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(54) Title: FUEL COMPOSITION FOR FUEL CELLS

(57) **Abrégé/Abstract:**

A hydride containing fuel composition for a liquid fuel cell. The composition comprises an alkaline liquid phase and at least two hydride compounds. The solubility of the first hydride compound in the liquid phase is higher than the solubility of a second hydride compound in the liquid phase and the opposite is true for the anodic oxidation products thereof.



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(54) Title: FUEL COMPOSITION FOR FUEL CELLS

(57) Abstract: A hydride containing fuel composition for a liquid fuel cell. The composition comprises an alkaline liquid phase and at least two hydride compounds. The solubility of the first hydride compound in the liquid phase is higher than the solubility of a second hydride compound in the liquid phase and the opposite is true for the anodic oxidation products thereof.



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FUEL COMPOSITION FOR FUEL CELLSCROSS-REFERENCE TO RELATED APPLICATIONS

[0001] The present application claims priority under 35 U.S.C. § 119(e) of U.S. provisional application No. 60/663,730, filed March 22, 2005, the entire disclosure whereof is expressly incorporated by reference herein.

BACKGROUND OF THE INVENTION1. Field of the Invention

[0002] The present invention relates to hydride-containing fuel compositions for direct and indirect liquid fuel cells.

2. Discussion of Background Information

[0003] Fuel cells are electrochemical power sources wherein electrocatalytic oxidation of a fuel (e.g., molecular hydrogen or methanol) at an anode and electrocatalytic reduction of an oxidant (often molecular oxygen) at a cathode take place simultaneously. Conventional fuels such as hydrogen and methanol pose several storage and transportation problems, in particular, for portable fuel cells (e.g., for use with portable electric and electronic devices such as laptops, cell phones, and the like).

[0004] Borohydride (and other hydride) based fuels are of particular interest for portable fuel cells, due to their very high specific energy capacity (see, e.g., J. of Electrochem. Soc., 150, (3), A398-402, 2003). This type of fuels may be used either directly as the fuel or indirectly as a generator of hydrogen (which is oxidized at the anode), e.g., as part of a portable proton exchange membrane (PEM) fuel cell (see, e.g., US 20010045364 A1, US 20030207160 A1, US 20030207157 A1, US 20030099876 A1, and U.S. Patent Nos. 6,554,877 B2 and 6,562,497 B2). The disclosures of all of the above documents are expressly incorporated by reference herein in their entireties.

[0005] There are several factors which have to be taken into account when assessing the performance of a hydride based fuel. One of these factors is the efficiency of the fuel. Fuel efficiency can be determined, for example, by comparing the actual energy density (Wh/volume unit fuel) provided in a given fuel cell to the theoretical energy density. The absolute value of the energy density is also one of the indicators of fuel performance. In this regard, it is to be taken into account that while a high (boro)hydride concentration in the fuel may afford a desired high energy density of the fuel, in some

situations a high (boro)hydride concentration in the liquid phase of the fuel may also be disadvantageous. For example, the corresponding fuel may be chemically too aggressive and as a result thereof, may damage one or more of the structural components of the fuel cell, in particular, the anode. Accordingly, one has to find a compromise between energy density of the fuel and compatibility of the fuel with the components of the fuel cell and/or find ways to avoid significant damage to the fuel cell components despite a relatively high (boro)hydride concentration in the liquid phase of the fuel.

