CAVITATION ASSISTED SONOCHEMICAL HYDROGEN PRODUCTION SYSTEM

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Abstract
Embodiments of the invention are directed to an electrolyte composition of matter comprising an aqueous solvent, at least one inorganic salt dissolved in said solvent, and at least one organic acid or salt thereof, the acid being substantially soluble in the aqueous solvent and being present at a concentration sufficient to support cavitation-assisted electrolytic combustion. Low molecular weight organic acids and inorganic salts are demonstrated herein as being useful for generating hydrogen.
FIG. 5
Effects of Cavitation on Hydrogen Production (Over 10 Minutes)

FIG. 12
FIG. 13

Citric Acid

Flow of Hydrogen (SLPM)

Time (Minutes)

0.00 0.50 1.00 1.50 2.00 2.50 3.00 3.50 4.00

- 0.1M
- 1.0M
Propanoic Acid

FIG. 15
Malonic Acid

Time (Minutes)

Flow of Hydrogen (SLPM)

- 0.1M
- 1.0M

FIG. 16
CAVITATION ASSISTED SONOCHEMICAL HYDROGEN PRODUCTION SYSTEM

CROSS REFERENCE TO RELATED APPLICATIONS

[0001] This application is a continuation-in-part of U.S. Ser. No. 12/166,979 filed Jul. 2, 2008, pending, and to which priority is claimed. This application also claims priority to U.S. Provisional Application No. 61/450,569, filed Mar. 8, 2011. Both documents are incorporated herein by reference in their entirety.

FIELD OF THE INVENTION

[0002] The present invention generally relates to efficient generation of hydrogen and more specifically to in-situ hydrogen generation.

BACKGROUND OF THE INVENTION

[0003] Water is composed of two parts hydrogen and one part oxygen by mass or volume. Decomposed by any means, two moles of water will produce one mole of oxygen gas (O2) and two moles of hydrogen gas (H2) at a given input of energy E1. When combined together through any means, hydrogen and oxygen react to form water, releasing a given output of energy E2. By all known principles of physics and chemistry, E1=E2 and thus by thermodynamics the process is not favored in direct action. For hydrogen to be useful as an energy source and economical to use, a means must be created to either reduce the dissociation energy of water, or provide energy in some other fashion in the process, for example with catalytic enhancement, or all the above.

[0004] Hydrogen can be manufactured by a variety of means (including, but not limited to chemical, electrical, thermal, radiolysis, etc.) from a variety of chemical substances (including, but not limited to, water, hydrocarbons, plants, rocks, etc.). In the present invention water is used as the hydrogen source and a catalytic combination of electrolysis and cavitation is used to generate the hydrogen. The method of cavitation may be a variety of means (acoustical, hydrodynamic inertial, non-inertial, mechanical, electromagnetic, etc.), or any combination thereof.

[0005] Hydrogen, being the most abundant element on earth as well as in the Universe, holds particular promise as a fuel source, both on earth as well as in space. Hydrogen can power homes and factories, transportation modes (planes, trains, and vehicles). Thus, hydrogen can serve to eliminate carbon fuels completely in the electrical cycle, thus bringing about a net subtraction by the contribution of anthropomorphic processes to terrestrial climate change. There are four significant "hurdles" cited by numerous reviews to the use of hydrogen. Each is noted as follows.

[0006] 1. Production—How to produce massive amounts of hydrogen in an efficient, safe, environmentally 'friendly' fashion.

[0007] 2. Storage—How to store the low density, flammable gas.

[0008] 3. Distribution—Hydrogen, being difficult to store, is thus difficult to transport.

[0009] 4. Use—How can hydrogen be used is a bigger hurdle in light of the prior two items.

[0101] Accordingly what is needed is a method and system to overcome the problems encountered in the prior art and to provide an economical method and apparatus to produce hydrogen.

SUMMARY OF THE INVENTION

[0111] A method and an apparatus to generate hydrogen gas as H2 from a hydrogen containing liquid such as water. In one embodiment, the structure is a electrolytic cell configured with catalytic enhancements to maximize the volume and mass of hydrogen produced, and minimize the energy input, thus minimizing cost of operation. This device is particularly configured to enhance catalytically the decomposition of water and the formation of hydrogen gas by: 1) the container apparatus configuration of electric and magnetic fields; 2) the use of sonocatalysts and cavitation; and 3) the use of applicable solutes and solvents in the device that change the pH, ionic state, and the chemical potential of the device solution.

[0102] The cavitation may be generated by a variety of means including but not limited to, acoustic energy, hydrodynamic (inertial, non-inertial), mechanical, electromagnetic energy, etc., or any combination thereof.

[0113] There are four significant "hurdles" cited by numerous reviews to the use of hydrogen. Each is noted as follows.

[0114] 1. Production—How to produce massive amounts of hydrogen in an efficient, safe, environmentally 'friendly' fashion. This patent is capable of producing hydrogen from water, and by any fashion in its recombination with oxygen to reform water, producing no pollution whatsoever and returning water back to its original form.

[0125] 2. Storage—How to store the low density, flammable gas. This patent eliminates the need for storage, by creating a scalable process to generate hydrogen from water in-situ wherever it is needed. It thus eliminates the need for dangerous, costly, and hazardous storage and transport issues.

[0116] 3. Distribution—Hydrogen, being difficult to store, is thus difficult to transport. Again, this patent eliminates the need for storage and thus transport, by creating a scalable process to generate hydrogen from water in-situ wherever it is needed. There is no need for dangerous, costly, and hazardous storage, distribution, and transport issues.

[0117] 4. Use—How can hydrogen be used is a bigger hurdle in light of the prior two items. With the elimination of those two items, the relative cost of the use of fuel cells becomes economical even to the middle class. Without the need for refueling, or by minimizing the need for refueling, the ability to use fuel cells will become ubiquitous to modern life.

[0118] A method and apparatus of producing hydrogen is disclosed comprising applying an electrical current to flow through an aqueous solution. Cavitation is generated within the aqueous solution, where the cavitation lowers an amount of energy required to break chemical bonds of said aqueous solution.

[0119] Embodiments of the invention are also directed to classes of chemical moieties that enhance the hydrogen content of the electrolyte and can be used to generate hydrogen in the process and apparatus described herein. An embodiment of the electrolyte composition of matter comprises an aqueous solvent, at least one inorganic salt dissolved in said solvent; and at least one organic acid or salt thereof; the acid being substantially soluble in the aqueous solvent and present in a concentration sufficient to support cavitation-assisted
Another embodiment of the invention includes the electrolyte composition wherein the aqueous solvent comprises at least 50% of said electrolyte on a wt/vol. basis. Another embodiment of the invention includes the electrolyte composition wherein the aqueous solvent comprises from about 50% to about 95% of said electrolyte on a weight to volume basis. Another embodiment of the invention includes the electrolyte composition wherein a noble gas is dissolved in the aqueous solvent. Another embodiment of the invention includes the noble gas Argon. Another embodiment of the invention includes the noble gas saturating the solvent.

Another embodiment of the invention includes the electrolyte composition wherein the inorganic salt comprises a cation selected from the group consisting of sodium, lithium and potassium. Another embodiment of the invention may include the inorganic salt comprising an anion selected from the group consisting of fluoride, chloride, bromide and iodide. Another embodiment of the invention includes the electrolyte composition wherein the inorganic salt comprises NaCl. Another embodiment of the invention includes the electrolyte composition wherein the inorganic salt comprises NaI.

Another embodiment of the invention includes the electrolyte composition wherein the organic acid is present in the electrolyte at a wt/vol. concentration of from about 1% to about 10%. Another embodiment of the invention includes the electrolyte composition wherein the organic acid comprises a plurality of carboxyl groups.

Another embodiment of the invention includes the electrolyte composition wherein the organic acid is substantially soluble in the electrolyte solution. “Substantially soluble” means the organic acid is at least 10 wt. % soluble in the electrolyte solution. Another embodiment of the invention includes the electrolyte composition wherein said at least one organic acid solubility is from about 10 weight % to about 90 weight % soluble.

Another embodiment of the invention includes the electrolyte composition wherein the organic acid comprises a main carbon chain having from one to eight carbons in the main chain (“C1-C8”).

