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(54) **COMPOSITION FOR PRODUCING HYDROGEN RICH WATER**

ZUSAMMENSETZUNG ZUR HERSTELLUNG VON WASSERSTOFFREICHEM WASSER

COMPOSITION POUR PRODUIRE DE L'EAU RICHE EN HYDROGÈNE

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Description

BACKGROUND OF THE INVENTION

[0001] Molecular hydrogen has been found to be of potential therapeutic use for a variety of diseases and injuries.

[0002] For example, H_2 has been shown to have applications as a method for reducing wrinkles in the skin (J. Photochem. Photobiol. B. 2012; 106:24-33), treating atopic dermatitis (Evid. Based Complement. Alternat. Med. 2013; 2013:538673), and as a post-treatment regimen for radiation therapies (Biochem. J., 2012, 442(1); 49-56). Hydrogen rich water represents one way in which molecular hydrogen can be administered to subjects. Common electrolytic and base metal methods of producing hydrogen-rich water typically result in an alkaline solution with a low H_2 concentration.

[0003] Creating ready-to-drink containers of H_2 (and thus hydrogen rich water) has its technical challenges. The equipment often used to saturate water with H_2 gas in sufficient volumes is both expensive and largely ineffective. When utilized, H_2 can be dissolved at a maximum concentration of 0.8 mM or 1.6 ppm under SATP conditions as per Henry's law. In order to retain this concentration of H_2 for any period of time, the container cannot have any headspace or the drink must be supersaturated to allow H_2 dissipation into the headspace to reach equilibrium. Even when no headspace is present, the level of H_2 in the container quickly falls to ~1 ppm and will continue to fall towards 0 ppm depending on the containment technology, level of headspace, and the initial concentration, as seen by other commercial products on the market. Some products retain almost no H_2 by the time they reach consumers. For example, the Japanese government recently evaluated consumer goods containing H_2 and found that most had no detectable level of H_2 present. (http[[://]]www.kokusen.go.jp/news/data/n-20161215_2.html).

[0004] JP 2015 205791 A describes a powdery hydrogen generation agent, the agent comprising metal magnesium powder, organic acid powder, and binder.

[0005] US 2016/113865 A1 describes effervescent tablets that the consumer can add to water to generate hydrogen-rich water just prior to drinking. The effervescent tablets include a base metal and an edible acid that, within about 5-10 minutes of mixing, generate a palatable aqueous solution having about 0.8 mM to about 3 mM hydrogen and a pH of 8-10.

[0006] Accordingly, there is a need for new compositions for producing hydrogen-rich water which maximize the dissolved hydrogen concentration.

SUMMARY OF THE INVENTION

[0007] The invention is defined by the appended claims.

[0008] The invention provides compositions for producing hydrogen rich water, nutraceuticals, cosmetics, pharmaceuticals, and other products. In one embodiment, the invention provides a composition, wherein the composition is a tablet, including magnesium metal of 75 μm (200 mesh) or smaller, at least one water-soluble acid, and a binding agent. The magnesium metal and at least one water-soluble acid may be present in amounts sufficient to maintain a pH of less than 7, e.g., at a specific time period after reaction, and are present in amounts sufficient to maintain a concentration of at least 0.5 mM H_2 after reaction in 50 mL water in a container e.g., a sealed or an open container, e.g., at least 0.5 mM H_2 after reaction in 100 mL water or at least 0.5 mM H_2 after reaction in 500 mL water. The composition may also include a lubricant.

[0009] In another aspect, the invention provides a composition containing magnesium metal of 75 μm (200 mesh) or smaller, at least one water-soluble acid, and a binding agent, where the at least one water-soluble acid has a solubility of at least 0.01 g/mL in water. In certain embodiments, the composition disintegrates in less than 5 minutes, in particular less than 2 minutes. The composition produces at least 0.5 mM H_2 after contact with 50 mL water in a container at atmospheric pressure and room temperature, e.g., at least 0.5 mM H_2 after reaction in 100 mL water or at least 0.5 mM H_2 after reaction in 500 mL water. The composition may also include a lubricant.

[0010] In certain embodiments of the above aspects, the composition disintegrates in less than 5 minutes, e.g., in less than 2 minutes. In certain embodiments, the disintegrated composition maintains a pH of less than 7 at 10 minutes after being contacted with water and produces at least 0.5 mM H_2 after contact with 50 mL water in a container at atmospheric pressure and room temperature, e.g., at least 0.5 mM H_2 after reaction in 100 mL water or at least 0.5 mM H_2 after reaction in 500 mL water.

[0011] In another aspect, the invention provides a composition containing magnesium metal of 75 μm (200 mesh) or smaller, at least one acid, and a binding agent where the composition disintegrates in less than 5 minutes to maintain a pH of less than 7 10 minutes after disintegration and at least 0.5 mM H_2 after contact with 50 mL water in a container at atmospheric pressure and room temperature, e.g., at least 0.5 mM H_2 after reaction in 100 mL water or at least 0.5 mM H_2 after reaction in 500 mL water.

[0012] In certain embodiments of any of the above aspects, the composition passes a pharmaceutical test for friability. In certain embodiments, the pH of the water is less than 7 at 10, 15, 20, 30, or 45 minutes after the composition is contacted with water. In certain embodiments, the pH of the water is less than 7 at least 1 hour after the composition is

contacted with water. In certain embodiments, the container is open to the atmosphere. In certain embodiments, the container is closed. In certain embodiments, when the container is closed the pH remains less than 7 at 7 days after contact with water. In certain embodiments, the magnesium in the composition reacts to produce H_2 as it disintegrates in water, i.e., that rate of disintegration and rate of consumption of magnesium are substantially the same.

[0013] The amount of magnesium metal is, for example, 5 - 500 mg, e.g., 5 - 100 mg. The amount of acid is, for example, 30 - 4000 mg, e.g., 200 - 400 mg. In certain embodiments, the magnesium metal and acid are present in amounts sufficient to maintain a pH of between 4 and 6, and/or the magnesium metal and acid are present in amounts sufficient to produce a concentration of at least 2 mM H_2 in 50 mL of water in a container, e.g., a sealed or an open container, e.g., at least 2 mM H_2 after reaction in 100 mL water or at least 2 mM H_2 after reaction in 500 mL water. In certain embodiments, the magnesium metal includes flakes, e.g., 44 μm or smaller (-325 mesh) flakes. In other embodiments, the magnesium metal is crushed, of 75 μm (200 mesh) or smaller. In some embodiments, the at least one acid is an edible acid. The edible acid is, for example, maleic acid, succinic acid, malic acid, fumaric acid, formic acid, citric acid, ascorbic acid, oxalic acid, tartaric acid, or a combination thereof. Exemplary edible acids are tartaric acid and malic acid. In some embodiments, the acid is a cosmetically or pharmaceutically acceptable acid. The cosmetically or pharmaceutically acceptable is, for example, acetic acid, adipic acid, alginic acid, aspartic acid, benzenesulfonic acid, benzoic acid, boric acid, butyric acid, camphoric acid, camphersulfonic acid, cyclopentanepropionic acid, digluconic acid, dodecylsulfric acid, ethanesulfonic acid, glucoheptonic acid, glycerophosphoric acid, hemisulfuric acid, heptonic acid, hexanoic acid, hydrobromic, hydrochloric acid, hydroiodiic acid, 2-hydroxy-ethanesulfonic acid, lactobionic acid, lactic acid, lauric acid, lauryl sulfuric acid, malonic acid, methanesulfonic acid, 2-naphthalenesulfonic acid, nicotinic acid, nitric acid, oleic acid, palmitic acid, pamoic acid, pectic acid, persulfuric acid, 3-phenylpropionic acid, phosphoric acid, picric acid, pivalic acid, propionic acid, stearic acid, sulfuric acid, tartric acid, thiocyanic acid, toluenesulfonic acid, undecanoic acid, valeric acid, or a combination thereof. Other acids include acetylsalicylic acid and 5-aminosalicylic acid. Examples of binding agents are mannitol, xylitol, maltose, dextrose, and lactose. Exemplary binding agents are dextrose and lactose. In certain embodiments, when the acid is tartaric citric, or ascorbic acid, the amount of magnesium is greater than 20 mg, e.g., at least 50 mg, or when the acid is acetylsalicylic acid and 5-aminosalicylic acid, the amount of magnesium is greater than 20 mg, e.g., at least 50 mg.

[0014] The composition may further include a nutritional supplement, e.g., a magnesium salt, sweetener, flavoring agent, coloring agent, fragrance, essential oil, water-soluble lubricant, or polysaccharide. Exemplary polysaccharides include cellulose and its derivatives, e.g., methyl cellulose or hydroxypropyl methyl cellulose, starch, apple powder, lemon powder, lime powder, grapefruit powder, psyllium husk, and pectin. Exemplary lubricants include sodium stearyl fumarate and stearic acid, in particular sodium stearyl fumarate.

[0015] The invention also provides a kit including a composition of the invention and a sealable container capable of holding between 100 mL and 2 L of water, e.g., between 150-750 mL of water. In certain embodiments, the container is double walled.

[0016] The invention further provides a method of producing hydrogen rich water by contacting a composition of the invention with water in a container so that the composition disintegrates and the magnesium metal and at least one acid react, e.g., to produce H_2 in the water at a concentration of at least 0.5 mM H_2 and maintain a pH of less than 7 at 10 minutes after disintegration at atmospheric pressure and room temperature. In certain embodiments, the water includes fruit juice, e.g., a juice containing pectin. In other embodiments, the concentration of H_2 is at least 1 mM. In certain embodiments, a pH of less than 7 is present at 1 hour after disintegration.