[0006] Another factor which may affect the performance of a hydride based fuel is solubility. For example, the main oxidation reaction of a borohydride compound at the anode of a fuel cell can be represented as follows:



[0007] Accordingly, during use of a liquid fuel cell, a starting material such as, e.g., a borohydride compound is converted to an anodic oxidation product such as, e.g., a metaborate. The solubility of the starting material and that of the oxidation product in the liquid phase of the fuel may differ substantially. This difference in solubility may affect the fuel efficiency and thus, the performance of the fuel cell. By way of non-limiting example, NaBH_4 has a relatively high solubility in an alkaline solution whereas its oxidation product, NaBO_2 , is less soluble in this solution. If a relatively high concentration of NaBH_4 is present in the initial fuel, the fuel cell will have a high level of activity at the beginning of the discharging process, and the activity will gradually decrease as more and more NaBH_4 is present in oxidized form. In addition, the concentration of oxidation product increases at the same rate as the BH_4^- concentration decreases, and due to the high initial concentration of sodium borohydride, the less soluble sodium metaborate starts precipitating at a relatively early stage of the discharge process. The metaborate precipitate may block the anode, membranes and other components of the fuel cell and may thereby aggravate the decrease in the activity of the fuel caused by the decrease in the BH_4^- concentration in the liquid phase of the fuel.

[0008] The situation with potassium borohydride is opposite to that with sodium borohydride. Potassium borohydride has a relatively low solubility in caustic solution (in particular, at room temperature) whereas its oxidation product, potassium metaborate is significantly more soluble in caustic solution than potassium borohydride. Due to the relatively low solubility of the potassium borohydride in caustic solution, the initial BH_4^- concentration in the liquid phase of the fuel cannot be made as high as in the case of

sodium borohydride, wherefore it is more difficult to obtain a high current. On the other hand, the spent fuel (containing potassium metaborate) does not exhibit any significant problems due to precipitation of oxidation product.

[0009] It would be advantageous to have available a fuel for a liquid fuel cell which allows a desirable high hydride concentration in the liquid phase of the fuel and provides a high energy density and/or other advantages without causing problems such as, e.g., premature precipitation of the anodic oxidation product of the employed hydride from the liquid phase of the fuel and/or damage to the components of the fuel cell.

SUMMARY OF THE INVENTION

[0010] The present invention provides a hydride containing fuel composition for a liquid fuel cell. The composition comprises an alkaline liquid phase and at least a first hydride compound and a second hydride compound. The solubility of the first hydride compound in the liquid phase is higher than the solubility of the second hydride compound in the liquid phase and the solubility of the anodic oxidation product of the first hydride compound in the liquid phase is lower than the solubility of the anodic oxidation product of the second hydride compound in the liquid phase.

[0011] In one aspect of this composition, the first hydride compound and the second hydride compound may independently be selected from hydrides, borohydrides (including polyborohydrides and cyanoborohydrides) and aluminum hydrides of metals, preferably of alkali and alkaline earth metals, Zn and Al, ammonium, and alkylamine-BH₃ complexes, for example, from NaBH₄, KBH₄, LiBH₄, NH₄BH₄, Be(BH₄)₂, Ca(BH₄)₂, Mg(BH₄)₂, Zn(BH₄)₂, Al(BH₄)₃, a polyborohydride such as, e.g., a compound of formula MB₃H₈, M₂B₁₀H₁₀, MB₁₀H₁₃, M₂B₁₂H₁₂ or M₂B₂₀H₁₈ wherein M = Li, Na, K, NH₄, Be_{1/2}, Ca_{1/2}, Mg_{1/2}, Zn_{1/2} or Al_{1/3}, (CH₃)₂NHBH₃, NaCNBH₃, LiH, NaH, KH, CaH₂, BeH₂, MgH₂, NaAlH₄, LiAlH₄ and KAlH₄.

[0012] In another aspect, the first hydride compound and/or the second hydride compound may be selected from borohydrides, e.g., from borohydrides of alkali and alkaline earth metals. For example, the first hydride compound and/or the second hydride compound may be selected from NaBH₄ and KBH₄.

[0013] In another aspect of the composition, the molar ratio of the first hydride compound and the second hydride compound may be from about 95 : 5 to about 5 : 95, e.g., from about 90 : 10 to about 10 : 90, from about 80 : 20 to about 20 : 80, from about 75 : 25 to about 25 : 75, or from about 60 : 40 to about 40 : 60.