Another embodiment of the invention includes the electrolyte composition wherein the organic acid is selected from the group consisting of Acetic acid, Citric acid, Fumaric acid, Malic acid, Malonic acid, Mandelic acid, Maleic acid, Iso-citric acid, Formic acid, Lactic acid, Glycolic acid, Glyceric acid, Hexanoic acid, Heptanoic acid, Octanoic acid, Pentanoic acid, Nonanoic acid, Propanoic acid, 3-Hydroxypropanoic acid, Tartaric acid and 2-Hydroxypropanedioic acid.

Another embodiment of the invention includes the electrolyte composition wherein the at least one organic acid is citric acid. The citric acid may be present in the electrolyte at a concentration of from about 0.05 M to about 5.0 M. In a preferred embodiment, the electrolyte composition has a citric acid concentration of about 0.1 M. Yet another embodiment of the invention includes the electrolyte composition wherein the concentration of citric acid is about 0.1M, NaCl is about 0.5 to 5 weight %, and NaI is present in a trace amount.

Another embodiment of the invention includes the electrolyte composition wherein the electrolyte is saturated with an amount of Argon sufficient to promote cavitation-assisted electrolytic combustion.

Another embodiment of the invention includes the electrolyte composition wherein the organic acid comprises the structural formula:

$$C_nH_{2n+1}COOR$$

wherein R can be individually any of H or any alkaline earth metal; and

Another embodiment of the invention includes the electrolyte composition wherein the organic acid comprises the structural formula:

$$ROOC-C_nH_{2n+1}COOR$$

wherein R can be individually any of H or any alkaline earth metal; and

n is from 2 to 8.

Another embodiment of the invention includes the electrolyte composition wherein the organic acid comprises the structural formula:

$$ROOC-(CH_{2})_n-C(R)_{2}-O(CR)_{2}(CH_{2})_n-COOR$$

wherein R can individually be any of H or any alkaline earth metal;

n may be from 1 to 3.

Another embodiment of the invention includes the electrolyte composition wherein the organic acid is an alpha-hydroxy acid or a beta-hydroxy acid substantially soluble in water.

Another embodiment of the invention includes an electrolyte composition of matter comprising an aqueous solvent, at least one inorganic salt dissolved in said solvent, and at least one organic acid or salt thereof, said acid having the following structural formula

$$C_nH_{2n+1}COOR$$

wherein R can individually be any of H or any alkaline earth metal; and

n is from 2 to 8.

The foregoing and other features and advantages of the present invention will be apparent from the following more particular description of the preferred embodiments of the invention, as illustrated in the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a diagram of a first embodiment of a hydrogen production system according to the present invention.

FIG. 2 is a diagram of a second embodiment of a hydrogen production system according to the present invention.

FIG. 3 is a diagram of a conical funnel member of FIG. 2.

FIG. 4 is a diagram of a third embodiment of a hydrogen production system according to the present invention.

FIG. 5 is a diagram of a first cavitation subsystem according to the present invention.

FIG. 6 is a diagram of a second cavitation subsystem according to the present invention.

FIG. 7 is a diagram of the major factors affecting hydrogen production.

FIG. 8 is a corner perspective of a computer-aided drawing of a single-cell sonoelectrochemical apparatus of the fourth embodiment.
FIG. 9 is a similar drawing taken from an elevation and facing the transverse transducer housing, with the gas collection tube, anode and bottom transducer housing made partially transparent. FIG. 10 is a similar drawing taken from an elevation and rotated 90 degrees from FIG. 9. FIG. 11 is an exploded view of the apparatus of FIGS. 8-10. FIG. 12 is a graph of the hydrogen data produced using this embodiment. FIGS. 13-16 are graphical depictions of the hydrogen versus time data collected for a series of organic acids that were tested in a cavitation-assisted electrolysis embodiment of the current invention.

**Detai**

**Detailed Description of Preferred Embodiments**

[0051] It should be understood that the following embodiments are only examples of the many advantageous uses of the innovative teachings herein. In general, statements made in the specification of the present application do not necessarily limit any of the various claimed inventions. Moreover, some statements may apply to some inventive features but not to others. In general, unless otherwise indicated, singular elements may be in the plural and vice versa with no loss of generality.

**Defined Terms**

[0052] In this patent the following definitions apply when these words are used:

[0053] Cavitation—Cavitation is the phenomenon of formation (irrespective of mechanism) of vapor bubbles in a fluid, in the region where the pressure of the fluid falls below its vapor pressure. Cavitation can be divided into two classes of behavior: inertial (or transient) cavitation, and non-inertial cavitation. Inertial cavitation is the process where a void or bubble in a liquid rapidly collapses, producing a shock wave. Non-inertial cavitation is the process where a bubble in a fluid is forced to oscillate in size or shape due to some form of energy (such as acoustic fields) input.

[0054] Acoustic Energy.—For the purposes of this patent, ultrasonic acoustic energy refers to those frequencies from 16 kHz up to and including 2 MHz. "Power ultrasound" is commonly understood to include the frequency range of from 20 kHz to 100 kHz which is where cavitation occurs. Above 100 kHz ultrasound is useful primarily for clinical imaging. Also for the purposes of this patent, acoustic energy, as well as any radiation of any frequency or wavelength in the electromagnetic spectrum, may be employed as a single frequency (wavelength) or any frequency combination thereof (as a discrete sum, difference, harmonics, sub-harmonics, overtones, series, etc.).

[0055] "Electrolysis" as it is used herein refers to Applicant's general protocol for producing hydrogen, but use of the term is not an admission that the process is electrolysis. Applicants have demonstrated herein that the hydrogen is produced by a sonochemical process, and not that of pure electrolysis. However, the term "electrolysis" is sometimes used to refer in shorthand manner to the sonochemical hydrogen production process developed hereunder.

[0056] The term "extractor" is used interchangeably with "apparatus" to indicate the hydrogen production cell embodiments described herein.

[0057] The term "cavitation-assisted electrolytic combustion" is the process of converting any suitable hydrocarbon soluble in the electrolyte into hydrogen and/or carbon dioxide in the various embodiments of the apparatus disclosed herein. It is not to be inferred that the process steps are limited to the apparatus described herein, but that the method may be adapted to any suitable cavitation-assisted electrolysis device by one of ordinary skill, given these teachings. That is, the process is separate and distinct from the apparatus.

**First Embodiment of Hydrogen Production System**

[0058] FIG. 1 is a cross-sectional side view of the hydrogen production system 100 according to the present invention. Hydrogen production system 100 consists of a container apparatus 102 in the fashion of an electrolytic cell capable of storing a volume of a solution 160. Solution 160 is comprised of a solvent and solute. The solvent is preferably water or another aqueous solution containing hydrogen. The solute is a chemical compound capable of carrying an electrical charge i.e., an electrolyte. The sides of container apparatus 102 are preferably non-electrically conductive. Two electrically-conductive pieces 130 and 132 are held above the bottom member 105 of container apparatus 120 by supporting members 106 and 108, respectively. The electrically-conductive piece 130 is connected to the negative terminal 112 of power supply 110. Thus, the electrically-conductive piece 130 is a cathode. Likewise, the electrically-conductive piece 132 is connected to the positive terminal 114 of power supply 110. Thus, the electrically-conductive piece 132 is an anode. A hollow, cylindrical tube 120 is connected to and passes through top member 104 of container apparatus 102. The bottom of tube 120 is below the bottom of cathode 130 but not touching bottom member 105 of container apparatus 102. The bottom of tube 122 is flared outward and positioned so that the bottom of tube 122 is below the bottom of anode 132 but not touching bottom member 105 of container apparatus 102. Finally, a transducer 140 is connected to one side of container apparatus 102. Wires 142 connect transducer 140 to power supply 110.

[0059] As previously mentioned, power supply 110 causes cathode 130 to be negatively charged and anode 132 to be positively charged. As a result, an electrical current is created between cathode 130 and anode 132. The electrical current electrolyzes solution 160 and causes hydrogen to form around cathode 130 and oxygen to form around anode 132. Tube 120 funnels the hydrogen out of container apparatus 102 for use further use (shown by arrow 150), such as to provide fuel for hydrogen fuel cells or to directly power an engine. Likewise tube 122 funnels the oxygen out of container apparatus 102 (shown by arrow 155). As solution 160 is electrolyzed and the constituent gases are removed from the system 100, additional solution can be added through an inlet 170.

