electrolyte is selected from the group consisting of sodium sulfate, potassium sulfate, calcium sulfate, magnesium sulfate, sodium nitrate, potassium nitrate, sodium bicarbonate, sodium carbonate, potassium bicarbonate, potassium carbonate, calcium carbonate, and magnesium carbonate.

[0251] The method according to any one of embodiments 1-16, wherein the aqueous electrolyte comprises sea water and/or sea salt.

[0252] The method according to any one of embodiments 1-16, wherein the electrolyte is saturated in solution.

[0253] The method according to any one of embodiments 1-16, further comprising maintaining a saturated electrolyte solution.

[0254] The method according to any one of embodiments 1-16, wherein the aqueous electrolyte is sodium sulfate or potassium sulfate.

[0255] The method according to any one of embodiments 1-16, further comprising isolating bicarbonate or carbonate from the solution.

[0256] The method according to any one of embodiments 1-16, further comprising isolating bicarbonate or carbonate from the solution by precipitating bicarbonate or carbonate from the solution.

[0257] The method according to any one of embodiments 1-16, further comprising the step of concentrating the solution.

[0258] The method according to any one of embodiments 1-16, further comprising chilling the solution to a temperature ranging from about 0° C. to about 10° C. to precipitate carbonate or bicarbonate from the solution.

[0259] The method according to any one of embodiments 1-16, further comprising adding a salt comprising calcium ions to isolate calcium carbonate from the solution.

[0260] The method according to any one of embodiments 1-16, further comprising adding a salt comprising magnesium ions to isolate magnesium carbonate from the solution.

[0261] The method according to any one of embodiments 1-16, further comprising spraying the solution to precipitate carbonate or bicarbonate from the solution.

[0262] The method according to any one of embodiments 1-16, further comprising regenerating electrolyte supplied to the electrolysis unit by reacting the acid removed from the anode region of the electrolysis cell with sodium chloride to produce hydrochloric acid and the original electrolyte salt.

[0263] The method according to any one of embodiments 1-16, further comprising collecting and concentrating the base.

[0264] The method according to any one of embodiments 1-16, wherein the acid is sulfuric acid.

[0265] The method according to any one of embodiments 1-16, wherein the base is sodium hydroxide.

[0266] The method according to any one of embodiments 1-16, wherein a difference of at least 6 pH units is maintained between the anode region and cathode region by supplying electrolyte utilizing bi-direction flow.

[0267] The method according to any one of embodiments 1-16, wherein a difference of at least 6 pH units is maintained between the anode region and cathode region utilizing convection currents generated by rising hydrogen gas in the cathode region and rising oxygen gas in the anode region.

[0268] The method according to any one of embodiments 1-16 further comprising concentrating cations from the electrolyte and delivering the cations the cathode region.

[0269] The method according to any one of embodiments 1-16 further comprising concentrating anions from the electrolyte and delivering the anions the anode region.

[0270] The method according to any one of embodiments 1-16, further comprising the step of separating the anode and the cathode region with a porous glass frit.

[0271] Although the present invention has been described with reference to preferred embodiments, workers skilled in the art will recognize that changes may be made in form and detail without departing from the spirit and scope of the invention.

1. A method of generating renewable hydrogen and sequestering gaseous carbon dioxide comprising:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one water electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;
b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;
c) producing hydrogen gas and hydroxide ions at the cathode, wherein the hydroxide ions are present in the form of a base;
d) collecting the hydrogen gas product;
e) collecting the oxygen gas product;
f) removing some or all of the acid from the anode region;
g) removing some or all of the base from the cathode region; and
h) contacting the hydroxide ions in the base with a source of gaseous carbon dioxide to sequester carbon dioxide in solution as bicarbonate or carbonate or a mixture thereof.

2. The method according to claim **1**, wherein the source of gaseous carbon dioxide is atmospheric carbon dioxide.

3. The method according to claim **1**, wherein the source of gaseous carbon dioxide is a gas stream.

4. The method according to claim **1**, further comprising isolating bicarbonate or carbonate from the solution.

5. The method according to claim **4**, wherein isolating bicarbonate or carbonate from the solution comprises precipitating bicarbonate or carbonate from the solution.