[0017] The invention further provides a non-therapeutic method of administering hydrogen to a subject by providing the subject with a composition containing hydrogen produced from a composition of the invention, wherein the composition is a tablet. In some embodiments, the composition containing hydrogen is a nutraceutical or topical formulation. In one embodiment, the nutraceutical is a beverage.

[0018] The invention further provides compositions enriched with hydrogen with the hydrogen gas dissolved in a carrier at a concentration of at least 0.5 mM, wherein the pH of the composition is less than 7 and the composition is produced by contacting a composition of any one of claims 1-11 with a carrier. In some embodiments, the carrier is edible, cosmetic, or pharmaceutical grade. In some embodiments, the carrier is an aqueous liquid, cream, lotion, foam, paste, or gel. In some embodiments, the composition is a beverage. In certain embodiments, the maximum concentration of hydrogen is 20 mM. In some embodiments, the composition has a pH of 4-6. In one embodiment, the pH is 4.6 or lower. In some embodiments, the composition contains a nutritional supplement. In one embodiment, the nutritional supplement contains magnesium ions, potassium ions, or calcium ions. In some embodiments, the composition contains a sweetener, flavoring agent, coloring agent, fragrance, essential oil, or polysaccharide. In some embodiments, the composition contains a binding agent or water-soluble lubricant.

[0019] The invention further provides compositions for producing acidic hydrogen rich water. In one embodiment, the invention provides a composition, wherein the composition is a tablet, including magnesium metal of 75 μm (200 mesh) or smaller, an edible acid, and a binding agent. In general, the magnesium metal and edible acid are present in amounts sufficient to produce a pH of less than 7 and at least 0.5 mM H_2 after reaction in 500 mL of water in a sealed container.

The invention also provides a kit including this composition of the invention, wherein the composition is a tablet, and a sealable container capable of holding between 200 mL and 2L of water, e.g., between 250-750 mL of water. In certain embodiments, the container is double walled. The invention further provides a method of producing hydrogen rich water by contacting this composition of the invention, wherein the composition is a tablet, with water in a sealable container so that the composition, wherein the composition is a tablet, disintegrates and the magnesium metal and acid react to produce H_2 in the water at a concentration of at least 0.5 mM H_2 and a pH of less than 7, e.g., between pH is 4-6. In certain embodiments, the water comprises fruit juice, e.g., a juice containing pectin. In other embodiments, the concentration of H_2 is at least 1 mM. The amount of magnesium metal is, for example, 5 - 100 mg. In certain embodiments, the magnesium metal and edible acid are present in amounts sufficient to produce a pH of between 4 and 6, and/or the magnesium metal and edible acid are present in amounts sufficient to produce at least 2 mM H_2 in 500 mL of water in the sealed container. In certain embodiments, the magnesium metal is powdered, of 75 μm (200 mesh) or smaller. In other embodiments, the magnesium metal includes flakes, e.g., 44 μm or smaller (-325 mesh) flakes. The edible acid is for example, selected from the group consisting of maleic acid, succinic acid, malic acid, fumaric acid, formic acid, citric acid, ascorbic acid, and oxalic acid. Examples of binding agents are mannitol, xylitol, maltose, and lactose. The composition may further include a vitamin, mineral, e.g., a magnesium salt, sweetener, flavoring agent, water soluble lubricant, or polysaccharide. Exemplary polysaccharides include methyl cellulose, starch, apple powder, lemon powder, lime powder, grapefruit powder, psyllium husk, and pectin.

Definitions

[0020] As used herein, the term "cosmetic" refers to a composition that is applied to the all or a part of the human body, e.g., hands, face, arms, or legs, for cleansing, beautifying, promoting attractiveness, or altering the appearance.

[0021] As used herein, the term "cosmetically acceptable" refers to a composition having ingredients which are acceptable for human topical use.

[0022] As used herein, the term "nutraceutical" refers to a composition having ingredients suitable at least for human consumption. Pharmaceutical grade ingredients may optionally be employed, as described in, e.g., "Remington: The Science and Practice of Pharmacy" (22nd ed.), ed. L.V. Allen, Jr., 2013, Pharmaceutical Press, Philadelphia, PA.

[0023] As used herein, the term "passes a pharmaceutical test of friability" refers a composition that decreases in mass by at most 1% after 100 revolutions in a rotating drum of a friability tester, e.g., from Copley Scientific.

[0024] As used herein, the term "pharmaceutically acceptable" refers to a composition having ingredients which are subject to the U.S. Food and Drug Administration's pharmaceutical purity standards and further regulated by standards set by the U.S. Pharmacopoeia; this standard is 99.9% purity of a particular ingredient.

[0025] As used herein, the term "subject" refers to any animal capable of being treated topically, orally, inhalation, or intravenously with a composition containing or used to generate H_2. Animals include fish, reptiles, birds (e.g., chicken, turkey), and mammals. Mammals capable of being treated with compositions of the invention include primates (e.g., humans, apes), livestock (e.g., cows, pigs, sheep), beasts of burden (e.g., ox, horse, llama), and companion animals (e.g., dogs, cats).

DETAILED DESCRIPTION OF THE INVENTION

[0026] The present invention provides a composition, wherein the composition is a tablet, that disintegrates in water to produce hydrogen rich water. By using a composition of the invention, e.g., in a ready-to-drink container, supersaturated levels of H_2 can be achieved, considerably more than can be achieved by addition of pure H_2 gas. In contrast to prior compositions, an advantage of the present invention is the ability to produce a hydrogen-enriched composition that contains a supersaturated amount of H_2 in an open container, i.e., at atmospheric pressure. In addition, the present invention provides compositions that pass a pharmaceutical friability test but still produce high levels of H_2. A further advantage of the invention is that the compositions can react quickly, e.g., in less than 2 minutes, to produce a usable, e.g., drinkable, hydrogen-enriched product having H_2 levels significantly higher than prior compositions.

[0027] The composition contains magnesium metal, i.e., elemental magnesium, an acid, and a binding agent and optionally a lubricant. In water or a water-containing carrier, the magnesium metal and acid react to produce H_2, which dissolves in the water, and magnesium ions. An advantage of the present invention is that the composition contains sufficient acid to maintain an acidic pH during H_2 production. When insufficient acid is employed, the pH of the reaction will increase, e.g., until the solution is alkaline, causing the reaction to cease prior to reaching high levels of H_2. Without wishing to be bound by theory, at high pH, the production of H_2 ceases due to passivation from hydroxides and carbonates acting as ligands with the unreacted magnesium particles. When this occurs, less of the magnesium metal will react, thereby reducing the available H_2 produced while leaving unacceptable levels of residual solids from the composition behind in the container. Use of an acid is also advantageous, as a low pH, e.g., 4.6 or lower, aids in reducing microbial growth and therefore reducing the possibility of contamination. Thus, in certain embodiments, the invention provides

compositions that produce a hydrogen-enriched product having an acidic pH during use or storage.

Magnesium Metal

[0028] Each composition contains a sufficient mass of magnesium to produce a sufficient volume of H_2 in the volume of water to which it is added. Accordingly, the composition contains a sufficient mass of magnesium to produce at least 0.1 mmol of H_2, e.g., at least 0.5 mmol, 1 mmol, 2 mmol, 3 mmol, 5mmol, or 10 mmol of H_2, in at least 50 and optionally 75, 100, 125, 150, 175, 200, 225, 250, 500, 750, 1000, 1500, or 2000 mL of water. Suitable masses of magnesium metal include 5 - 1000 mg, e.g., 5 - 500 mg, 5 - 450 mg, 10 - 400 mg, 20 - 350 mg, 30 - 300, 40 - 250 mg, 50 - 200 mg, 60 - 100 mg, or about 70 mg or 80 mg of magnesium.

[0029] The physical form, e.g., size and shape, of the magnesium may be used to control the rate of reaction. Particles may be spherical, spheroidal, granular, or flaked. Smaller particles and particles with higher surface area to volume ratios react with faster kinetics. Mixtures of various sizes may also be employed. Flaked magnesium has a higher surface area to volume ratio than granular magnesium. In certain embodiments, flaked magnesium of 44 μm or smaller (-325 mesh) may be employed in the composition. Alternatively, or in combination, larger sized magnesium or magnesium with a lower surface area to volume ratio relative to flaked magnesium may be employed. Magnesium of 75 μm or smaller (-200 mesh) is employed. In other embodiments, magnesium of 44 μm or smaller (-325 mesh), or smaller mesh is employed. In certain embodiments, the magnesium is supplied in two sizes, e.g., 75 μm or smaller (-200 mesh) and 44 μm or smaller (-325 mesh), with the smaller size being 20-50% of the total and the larger size being the balance.