[0060] Transducer 140 produces acoustic energy waves 144 which transmit through and cause cavitation in solution 160. This cavitation decreases the energy required to break the chemical bonds of solution 160. As a result, in the presence of cavitation, a greater amount of hydrogen is produced at cathode 130 at a given voltage than in the absence of cavitation. Alternatively, in the presence of cavitation, the same amount of hydrogen is produced at cathode 130 at a lower voltage than in the absence of cavitation.
Hydrogen production system 100 is designed to be portable. In one embodiment, hydrogen production system 100 is sized approximately 8” in length by 8” in width by 8” in height so that it can fit as an engine component in a vehicle. However, it is clear to one skilled in the art that hydrogen production system 100 and its components can be scaled larger or smaller without affecting the spirit and scope of the present invention. Likewise, it is clear to one skilled in the art that hydrogen production system 100 and its components can take on many different shapes without affecting the spirit and scope of the present invention. FIG. 1 shows one embodiment of the present invention where container apparatus 102 is shaped to allow maximum transmittal of sound waves 144 through solution 160. Finally, it is clear to one skilled in the art that any number of transducers 140 may be placed at various locations on container apparatus 102 and used to produce acoustic energy waves 144 in order to maximize the creation of cavitation within solution 160.

Second Embodiment of Hydrogen Production System

FIG. 2 is a cross sectional side view of another embodiment, referred to as hydrogen production system 200, of the present invention. Hydrogen production system 200 consists of a container apparatus 202 in the fashion of an electrolytic cell capable of storing a solution 160. The sides of container apparatus 102 are preferably non-electrically conductive. A hollow, cylindrical, electrically conductive piece 230 is held above the bottom member 207 of container apparatus 202 by supporting members 232. A second electrically conductive member 234 is held above the bottom member 207 of container apparatus 202 by supporting member 205. Electrically conductive piece 230 is connected to the positive terminal 214 of power supply 210. Thus, electrically conductive piece 230 is an anode. Likewise, electrically conductive piece 234 is connected to the negative terminal 212 of power supply 210. Thus, electrically conductive piece 234 is a cathode. A hollow, cylindrical tube 220 is connected to and passes through top member 206 of container apparatus 202. The bottom of tube 220 is flared outward and positioned so that some portion of cathode 234 is within the tube 220. Finally, a transducer 240 is connected to one side of container apparatus 202. Wires 242 connect transducer 240 to power supply 210.

Power supply 210 causes cathode 234 to be negatively charged and anode 230 to be positively charged. As a result, an electrical current is created between cathode 234 and anode 230. The cylindrical shape of anode 230 and the position of cathode 234 along the axis of anode 230 takes advantage of the electrical field produced by cathode 234 and anode 230 and helps to maximize the flow of electricity between cathode 234 and anode 230.

As previously described, the electrical current flowing between cathode 234 and anode 230 electrolyzes solution 160 and causes hydrogen to form around cathode 234 and oxygen to form around anode 230. Tube 250 funnels the hydrogen out of container apparatus 202 for further use (shown by arrow 250). Referring to FIG. 3, a conical piece 310 is placed on top of anode 230. Conical piece 310 funnels oxygen out of container apparatus 202 (shown by arrow 340). Referring back to FIG. 2, as solution 160 is electrolyzed and the constituent gases are removed from the system 100, additional solution can be added through an inlet 280.

Hydrogen production system 200 is the same as hydrogen production system 100 in that transducer 240 produces sound waves 244 which transmit through and cause cavitation in solution 160. This cavitation decreases the energy required to break the chemical bonds of solution 160 via electrolysis. As a result, in the presence of cavitation, a greater amount of hydrogen is produced at cathode 234 at a given voltage than in the absence of cavitation. Alternatively, in the presence of cavitation, the same amount of hydrogen is produced at cathode 234 at a lower voltage than in the absence of cavitation.

Hydrogen production system 200 is designed to be portable. In one embodiment, hydrogen production system 200 is sized approximately 8” in length by 8” in width by 8” in height so that it can fit as an engine component in a vehicle. However, it is clear to one skilled in the art that hydrogen production system 200 and its components can take on many different shapes without affecting the spirit and scope of the present invention. Likewise, it is clear to one skilled in the art that hydrogen production system 200 and its components can take on many different shapes without affecting the spirit and scope of the present invention. FIG. 2 shows one embodiment of the present invention where container apparatus 202 is shaped to allow maximum transmittal of acoustic energy waves 244 through solution 160. Finally, it is clear to one skilled in the art that numerous transducers 240 may be placed at various locations on container apparatus 202 and used to produce acoustic energy waves 244 in order to maximize the creation of cavitation within solution 160.

Third Embodiment of Hydrogen Production System

FIG. 4 is a cross sectional side view of another embodiment, referred to as hydrogen production system 400, of the present invention. Hydrogen production system 400 consists of a cylindrically-shaped container apparatus 402 in the fashion of an electrolytic cell capable of storing a solution 160. Container apparatus 402 has an electrically conductive inner wall 403 and a non-electrically conductive outer wall 470. An electrically conductive piece 430 is held above the bottom member 407 of container apparatus 402 by supporting member 405. Electrically conductive inner wall 403 is connected to the positive terminal 414 of power supply 410. Thus, conductive inner wall 403 is an anode. Electrically conductive piece 430 is connected to the negative terminal 412 of power supply 410. Thus, electrically conductive piece 430 is a cathode. A hollow, cylindrical tube 420 is connected to and passes through the top member 406 of container apparatus 402. The bottom of tube 420 is flared outward and position so that some portion of cathode 430 is within the tube 420. Finally, a transducer 440 is connected to bottom member 407 of container apparatus 402. Wires 444 connect transducer 440 to power supply 410.

Power supply 410 causes cathode 430 to be negatively charged and anode 430 to be positively charged. As a result, an electrical current is created between cathode 430 and anode 430. The cylindrical shape of anode 430 and the position of cathode 430 along the axis of anode 430 takes advantage of the electrical field produced by cathode 430 and anode 430 and helps to maximize the flow of electricity between cathode 430 and anode 430.

As previously described, the electrical current flowing between cathode 430 and anode 430 electrolyzes solution 160 and causes hydrogen to form around cathode 430 and oxygen to form around anode 430. Tube 450 funnels the hydrogen out of container apparatus 402 for further use (shown by arrow 450). Conically-shaped top member 480 of
container apparatus 402 funnels oxygen out of container apparatus 402 (shown by arrow 455). As solution 160 is electrolyzed and the constituent gases are removed from the system 400, additional solution can be added through an inlet 490.

[0070] Hydrogen production system 400 is the same as hydrogen production systems 100 and 200 in that transducer 440 produces acoustic energy waves 442 which transmit through and cause cavitation in solution 160. This cavitation decreases the energy required to break the chemical bonds of solution 160 via electrolysis. As a result, in the presence of cavitation, a greater amount of hydrogen is produced at cathode 430 at a given voltage than in the absence of cavitation. Alternatively, in the presence of cavitation, the same amount of hydrogen is produced at cathode 430 at a lower voltage than in the absence of cavitation.

[0071] Hydrogen production system 400 is designed to be portable. In one embodiment, hydrogen production system 400 is sized approximately 8" in length by 8" in width by 8" in height so that it can fit as an engine component in a vehicle. However, it is clear to one skilled in the art that hydrogen production system 400 and its components can be scaled larger or smaller without affecting the spirit and scope of the present invention. Likewise, it is clear to one skilled in the art that hydrogen production system 400 and its components can be adapted to a variety of different shapes which do not affect the spirit and scope of the present invention. Finally, it is clear to one skilled in the art that any number of transducers 440 may be placed on container apparatus 402 and used to produce sound waves 442 in order to maximize the creation of cavitation within solution 160.

[0072] Throughout the descriptions of hydrogen production systems 100, 200, and 400, a cylindrical tube, tube 120, 250, and 420, is used to capture hydrogen formed around the cathode and direct the hydrogen out of the systems. It will be clear to one skilled in the art that tubes 120, 250, and 450 can be replaced by any means to capture and direct the hydrogen. Such means include, but are not limited to, tubes and similarly shaped conduits, membrane filtering, diffusive evaporation, differential pressures, and channeling solution flow.

Embodiments of Cavitation Sub-System

[0073] Throughout the descriptions of hydrogen production systems 100, 200, and 400, transducers 140, 240, and 440 are used to produce acoustic energy waves 144, 244, and 442 which cause cavitation within solution 160. It will be clear to one skilled in the art that transducers 140, 240, and 440 may be replaced by any means for generating cavitation. Such means for creating cavitation include, but are not limited to, acoustic means, mechanical means, hydrodynamic means, electromagnetic means, and ionizing radiation means.