[0014] In yet another aspect, the composition may comprise hydride compounds in a total concentration of at least about 0.5 mole per liter of composition, e.g., in a total concentration of at least about 1 mole per liter, at least about 2 moles per liter, at least about 3 moles per liter, at least about 4 moles per liter, or at least about 5 moles per liter of composition. Of course, if a concentrate is to be provided, the concentration of the hydride compounds may as high as, e.g., at least about 6 moles per liter, at least about 8 moles per liter, or at least about 10 moles per liter of composition.

[0015] In a still further aspect, the liquid phase of the composition may comprise hydroxide ions. By way of non-limiting example, the hydroxide ions may be present in a concentration of at least about 0.01 mole per liter, e.g., at least about 0.05 mole per liter, at least about 0.1 mole per liter, at least about 0.5 mole per liter, at least about 1 mole per liter, at least about 1.5 moles per liter, at least about 2 moles per liter, at least about 3 moles per liter, at least about 4 moles per liter, or at least about 5 moles per liter and/or in a concentration that is not higher than about 7 moles per liter of liquid phase. Especially if a concentrate is to be provided, the hydroxide ion concentration may even be higher, e.g., up to about 14 moles per liter, e.g., up to about 12 moles per liter.

[0016] In another aspect, the liquid phase may comprise at least one hydroxide ion providing compound dissolved therein, said hydroxide ion providing compound being selected from hydroxides of alkali metals, alkaline earth metals, Zn and Al, and from ammonium hydroxide, e.g., from one or more of LiOH, NaOH, KOH, RbOH, CsOH, Ca(OH)₂, Mg(OH)₂, Ba(OH)₂, Zn(OH)₂, Al(OH)₃ and NH₄OH. For example, the liquid phase may comprise dissolved therein NaOH and/or KOH.

[0017] In another aspect of the composition of the present invention, the liquid phase may comprise at least one solvent, e.g., at least two solvents, which are selected from water and (preferably water-miscible and/or water-soluble) substances, for example, from (cyclo)aliphatic alcohols having up to about 6 carbon atoms and up to about 6 hydroxy groups, C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols), poly(C₂₋₄ alkylene glycols), mono-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), di-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), ethylene oxide/propylene oxide block copolymers, ethoxylated aliphatic polyols, propoxylated aliphatic polyols, ethoxylated and propoxylated aliphatic polyols, aliphatic ethers having up to about 6 carbon atoms, aliphatic ketones having up to about 6 carbon atoms, aliphatic aldehydes having up to about 6 carbon atoms, C₁₋₄-alkyl esters of C₁₋₄ alkanolic (aliphatic) acids and primary, secondary and tertiary aliphatic amines having a total of up to about 10 carbon atoms.

By way of non-limiting example, the liquid phase may comprise one or more of water, methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol (or any other sugar alcohol), glycerol, acetone, methyl ethyl ketone, diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran, diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine.

[0018] In another aspect, the liquid phase may comprise water, either alone or in combination with one or more substances selected from (cyclo)aliphatic alcohols having up to about 6 carbon atoms and up to about 6 hydroxy groups, C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols), poly(C₂₋₄ alkylene glycols), mono-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), di-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), ethylene oxide/propylene oxide block copolymers, ethoxylated aliphatic polyols, propoxylated aliphatic polyols, ethoxylated and propoxylated aliphatic polyols, aliphatic ethers having up to about 6 carbon atoms, aliphatic ketones having up to about 6 carbon atoms, aliphatic aldehydes having up to about 6 carbon atoms, C₁₋₄-alkyl esters of C₁₋₄ alkanic (aliphatic) acids and primary, secondary and tertiary aliphatic amines having a total of up to about 10 carbon atoms.

[0019] In another aspect, the composition of the present invention may comprise two hydride compounds. In a still further aspect, it may comprise at least three hydride compounds.