6. The method according to claim **4**, further comprising the step of concentrating the solution.

7. The method according to claim **1**, further comprising chilling the solution to a temperature ranging from about 0° C. to about 10° C. to precipitate carbonate or bicarbonate from the solution.

8. The method according to claim **1**, further comprising adding a salt comprising calcium ions to isolate calcium carbonate from the solution.

9. The method according to claim **1**, further comprising adding a salt comprising magnesium ions to isolate magnesium carbonate from the solution.

10. The method according to claim **1**, further comprising spraying the solution to precipitate carbonate or bicarbonate from the solution.

11. The method according to claim **1**, further comprising applying to the water electrolysis cell a counter-current flow adapted to introduce concentrated electrolyte into a central feed chamber separated by semi-permeable membranes from the anode and cathode regions, such that the direction of flow of the electrolyte is opposite to the direction of flow of acid and base produced at the anode and cathode regions.

12. The method according to claim **1**, further comprising maintaining a separate anode and cathode region by providing at least one ion selective membrane positioned between the anode region and the cathode region of the electrolysis cell.

13. The method according to claim **1**, further comprising maintaining a separate anode and cathode region by providing at least one non-selective, semi-permeable membrane positioned between the anode region and the cathode region of the electrolysis cell.

14. The method according to claim **1**, further comprising continuously supplying fresh electrolyte to the electrolysis cell.

15. The method according to claim **1**, further comprising supplying fresh electrolyte to the electrolysis cell in a batchwise manner.

16. The method according to claim **1**, further comprising maintaining a pH difference between the anode region and the cathode region of at least 6 pH units.

17. The method according to claim **1**, wherein the electricity supplied from the source ranges from about 1.2 to about 10.0 volts.

18. The method according to claim **1**, wherein the voltage supplied from the energy source is at least 1.2 volts.

19. The method according to claim **1**, wherein a pH of the base produced at the cathode region ranges from about pH=8 to about pH=14.

20. The method according to claim **1**, further comprising applying positive pressure to the electrolysis cell to remove the acid from the anode region and the base from the cathode region.

21. The method according to claim **1**, further comprising applying a gravity feed to the electrolysis cell to remove the acid from the anode region and the base from the cathode region.

22. The method according to claim **1**, further comprising continuously pumping acid from the anode region to an acid storage region and the base from the cathode region to a base storage region.

23. The method according to claim **1**, wherein the electrical source is a renewable energy source.

24. The method according to claim **23**, further comprising recycling the hydrogen gas to the renewable energy source to be utilized as a fuel.

25. The method according to claim **23**, wherein the renewable energy source is a fuel cell powered by renewable hydrogen, a synthetic fuel, biomass, or biofuel.

26. The method according to claim **23**, wherein the renewable energy source is any one of a wind turbine, solar cell, hydroelectric generator, biofuel, geothermal, or oceanic.

27. The method according to claim **1**, further comprising recycling the hydrogen gas to generate renewable electricity.

28. The method according to claim **1**, further comprising recycling the hydrogen gas and the oxygen gas to regenerate water and electricity supplied to the electrolysis unit.

29. The method according to claim **1**, wherein the source does not consume fossil based fuels.

30. The method according to claim **1**, wherein the electrical source is a nuclear generator.

31. The method according to claim **1**, wherein the aqueous electrolyte comprises an alkali salt substantially free of chloride ions.

32. The method according to claim **1**, wherein the aqueous electrolyte comprises a sodium salt substantially free of chloride ions.

33. The method according to claim **1**, wherein the aqueous electrolyte is selected from the group consisting of sodium sulfate, potassium sulfate, calcium sulfate, magnesium sulfate, sodium nitrate, potassium nitrate, sodium bicarbonate, sodium carbonate, potassium bicarbonate, potassium carbonate, calcium carbonate, and magnesium carbonate.