[0074] FIGS. 1, 2 and 4 show embodiments of the present invention where the cavitation is produced by a specific acoustic means, namely, by using a transducer to pass acoustic energy waves through solution 160. However, other acoustic means can be used to produce the cavitation. It will be understood by one having skill in the art that such acoustic means includes, but is not limited to, transducers, microphones, and speakers.

[0075] An example of a mechanical means to cause cavitation within hydrogen production systems 100, 200, and 400 includes, but is not limited to, a propeller system contained within container apparatus 102, 202, and 402, which causes cavitation as the propeller spins on its axis. FIG. 5 shows a cross sectional view of such a propeller system. As shown, propeller blades 520 spin about the axis of propeller system 510 causing cavitation to be produced in solution 160. Propeller system 510 may be powered by power source 110, 210, or 410. It will be understood by one having skill in the art that other mechanical means can be used to produce the cavitation. Such mechanical means include, but are not limited to, a propeller system, pistons, shock tubes, and light gas guns.

[0076] An example of a hydrodynamic means to cause cavitation within hydrogen production systems 100, 200, and 400 includes, but is not limited to, the injection of a compressed gas, for example, compressed air, into container apparatus 102, 202, and 402 to cause cavitation. FIG. 6 shows a cross sectional view of such a compressed gas injection system. As shown, compressed gas injection system 610 is affixed to container apparatus 102, 202, or 402. Compressed gas travels (indicated by arrows 640) from a compressor (not shown) through tube 630 to compressed gas injection system 610. The compressed gas flows through tubes 620 and is introduced into solution 160 as bubbles, i.e. cavitation. In one embodiment, compressed gas injection system 610 may be separated from solution 160 by a porous membrane that permits the transfer of the compressed gas through the membrane while preventing solution 160 from entering compressed air system 610. An example of such a membrane is Gore-Tex. It will be understood by those having skill in the art that other hydrodynamic means can be used to produce the cavitation. Such hydrodynamic means include, but are not limited to, a compressed gas injector system and any device capable of transferring momentum into solution 160 without transferring mass into solution 160, for example, a shock plate or paint shaker.

[0077] An example of an electromagnetic means to cause cavitation within the hydrogen production systems 100, 200, and 400 includes, but is not limited to, a laser beam directed to pass into solution 160 so as to produce a shock wave that causes cavitation within solution 160. It will be understood by those having skill in the art that other electromagnetic means can be used to produce cavitation. Such electromagnetic means include, but are not limited to, a laser beam, x-rays, gamma rays, high speed electrons, electric arc, magnetic compression, plasma generation, and electromagnetic radiation arising from any type of electron or proton reaction.

[0078] Finally, an example of an ionizing radiation means to cause cavitation within the hydrogen production systems 100, 200, and 400 includes, but is not limited to, passing high energy protons into solution 160 where cavitation is formed around the protons. Generally, ionizing radiation is any radiation that is capable of removing an electron from a chemical bond. Therefore, it will be understood by those having skill in the art that such ionizing radiation means include, but are not limited to, all electromagnetic radiation greater in energy than ultraviolet radiation and high energy particles such as photos, protons, neutrons, and charged and uncharged nuclei.

[0079] Throughout the descriptions of hydrogen production systems 100, 200, and 400, as well as the examples of the various means of causing cavitation, cavitation is said to occur within solution 160. It will be understood by those having skill in the art that causing cavitation "within" solution 160 means causing cavitation within the electrolytic zone.

[0080] FIG. 7 is a diagram of the major factors affecting the production of hydrogen according to the present invention. Solution factors 710 are the major factors affecting solution 160. These solution factors include a solvent and solute. As
previously described, the solvent is water or another aqueous solution containing hydrogen. The solute is a chemical compound, such as acid (such as HCl or HCl), base (NaOH), or salt (such as KCl or NaCl), and is held at a particular density per volume of solvent in order to maximize the electrical conductivity of the solution. The solution has a particular pH, and it is held at a particular temperature and pressure, whether in hydrogen production systems 100, 200, or 400, to minimize the energy required to break the chemical bonds of the solvent. Finally, the solution has a particular ionic and ionic state (chemical potential).

[0081] Power factors 720 are the major factors affecting the delivery of power to cathodes 130, 234, and 430, and anodes 132, 230, and 403. It will be readily apparent to one skilled in the art that the power factors 720 include voltage applied, current applied, and total power applied. Additionally, although hydrogen production systems 100, 200, and 400, have been shown with a single cathode and single anode, it is apparent to one skilled in the art that the number of voltage/current applications points can be increased without affecting the spirit and scope of the present invention. Likewise it is apparent to one skilled in the art that the sizes and shapes of cathodes 130, 234, and 430 and anodes 132, 230, and 403 can change without affecting the spirit and scope of the present invention. Finally, it is apparent to one skilled in the art that power supplies 110, 210, and 410 can be any power producing device, such as a battery, solar panel, or fuel cell.

[0082] Material Composition factors 730 are the major factors affecting the materials of the hydrogen production systems 100, 200, and 400. The materials comprising cathodes 130, 234, and 430, and anodes 132, 230, and 403 are selected to maximize electrical conductivity. Such materials include, but are not limited to, metals such as copper, platinum, and high order non-linear crystals including, but not limited to, lithium nitrate and lithium tantalate.

[0083] The catalytic factors 740 employed to enhance and catalyze the production of hydrogen are the major factors affecting the energy balance within solution 160. The non-energy input catalytic factors lowering the necessary electrolytic input energy $\Delta E_1$ to $\Delta E_2$ include but are not limited to: (1) process temperature (as a function of $\Delta E_{cav}$, $\Delta E_{p}$, partial molar concentrations of species), (2) container properties (composition, shape), (3) solution properties (solute/solvent composition [species, concentrations, etc.], pH, chemical potential, pressure, catalytic agents added [supported catalysts, gases such as noble gases, etc.]), (4) electrode properties (composition [elemental, isotopic, chemical], shape, microsurface [crystal planes, etc.], macrosurface [holes, edges, etc.]), and (5) structure of applied electromagnetic field [energized, unenergized]).

[0084] Referring to Table 1, a set of equations is set forth showing that even in the presence of cavitation, the energy required to perform the electrolysis of solution 160 to produce hydrogen is greater than the energy that is produced when that hydrogen is recombined with oxygen. Thus, it is apparent to one skilled in the art that the teachings described herein are not directed to a perpetual energy device. Rather, because of the net energy loss that results from the electrolysis of solution 160, energy is introduced into systems 100, 200, and 400 as represented by power supplies 110, 210, and 410 to drive the electrolysis and catalytic processes.

### Table 1

<table>
<thead>
<tr>
<th>Step</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Electrolysis (decomposition) of water requires energy input: $\Delta E_{elec}$</td>
</tr>
<tr>
<td>2</td>
<td>Formation of water requires energy output: $\Delta E_{form}$</td>
</tr>
<tr>
<td>3</td>
<td>By the First Law of Thermodynamics, electrolysis is not fully reversible since the heat and entropy losses cannot be fully accounted for. Thus, we have the result: $\Delta E_1 &gt; \Delta E_2$ always. As a result, the process of electrolysis/water reformation, as well as the process described herein cannot be termed a “Perpetual Motion (or Energy) Machine” of any kind.</td>
</tr>
<tr>
<td>4</td>
<td>The thermodynamic efficiency relation $\epsilon = \Delta E_2 / \Delta E_1 = 100%$ gives a guide to the relative efficiency of the electrolysis/water reformation process. An eventual efficiency of 80% or more is in possible.</td>
</tr>
<tr>
<td>5</td>
<td>$\Delta E_1$ (energy consumed per mole $H_2O$ or $H_2$ to decompose water to $H_2$ gas) may be represented in the present invention by the quantity $\Delta E_2$, which is: $\Delta E_2 = \Delta E_{elec} - \Delta E_{cav} - \Delta E_{co}$ where the electrolysis term represents only the electrical energy input from the electrodes as electrolysis, the cavitation term represents only the electrical energy input from acoustical energy (or any means) to cause or sustain cavitation, and the “other” term represents any energy input for heating, cooling, stirring, or measurement. Here energy is represented as the total energy (power) input as the function of current and voltage by Ohm’s Law.</td>
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<td>6</td>
<td>In the absence of catalytic factors 740, $\Delta E_{elec} = \Delta E_1$. However, for the process described herein to be valid, $\Delta E_{elec}$ must be less than $\Delta E_1$: $\Delta E_1 &gt; \Delta E_{elec}$ since the process described herein is a catalytic process which lowers the necessary energy to form hydrogen gas. Thus, the overall equation is: $\Delta E_2 = \Delta E_{elec} - \Delta E_{cav} - \Delta E_{co} &lt; \Delta E_2$ which requires the value $\Delta E_1$ to approach $\Delta E_2$. Since $\Delta E_1 &gt; \Delta E_2$ always, the equation $\Delta E_1 &gt; \Delta E_2$ is valid.</td>
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</table>