[0020] The present invention also provides a hydride containing fuel composition for a liquid fuel cell, which composition comprises an alkaline liquid phase and at least a first hydride compound and a second hydride compound. The solubility of the first hydride compound in the liquid phase is higher than the solubility of the second hydride compound in the liquid phase and the solubility of the anodic oxidation product of the first hydride compound in the liquid phase is lower than the solubility of the anodic oxidation product of the second hydride compound in the liquid phase. The alkaline liquid phase further has a hydroxide ion concentration of at least about 0.5 mole per liter and comprises dissolved therein one or more of LiOH, NaOH, KOH, RbOH, CsOH, Ca(OH)₂, Mg(OH)₂, Ba(OH)₂, Zn(OH)₂, Al(OH)₃ and NH₄OH. The first hydride compound and the second hydride compound are independently selected from NaBH₄, KBH₄, LiBH₄, NH₄BH₄, Be(BH₄)₂, Ca(BH₄)₂, Mg(BH₄)₂, Zn(BH₄)₂, Al(BH₄)₃, polyborohydrides, (CH₃)₂NHBH₃, NaCNBH₃, LiH, NaH, KH, CaH₂, BeH₂, MgH₂, NaAlH₄, LiAlH₄ and KAlH₄.

[0021] In one aspect of this composition, the first hydride compound and the second hydride compound may independently be selected from NaBH_4 , KBH_4 , LiBH_4 , $\text{Be}(\text{BH}_4)_2$, NH_4BH_4 , a polyborohydride, $\text{Ca}(\text{BH}_4)_2$, $\text{Mg}(\text{BH}_4)_2$, $\text{Zn}(\text{BH}_4)_2$, $\text{Al}(\text{BH}_4)_3$, $(\text{CH}_3)_2\text{NHBH}_3$ and NaCNBH_3 . For example, the first hydride compound and the second hydride compound may be NaBH_4 and KBH_4 .

[0022] In another aspect, the molar ratio of the first hydride compound and the second hydride compound may be from about 95 : 5 to about 5 : 95, e.g., from about 75 : 25 to about 25 : 75.

[0023] In yet another aspect, the composition may comprise the hydride compounds in a total concentration of at least about 0.5 mole per liter of composition, e.g., in a total concentration of at least about 1 mole per liter, at least about 2 moles per liter, or at least about 3 moles per liter of composition.

[0024] In a still further aspect of the composition, the hydroxide ion concentration in the liquid phase may be at least about 1 mole per liter, e.g., at least about 1.5 moles per liter, at least about 2 moles per liter, at least about 3 moles per liter, at least about 4 moles per liter, or at least about 5 moles per liter and/or may be not higher than about 7 moles per liter.

[0025] In another aspect of the composition, the liquid phase may comprise dissolved therein NaOH and/or KOH .

[0026] In another aspect, the liquid phase may comprise at least one, e.g., at least two, solvents selected from water, (cyclo)aliphatic alcohols having up to about 6 carbon atoms and up to about 6 hydroxy groups, C_{2-4} alkylene glycols, di(C_{2-4} alkylene glycols), poly(C_{2-4} alkylene glycols), mono- C_{1-4} -alkyl ethers of C_{2-4} alkylene glycols, di(C_{2-4} alkylene glycols) and poly(C_{2-4} alkylene glycols), di- C_{1-4} -alkyl ethers of C_{2-4} alkylene glycols, di(C_{2-4} alkylene glycols) and poly(C_{2-4} alkylene glycols), ethylene oxide/propylene oxide block copolymers, ethoxylated aliphatic polyols, propoxylated aliphatic polyols, ethoxylated and propoxylated aliphatic polyols, aliphatic ethers having up to about 6 carbon atoms, aliphatic ketones having up to about 6 carbon atoms, aliphatic aldehydes having up to about 6 carbon atoms, C_{1-4} -alkyl esters of C_{1-4} alkanolic (aliphatic) acids and primary, secondary and tertiary aliphatic amines having a total of up to about 10 carbon atoms. For example, the liquid phase may comprise one or more of water, methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol, glycerol, acetone, methyl ethyl ketone, diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran,

diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine.