Acids

[0030] Any water-soluble acid may be employed in the invention. The acid may be edible, or otherwise of cosmetic or pharmaceutical grade. Examples of edible acids include, but are not limited to, maleic acid, succinic acid, malic acid, fumaric acid, formic acid, citric acid, ascorbic acid, oxalic acid, tartaric acid, and combinations thereof. Examples of cosmetic or pharmaceutical grade acids include acetic acid, adipic acid, alginic acid, aspartic acid, benzenesulfonic acid, benzoic acid, boric acid, butyric acid, camphoric acid, camphersulfonic acid, cyclopentanepropionic acid, digluconic acid, dodecylsulfric acid, ethanesulfonic acid, glucoheptonic acid, glycerophosphoric acid, hemisulfuric acid, heptonic acid, hexanoic acid, hydrobromic, hydrochloric acid, hydroiodiic acid, 2-hydroxy-ethanesulfonic acid, lactobionic acid, lactic acid, lauric acid, lauryl sulfuric acid, malonic acid, methanesulfonic acid, 2-naphthalenesulfonic acid, nicotinic acid, nitric acid, oleic acid, palmitic acid, pamoic acid, pectic acid, persulfuric acid, 3-phenylpropionic acid, phosphoric acid, picric acid, pivalic acid, propionic acid, stearic acid, sulfuric acid, tartric acid, thiocyanic acid, toluenesulfonic acid, undecanoic acid, valeric acid, their stereoisomers, all forms of alpha acids (e.g., α-lupulic acid), polycarboxylic acids, a Lewis acid, e.g., $AlCl_3$, and combinations thereof. Other acids include acetylsalicylic acid and 5-aminosalicylic acid. The acid will be present in an amount to react with the magnesium metal and optionally to maintain a pH of less than 7 when the composition is placed in water. It is preferable that the amount of acid chosen is sufficient to maintain a pH of less than 6, e.g., between 4 and 6, for the duration of typical beverage consumption e.g., at least 30 minutes or 1 hour. In certain embodiments, the number of moles of acid protons in the acid is at least 10, 20, 30, 40, 50, 75, or 100% greater than the number of moles of magnesium metal present. Suitable masses of the acid include 30 - 4000 mg, e.g., 100 - 1000 mg, 50 - 900 mg, 100 - 800 mg, 150 - 700 mg, 200 - 600 mg, 250 - 500 mg, 300 - 400 mg, or about 340 mg of acid. An exemplary edible acid is malic acid. Another exemplary edible acid for use in compositions of the invention is tartaric acid. Tartaric acid is highly water-soluble, having a solubility of 0.125 g/mL in water. Acids with a solubility of between about 0.01 - 1 g/mL, e.g., between about 0.02 - 0.9 g/mL, between about 0.03 - 0.8 g/mL, between about 0.04 - 0.7 g/mL, between about 0.05 - 0.6 g/mL, between about 0.06 - 0.5 g/mL, between about 0.07 - 0.4 g/mL, between about 0.08 - 0.3 g/mL, between about 0.09 - 0.2 g/mL, between about 0.1 - 0.2 g/mL, between about 0.11 - 0.5 g/mL, or between about 0.12 - 0.3 g/mL, are suitable for use in compositions of the invention. When highly water-soluble acids are used in a composition of the invention, the composition is able to quickly disintegrate upon contact with water, e.g., resulting in a more complete reaction with magnesium. This fast dissolution has the benefit of allowing the pH to remain below 7 on a timescale commensurate with beverage consumption, e.g., 1-2 hours. Other such acids, both edible and cosmetic and/or pharmaceutical grade, are known in the art.

[0031] The physical form, e.g., size and shape, of the acid may be used to control the rate of the reaction. For example, acids that are solids at room temperature, e.g., malic acid or tartaric acid, can be processed to control the size of the acid particles used to produce a composition of the invention. Smaller particles and particles with higher surface area to volume ratios react with faster kinetics. Milled acid particles may be used in a variety of mesh sizes, e.g., 400 μm (40 mesh) through 5 μm (2500 mesh). Without being bound by theory, the rate of the dissolution of the composition is believed to be linearly dependent on mesh size. Compositions of the invention made with larger acid particles, e.g., 400-250 μm (40-60 mesh), dissolve more slowly than those made with finer, e.g., 125 μm (120 mesh) through 5 μm (2500 mesh) acid particles. Mixtures of various sizes of acid particles may also be employed. Acid particles with con-

trollable sizes may be produced by a number of different techniques, including, but not limited to, micronizing, ball milling, or tumbling. Other methods of producing acid particles with controllable sizes are known in the art.

Binding Agents

[0032] Any binding agent capable of disintegrating in water may be employed. Examples of binding agents include sugars such as maltose, dextrose, and lactose, and sugar alcohols such as mannitol and xylitol. Exemplary binding agents for compositions of the invention include lactose and dextrose. Other binding agents for compositions are known in the art. The amount of binding agent is, for example, between 10 and 50% of the weight of the composition, e.g., between 20-30%. Compositions of the invention may include a single binding agent, such as lactose, or may be made from a combination of two or more binding agents to control the physical properties of the composition.

[0033] The binding agent may be edible, or otherwise be of cosmetic or pharmaceutical grade as is known in the art, e.g., in Remington (Remington: The Science and Practice of Pharmacy, (22nd ed.) ed. L.V. Allen, Jr., 2013, Pharmaceutical Press, Philadelphia, PA).

Additional Components

[0034] The composition may also include other ingredients such as a nutritional supplement, sweetener, flavoring agent, coloring agent, fragrance, essential oil, lubricant, polysaccharide, or coating. Compositions of the invention may contain nutritional supplements, e.g., vitamins, minerals, and/or herbal extracts. For example, the composition may contain a magnesium, potassium, or calcium salt. Suitable sweeteners are known in the art, e.g., sucrose, mannose, sucralose, aspartame, saccharin, stevia, monk fruit extract, and acesulfame K. The composition may also include any food grade coloring, e.g., FD&C dyes, and/or flavoring, such as a fruit flavoring. The composition may further include an essential oil, e.g., grapeseed oil, oil of wintergreen, lavender oil. Other essential oils are known in the art. A composition may further include a fragrance, e.g. eucalyptus. A composition may also contain a polysaccharide, such as pectin, psyllium fiber, cellulose, and its derivatives, e.g., methyl cellulose or hydroxypropyl methyl cellulose, various starches, apple powder, lemon powder, lime powder, or grapefruit powder. Polysaccharides may increase the amount of H_2 retained after reaction. A composition may further include a water-soluble lubricant such as micronized sodium stearyl fumarate or finely prepared stearic acid, e.g., 5 micron. The composition may also have a water-penetrable coating, such as a soluble surfactant, to control the rate at which the composition dissolves. The soluble surfactant coating may be a triblock co-polymer, e.g., a poloxamer, e.g., Poloxamer 407, or a non-ionic polymer surfactant suitable for pharmaceutical use, e.g., glucosides. For example, the composition may have a coating that dissolves in under 5 minutes, e.g., under 1 minute, to allow the user to close a container before the composition begins to disintegrate and H_2 production begins.

[0035] Previous attempts to produce effervescent compositions, e.g., tablets, using typical lubricants such as sodium lauryl sulfate as described in U.S. Patent Publication 2016/0113865, and sodium stearyl fumarate, proved unsuccessful in producing tablets that rapidly disintegrated. This was due to the use of a higher amount of lubricant needed to form the tablets. The use of large quantities of non-micronized lubricant resulted in a tablet which had a slow disintegration time, which further created excess undissolved residues in a container and a foul taste. In contrast, compositions of the invention can make use of much less lubricant, resulting in faster reaction kinetics, a satisfactory amount of residue, and a palatable taste.

Forms of Composition

[0036] The composition is formed into a tablet. A tablet may be of any suitable shape. For example, the tablet may be a disk, a sphere, or an ovoid. A single tablet will typically include the amount of magnesium and acid required to produce the desired amount of H_2 in a given volume of water, e.g., 50, 150, or 500 mL. However, a combination of multiple, smaller tablets may be employed. For example, tablets may be sized to provide sufficient H_2 in 250 mL, and multiple tablets may be employed for larger volumes. As the reaction of magnesium metal and acid is activated by water, the compositions of the invention will typically be stored in water-resistant packaging, such as foil or plastic. The components of the tablet will typically also be non-hygroscopic, but hygroscopic ingredients may be employed if the tablet is packaged dry in a waterproof container or wrapper. Tablets may be formed by methods known in the art.

[0037] A consideration when forming compositions of the invention into tablets is the tablet physical properties, e.g., friability. Friability is defined as the tendency for a tablet to chip, crumble or break following compression or other handling. Friability of tablets is assessed using a rotating drum and measuring the percent mass loss of tablets after rolling around the drum for a fixed number of drum revolutions. For a tablet to successfully pass friability testing, the tablet's mass can only decrease by 1% after 100 revolutions in the rotating drum. For compositions of the invention, friability is controlled by the type and grain size of acid used in the composition, the type and grain size of the binding agent, type and grain

size of the lubricant, and the pressure at which the tablets were pressed in the die. The use of more finely meshed particles typically results in tablets that are highly friable. As a result of this, tablets made of finely meshed particles are often made under higher pressure to ensure they do not fall apart; this has the effect of making the tablet very hard, reducing the speed at which it can disintegrate upon contact with water. Thus, tablets using fine mesh acid may be made of a highly water-soluble acid, such as tartaric acid, in order to sustain the hydrogen generation reaction.

[0038] The dissolution time of the composition, and thus the measured concentration of H_2, is controlled by the percent mass of the binding agent, the percent mass and type of lubricant, the acid to magnesium ratio, the physical properties of both the magnesium and the acid, e.g., mesh size and the physical conditions the composition is placed in. Compositions of the invention will typically disintegrate when contacted with water in a container in less than 5 minutes, e.g., less than 4 minutes, less than 3 minutes, less than 2 minutes, or less than 1 minute.

[0039] The temperature of the water the composition is placed in affects how quickly the composition disintegrates. Hot water will cause the composition to disintegrate quickly, but not hold a high concentration of hydrogen gas. Colder water increases the solubility of hydrogen in water, but does not cause rapid disintegration of the composition. A suitable temperature for the production of hydrogen from a composition of the invention is approximately room temperature, e.g., between 15°C -25°C, e.g., 15°C, 16°C, 17°C, 18°C, 19°C, 20°C, 21°C, 22°C, 23°C, 24°C, or 25°C.