7 Generally, there are two kinds of catalytic factors: non-energy input catalytic factors which are based on no energy input (e.g. electrode materials, configurations, etc.); and energy input catalytic factors which are based on energy input (e.g. cavitation, heating, cooling, stirring, etc.). Examples of both kinds of catalytic factors are set forth in catalytic factors 740.
Referring back to FIG. 7, the energy input factors lowering the electrolytic input energy $\Delta E_{\text{cat}}$ include, but are not limited to: (1) $\Delta E_{\text{cat}}$ (energy necessary for the temperature control and measurement, mechanical, stirring, etc.), and (2) $\Delta E_{\text{com}}$ (cavitator properties [size, shape, composition], configuration [number, density per unit area/volume, etc.], power input [V, I], acoustic frequency spectrum input, electromagnetic frequency spectrum input). As described above, a cavitator can be any device capable of causing cavitation.

It has been advantageously shown that the following factors in one embodiment, hydrogen production system 400, greatly increase hydrogen production in the present invention: (1) the use of a specific acoustical spectrum to maximize cavitation in solution 160; (2) the use of sodium or potassium iodide salt in solution 160 to maximize the conductivity and chemical potential of solution 160; (3) the dissolution of an effective amount of noble gas into solution 160, such that the noble gas is completely dissolved in the solution, to electromagnetically enhance the production of cavitation thus maximizing the generation of hydrogen gas—in the present embodiment, the noble gas is preferably argon and an effective amount of noble gas to be completely dissolved in solution 160 is up to five percent (5%) at Standard Temperature and Pressure; (4) the shape and configuration of the electrodes, which for hydrogen production system 400 comprise the electrically conductive inner wall 403 and electrically conductive inner piece 430, to (i) maximize the mechanical separation of the hydrogen and oxygen gas products and (ii) maximize the electrolysis electric field by use of the cylindrical electrode configuration (which maximizes the electric field by a multiplicative ratio of the inner and outer radii); and (5) the shape of the container, for example, hydrogen production system 400 comprises an electrically conductive inner wall 403 contained within an non-electrically conductive outer wall 470 so as to electrically isolate the function of the hydrogen production system 400 from the outside world.

Likewise, although it is clear to one skilled in the art that the solution 160 may be exposed to any temperature and/or pressure and that solution 160 may be contained within either a sealed or unsealed container, it has been advantageously shown for one embodiment, hydrogen system 400, that the hydrogen production using the teachings described herein is preferably performed in a sealed, but not pressurized, container at approximately Standard Temperature and Pressure (STP).

Fourth Embodiment of Hydrogen Production System

A fourth embodiment of the invention is the apparatus shown in FIGS. 8-11. Apparatus 500 is generally described as having the overall configuration and components of an electrolytic cell, with the addition of two ultrasound transducers positioned orthogonally to each other. It is to be emphasized that the apparatus is not used to conduct electrolysis per se, but is used to create a sono electrochemical reaction process. Therefore, the device is called "sono electrolytic cell" 500.

With attention directed to FIG. 8, sono electrolytic cell 500 is shown in angular perspective. In operation, sono electrolytic cell 500 would be situated inside a container for holding liquids such as aqueous electrolyte. However, for purposes of clarity the container is omitted from the drawings. The cell is comprised of a cylindrical anode 520 mounted on anode support plate 525, a cylindrical cathode 530 (best seen in FIGS. 9-10) located inside the anode 520; an ultrasonic bottom transducer 540 mounted under the anode support plate 525 and oriented to transmit along the axis of the cathode 530; and a transverse transducer 550 mounted at a 90 degree angle to the bottom transducer 540. Each transducer has a pair of terminals $541a$ and $541b$ for positive and negative leads, respectively. Not shown in these figures are the electronics for driving the transducers, power supply for the electrodes, or a gas removing means for removing any evolving Hydrogen.

The supporting skeleton comprises four support plates 510, 512, 514 and 525. Base support plate 510 has various holes therethrough for receiving and/or supporting various structural and functional components such as the tie rods 527 and anode 520. Lower anode support plate 525 has a stepped cutout 526 that serves to support the lower end of anode 520. Upper anode support plate 512 has a similar cutout that allows the two anode support plates 512, 525 to “sandwich” the anode when the opposing nuts 528 are tightened thereby clamping the anode in place between upper and lower anode support plates 512, 525, respectively. Spacer supports 513 are non-conductive tubes that thread over support rods 527 and provide additional structural rigidity to the device. Gas collection tube support plate 514 is the fourth support plate and is located above the upper anode support plate. It has a cutout for the gas collection tube 515, which extends from the top of bottom transducer housing 544 to a gas collection tube adapter (not shown). Gas collection tube 515 surrounds the inner electrode (cathode 530) and is located between the inner diameter of the anode and the cathode. In one embodiment the tube may be 1" in diameter, in another embodiment it is 2". The functions of the gas collection tube 515 are to collect hydrogen gas evolved in the electrolyte volume around the cathode 530, to direct the gas upwards either entrained in fluid flow or as bubbles, and it may also have a focusing effect on the electromagnetic fields generated by the electrodes. The support plates may also have orienting criteria such as a notch 516. The plates may be oriented horizontally as shown, or vertically. The plates in this case are made from NYLON® (DuPont, Wilmington, Del.) approximately 3/16 inch in thickness, although other materials are equally suitable so long as they can maintain some structural rigidity. The four plates are held in horizontal orientation by four tie rods 527 which are also NYLON, and are threaded at their ends to accept nuts 528. There are also power leads (anode lead (not shown)) and a cathode lead 532). Cathode power is defined as being negative, and the anode is positive. Power to the bottom and transverse transducers was applied through twin leads (not shown in this embodiment).

FIGS. 9 and 10 show the anode 520 and gas collection tube 515 as partially transparent so that the inner cathode may be seen. The anode 520 and cathode 530 may be made from any suitable electrically conductive metals commonly used in electrolysis. The cylindrical anode 520 was made from solid copper pipe having dimensions 5.4 cm OD by 5.1 cm ID, and a height of 6 cm; Graining, Copper stream line tube, Fulton, MS Type M NSE/ANSI, 61-G. The cathode 530 is two pieces, a central 14-gauge copper wire (2 mm OD), and a cylindrical solid copper mesh 533 slipped over it, approximately 2 mm OD, 2 mm ID, height 6 cm; 99.9% pure copper mesh; 0.010 thick; Stock no. 6095, K&S Engineering, Chicago, Ill. The cathode shape is that of a concentric cylinder having a fixed diametric ratio of approximately 1:25 relative to and inside the anode, as measured according to the inside
diameter of the outer anode compared to the outside diameter of the inner cathode. The cathode was located concentrically along the axis of the anodic volume. The copper wire was sourced from Home Depot, and is 99.9% pure copper wire—14 gauge; 600 volt; WV-1 rated; Issue No. YM-680, 590.

**0092** FIG. 11 shows how the transverse transducer 550 was supported. Transverse transducer housing cap 553 covers and is in direct contact with transverse transducer 550. Mesh 551 is provided for terminals 541a, b to penetrate. Mesh 551 is glued to transverse transducer support plate 555. Cap 553 is similarly affixed to plate 555 thereby resulting in transverse transducer 550 being contained and supported in a vertical manner and aiming directly at the side of anode 520. Bottom transducer 540 is contained within the bottom transducer housing base 546 and the bottom transducer housing cap 544. They are both threaded to receive each other. Split ring 543 and mesh 545 support the transducer 540. The top of bottom transducer housing cap 544 is affixed to the bottom of lower anode support plate 525 so that when the bottom transducer housing base 546 is screwed in, it retains all the elements snugly.

**0093** A DC power supply (30 volts/3 amps) (3 channel programmable BK Precision Model 903) was used to power the sono-electrolytic process. A frequency generator drives the transducers.