34. The method according to claim **1**, wherein the electrolyte is saturated in solution.

35. The method according to claim **1**, further comprising maintaining a saturated electrolyte solution.

36. The method according to claim **1**, wherein the aqueous electrolyte is sodium sulfate or potassium sulfate.

37. The method according to claim **1**, further comprising regenerating electrolyte supplied to the electrolysis unit by reacting the acid removed from the anode region of the electrolysis cell with sodium chloride to produce hydrochloric acid and the original electrolyte salt.

38. The method according to claim **1**, further comprising collecting and concentrating the base.

39. A base produced according to the method of claim **1**.

40. A building material comprising carbonate produced according to the method of claim **1**, wherein the building material is any one of a dry wall product, filled polyvinyl chloride, tile, grout, synthetic stone, filled resin, or an adhesive.

41. The method according to claim **1**, further comprising contacting the acid produced at the anode region with a mineral compound to form a carbon dioxide sequestering material.

42. The method according to claim **42**, wherein the mineral compound is any one of talc, clay mineral sepiolite, clay minerals, serpentine, asbestos, or mining byproducts.

43. The method according to claim **1**, further comprising contacting the acid produced at the anode region with the gaseous carbon dioxide in an electrochemical cell to produce reduced carbon compounds having a general formula of $CmHxO_2n$, wherein m an integer between 1 and 6, x is an integer between 0 and 24, and n is an integer between 0 and 6.

44. The method according to claim **43**, wherein the reduced carbon compound is any one of formic acid, oxalic acid, formaldehyde, or methanol.

45. The method according to claim **1**, further comprising chemically reducing carbon dioxide by reacting hydrogen gas produced by the system with carbon dioxide trapped by the system to produce carbon monoxide, a precursor for other synthetic processes.

46. The method according to claim **1**, wherein an amount of carbon dioxide sequestered is greater than an amount of carbon dioxide generated by the electrical source.

47. The method according to claim **1**, wherein a difference of at least 6 pH units is maintained between the anode region and cathode region by supplying electrolyte utilizing bi-direction flow.

48. The method according to claim **1**, wherein a difference of at least 6 pH units is maintained between the anode region and cathode region utilizing convection currents generated by rising hydrogen gas in the cathode region and rising oxygen gas in the anode region.

49. The method according to claim **1** further comprising concentrating cations from the electrolyte and delivering the cations to the cathode region.

50. The method according to claim **1** further comprising concentrating anions from the electrolyte and delivering the anions to the anode region.

51. A process of producing renewable hydrogen from water and bicarbonate from gaseous carbon dioxide comprising:

supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

producing hydrogen gas and hydroxide ions at the cathode region;

removing some or all of the base comprising hydroxide ions from the cathode region of the electrolysis cell; and

contacting the hydroxide ions with gaseous carbon dioxide to produce a solution comprising bicarbonate, carbonate or a mixture thereof; and

isolating bicarbonate from the solution.

52. A process of producing renewable hydrogen from water and carbonate from gaseous carbon dioxide comprising:

supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

producing hydrogen gas and a base comprising hydroxide ions at the cathode region;

removing some or all of the base comprising hydroxide ions from the cathode region;

contacting the hydroxide ions with gaseous carbon dioxide to produce a solution comprising bicarbonate, carbonate, or a mixture thereof; and

isolating carbonate from the solution.

53. A method of producing renewable hydrogen and carbon dioxide neutral or carbon dioxide negative acid comprising:

a) supplying a direct current from a renewable electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the cathode region, wherein the protons are present in the form of an acid;

c) removing some or all of the acid from anode region;

d) collecting the acid in a reservoir; and

e) concentrating the acid.

54. The method according to claim **53**, wherein a pH of the cathode region ranges from about pH=0 to about pH=5.

55. The method according to claim **53**, wherein the acid is sulfuric acid.

56. A method of producing renewable hydrogen and a carbon dioxide neutral or carbon dioxide negative base comprising:

a) supplying a renewable electric current from a source to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

c) removing some or all of the base from the cathode region;

d) collecting the base in a reservoir; and

e) concentrating the base.