[0027] In another aspect, the liquid phase may comprise water. For example, the liquid phase may comprise water and one or more of methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol, glycerol, acetone, methyl ethyl ketone, diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran, diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine.

[0028] The present invention also provides a hydride containing mixture for use in a fuel for a liquid fuel cell, which mixture comprises at least a first hydride compound and a second hydride compound, which compounds are independently selected from hydrides, borohydrides (including polyborohydrides and cyanoborohydrides) and aluminum hydrides of alkali and alkaline earth metals, Zn, Al, ammonium and dialkylamines and wherein the molar ratio of the first hydride compound and the second hydride compound is from about 95 : 5 to about 5 : 95.

[0029] In one aspect of the mixture, the molar ratio of the hydride compounds may be from about 80 : 20 to about 20 : 80.

[0030] In another aspect of the mixture, the first hydride compound and the second hydride compound may independently be selected from NaBH_4 , KBH_4 , LiBH_4 , $\text{Be}(\text{BH}_4)_2$, NH_4BH_4 , a polyborohydride, $\text{Ca}(\text{BH}_4)_2$, $\text{Mg}(\text{BH}_4)_2$, $\text{Zn}(\text{BH}_4)_2$, $\text{Al}(\text{BH}_4)_3$, $(\text{CH}_3)_2\text{NHBH}_3$ and NaCNBH_3 . For example, the first hydride compound and/or the second hydride compound may be selected from borohydrides, e.g., borohydrides of alkali and alkaline earth metals. In particular, the first hydride compound and/or the second hydride compound may be NaBH_4 or KBH_4 .

[0031] The present invention also provides a hydride containing fuel composition for a direct liquid fuel cell, which composition comprises at least two different hydride compounds. The at least two different hydride compounds are selected such that the composition provides, under otherwise identical circumstances, a higher fuel efficiency than an otherwise identical fuel composition that comprises only one of these different hydride compounds in a molar amount which is identical with a total molar amount of the at least two different hydride compounds. In other words, there is a synergism.

[0032] In one aspect of the composition, a first hydride compound of the at least two different hydride compounds may have a higher solubility in the liquid phase of a liquid fuel than a second hydride compound of the at least two different hydride compounds,

and the anodic oxidation product of the first hydride compound may have a lower solubility in the liquid phase of the liquid fuel than the anodic oxidation product of the second hydride compound.

[0033] In another aspect, the first hydride compound and the second hydride compound may independently be selected from hydrides, borohydrides (including polyborohydrides) and aluminum hydrides of alkali and alkaline earth metals, Zn, Al, ammonium and dialkylamines, e.g., from NaBH_4 , KBH_4 , LiBH_4 , $\text{Be}(\text{BH}_4)_2$, NH_4BH_4 , polyborohydrides, $\text{Ca}(\text{BH}_4)_2$, $\text{Mg}(\text{BH}_4)_2$, $\text{Zn}(\text{BH}_4)_2$, $\text{Al}(\text{BH}_4)_3$, $(\text{CH}_3)_2\text{NHBH}_3$ and NaCNBH_3 . For example, the first hydride compound and/or the second hydride compound may be selected from borohydrides, e.g., from borohydrides of alkali and alkaline earth metals. In particular, at least one of the first hydride compound and the second hydride compound may be NaBH_4 or KBH_4 .

[0034] In yet another aspect, the molar ratio of the first hydride compound and the second hydride compound may be from about 95 : 5 to about 5 : 95, e.g., from about 90 : 10 to about 10 : 90, from about 80 : 20 to about 20 : 80, from about 75 : 25 to about 25 : 75, or from about 60 : 40 to about 40 : 60.

[0035] The present invention also provides a method of increasing the fuel efficiency of a hydride compound containing liquid fuel composition for a direct liquid fuel cell, wherein the method comprises employing at least two different hydride compounds in the liquid fuel, the at least two different hydride compounds being selected such that a fuel comprising these different hydride compounds has a higher efficiency than is obtainable with a fuel that comprises only one of these hydride compounds in a molar amount which is identical with a total molar amount of the at least two different hydride compounds.