**0094** The electrolyte used to generate the attached hydrogen production data was an aqueous solution of citric acid, NaCl and NaI. 121.731 g NaCl, ACS grade reagent, Aqua Solutions, Deer Park, Tex., Cat. No. 52075-2KG was dissolved in 2 liters of water purified by reverse osmosis. Next, 20,500 g Citric Acid—ACS grade (ACROS, Cat. No. 42356-0020) was dissolved in the same solution. Then 3.54 mg Reagent Grade NaI from MP Biomedicals, Solon, Ohio, Cat No. 193979 was dissolved in the electrolyte solution. Argon was bubbled through the solution prior to use sufficient to displace other dissolved gases.

**0095** Hydrogen was produced according to the following electrolysis protocol. All potentials mentioned are direct current (DC) unless otherwise noted. First, the electrolyte solution was “charged” or brought to potential. Priming or charging is the process of applying an electric potential to the solution which retains a portion of the charge throughout and after the reaction has concluded. It is currently understood that the solution possesses a complex dielectric function $\varepsilon$ and thus functions similar to a resistive capacitive network. The charging step is required of all methods to induce hydrogen production. Bringing the solution to potential alleviates the delay normally associated with initializing electrolysis. On a molecular level, this causes the ion channels to start “flowing,” and promotes electron exchange. The initial runs to bring the solution to potential are relatively straight-forward. The electrolyte in the apparatus was brought to a set current of 1.5 Amps with the voltage set at 20V. When the current approached the set value of 1.5 Amps, the voltage was observed to be between 6-8 volts. The solution was held at these values for approx. 5 minutes and then the power applied was turned off. This particular procedure was carried out at least once, sometimes twice. At this point, the solution was considered to be charged (at potential).

**0096** The positive power lead was attached to the anode; the negative lead was attached to the cathode. The transducers were attached to the function/frequency generator (if using cavitation). The power supply was set to float with a voltage ceiling of 20V, while the anode was set at a fixed value (which ranges from 250 mA to 2.0 A). Any suitable function generator can be used to drive the transducers, but preferred generators include a PROTEK B8012, or a QUAKKO 5000 digital signal generator. The transducers were set at 3.3 V, and drew about 10-20 mA. Frequencies were set at 38.248 kHz for the transverse/horizontal transducer, and 76 kHz for the bottom transducer. The transverse transducer 550 was located 2.6 cm from the center of the cell; the bottom transducer 540 was located 5.2 cm from the center measured from the face of the transducer. Both transducers were oriented towards the center of the cell. The central area of the cell is thus considered the “reaction zone” for purposes of this apparatus 500. The transducer used in the present invention was a Pizo Air Transducer, Part No. SMUTF40TR18A, Stemine (Steiner & Martins, Inc.), Miami, Fla. Hydrogen was produced in the quantities indicated in the attached graph (FIG. 12).

**Chemical Embodiments**

**0097** The electrolysis of pure water (H$_2$O) occurs when H$^+$ ions flow to the cathode to combine with electrons to produce hydrogen gas in a reduction reaction. Likewise, hydroxide ions (OH—) flow to the anode to release electrons and an H$^+$ ion to produce oxygen gas in an oxidation reaction. When NaCl is added to create an electrolyte capable of carrying significantly more charge (higher charge density), the reaction shifts. The standard electrolysis reaction of an aqueous solution of NaCl results in water molecules being reduced to OH— and hydrogen gas (H$_2$) at the cathode, with the chloride anions still being oxidized to chlorine gas at the anode. The end result is a steadily more basic solution of NaOH with H$_2$ gas being evolved. However, the embodiments of this system do not exhibit chlorine gas being evolved under normal operating conditions.

**0098** The field of sonochemistry has established that ultrasound energy transferred to liquid H$_2$O creates cavitation (Mason, Sonochemistry 2:4 (1999)). The cavitation of water decomposes H$_2$O into H$^*$ and OH$^*$ radicals, amongst others, and the subsequent H$^*$ radicals recombine to form H$_2$ gas (Id, pp. 16-17).

**0099** Embodiments of the invention are directed to an electrolyte composition of matter comprising an aqueous solvent, at least one inorganic salt dissolved in said solvent, and at least one organic acid or salt thereof, the at least one acid being substantially soluble in the aqueous solvent and present in a concentration sufficient to support cavitation-assisted electrolytic combustion.

**0100** In the electrolyte composition of the current invention, citric acid (C$_6$H$_8$O$_7$) was added to a water-based NaCl electrolyte to result in a 0.1M solution. Upon application of both electrolysis and cavitation, hydrogen and carbon dioxide gases were evolved in relatively pure amounts. No carbon monoxide or other unwanted side products have been detected. The inventors currently believe that one possible equation that explains the overall reaction chemistry is the following:

$$C_6H_8O_7 + 5 H_2O \rightarrow 9 H_2 + 6 CO_2$$

This has been determined experimentally through measurement of the ratios of the gases in the resultant gaseous product recovered. Gas analysis performed by the Rowland/Blake Research Group, Dept. of Chemistry, U. California/Irvine, showed a H$_2$/CO$_2$ ratio of 7.76 from a combined sample of gas evolved from the citric acid-based electrolyte in the apparatus of the embodiments disclosed herein. The gas was a
combination of four separate samplings taken from the experimental gas manifold system described above in the fourth embodiment during a one- to two-hour run during which time the citric acid-based electrolyte of the current invention was subjected to the sonoelectrolytic process. Th

cell differed slightly from that of the fourth embodiment in that it had a single transducer, and no gas collection tube. All other components and operating criteria are substantially identical.

[0101] The chemical pathway is likely radical-initiated, and bears indications of both oxidation (production of CO₂ at the anode) and reduction (production of hydrogen gas at the cathode) occurring in possibly separate pathways. There is also some possibility that pyrolysis of the alkane component of citric acid is occurring given that hydrogen is being evolved in a high-temperature environment. However, other mechanisms are also possible, and the embodiments of the invention do not depend upon any particular mechanism being ultimately proven to be the scientifically correct one. We call this process “cavitation-assisted electrolytic combustion.”

[0102] The aqueous solvent of the electrolyte composition comprises at least about 50% of the electrolyte on a weight-to-volume basis. Since the water molecules of the electrolyte dissociate into radical species under the effect of cavitation, and those radicals have catalytic effect in the conversion of organic acids to hydrogen and carbon dioxide, the concentration of water sufficient to support cavitation-assisted electrolytic combustion is believed to be at least 50%. That is, the proportion of water is at least about half; more preferably at least about 60%, and most preferably at least about 70% water. A preferred range of water concentration is from about 50% to about 95% of said electrolyte on a weight to volume basis.

[0103] Embodiments of the invention may include a noble gas dissolved in the aqueous solvent. It is believed that an amount of noble gas, such as Argon, for example, may be advantageous in operation of the invention. Argon has been dissolved in the electrolyte by bubbling Argon through solution of electrolyte prior to use, a process known as “sparging”. Sparging has the effect of excluding other gases such as oxygen, carbon dioxide, nitrogen etc. that are present in the air. Sparging includes the step of degassing by applying vacuum to the electrolyte prior to bubbling Argon. This has the effect of accelerating the displacement of air with Argon by first reducing the partial pressures of all the dissolved gases. Bubbling in Argon then saturates the liquid with the heavier, inert Argon. Other noble gases may also be substituted and are well-known such as Helium, Neon, Krypton, etc. Saturation of a solution is typically attained after about 30 minutes of bubbling with a multi-bubble fit. For Argon, saturation is believed to be reached at about 5% weight-to-volum, although the sparging process is effective in replacing most of the dissolved air with Argon.

[0104] Embodiments of the invention include an inorganic salt such as sodium chloride, sodium iodide, or another inorganic salt capable of conducting charge through an aqueous solution. Salts comprise a cation (positively-charged ionic species) and an anion (negatively-charged ionic species). A preferred cation is selected from the group consisting of sodium, lithium and potassium. Embodiments of sodium cation useful in the present invention are sodium chloride (table salt) and sodium iodide. Embodiments of lithium include lithium chloride and lithium iodide. Embodiments of potassium include potassium iodide, potassium chloride. As mentioned above in reference to the salt discussion, an anion is also necessary for the electrolyte. Preferred anions include but are not limited to an anion selected from the group consisting of fluoride, chloride, bromide and iodide.

[0105] Embodiments of the invention also include at least one organic acid present in the electrolyte at a weight-to-volume concentration of from about 1% to about 10%. A carboxylic acid by definition contains at least one carboxyl (COOH) moiety, but multi-carboxyl acids are well-known and include citric acid, a tri-carboxylic acid. Other exemplary multi-carboxylic acids include fumaric (2), maleic (2), malonic (2) and tartaric (2).