[0040] The compositions will maintain an acidic hydrogen-enriched product, e.g., having a pH less than 7, e.g., 4-6, for at least a period of time after being contacted with water. An acidic pH may be maintained throughout the course of the typical time scale for use of the hydrogen-enriched product, such as 10 minutes after the composition is contacted by water. For example, the composition of the invention maintains a pH of less than 7 for a period of time of at least 5 minutes, e.g., 5 - 300 minutes, 10 - 250 minutes, 15- 200 minutes, 20 - 150 minutes, 25 - 120 minutes, 30 - 100 minutes, 50 - 90 minutes, e.g., at least 5 minutes, at least 10 minutes, at least 15 minutes, at least 20 minutes, at least 25 minutes, at least 30 minutes, at least 35 minutes, at least 40 minutes, at least 45 minutes, at least 50 minutes, at least 55 minutes, at least 60 minutes, at least 70 minutes, at least 80 minutes, at least 90 minutes, at least 100 minutes, at least 110 minutes, at least 120 minutes, at least 130 minutes, at least 140 minutes, at least 150 minutes, at least 160 minutes, at least 170 minutes, at least 180 minutes, at least 190 minutes, at least 200 minutes, at least 250 minutes, or at least 300 minutes, e.g., at least 0.5 hours, at least 1 hour, at least 1.5 hours, at least 2 hours, at least 2.5 hours, at least 3 hours, at least at least 3.5 hours, at least 4 hours, at least 4.5 hours, or at least 5 hours. Compositions may also maintain a pH that is less than 7 for an extended period of time, e.g., at one day, seven days, 30 days, or 6 months after the composition is contacted with water. Furthermore, after this timeframe, the pH of the hydrogen enriched product may become alkaline, e.g., greater than 7.

[0041] For compositions designed for use as a cosmetic additive to a shower or bath, the rate of dissolution of the tablet is an important consideration. The composition has to dissolve slowly enough in the water in order to produce a consistent level of H_2 for the duration of the time in the shower or bath. An additional constraint is the thermodynamics of the dissolution reaction, as the reaction is exothermic and produces small amounts of magnesium hydroxide. If the reaction proceeds too quickly, the temperature of the resulting bath could become too hot or produce too much magnesium hydroxide; both of these effects can harm the skin. The addition of polysaccharides to the compositions has been shown to have an effect on the dissolution rate while maximizing retained H_2. The added polysaccharide can be a fibrous polysaccharide, such as cellulose and its derivatives, e.g., hydroxypropyl methylcellulose (HPMC, known as hypromellose). Alternatively or additionally, the addition of a soluble surfactant, such as a triblock co-polymer (e.g., Poloxamer 407) can be used to slow down the dissolution of the tablet, ensuring sufficient consumption of the magnesium and maximizing the length of time that H_2 is dissolved in the water.

[0042] Liquids enriched with hydrogen for use as a cosmetic or beauty spray can have a larger acid content, and the resulting composition can take advantage of the skin's naturally occurring pH of 4.5-5.2 (Lambers et al., Int. J. Cosmet. Sci., 2006, 28, 359-370) to further enhance the H_2 concentration. H_2 has been shown to provide numerous benefits to the skin, and using the beauty spray as a cleanser to return the skin to its natural pH may have further health benefits.

Carriers

[0043] Compositions of the invention, wherein the compositions are tablets, are used by contacting them with a carrier such as water or other aqueous liquid. The water may be pure, e.g., deionized, or may contain other dissolved ions, e.g., spring or tap water. The water may also contain other ingredients, e.g., it can be or contain fruit juice, or may contain other dissolved gases, e.g., carbonated water, or dissolved solids, e.g., table sugar or salt. An exemplary fruit juice is lemon juice.

[0044] The volume of carrier is selected based on the application to be enriched with hydrogen. When a composition of the invention is used to produce a beverage, the volume of liquid, e.g., water or fruit juice, to be enriched is from about 100 mL to 2L, e.g., about 100 mL, about 150 mL, about 200 mL, about 250 mL, about 300 mL, about 350 mL, about 400 mL, about 450 mL, about 500 mL, about 550 mL, about 600 mL, about 650 mL, about 700 mL, about 750 mL, about 800 mL, about 850 mL, about 900 mL, about 950 mL, about 1 L, about 1.5 L, or about 2 L. When a composition of the

invention is used to produce a cosmetic product, the amount of water is from about 50 mL to 500 mL, e.g., about 50 mL, about 100 mL, about 150 mL, about 200 mL, about 250 mL, about 300 mL, about 350 mL, about 400 mL, about 450 mL, or about 500 mL.

[0045] Alternatively, the water may be present in a topical carrier such as a cream, lotion, foam, paste, or gel such that H_2 can be effectively delivered to the skin. Methods of producing water-soluble topical carriers are well-known in the art, e.g., as described in Remington (Remington: The Science and Practice of Pharmacy, (22nd ed.) ed. L.V. Allen, Jr., 2013, Pharmaceutical Press, Philadelphia, PA) and in the cosmetics industry. During or after reaction of the composition of the invention with water, the carrier can be stirred, mixed, or agitated to ensure uniform consistency.

Containers

[0046] Various containers may be used to contact the composition with a volume of water. In one embodiment, the container has a lid that may be used to seal the container, e.g., shortly after introducing a composition into a volume of water. A sealed container retains H_2 produced while the reaction proceeds to completion. Alternatively, H_2 can be produced in an open container. An example of a suitable container is a double walled, double gasketed stainless steel bottle.

METHODS OF USE

[0047] Compositions of the invention, wherein the compositions are tablets, are used by contacting them with a carrier that facilitates the dissolution of the composition. An exemplary carrier is water. Typically, the amount of water used to dissolve the composition is between 50 mL and 2 L, e.g., 50 mL, 150 mL, 250 mL, 355 mL, 500 mL, 750 mL, or 1L. The user can add the composition to the water or other carrier in a sealable container and allow the reaction to proceed for 1 or more minutes depending on the temperature of the water, e.g., 1-2 minutes, at least 5 min, 10 min, 15 min, 30 min, 45 min, 60 min, 90 min, or 12 h. In certain embodiments, it is preferred that the composition react in less than 2 minutes. Preferably, the tablet and volume of water produce a concentration of at least 0.5 mM, e.g., at least 1 mM, at least 3 mM, at least 5 mM, or at least 10 mM, e.g., between 0.5 - 20 mM, 1-15 mM, or 5 - 10 mM. The inclusion of a polysaccharide, either in the composition, or in the water or carrier, e.g., in fruit juice, may increase the concentration of H_2 relative to the reaction in the absence of the polysaccharide, either locally near the polysaccharide or in the H_2 enriched composition as a whole.

[0048] As is known in the art, the consumption of hydrogen rich water aids in the treatment of various disorders including Parkinson's disease (Yoritaka et al., BMC Neurology, 2016, 16:66), depression (Zhang et al. Sci. Rep. 2016; 6:23742), periodontitis (Azuma et al. Antioxidants (Basel). 2015; 4(3):513-22), diabetes type II, metabolic syndrome, chronic renal failure, inflammation, rheumatoid arthritis, interstitial cystitis, cerebral ischemia, hyperlipidemia, chronic hepatitis B, and others as described in Ichihara et al. (Med. Gas Res. (2015) 5:12). Accordingly, the compositions of the present invention may be consumed by subjects suffering from any of these disorders to treat the disorder or alleviate one or more symptoms thereof.

[0049] Additionally, H_2 has been shown to be an effective treatment for a variety of dermatological conditions. For example, when a composition of the invention is used to make a hydrogen enriched aqueous liquid, the pH of the resulting aqueous liquid can be adjusted to create a "beauty water" with a pH of 4.5-5.5, which has numerous health benefits (Lambers et al., Int. J. Cosmet. Sci. 2006, 28, 359-370). This "beauty water" has been used as a carrier base for ionic magnesium topical cosmetics, with the lower pH and H_2 content effectively facilitating magnesium absorption through the skin (Magnes. Res. 2016; 29(2):35-42). In another example, H_2 containing products have been shown to be a promising treatment for topical skin conditions such as wrinkles, atopic dermatitis, and UV-induced burns to the skin (Mol. Cell. Toxicol. 2013, 9(1), 15-21). For topical indications, the compositions of the invention can be directly incorporated into a dermatological carrier such as a cream, lotion, foam, paste, or gel.

[0050] Hydrogen containing products produced from in-situ generation of H_2 can be used to improve the health of certain livestock animals, in particular, dairy cows. It is believed that H_2 has potential to increase the usable lifespan and longevity of dairy cows, resulting in increased milk production.

[0051] Compositions of the invention may also be used to produce hydrogen gas that is inhaled, e.g., by breathing the gas as it evolves either from an open container or via a cannula or nasal tube.

HYDROGEN-ENRICHED ACIDIC COMPOSITIONS

[0052] A composition of the invention can be used in the manufacture of a number of consumer products, including, but not limited to, edible foodstuffs and nutraceuticals (e.g., beverages), and skin care products, e.g., lotions, bath bombs, or shower tablets, for effective delivery of H_2 to the skin. In certain embodiments, the hydrogen enriched composition is a beverage in an open container. For topical compositions, a composition of the invention can be directly incorporated

into a pharmaceutical grade or cosmetic grade topical carrier such as a cream, lotion, foam, paste, or gel. Topical compositions containing H_2 can be soaked in, rolled, rubbed on, or sprayed directly onto the skin.