57. The method according to claim **56**, wherein a pH of the base region ranges from about pH=8 to about pH=14.

58. The method according to claim **56**, wherein the base is sodium or potassium hydroxide.

59. A method of generating and maintaining separate regions of concentrated hydronium ions and concentrated hydroxide ions comprising:

a) contacting a cathode region including at least one cathode adapted to generate hydrogen gas and hydroxide ions and an anode region including at least one anode adapted to generate oxygen gas and hydronium with an aqueous electrolyte;

b) applying a DC voltage between 1.2 and 10 volts to the anode and cathode;

c) removing the hydrogen gas and hydroxide ions from the cathode region; and

d) removing the oxygen gas and hydronium ions from the anode region.

60. The method of claim **60**, further comprising the step of supplying the cathode and anode regions with fresh electrolyte utilizing bi-directional flow.

61. The method of claim **60** further comprising the step of creating convection currents within the anode and the cathode regions.

62. The method of claim **60**, further comprising the step of separating the anode and the cathode region with a porous glass frit.

63. A method of generating renewable hydrogen and producing a carbon dioxide sequestering compound comprising the steps of:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, the anode and the cathode regions electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) removing some or all of the acid from the anode region;

d) concentrating the acid, wherein the acid has a pH ranging from about pH=0 to about pH=5; and

e) contacting the acid with a material that when exposed to a strong acid is converted to a carbon dioxide sequestering solution.

64. The method according to claim **63**, wherein the material is any one of a mineral clay sepiolite, serpentine, talc, asbestos, or a mining byproduct.

65. The method according to claim **63**, wherein the acid is sulfuric acid.

66. The method according to claim **63**, further comprising the step of adding base to the carbon dioxide sequestering solution.

67. The method according to claim **63**, further comprising the step of contacting the carbon dioxide sequestering solution with a source of gaseous carbon dioxide.

68. The method according to claim **67**, further comprising the step of precipitating and processing magnesium salts from the carbon dioxide sequestering solution.

69. A method of generating renewable hydrogen and producing pressurized carbon dioxide gas from atmospheric or gas stream carbon dioxide comprising:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) producing hydrogen gas and hydroxide ions at the cathode, wherein the hydroxide ions are present in the form of a base;

d) removing some or all of the acid from the anode region;

e) removing some or all of the base from the cathode region;

f) contacting the base comprising hydroxide ions with gaseous carbon dioxide to produce a solution comprising bicarbonate, carbonate, or a mixture thereof;

g) contacting the solution with acid produced at the anode region of the cell to produce carbon dioxide gas under pressure; and

h) collecting the pressurized carbon dioxide gas.

70. The method according to claim **69**, further comprising producing super critical carbon dioxide from the collected pressurized carbon dioxide gas.

71. A method of generating renewable hydrogen and producing urea fertilizer, the method comprising the steps of:

a) supplying an electric current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

d) removing some or all of the acid from the anode region;

e) removing some or all of the base from the cathode region;

f) contacting the base with a gaseous source of carbon dioxide to produce a feedstock comprising bicarbonate or carbonate or a mixture thereof;

g) contacting the acid produced at the anode region with the feedstock to produce carbon dioxide gas; and

h) contacting the carbon dioxide gas with a source of anhydrous ammonia under pressure to produce urea.

72. A method of generating renewable hydrogen and producing urea fertilizer, the method comprising the steps of:

a) supplying an electric current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

d) removing some or all of the acid from the anode region;

e) removing some or all of the base from the cathode region;

f) contacting the base with a gaseous source of carbon dioxide to produce a feedstock comprising bicarbonate or carbonate or a mixture thereof;

g) contacting the acid produced at the anode region with the feedstock to produce carbon dioxide gas; and

h) reacting the carbon dioxide gas and hydrogen produced at the cathode with nitrogen gas in an electrochemical process to produce urea.