[0106] Embodiments of the invention also include at least one organic acid that is substantially soluble in the electrolyte solution. “Substantially soluble” means that the acid’s solubility in water at standard pressure and temperature is sufficient to generate hydrogen in the cavitation-assisted electrolytic combustion process. A preferred lower solubility range of the at least one organic acid is at least 10 wt. % soluble in said electrolyte solution. A more preferred range is from about 10 to about 90 weight % soluble. Low molecular weight organic acids seem to have excellent solubility in water, particularly those having a main carbon chain of from C₃-C₈.

[0107] Embodiments of the invention may include one or more organic acids selected from, but limited to, the following group: Acetic acid, Citric acid, Formic acid, Malic acid, Malonic acid, Mandelic acid, Maleic acid, Izo-citric acid, Fumaric acid, Lactic acid, Glycolic acid, Glycereic acid, Hexanoic acid, Heptanoic acid, Octanoic acid, Pentanoic acid, Nonanoic acid, Propanoic acid, 3-Hydroxypropanoic acid and 2-Hydroxypropionic acid. This list is for exemplary purposes only, and is not to be taken as a limitation of the invention. Other organic acids that are substantially soluble in water, such as the classes of alpha- and beta-hydroxy acids, also come within the scope of the current invention.

[0108] An embodiment of the invention includes citric acid, and citric acid having a concentration in the electrolyte of from about 0.05 M to about 5.0 M. A particularly preferred concentration is about 0.1 M. A preferred citric acid-based electrolyte has about 0.1M citric acid, NaCl at about 0.5 to 5 weight %, and NaN₃ in a trace amount. Another preferred embodiment includes saturating the electrolyte with Argon sufficient to promote cavitation-assisted electrolytic combustion.

[0109] Other embodiments of the electrolyte invention herein includes the electrolyte composition wherein the at least one organic acid comprises the structural formula:

\[ \text{C}_n\text{H}_{2n+1} \text{COOR} \]

wherein R can be indvidually any of H or any alkali earth metal; and n is from 2 to 8.

[0110] Other embodiments of the electrolyte invention herein includes the electrolyte composition wherein the at least one organic acid comprises the structural formula:

\[ \text{ROOC} - \text{C}_n\text{H}_{2n+1} \text{COOR} \]

wherein R can be any of H or any alkali earth metal; and n is from 2 to 8.

[0111] Other embodiments of the electrolyte invention herein include the electrolyte composition of claim 1 wherein the at least one organic acid comprises the structural formula:

\[ \text{ROOC} - \text{(CHR)}_3 - \text{(CHR)}_2 - \text{C}(\text{OH}) - \text{(CHR)}_2 \text{COOR} \]

wherein R can be any of H or any alkali earth metal; and n is from 2 to 8.
wherein R can be any of H or any alkaline earth metal; R2 can be either OH or COOR; and n may be from 1 to 3.

[0112] Other embodiments of the electrolyte invention herein includes the electrolyte composition wherein the at least one organic acid is either an alpha-hydroxy acid or a beta-hydroxy acid substantially soluble in water. Examples of alpha-hydroxy acids include but are not limited to glycolic acid, lactic acid, 2-hydroxyisobutyric acid, 2-hydroxy-2-phenyl propanoic acid, mandelic acid, 2-hydroxy-3,3-dimethyl-4-butyrolactone, and 2-methylthiobutyric acid. A non-limitative list of beta-hydroxy acids useful with this invention includes 3-hydroxyhexanoic acid, 3-hydroxyoctanoic acid, 3-hydroxydecanoic acid, 3-hydroxydecanoic acid, 3-hydroxyexaprylic acid, salicylic acid, 3-hydroxybutanoic acid, 3-hydroxypentanoic acid, 3-hydroxypropionic acid, and mixtures thereof.

[0113] Embodiments of the invention are directed to additional chemical species tested including and in addition to citric acid. The test protocol for the data in the table below is as follows: Protocol: Examination of The Gas Production of Various Carboxylic Acids

[0114] The purpose of this set of experiments was to establish the hydrogen gas production potential of four carboxylic acids and/or their sodium salts in comparison to the values achieved with citric acid.

Methods:

[0115] For this set of experiments, the only two variables were the type and amount of carboxylic acid that was added to each aliquot of LSF. The amount of sodium chloride and sodium iodide remained constant. To ensure the overall consistency of the salts, a master batch of salt water solution consisting of 1152 g sodium chloride (NaCl) per 5 gal (18.9 L) DI H2O was used for these tests. Aliquots were withdrawn from this batch and specific amounts of carboxylic acids were added to achieve 0.1 M and 1.0 M concentrations of each aliquot, respectively. Each acid concentration was tested for its ability to generate hydrogen gas per this protocol. The results are shown in FIGS. 10-13 for citric, acetic, propanoic and malonic acids.

[0116] Preparation of 5 gal (18.9 L) batch salt solution:

[0117] 5 gal (18.9 L) of deionized water was measured out into a 5 gal (18.9L) Samson Stackable API HDPE Kirk container. 1152 g of ACS Reagent Grade NaCl (CAS #7647-14-5) was weighed out into a round Pyrex glass watch sitting on an A&D Ex-300A electronic balance and 3.37 g of ACS Reagent Grade NaI (CAS #7681-82-5) was likewise weighed out into a round Pyrex glass watch sitting on a Sauter RE 1614 electronic balance; once weighed, both salts were added to the 5 gal (18.9 L) container. This batch solution is gently mixed.

Hydrogen Extractor Runs:

[0118] The sono-electrolytic cell (aka “extractor”) used herein was similar to the extractor described in the fourth embodiment, operated in accordance with the following specific settings or conditions. Each aliquot of solution at their tested concentrations was added to the tank of the extractor. The electrical input to the extractor was a set current (i_0) of 500 mA with a voltage celling of 30 V, provided by a BK Precision 1901 Switching Mode Power Supply 1-30 VDC/30 A power supply. The acoustic input was provided by one pair of transducers, as shown in FIG. 8 and described in the fourth embodiment. The transverse transducer produced 38.248 kHz; the bottom transducer produced 76 kHz. Both transducers were connected to a 20 mHz 100 MS a/s Rigol DG1022 Dual Channel Function/Arbitrary Waveform Generator. The output (hydrogen and carbon dioxide gases) were pulled through a collection manifold fitted with a Pall PTPO45LHP-SAMP membrane by a Parker 12V T1-114D-12-1 diaphragm pump. The Parker pump is powered by a Power Design, Inc. Model 6050 Universal power supply.

The duration of each run was 5 minutes and the following data was taken once a minute on the minute during each run: observed current (i_0) and observed voltage (V_0) from a Fluke 8845A 6½ Digit Precision Multimeter; the flow rate of hydrogen being produced was measured in standard liters per minute (SLPM) with an Alicat Scientific M-10SLPM-D15M H2, calibrated mass flowmeter. Background air leakage was approximated in a series of four control runs, and appears in the graph depicted in FIG. 10. U.S. patent application Ser. No. 13/170, 115 filed on Jun. 27, 2011 describes additional sonoelectrochemical apparatus capable of carrying out these runs, and is incorporated herein by reference in its entirety.

[0119] The following organic acid-electrolyte solutions were tested for their ability to be converted to hydrogen in the apparatus described herein.

Solution #1—Citric Acid

[0120] 19.2 g of ACS Reagent Grade citric acid (CAS #77-92-9) was weighed out into a round Pyrex watch glass sitting on an A&D Ex-300A electronic balance and added to a 1 L aliquot of batch salt solution to achieve a 0.1 M solution. The 0.1M solution was placed into the hydrogen extractor and run according to the general operating protocol described under the Fourth Embodiment, supra. After the initial run, an additional 172.8 g of citric acid is added to the 0.1 M solution to achieve a 1.0 M solution and placed into the hydrogen extractor for the second run. At the termination of the run, the solution was discarded. Results are shown in FIG. 10. The lower line is the average DI water run. It approximates air leakage. The average SLPM (standard liters per minute) of DI water was 2.47±0.36. Any result greater than 2.83 was deemed evidence of gas evolution. Citric acid had average SLPMs of 0.1M and 1.0M of 3.28 and 3.23, respectively.