[0053] For consumer products designed to be ingested within the human body, e.g., nutraceuticals, e.g., beverages, the acid used in the production of the composition of the invention must be safely consumable, as with the edible acids described herein (e.g., malic acid or tartaric acid). The acid used in a composition of the invention to be used in the manufacture of consumer products designed for topical administration may be any pharmaceutically or cosmetically acceptable acid and its counterion that are considered "generally regarded as safe" as defined us the U.S. Food and Drug Administration for human and veterinary use. Representative acids include acetic acid, adipic acid, alginic acid, aspartic acid, benzenesulfonic acid, benzoic acid, boric acid, butyric acid, camphoric acid, camphersulfonic acid, cyclopentanepropionic acid, digluconic acid, dodecylsulfric acid, ethanesulfonic acid, glucoheptonic acid, glycerophosphoric acid, hemisulfuric acid, heptonic acid, hexanoic acid, hydrobromic, hydrochloric acid, hydroiodiic acid, 2-hydroxyethanesulfonic acid, lactobionic acid, lactic acid, lauric acid, lauryl sulfuric acid, malonic acid, methanesulfonic acid, 2-naphthalenesulfonic acid, nicotinic acid, nitric acid, oleic acid, palmitic acid, pamoic acid, pectic acid, persulfuric acid, 3-phenylpropionic acid, phosphoric acid, picric acid, pivalic acid, propionic acid, stearic acid, sulfuric acid, tartric acid, thiocyanic acid, toluenesulfonic acid, undecanoic acid, valeric acid, their stereoisomers, all forms of alpha acids (e.g., α-lupulic acid), polycarboxylic acids, a Lewis acid, e.g., $AlCl_3$, or combinations thereof. Other such acids are known in the art.

[0054] The hydrogen enriched water produced from a composition of the invention has a dissolved H_2 concentration between 0.5 mM and 20 mM, e.g., between 1 mM and 15 mM, between 1 and 10 mM, between 1 mM and 4 mM, between 1 mM and 3 mM, between 1 mM and 2 mM, between 1.5 mM and 4 mM, or between 2 mM and 3 mM, e.g., about 0.5 mM, about 1 mM, about 1.5 mM, about 2 mM, about 3 mM, about 4 mM, about 5 mM, about 6 mM, about 7 mM, about 8 mM, about 9 mM, about 10 mM, about 15 mM, or about 20 mM. In other embodiments, the concentration is between 1 ppm and 3 ppm, between 2 ppm and 4 ppm, between 3 ppm and 6 ppm, between 4 ppm and 8 ppm, between 5 ppm and 10 ppm, between 6 ppm and 12 ppm, or between 5 ppm and 15 ppm.

[0055] The acid content of the composition used to enrich the water with hydrogen may be sufficient to maintain a pH of less than 7, e.g., less than 6, e.g. between 4-6, while consuming a sufficient amount of the magnesium. A hydrogen enriched composition may also include a nutritional supplement, e.g., a magnesium salt, sweetener, flavoring agent, coloring agent, fragrance, essential oil, water-soluble lubricant, or a polysaccharide.

EXAMPLES

Example 1

[0056] In this example, hydrogen enriched water was created by dissolving the following two compositions in separate open containers and monitoring the evolved hydrogen concentration as a function of time.

Sample composition #1 - "F6" - dissolved in 500 mL of water held at 17°C

80 mg magnesium, 44 μm or smaller (-325 mesh), flaked

120 mg tartaric acid, 125 μm (120 mesh)

200 mg malic acid, 125 μm (120 mesh)

200 mg dextrose

6 mg sodium stearyl fumarate

Sample composition #2 - "F1" - dissolved in 500 mL of water held at 17°C

55 mg magnesium, 75 μm or smaller (-200 mesh), crushed

25 mg magnesium, 44 μm or smaller (-325 mesh), flaked

340 mg malic acid, 250 μm (60 mesh)

160 mg lactose

6 mg sodium stearyl fumarate

[0057] The F6 composition containing milled 125 μm (120 mesh) acid particles dissolved faster (in approximately 1.75 min) than the F1 composition with larger 250 μm (60 mesh) grained acid particles (approximately 3.5 min). If the acids are only milled to 250 μm (60 mesh) in the F6 composition, the dissolution time of the tablet is approximately 3 min. Both the F6 and F1 compositions passed the minimum pharmaceutical test for friability. Further experimental data using sub-10 micron milled tartaric acid particles in the F6 composition instead of 125 μm (120 mesh) particles resulted in a tablet dissolution time of 45 seconds. Further, by using dextrose instead of lactose as the binder, the dissolution time per tablet decreased by approximately 30 second while allowing friability to stay within acceptable limits.

[0058] The hydrogen concentration achieved by the F6 composition after it fully disintegrated was 9 ppm. For the F1 composition, the hydrogen concentration after full disintegration was 3.5 ppm. A similar composition that did not pass friability testing provided a peak hydrogen concentration of 12 ppm after about 75 seconds. All concentration data were an average of approximately 20 individual tablets of each composition.

[0059] In a second experiment, the dissolution of a composition containing both tartaric acid and malic acid ("F35") with the following ingredients was investigated:

60 mg magnesium, 44 μm or smaller (-325 mesh), flaked

90 mg tartaric acid

150 mg malic acid

150 mg dextrose

6 mg sodium stearyl fumarate

[0060] In identical water conditions as F1 and F6 (e.g., 500 mL water in open container at 17°C), the concentration was 5.3 ppm after approximately 80-90 s of reaction time. The measured concentration was an average of approximately 20 tablets.

Example 2

[0061] An exemplary tablet for use in creating a hydrogen enriched ready-to-drink beverage includes the following components:

30 mg 75 μm or smaller (-200 mesh) magnesium

30 mg 44 μm or smaller (-325 mesh) magnesium, flaked

90 mg tartaric acid

150 mg malic acid

150 mg dextrose

5.5 mg 5 μm (2500 mesh) stearic acid

Example 3

[0062] A tablet for use for producing a hydrogen enriched beverage in a closed container includes the following ingredients:

55 mg 75 μm or smaller (-200 mesh) magnesium

25 mg 44 μm or smaller (-325 mesh) magnesium, flaked

310 mg malic acid

100 mg magnesium malate

160 mg lactose

7 mg sodium stearyl fumarate

[0063] These ingredients were pressed into a tablet using a hand operated mechanical tablet press. This tablet, when dissolved in water in an airtight 500 mL container, produces H_2 gas. In a standard soda bottle (500 mL), the H_2 concentration reached 1.6 ppm (0.8 mM) within 15 minutes, 4 ppm (2 mM) within 2 hours, and exceeded 6 ppm (3 mM) within 12 hours. In a double walled, double gasketed stainless steel bottle, the concentration reached 2.8 ppm (1.4 mM) at 15 minutes, 3.8 ppm (1.9 mM) at 1 hour, and exceeded 7 ppm (3.5 mM) at 12 hours. The pH of the final solution when the tablet is added to plain water is between 4-6.

[0064] When fruit juice, including fruit juices high in pectin such as lemon, lime, apple, and orange, is used as or in addition to the liquid, the concentration of H_2 in the foam at the top of the liquid can exceed 20 ppm (10 mM). An increase in H_2 concentration was also observed with premixed pectin (Certo®) and psyllium husk.

Example 4

[0065] A tablet configured for use in a beverage for producing high concentration of H_2 includes the following components:

30 mg magnesium

200 mg malic acid

Sufficient amounts of both binding agent and lubricant

These ingredients may be pressed into a suitable tablet shape in a tableting die of 9-11 mm in diameter.

Example 5

[0066] A tablet configured for use in a cosmetic, shower, or bathtub use include the following components:

480 mg magnesium

720 mg tartaric acid

1200 mg malic acid

Sufficient amounts of both binding agent and lubricant to bring the tablet mass to 3600 mg.

These ingredients may be pressed into a suitable tablet shape in a tableting die of 24 mm in diameter.

Example 6

[0067] A second tablet configured for use in a cosmetic, shower, or bathtub use include the following components:

240 mg 44 μm or smaller (-325 mesh) magnesium

360 mg 180 μm (80 mesh) (or lower) tartaric acid

600 mg 180 μm (80 mesh) (or lower) malic acid

Sufficient amounts of both binding agent and lubricant to bring the tablet mass to 1800 mg.

These ingredients may be pressed into a suitable tablet shape in a tableting die of 18 mm in diameter.

Example 7

[0068] A tablet configured for use in a beverage or cosmetic spray includes the following components:

80 mg 44 μm or smaller (-325 mesh) magnesium

120 mg 180 μm (80 mesh) (or lower) tartaric acid

200 mg 180 μm (80 mesh) (or lower) malic acid

Sufficient amounts of both binding agent and lubricant to bring the tablet mass to 600 mg.

These ingredients may be pressed into a suitable tablet shape in a tableting die of 12 mm in diameter.

Example 8

[0069] A tablet configured for use in a beverage or cosmetic spray includes the following components:

60 mg magnesium

90 mg tartaric acid

200 mg malic acid

Sufficient amounts of both binding agent and lubricant to bring the tablet mass to 4500 mg.

These ingredients may be pressed into a suitable tablet shape in a tableting die of 12 mm in diameter.

Example 9

[0070] A tablet configured for use exclusively in a cosmetic or beauty spray includes the following components:

25-40 mg magnesium

A sufficient amount of acid, in an amount higher than the tablets used to produce an enriched beverage.

Sufficient amounts of both binding agent and lubricant

These ingredients may be pressed into a suitable tablet shape in a tableting die of 9 mm in diameter.

Example 10

[0071] An advantage of a tablet comprising finely flaked magnesium particles is that molecules of H_2 are evolved one at a time. When sufficient acid is present, and the mass of magnesium in the tablet is appropriate for the volume of liquid to be saturated with H_2 (at least 80 mg of magnesium and 300 mg total acid per 500 mL of liquid), the H_2 will continuously evolve, creating bubbles of H_2 first in the picometer size range, before they coalesce to nanometer sized bubbles, then micrometer sized bubbles, and then larger bubbles. Nanometer sized bubbles are able to saturate an aqueous solution to a higher level than bubbles of other sizes and therefore can create a higher pressure of H_2 in the liquid. This is because larger bubbles dissipate out of solution, but nanometer sized bubbles are more stable and the physiochemical properties of the nanometer sized bubbles are different than the individually dissolved H_2 molecules, which changes the relation with Henry's Law and the gas's fugacity coefficient. The increased pressure stops the reaction as per Le Chatelier's principle, leaving sub-micrometer sized magnesium flakes suspended in solution. As the H_2 bubbles coalesce and further dissipate, the pressure of the system drops, and an equal amount of magnesium reacts to further produce H_2.