[0036] In one aspect of the method, a first hydride compound of the at least two different hydride compounds may have a higher solubility in the liquid phase of the fuel composition than a second hydride compound of the at least two different hydride compounds, and an anodic oxidation product of the first hydride compound may have a lower solubility in the liquid phase of the fuel composition than an anodic oxidation product of the second hydride compound.

[0037] In another aspect of the method, the first hydride compound and the second hydride compound may independently be selected from hydrides, borohydrides and aluminum hydrides of alkali and alkaline earth metals, Zn, Al, ammonium and dialkylamines, e.g., from NaBH_4 , KBH_4 , LiBH_4 , $\text{Be}(\text{BH}_4)_2$, NH_4BH_4 , polyborohydrides, $\text{Ca}(\text{BH}_4)_2$, $\text{Mg}(\text{BH}_4)_2$, $\text{Zn}(\text{BH}_4)_2$, $\text{Al}(\text{BH}_4)_3$, $(\text{CH}_3)_2\text{NHBH}_3$ and NaCNBH_3 . For example, the

first "hydride compound" and/or the second hydride compound may be selected from borohydrides, e.g., from borohydrides of alkali and alkaline earth metals. In particular, the first hydride compound and/or the second hydride compound may be NaBH_4 or KBH_4 .

[0038] In yet another aspect of the method of the invention, the molar ratio of the first hydride compound and the second hydride compound may be from about 95 : 5 to about 5 : 95, e.g., from about 90 : 10 to about 10 : 90, from about 80 : 20 to about 20 : 80, from about 75 : 25 to about 25 : 75, or from about 60 : 40 to about 40 : 60.

[0039] The present invention also provides a hydride containing fuel composition for a liquid fuel cell, which composition comprises an alkaline liquid phase and one or more hydride compounds. The one or more hydride compounds are dissolved in the liquid phase (e.g., at about room temperature) in a total concentration of at least about 0.5 mole per liter of liquid phase and the solubility of the anodic oxidation products of the one or more hydride compounds in the liquid phase at about room temperature (e.g., at about 25°C) is such that after an anodic oxidation of about 80 mole-% of the one or more hydride compounds substantially no oxidation products precipitate (at about room temperature). "Substantially no precipitate" as used herein and in the appended claims is intended to mean less than about 10 %, preferably less than about 5 %, e.g., less than about 1 % of the formed anodic oxidation product is present in undissolved form.

[0040] In one aspect of the composition, substantially no oxidation products precipitate after an anodic oxidation of about 90 mole-%, or even after an anodic oxidation of about 95 mole-% of the one or more hydride compounds and/or the one or more hydride compounds may be dissolved in the liquid phase in a total concentration of at least about 1 mole per liter, e.g., in a total concentration of at least about 2 moles per liter, or at least about 3 moles per liter.

[0041] In another aspect of the composition of the present invention, the one or more hydride compounds may independently be selected from hydrides, borohydrides and aluminum hydrides of alkali and alkaline earth metals, Zn, Al, ammonium and dialkylamines, for example, from NaBH_4 , KBH_4 , LiBH_4 , $\text{Be}(\text{BH}_4)_2$, NH_4BH_4 , a polyborohydride, $\text{Ca}(\text{BH}_4)_2$, $\text{Mg}(\text{BH}_4)_2$, $\text{Zn}(\text{BH}_4)_2$, $\text{Al}(\text{BH}_4)_3$, $(\text{CH}_3)_2\text{NHBH}_3$ and NaCNBH_3 . In particular, the one or more hydride compounds may selected from (poly)borohydrides, e.g., from borohydrides of alkali and alkaline earth metals. For example, at least one of the one or more hydride compounds may be selected from NaBH_4 and KBH_4 .