73. A method of generating renewable hydrogen and producing carbon dioxide neutral or carbon dioxide negative agricultural lime comprising the steps of:

a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) producing hydrogen gas and hydroxide ions at the cathode, wherein the hydroxide ions are present in the form of a base;

d) removing some or all of the acid from the anode region;

e) removing some or all of the base from the cathode region;

f) contacting the base having a pH of greater than about pH=10 with gaseous carbon dioxide to produce a carbonate enriched feedstock;

g) contacting the feedstock with an aqueous composition comprising calcium ions; and

h) precipitating calcium carbonate from the feedstock to produce agricultural lime.

74. A method of generating renewable hydrogen and producing carbon dioxide neutral or carbon dioxide negative quick lime comprising:

a) supplying a direct current from a renewable or nuclear electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

d) removing some or all of the acid from the anode region;

e) removing some or all of the base from the cathode region;

f) contacting the base having a pH of greater than about pH=10 with gaseous carbon dioxide to produce a carbonate enriched feedstock;

g) contacting the carbonate enriched feedstock with an aqueous solution comprising calcium ions to generate calcium carbonate;

h) applying heat to the calcium carbonate to produce quick lime.

75. The method according to claim **74**, wherein carbon dioxide released from applying heat to the calcium carbonate is contacted with base to regenerate the carbonate feedstock.

76. The method according to claim **74**, wherein carbon dioxide released from applying heat to the calcium carbonate is sequestered and pressurized for use as supercritical carbon dioxide.

77. A method of generating renewable hydrogen and producing carbon monoxide from atmospheric or gas stream carbon dioxide comprising:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;

b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;

c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;

d) removing some or all of the acid from the anode region;

e) removing some or all of the base from the cathode region;

f) contacting the base with gaseous carbon dioxide to produce a solution comprising bicarbonate or carbonate or a mixture thereof;
g) contacting the solution with acid produced in the anode region of the cell to produce carbon dioxide gas under pressure;
h) collecting the pressurized carbon dioxide gas; and
i) reducing a portion of the carbon dioxide gas with a portion of the hydrogen gas generated at the cathode to produce carbon monoxide.

78. A method of generating renewable hydrogen and producing formic acid from atmospheric or gas stream carbon dioxide comprising:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;
b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;
c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;
d) removing some or all of the acid from the anode region;
e) removing some or all of the base from the cathode region;
f) contacting the base with a gaseous carbon dioxide to produce a solution comprising bicarbonate or carbonate or a mixture thereof;
g) contacting the solution with acid produced in the anode region of the cell to produce carbon dioxide gas under pressure;
h) collecting the pressurized carbon dioxide gas;
i) reducing a portion of the carbon dioxide gas with a portion of hydrogen gas generated at the cathode to produce carbon monoxide; and
j) reacting a portion of the carbon monoxide with methanol in the presence of the base produced at the cathode having a pH of at least 10 to produce methyl formate; and
k) hydrolyzing the methyl formate to produce formic acid.

79. A method of generating renewable hydrogen and producing formic acid from atmospheric or gas stream carbon dioxide comprising:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;
b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;
c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;
d) removing some or all of the acid from the anode region;
e) removing some or all of the base from the cathode region;
f) contacting the base having a with gaseous carbon dioxide to produce a solution comprising cesium bicarbonate; and
g) electrolyzing cesium bicarbonate to form formic acid.

80. A method of generating renewable hydrogen and producing formic acid from atmospheric or gas stream carbon dioxide comprising:

a) supplying a direct current from an electrical source at a predetermined voltage to a water electrolysis unit having at least one electrolysis cell including an aqueous electrolyte substantially free of chloride ions and an anode region adapted to generate oxygen gas and protons separated from a cathode region adapted to generate hydrogen gas and hydroxide ions, wherein the anode and the cathode regions are electrically connected by the electrolyte;
b) producing oxygen gas and protons at the anode region, wherein the protons are present in the form of an acid;
c) producing hydrogen gas and hydroxide ions at the cathode region, wherein the hydroxide ions are present in the form of a base;
d) removing some or all of the acid from the anode region;
e) removing some or all of the base from the cathode region;
f) contacting the base with gaseous carbon dioxide to produce a solution comprising bicarbonate or carbonate or a mixture thereof; and
g) hydrogenating the bicarbonate in the presence of a catalyst to produce formic acid.

* * * * *