[0072] This continuous reaction allows for the concentration of H_2 generated to be greater than 3 ppm when the composition is placed within an open container rather than a sealed bottle, e.g., under 1 atmosphere of pressure, but deliver a constant replenishment of H_2 to bring it the local concentration of H_2 to approximately 9 ppm. The speed of the tablet disintegrating and subsequent reaction of the magnesium and acid can be accelerated by choosing tablet components, e.g. coatings or binders, to control the reaction kinetics, with the reaction running to completion in no more

than 4 minutes, e.g., in the range of 1-2 minutes.

[0073] The use of cold water to dissolve compositions of the invention allows for the retention of an increased concentration of hydrogen but also causes a significant slowing down of the dissolution rate of the tablet, and subsequently the overall hydrogen generation reaction. For example, in water that is just above freezing (1 °C), tablets of the invention typically need 4-5 minutes to fully dissolve. Once the water is saturated, though, the dissolved hydrogen is retained for longer in the water at a higher concentration. The use of hot water to dissolve a composition of the invention results in a significant increase in the dissolution rate and subsequently faster evolution of hydrogen. For example, in hot water, e.g., greater than room temperature, tablets of the invention typically fully dissolve is 1 minute or less. However, the rate of bubble coalescence increases dramatically with the increasing temperature, thus reducing the retention time and overall stability of the enriched water.

[0074] Using current tableting technology and capabilities, the ideal water temperature range for dissolving a tablet made from a composition of the invention is between 12-20°C, depending on the final composition of the tablet.

Example 11

[0075] A composition of the invention, wherein the composition is a tablet, is able to dissolve and produce a semi-stable supersaturation of H_2 in an open, e.g., ambient pressure, container. Polysaccharides contained within the composition or present in the liquid carrier are able to form a boundary layer at the surface of the liquid. This boundary layer prevents the H_2 gas cloud which forms from the dissolution of the composition from quickly dissipating. For example, in a rigid container, the addition of a pH modifier, e.g., 2 tablespoons of lemon juice (which contains pectin) or vinegar, increases the available concentration of H_2. Use of vinegar results in a higher concentration of H_2 throughout the liquid. When water and lemon juice are placed in a standard soda bottle made primarily of polyethylene terephthalate (PET), the concentration of H_2 is increased at the top of the gas cloud by a factor of 6-7x. In an open glass bottle with the same solution of water and lemon juice, the concentration of H_2 gas produced increases by approximately 20%. When polysaccharides are used, a foam forms on the surface of the liquid which includes a higher concentration of H_2 relative to the rest of the liquid.

[0076] Open containers are able to quickly create a suspension of magnesium nanoparticles once the tablet is placed in the container. This increases the reaction rate for producing H_2. For example, in a tablet that fully reacts in 30 to 60 seconds, the bubbles formed at the top of the surface aggressively burst and resulted in a measured H_2 concentration of 1.6 ppm.

[0077] For a composition of the invention, 70-90 seconds is ideal reaction speed, often reaching 10 ppm of supersaturation within the gas cloud. The measured concentration of H_2 appears to linearly decrease with time, as shown in the data in Table 1, reaching the typical SATP concentration of 1.6 ppm after 8 minutes of dissolution.

Table 1. Measured H_2 concentrations after dissolving a composition of the invention

Time of Dissolution (s)	Measured H_2 concertation (ppm)
80-90	10
150	6
180	4.5-5
210	3.5-4
300	2.2-2.5
360	1.8
480	1.6

Example 12

[0078] In this example, H_2 enhanced water created by dissolving a composition of the invention, wherein the composition is a tablet, in an open container was transferred into a sealable swing top glass bottle and allowed to stabilize under pressure upon further dissolution of the composition. When the sealed bottle was opened, the measured concentration of H_2 was 5.3 ppm.

[0079] In a further experiment, H_2 enhanced water created by dissolving a composition of the invention, wherein the composition is a tablet, in an open container was transferred into a sealable PET soda bottle modified to include a pressure gauge for measuring the pressure of the interior of the bottle. As the reaction progressed, that bottle began to pressurize, creating a headspace within the bottle as H_2 bubbles were formed and subsequently dissipated. After 5

minutes of reaction, the measured pressure of the bottle was 172 kPa (25 psi), and after 30 minutes of reaction, the measured pressure of the bottle was 310 kPa (45 psi). At this pressure, the measured concentration of H_2 was 2.3 ppm.

[0080] The composition in both containers (open and sealed bottle) continued to react with micrometer particles. As the H_2 coalesced and dissipated, the pressure that had been contained inside the liquid transferred to the headspace, increasing the pressure in the container. Remarkably, the pressure reached by transferring the open container liquid into the PET bottle reaches and even exceeds the pressure created by dropping a tablet into a PET bottle and immediately sealing. The same tablet produces roughly 241 kPa (35 psi) when sealed immediately due to the reaction stopping as per Le Chatelier's principle, thus also confirming the supersaturation ability of the composition.

Example 13

[0081] One variable that exerts control over the production of hydrogen from a composition of the invention is the mass of magnesium used to react with the milled acid. The relationship mass of magnesium and dissolved hydrogen concentration is approximately linear - as the amount of magnesium used increases, the amount of hydrogen produced for a fixed mass of acid increases. Table 2 presents hydrogen concentration data for a variety of magnesium masses where the acid was milled to a fine mesh (~125 μm (120 mesh)) particle size and a dissolution time of approximately 60-75 s.

Table 2. Measured H_2 concentrations after dissolving a composition of the invention made with ~125 μm (120 mesh) acid

Mass of magnesium (mg)	Measured H_2 concertation (ppm)
80	9-12
70	N/A
60	4-5
55	3
50	2.6-3

[0082] The effect of milling the acids was also studied by performing laser diffraction size distribution measurements of the bubbles produced during the dissolution reaction. Using finely milled acids, the generated bubbles exhibited a bimodal distribution, with a first mode at a diameter of approximately 50-60 nm and the second mode at a diameter of 600 nm after compensating for noise. As noted in Example 10, nanometer sized bubbles are able to saturate an aqueous solution to a higher level than bubbles of other sizes and therefore can create a higher pressure of H_2 in the liquid, thus confirming the importance of using finely milled acids in producing compositions for enriching water with hydrogen.

[0083] Additional testing in an open container using a tablet made with a finely milled acid produced 70 mL of hydrogen gas out of a theoretical limit of 80 mL within the first 80-90 seconds. By the second minute of the reaction, only 6 mL of hydrogen gas had escaped from the surface of the liquid in the open container.

Claims

1. A composition comprising:

magnesium metal of 75 μm (200 mesh) or smaller;
at least one acid; and
a binding agent;
wherein the composition produces at least 0.5 mM H_2 after contact with 50 mL water in a container at atmospheric pressure and room temperature;
wherein the composition is a tablet and
wherein:

i) the composition disintegrates in less than 5 minutes;
ii) the at least one acid is water-soluble, and has a water solubility of at least 0.01g/mL; or,
iii) the composition maintains a pH of less than 7 10 minutes after contact.

2. The composition of claim 1, wherein the composition passes a pharmaceutical test for friability; or, wherein the

composition disintegrates in less than 2 minutes.

3. The composition of claim 1, wherein the pH of the water remains less than 7 for at least 10 minutes after the composition is contacted by water; or wherein the container is closed, and the composition maintains a pH of less than 7 at 7 days after contact with water.

4. The composition of claim 1, wherein:

a) the magnesium metal comprises flakes; preferably flakes of 44 μm or smaller (-325 mesh); and/or
b) the magnesium metal is crushed; and/or,
c) the amount of magnesium metal is 5 - 500 mg.

5. The composition of claim 1, wherein:

a) the at least one acid comprises an edible acid, preferably wherein the at least one edible acid is selected from the group consisting of maleic acid, succinic acid, malic acid, fumaric acid, formic acid, citric acid, ascorbic acid, oxalic acid, and tartaric acid, or a mixture thereof; or,
b) the at least one acid is a cosmetically or pharmaceutically acceptable acid, preferably acetic acid, adipic acid, alginic acid, aspartic acid, benzenesulfonic acid, benzoic acid, boric acid, butyric acid, camphoric acid, camphersulfonic acid, cyclopentanepropionic acid, digluconic acid, dodecylsulfric acid, ethanesulfonic acid, glucoheptonic acid, glycerophosphoric acid, hemisulfuric acid, heptonic acid, hexanoic acid, hydrobromic, hydrochloric acid, hydroiodiic acid, 2-hydroxy-ethanesulfonic acid, lactobionic acid, lactic acid, lauric acid, lauryl sulfuric acid, malonic acid, methanesulfonic acid, 2-naphthalenesulfonic acid, nicotinic acid, nitric acid, oleic acid, palmitic acid, pamoic acid, pectic acid, persulfuric acid, 3-phenylpropionic acid, phosphoric acid, picric acid, pivalic acid, propionic acid, stearic acid, sulfuric acid, tartric acid, thiocyanic acid, toluenesulfonic acid, undecanoic acid, valeric acid, their stereoisomers, all forms of alpha acids (e.g., α-lupulic acid), polycarboxylic acids, a Lewis acid, or a combination thereof.