[0042] In yet another aspect of the composition of the present invention, the one or more hydride compounds may comprise at least a first hydride compound and a second hydride compound and the solubility of the first hydride compound in the liquid phase may be higher than the solubility of the second hydride compound in the liquid phase and the solubility of the anodic oxidation product of the first hydride compound in the liquid phase may be lower than the solubility of the anodic oxidation product of the second hydride compound in the liquid phase.

[0043] In one aspect, the molar ratio of the first hydride compound and the second hydride compound may be from about 95 : 5 to about 5 : 95, e.g., from about 90 : 10 to about 10 : 90, from about 80 : 20 to about 20 : 80, from about 75 : 25 to about 25 : 75, or from about 60 : 40 to about 40 : 60.

[0044] In a still further aspect of the composition, the liquid phase may comprise hydroxide ions in a concentration of at least about 1 mole per liter, e.g., in a concentration of at least about 1.5 moles per liter, at least about 2 moles per liter and/or in a concentration of not higher than about 7 moles per liter.

[0045] In yet another aspect, the liquid phase may comprise at least one hydroxide ion providing compound dissolved therein, said hydroxide ion providing compound being selected from hydroxides of alkali metals, alkaline earth metals, Al and Zn and from ammonium hydroxide, such as, e.g., LiOH, NaOH, KOH, RbOH, CsOH, Ca(OH)₂, Mg(OH)₂, Ba(OH)₂, Zn(OH)₂, Al(OH)₃, and NH₄OH. In particular, the liquid phase may comprise dissolved therein NaOH and/or KOH.

[0046] In a still further aspect of the composition, the liquid phase may comprise at least one (e.g., at least two) of water, (cyclo)aliphatic alcohols having up to about 6 carbon atoms and up to about 6 hydroxy groups, C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols), poly(C₂₋₄ alkylene glycols), mono-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), di-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), ethylene oxide/propylene oxide block copolymers, ethoxylated aliphatic polyols, propoxylated aliphatic polyols, ethoxylated and propoxylated aliphatic polyols, aliphatic ethers having up to about 6 carbon atoms, aliphatic ketones having up to about 6 carbon atoms, aliphatic aldehydes having up to about 6 carbon atoms, C₁₋₄-alkyl esters of C₁₋₄ alkanolic (aliphatic) acids and primary, secondary and tertiary aliphatic amines having a total of up to about 10 carbon atoms, for example, at least one of water, methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol, glycerol, acetone, methyl ethyl ketone,

diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran, diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine.

[0047] In another aspect, the liquid phase may comprise water, alone or in combination with one or more solvents selected from (cyclo)aliphatic alcohols having up to about 6 carbon atoms and up to about 6 hydroxy groups, C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols), poly(C₂₋₄ alkylene glycols), mono-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), di-C₁₋₄-alkyl ethers of C₂₋₄ alkylene glycols, di(C₂₋₄ alkylene glycols) and poly(C₂₋₄ alkylene glycols), ethylene oxide/propylene oxide block copolymers, ethoxylated aliphatic polyols, propoxylated aliphatic polyols, ethoxylated and propoxylated aliphatic polyols, aliphatic ethers having up to about 6 carbon atoms, aliphatic ketones having up to about 6 carbon atoms, aliphatic aldehydes having up to about 6 carbon atoms, C₁₋₄-alkyl esters of C₁₋₄ alkanic (aliphatic) acids and primary, secondary and tertiary aliphatic amines having a total of up to about 10 carbon atoms. By way of non-limiting example, the liquid phase may comprise water and one or more (e.g., two) of methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol, glycerol, acetone, methyl ethyl ketone, diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran, diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine, e.g., water and at least one of methanol, ethanol, propanol, isopropanol and ethylene glycol.

[0048] The present invention also provides a liquid fuel cell which comprises a fuel composition of the present invention, including the various aspects thereof.

[0049] The present invention also provides a fuel cartridge for (re)filling a liquid fuel cell, which cartridge comprises a fuel composition of the present invention, including the various aspects thereof.