Solution #2—Acetic Acid

[0121] The same general procedure for the citric acid runs was repeated for the remainder of the runs, except as follows. 6.0 g of ACS Grade 99.74% glacial acetic acid was used to make a 0.1 M solution. The solution was placed into the hydrogen extractor and run. After the initial run, an additional 54 g of acetic acid was added to the 0.1 M solution to achieve a 1.0 M solution and placed into the hydrogen extractor for the second run. Results are shown in FIG. 11. Acetic acid had SLPMs of 3.88/3.56.

Solution #3—Propanoic Acid

[0122] The same general procedure for the citric acid runs was repeated, except as follows. 7.4 g of ACS Grade 99.9% propanoic acid was used to achieve a 0.1 M solution. After the initial run, an additional 66.6 g of propanoic acid was added to the 0.1 M solution to achieve a 1.0 M solution. Results are shown in FIG. 12. Propanoic acid had SLPMs of 4.12/3.65.

Solution #4—Malonic Acid

[0123] The same general procedure for the citric acid runs was repeated, except as follows. 10.4 g of ACS Grade 99.54%
malonic acid is used to achieve a 0.1 M solution. An additional 93.6 g of malonic acid was added to the 0.1 M solution to achieve a 1.0 M solution. Results are shown in FIG. 13. Malonic acid had SLPs of 5.58/5.30.

[0124] Malonic acid seems to be the best generator of hydrogen under the limited testing conditions described herein. Although the acid salts sodium propanoate, sodium butanoate and sodium octanoate were run in the extractor, no data was collected after the runs were started due to noxious-apparing gases being generated immediately.

[0125] Embodiments of the invention are also directed to a method of generating hydrogen gas from water and a hydrocarbon feedstock, comprising providing a high-temperature electron-rich aqueous environment wherein water is dissociated for reaction with the feedstock and the hydrocarbon is converted to hydrogen gas and carbon dioxide. The hydrocarbon feedstock may be one or more organic acids, in combination or alone, or may include other water-soluble organic species having easily dissociable hydrogen atoms such as alcohols.

[0126] Embodiments of the invention are also directed to a method of generating hydrogen gas from water and a hydrocarbon feedstock, comprising creating cavitation within and supplying an external source of electrons to an aqueous electrolyte solution containing a hydrocarbon feedstock thereby producing hydrogen gas and carbon dioxide. As disclosed herein, cavitation is generated using acoustic transducers directed into the electrolyte solution as depicted in the various embodiments. The external source of electrons is supplied through a pair of concentric electrodes immersed in the electrolyte at varying voltage and current settings also in accordance with the various embodiments.

[0127] Additionally, it is self evident that the teachings and embodiments set forth herein are focused on minimizing the amount of input energy while maximizing the output of hydrogen gas. The most important factor affecting the total input energy is electrolysis voltage. Thus, it is self evident that requiring less input voltage for the same given amount (or greater) of hydrogen gas generated will result in requiring less input energy, thus, less input power. As a result of requiring less input power, the input-output thermodynamic difference is minimized and as a result a larger fraction of input power can be generated by energy sources such as solar cells, recharged batteries, etc., thus maximizing overall efficiency and quantity of hydrogen generated.

[0128] Although a specific embodiment of the invention has been disclosed, it will be understood by those having skill in the art that changes can be made to this specific embodiment without departing from the spirit and scope of the invention. Likewise, it will be understood by those having skill in the art that the teachings herein can be scaled in size to increase or decrease hydrogen production without affecting the scope and spirit of the present invention. The scope of the invention is not to be restricted, therefore, to the specific embodiment, and it is intended that the appended claims cover any and all such applications, modifications, and embodiments within the scope of the present invention.

What is claimed is:

1. An electrolyte composition of matter comprising: 
   an aqueous solvent; 
   at least one inorganic salt dissolved in said solvent; and 
   at least one organic acid or salt thereof, said at least one acid being substantially soluble in said aqueous solvent and present in a concentration sufficient to support cavitation-assisted electrolytic combustion.

2. The electrolyte composition of claim 1 wherein the aqueous solvent comprises at least about 50% of said electrolyte on a wt/vol. basis.

3. The electrolyte composition of claim 1 wherein the aqueous solvent comprises from about 50% to about 95% of said electrolyte on a weight to volume basis.

4. The electrolyte composition of claim 1 wherein a noble gas is dissolved in the aqueous solvent.

5. The electrolyte composition of claim 4 wherein the noble gas is Argon.

6. The electrolyte composition of claim 4 wherein the noble gas saturates the solvent.

7. The electrolyte composition of claim 1 wherein at least one inorganic salt comprises a cation selected from the group consisting of sodium, lithium and potassium.

8. The electrolyte composition of claim 1 wherein at least one inorganic salt comprises an anion selected from the group consisting of fluoride, chloride, bromide and iodide.

9. The electrolyte composition of claim 1 wherein said at least one inorganic salt comprises NaCl.

10. The electrolyte composition of claim 1 wherein said at least one inorganic salt comprises NaI.

11. The electrolyte composition of claim 1 wherein said at least one organic acid is present in said electrolyte at a wt/vol concentration of from about 1% to about 10%.

12. The electrolyte composition of claim 1 wherein said at least one organic acid comprises a plurality of carboxyl groups.

13. The electrolyte composition of claim 1 wherein said at least one organic acid is substantially soluble in said electrolyte solution.

14. The electrolyte composition of claim 12 wherein said at least one organic acid is at least 10 wt. % soluble in said electrolyte solution.

15. The electrolyte composition of claim 1 wherein said at least one organic acid solubility is from about 10 weight % to about 90 weight % soluble.

16. The electrolyte composition of claim 1 wherein said at least one organic acid comprises a main carbon chain of from C₂-C₈.

17. The electrolyte composition of claim 1 wherein said at least one organic acid is selected from the group consisting of Acetic acid, Citric acid, Formic acid, Malic acid, Malonic acid, Mandelic acid, Maleic acid, Icso-citric acid, Fumaric acid, Lactic acid, Glucolic acid, Glyclic acid, Hexanoic acid, Heptanoic acid, Octanoic acid, Pentanoic acid, Nonanoic acid, Propanoic acid, 3-Hydroxypropanoic acid and 2-Hydroxypropenoic acid.

18. The electrolyte composition of claim 1 wherein said at least one organic acid is citric acid.

19. The electrolyte composition of claim 18 wherein said citric acid is present in said electrolyte at a concentration of from about 0.05 M to about 5.0 M.

20. The electrolyte composition of claim 19 wherein the citric acid concentration is about 0.1 M.

21. The electrolyte composition of claim 1 wherein the concentration of citric acid is about 0.1 M, NaCl is about 0.5 to 5 weight %, and NaI is a trace amount.

22. The electrolyte composition of claim 21 wherein the electrolyte is saturated with an amount of Argon sufficient to promote cavitation-assisted electrolytic combustion.
23. The electrolyte composition of claim 1 wherein said at least one organic acid comprises the structural formula:

\[ C_\text{R} \text{H}_{2n+1} \text{COOR} \]

wherein \( R \) can be individually any of H or any alkaline earth metal; and

\( n \) is from 2 to 8.

24. The electrolyte composition of claim 1 wherein said at least one organic acid comprises the structural formula:

\[ \text{ROOC} \longrightarrow \text{C}_\text{H}_{2n+1} \longrightarrow \text{COOR} \]

wherein \( R \) can be any of H or any alkaline earth metal; and

\( n \) is from 2 to 8.

25. The electrolyte composition of claim 1 wherein said at least one organic acid comprises the structural formula:

\[ \text{ROOC} \longrightarrow (\text{CHR})_n \longrightarrow \text{C(R)}_2 \longrightarrow (\text{CHR})_n \longrightarrow \text{COOR} \]

wherein \( R \) can be any of H or any alkaline earth metal; \( R_2 \) can be either OH or COOR; and

\( n \) may be from 1 to 3.

26. The electrolyte composition of claim 1 wherein said at least one organic acid is an alpha-hydroxy acid substantially soluble in water.

27. The electrolyte composition of claim 1 wherein said at least one organic acid is a beta-hydroxy acid substantially soluble in water.

28. A method of generating hydrogen gas from water and a hydrocarbon feedstock, comprising providing a high-temperature electron-rich aqueous environment whereby water is dissociated for reaction with the feedstock and the hydrocarbon is converted to hydrogen gas and carbon dioxide.

29. A method of generating hydrogen gas from water and a hydrocarbon feedstock, comprising creating cavitation within and supplying an external source of electrons to an aqueous electrolyte solution containing a hydrocarbon feedstock thereby producing hydrogen gas and carbon dioxide.

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