6. The composition of claim 1, wherein the amount of the at least one acid is 30 - 4000 mg.

7. The composition of claim 1, wherein the binding agent is mannitol, xylitol, maltose, dextrose, or lactose.

8. The composition of claim 1, further comprising:

a) a water-soluble lubricant; preferably wherein the water-soluble lubricant is selected from sodium stearyl fumarate or stearic acid; and/or
b) a nutritional supplement, preferably wherein the nutritional supplement is a magnesium salt; and/or,
c) a sweetener or flavoring agent; and/or,
d) a coloring agent; and/or,
e) a fragrance; and/or,
f) an essential oil; and/or,
g) a polysaccharide; preferably wherein the polysaccharide is selected from the group consisting of cellulose and its derivatives, starch, apple powder, lemon powder, lime powder, grapefruit powder, psyllium husk, and pectin.

9. The composition of claim 1, wherein the magnesium metal and at least one acid are present in amounts sufficient to produce at least 2 mM H_2.

10. The composition of claim 1, wherein the at least one acid is water-soluble and has a solubility of at least 0.05 g/mL in water; or, wherein the at least one acid is water-soluble and has a solubility of at least 0.1 g/mL in water; or, wherein the at least one acid is water-soluble and has a solubility of at least 1 g/mL in water.

11. The composition of claim 1, wherein the pH of less than 7 is maintained at 30 minutes after contact with water; or, wherein the pH of less than 7 is maintained at 1 hour after contact with water.

12. A kit comprising a composition of any of claims 1-11 and a sealable container capable of holding between 100 mL and 2L of water; preferably wherein the container is double walled; optionally wherein the container is capable of holding between 250-750 mL of water.

13. A method of producing hydrogen rich water, the method comprising the steps of: contacting a composition of any one of claims 1-11 with water in a container so that the composition disintegrates and the magnesium metal and at least one acid react to produce H_2; optionally wherein the pH of less than 7 is sustained for at least 1 hour; and/or wherein the pH is 4-6; and/or wherein the water comprises fruit juice; and/or wherein the concentration of H_2 is at least 0.5 mM.

14. A non-therapeutic method of administering hydrogen to a subject, the method comprising providing a subject with a composition containing hydrogen produced from a composition of any one of claims 1-11; optionally wherein the composition containing hydrogen is a nutraceutical or topical formulation, preferably wherein the nutraceutical is a beverage.

15. A composition enriched with hydrogen, comprising hydrogen dissolved in a carrier at a concentration of at least 0.5 mM wherein the pH of the composition is less than 7 and the composition is produced by contacting a composition of any one of the claims 1-11 with a carrier;

optionally wherein the composition is edible, cosmetic or pharmaceutical grade;
optionally wherein the carrier is selected from an aqueous liquid, cream, lotion, foam, paste, or gel, or is a beverage;
optionally wherein the maximum concentration of hydrogen is 20 mM;
optionally wherein the pH is 4-6 or is 4.6 or lower;
optionally wherein the composition contains a nutritional supplement; or
optionally wherein the composition contains a sweetener, flavoring agent, coloring agent, fragrance, essential oil, polysaccharide, binding agent, or water-soluble lubricant.

Patentansprüche

1. Zusammensetzung, umfassend:

Magnesiummetall einer Größe von 75 μm (200 Mesh) oder kleiner,
mindestens eine Säure und
ein Bindemittel,
wobei die Zusammensetzung nach Kontakt mit 50 ml Wasser in einem Behälter bei Atmosphärendruck und Raumtemperatur mindestens 0,5 mM H_2 bildet,
wobei es sich bei der Zusammensetzung um eine Tablette handelt und wobei:

i) die Zusammensetzung innerhalb von weniger als 5 Minuten zerfällt,
ii) die mindestens eine Säure wasserlöslich ist und eine Löslichkeit in Wasser von mindestens 0,01 g/ml aufweist oder
iii) die Zusammensetzung 10 Minuten nach dem Kontakt einen pH-Wert von unter 7 aufrechterhält.

2. Zusammensetzung nach Anspruch 1, wobei die Zusammensetzung einen pharmazeutischen Test auf Brüchigkeit besteht oder wobei die Zusammensetzung in weniger als 2 Minuten zerfällt.

3. Zusammensetzung nach Anspruch 1, wobei der pH-Wert des Wassers nach dem Inkontaktbringen der Zusammensetzung mit Wasser mindestens 10 Minuten unter 7 bleibt oder wobei der Behälter geschlossen ist und die Zusammensetzung nach dem Kontakt mit Wasser 7 Tage einen pH-Wert von unter 7 aufrechterhält.

4. Zusammensetzung nach Anspruch 1, wobei:

a) das Magnesiummetall Flocken umfasst, vorzugsweise Flocken mit einer Größe von 44 μm oder kleiner (-325 Mesh), und/oder
b) das Magnesiummetall zerstoßen ist und/oder
c) die Menge an Magnesiummetall 5 - 500 mg beträgt.

5. Zusammensetzung nach Anspruch 1, wobei:

a) die mindestens eine Säure eine essbare Säure umfasst, wobei die mindestens eine essbare Säure vorzugs-

weise aus der aus Maleinsäure, Bernsteinsäure, Äpfelsäure, Fumarsäure, Ameisensäure, Citronensäure, Ascorbinsäure, Oxalsäure und Weinsäure oder einer Mischung davon bestehenden Gruppe ausgewählt ist, oder b) es sich bei der mindestens einen Säure um eine kosmetisch oder pharmazeutisch unbedenkliche Säure, vorzugsweise Essigsäure, Adipinsäure, Alginsäure, Asparaginsäure, Benzolsulfonsäure, Benzoesäure, Borsäure, Buttersäure, Camphersäure, Camphersulfonsäure, Cyclopentanpropionsäure, Digluconsäure, Dodecylsulfonsäure, Ethansulfonsäure, Glucoheptonsäure, Glycerophosphorsäure, Hemischwefelsäure, Heptonsäure, Hexansäure, Bromwasserstoffsäure, Salzsäure, Iodwasserstoffsäure, 2-Hydroxyethansulfonsäure, Lactobionsäure, Milchsäure, Laurinsäure, Laurylschwefelsäure, Malonsäure, Methansulfonsäure, 2-Naphthalinsulfonsäure, Nicotinsäure, Salpetersäure, Ölsäure, Palmitinsäure, Pamoasäure, Pektinsäure, Persulfonsäure, 3-Phenylpropionsäure, Phosphorsäure, Pikrinsäure, Pivalinsäure, Propionsäure, Stearinsäure, Schwefelsäure, Weinsäure, Thiocyansäure, Toluolsulfonsäure, Undecansäure, Valeriansäure, deren Stereoisomere, alle Formen von Alphasäuren (z. B. α-Lupulinsäure), Polycarbonsäuren, eine Lewis-Säure oder eine Kombination davon handelt.

6. Zusammensetzung nach Anspruch 1, wobei die Menge der mindestens einen Säure 30 - 4000 mg beträgt.

7. Zusammensetzung nach Anspruch 1, wobei es sich bei dem Bindemittel um Mannit, Xylit, Maltose, Dextrose oder Lactose handelt.

8. Zusammensetzung nach Anspruch 1, weiterhin umfassend:

a) ein wasserlösliches Schmiermittel, wobei das wasserlösliche Schmiermittel vorzugsweise aus Natriumstearylfumarat und Stearinsäure ausgewählt ist, und/oder
b) ein Nahrungsergänzungsmittel, wobei es sich bei dem Nahrungsergänzungsmittel vorzugsweise um ein Magnesiumsalz handelt, und/oder
c) einen Süßstoff oder Geschmackstoff und/oder
d) einen Farbstoff und/oder
e) einen Duftstoff und/oder
f) ein essentielles Öl und/oder
g) ein Polysaccharid, wobei das Polysaccharid vorzugsweise aus der aus Cellulose und deren Derivaten, Stärke, Apfelpulver, Zitronenpulver, Limettenpulver, Grapefruitpulver, Flohsamenschalen und Pektin bestehenden Gruppe ausgewählt ist.

9. Zusammensetzung nach Anspruch 1, wobei das Magnesiummetall und die mindestens eine Säure in Mengen vorliegen, die ausreichen, um mindestens 2 mM H_2 zu bilden.

10. Zusammensetzung nach Anspruch 1, wobei die mindestens eine Säure wasserlöslich ist und eine Löslichkeit in Wasser von mindestens 0,05 g/ml aufweist oder wobei die mindestens eine Säure wasserlöslich ist und eine Löslichkeit in Wasser von mindestens 0,1 g/ml aufweist oder wobei die mindestens eine Säure wasserlöslich ist und eine Löslichkeit in Wasser von mindestens 1 g/ml aufweist.

11. Zusammensetzung nach Anspruch 1, wobei der pH-Wert von weniger als 7 nach dem Kontakt mit Wasser 30 Minuten aufrechterhalten wird oder wobei der pH-Wert von weniger als 7 nach dem Kontakt mit Wasser 1 Stunde aufrechterhalten wird.

12. Kit, umfassend eine Zusammensetzung nach einem der Ansprüche 1-11 und einen verschließbaren Behälter, der zwischen 100 ml und 2 l Wasser aufnehmen kann, wobei der Behälter doppelwandig ist, wobei der Behälter gegebenenfalls 250-750 ml Wasser aufnehmen kann.

13. Verfahren zur Herstellung von wasserstoffreichem Wasser, wobei das Verfahren die folgenden Schritte umfasst: das Inkontaktbringen einer Zusammensetzung nach einem der Ansprüche 1 bis 11 mit Wasser in einem Behälter, so dass die Zusammensetzung zerfällt und das Magnesiummetall und die mindestens eine Säure unter Bildung von H_2 reagieren, wobei der pH-Wert von weniger als 7 gegebenenfalls für mindestens 1 Stunde aufrechterhalten wird und/oder wobei der pH-Wert 4 bis 6 beträgt und/oder wobei das Wasser Fruchtsaft umfasst und/oder wobei die Konzentration an H_2 mindestens 0,5 mM beträgt.

14. Nichttherapeutisches Verfahren zur Verabreichung von Wasserstoff an ein Subjekt, wobei das Verfahren die Bereitstellung einer Wasserstoff enthaltenden Zusammensetzung, die aus einer Zusammensetzung nach einem der

Ansprüche 1 bis 11 hergestellt wurde, für ein Subjekt umfasst, wobei es sich bei der Wasserstoff enthaltenden Zusammensetzung gegebenenfalls um ein Nutrazeutikum oder eine topische Formulierung handelt, wobei es sich bei dem Nutrazeutikum vorzugsweise um ein Getränk handelt.

15. Mit Wasserstoff angereicherte Zusammensetzung, umfassend Wasserstoff in einem Träger in einer Konzentration von mindestens 0,5 mM, wobei der pH-Wert der Zusammensetzung weniger als 7 beträgt und die Zusammensetzung durch Inkontaktbringen einer Zusammensetzung nach einem der Ansprüche 1-11 mit einem Träger hergestellt wird, wobei die Zusammensetzung gegebenenfalls essbar, kosmetischer oder pharmazeutischer Qualität ist,

wobei der Träger gegebenenfalls aus einer wässrigen Flüsigkeit, einer Creme, einer Lotion, einem Schaum, einer Paste oder einem Gel ausgewählt ist oder ein Getränk ist,
wobei die Höchstkonzentration an Wasserstoff gegebenenfalls 20 mM beträgt,
wobei der pH-Wert gegebenenfalls 4-6 oder 4,6 oder weniger beträgt,
wobei die Zusammensetzung gegebenenfalls ein Nahrungsergänzungsmittel enthält oder
wobei die Zusammensetzung gegebenenfalls einen Süßstoff, einen Geschmackstoff, einen Farbstoff, einen Duftstoff, ein essentielles Öl, ein Polysaccharid, ein Bindemittel oder ein wasserlösliches Schmiermittel enthält.

Revendications

1. Composition comprenant :

du magnésium métallique de 75 μm (200 mesh) ou moins ;
au moins un acide ; et
un liant ;
la composition produisant au moins 0,5 mM de H_2 après contact avec 50 ml d'eau dans un récipient à pression atmosphérique et température ambiante ;
la composition étant un comprimé et
où :

i) la composition se désintègre en moins de 5 minutes ;
ii) l'au moins un acide est hydrosoluble, et a une solubilité dans l'eau d'au moins 0,01 g/ml ; ou,
iii) la composition maintient un pH inférieur à 7 10 minutes après contact.

2. Composition selon la revendication 1, la composition passant un test pharmaceutique pour la friabilité ; ou, la composition se désintégrant en moins de 2 minutes.

3. Composition selon la revendication 1, dans laquelle le pH de l'eau reste inférieur à 7 pendant au moins 10 minutes après que la composition ait été mise en contact avec l'eau ; ou dans laquelle le récipient est fermé, et la composition maintient un pH inférieur à 7 pendant 7 jours après contact avec l'eau.

4. Composition selon la revendication 1, dans laquelle :

a) le magnésium métallique comprend des flocons ; de préférence des flocons de 44 μm ou moins (-325 mesh) ; et/ou
b) le magnésium métallique est broyé ; et/ou,
c) la quantité de magnésium métallique est de 5 à 500 mg.

5. Composition selon la revendication 1, dans laquelle :

a) l'au moins un acide comprend un acide comestible, de préférence dans laquelle l'au moins un acide comestible est choisi dans le groupe constitué de l'acide maléique, l'acide succinique, l'acide malique, l'acide fumarique, l'acide formique, l'acide citrique, l'acide ascorbique, l'acide oxalique et l'acide tartrique, ou un mélange de ceux-ci ; ou,
b) l'au moins un acide est un acide cosmétiquement ou pharmaceutiquement acceptable, de préférence l'acide acétique, l'acide adipique, l'acide alginique, l'acide aspartique, l'acide benzènesulfonique, l'acide benzoïque, l'acide borique, l'acide butyrique, l'acide camphorique, l'acide camphorsulfonique, l'acide cyclopentanepropionique, l'acide digluconique, l'acide dodécylsulfrique, l'acide éthanesulfonique, l'acide glucoheptonique, l'acide

glycérophosphorique, l'acide hémisulfurique, l'acide heptonique, l'acide hexanoïque, l'acide bromhydrique, l'acide chlorhydrique, l'acide iodhydrique, l'acide 2-hydroxy-éthanesulfonique, l'acide lactobionique, l'acide lactique, l'acide laurique, l'acide laurylsulfurique, l'acide malonique, l'acide méthanesulfonique, l'acide 2-naphtalènesulfonique, l'acide nicotinique, l'acide nitrique, l'acide oléique, l'acide palmitique, l'acide pamoïque, l'acide pectique, l'acide persulfurique, l'acide 3-phénylpropionique, l'acide phosphorique, l'acide picrique, l'acide pivalique, l'acide propionique, l'acide stéarique, l'acide sulfurique, l'acide tartrique, l'acide thiocyanique, l'acide toluènesulfonique, l'acide undécanoïque, l'acide valérique, leurs stéréoisomères, toutes les formes d'alpha-acides (par exemple, l'acide α-lupulique), des acides polycarboxyliques, un acide de Lewis, ou une combinaison de ceux-ci.

6. Composition selon la revendication 1, dans laquelle la quantité de l'au moins un acide est de 30 à 4000 mg.

7. Composition selon la revendication 1, dans laquelle le liant est le mannitol, le xylitol, le maltose, le dextrose ou le lactose.

8. Composition selon la revendication 1, comprenant en outre :

a) un lubrifiant hydrosoluble ; de préférence dans laquelle le lubrifiant hydrosoluble est choisi parmi le stéarylfumarate de sodium ou l'acide stéarique ; et/ou
b) un complément alimentaire, de préférence dans laquelle le complément alimentaire est un sel de magnésium ; et/ou,
c) un édulcorant ou un agent aromatisant ; et/ou,
d) un agent colorant ; et/ou,
e) un parfum ; et/ou,
f) une huile essentielle ; et/ou,
g) un polysaccharide ; de préférence dans laquelle le polysaccharide est choisi dans le groupe constitué de cellulose et ses dérivés, amidon, poudre à l'arôme de pomme, poudre à l'arôme de citron, poudre à l'arôme de citron vert, poudre à l'arôme de pamplemousse, cosse de psyllium et pectine.

9. Composition selon la revendication 1, dans laquelle le magnésium métallique et au moins un acide sont présents en quantités suffisantes pour produire au moins 2 mM H_2.

10. Composition selon la revendication 1, dans laquelle l'au moins un acide est hydrosoluble et a une solubilité d'au moins 0,05 g/ml dans l'eau ; ou, dans laquelle l'au moins un acide est hydrosoluble et a une solubilité d'au moins 0,1 g/ml dans l'eau ; ou, dans laquelle l'au moins un acide est hydrosoluble et a une solubilité d'au moins 1 g/ml dans l'eau.

11. Composition selon la revendication 1, dans laquelle le pH inférieur à 7 est maintenu 30 minutes après contact avec l'eau ; ou, dans laquelle le pH inférieur à 7 est maintenu 1 heure après contact avec l'eau.

12. Kit comprenant une composition selon l'une quelconque des revendications 1 à 11 et un récipient scellable pouvant contenir entre 100 ml et 2 l d'eau ; de préférence dans lequel le récipient est à double paroi ; facultativement dans lequel le récipient peut contenir entre 250 et 750 ml d'eau.

13. Procédé de production d'eau riche en hydrogène, le procédé comprenant les étapes de : mise en contact d'une composition selon l'une quelconque des revendications 1 à 11 avec de l'eau dans un récipient de sorte que la composition se désintègre et le magnésium métallique et au moins un acide réagissent pour produire H_2 ; facultativement dans lequel le pH inférieur à 7 est maintenu pendant au moins 1 heure ; et/ou dans lequel le pH est de 4 à 6 ; et/ou dans lequel l'eau comprend du jus de fruit ; et/ou dans lequel la concentration de H_2 est au moins 0,5 mM.

14. Procédé non thérapeutique d'administration d'hydrogène à un sujet, le procédé comprenant la fourniture à un sujet d'une composition contenant de l'hydrogène produit à partir d'une composition selon l'une quelconque des revendications 1 à 11 ; facultativement dans lequel la composition contenant de l'hydrogène est un nutraceutique ou une formulation topique, de préférence dans lequel le nutraceutique est une boisson.

15. Composition enrichie en hydrogène, comprenant de l'hydrogène dissous dans un véhicule à une concentration d'au moins 0,5 mM, dans laquelle le pH de la composition est inférieur à 7 et la composition est produite par mise en contact d'une composition selon l'une quelconque des revendications 1 à 11 avec un véhicule ; la composition étant facultativement de quantité comestible, cosmétique ou pharmaceutique ;

facultativement dans laquelle le véhicule est choisi parmi un liquide, une crème, une lotion, une mousse, une pâte ou un gel aqueux, ou est une boisson ;
facultativement dans laquelle la concentration maximale d'hydrogène est 20 mM ;
facultativement dans laquelle le pH est de 4 à 6 ou est 4,6 ou moins ;
la composition contenant facultativement un complément alimentaire ; ou
la composition contenant facultativement un édulcorant, un agent aromatisant, un agent colorant, un parfum, une huile essentielle, un polysaccharide, un liant ou un lubrifiant hydrosoluble.

REFERENCES CITED IN THE DESCRIPTION

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