[0050] The present invention also provides a method of increasing the fuel efficiency of a hydride compound containing liquid fuel composition for a direct liquid fuel cell. This method comprises selecting one or more hydride compounds and one or more solvents such that the one or more hydride compounds are soluble in the one or more solvents in a concentration of at least about 0.5 mole per liter (at about room temperature) and that after an anodic oxidation of about 80 mole-% of the one or more hydride compounds substantially all formed anodic oxidation products are soluble in the one or more solvents (at about room temperature).

[0051] In one aspect of the method, the one or more hydride compounds may comprise at least a first hydride compound and a second hydride compound and the solubility of the first hydride compound in the one or more solvents may be higher than the solubility of the second hydride compound in the one or more solvents and the solubility of the anodic oxidation product of the first hydride compound in the one or more solvents may be lower than the solubility of the anodic oxidation product of the second hydride compound in the one or more solvents.

[0052] The present invention also provides a method for increasing the fuel efficiency of a hydride compound containing liquid fuel composition for a direct liquid fuel cell, which method comprises the employment of at least two different hydride compounds (e.g., at least two different borohydride compounds) in the fuel composition. The at least two (e.g., two, three or four) different hydride compounds are selected such that a fuel composition comprising these different hydride compounds, under otherwise identical conditions, has a higher efficiency than an otherwise identical fuel composition which comprises only one of the at least two different hydride compounds in a molar amount which is identical with a total molar amount of the at least two different hydride compounds.

[0053] In one aspect of the method, a first hydride compound of the at least two different hydride compounds may have a higher solubility in the liquid phase of the liquid fuel composition than a second hydride compound of the at least two different hydride compounds and an anodic oxidation product of the first hydride compound may have a lower solubility in the liquid phase of the liquid fuel composition than an anodic oxidation product of the second hydride compound.

[0054] In another aspect of the method, the at least two hydride compounds may be present in the liquid phase of the fuel composition in a total concentration of at least about 1 mole per liter, e.g., at least about 2 moles per liter.

[0055] The present invention also provides a method for improving the performance of a liquid fuel cell which comprises a hydride compound containing liquid fuel composition. The method comprises the employment of at least two different hydride compounds (e.g., two or more different borohydride compounds) in the liquid fuel composition, the at least two different hydride compounds being selected such that under otherwise identical conditions the fuel composition comprising the at least two different hydride compounds is chemically less aggressive toward at least one structural component of the fuel cell (e.g., the anode) than, and shows at least the same efficiency as a liquid fuel composition which comprises only one of these hydride compounds in a

molar amount which is identical with a total molar amount of the at least two different hydride compounds.

[0056] According to one aspect of the present invention, the performance of a fuel for a liquid fuel cell may be optimized by providing a relatively high initial concentration of dissolved (boro)hydride compound(s) while at the same time preventing the precipitation of substantial amounts of anodic oxidation products and/or while preventing any significant damage to the structural components of the fuel cell, in particular the anode, caused by a chemically too aggressive fuel. To this end, the present invention employs several basic concepts which can be used individually or in combination. A first concept comprises the use of a mixture of two or more hydride compounds which differ in their solubilities in the liquid component of the fuel and also differ in the solubilities of their anodic oxidation products, with the more soluble hydride compound affording the less soluble oxidation product. A second concept comprises the use of a mixture of at least two different solvents which are selected so as to afford a desirably high solubility of one or more hydride compounds and also a desirably high solubility of the anodic oxidation product(s) of these one or more hydride compounds. In this case, if more than one hydride compound is employed, the solubilities of the hydride compounds and the anodic reaction products may be similar or different, as long as the solvent mixture can dissolve both the hydride compounds and the anodic oxidation products thereof to a desirable high extent. A third concept balances the concentrations of the various (metal) cations in the liquid phase of the fuel (which may be a suspension) in order to achieve a desirable high (boro)hydride concentration in the liquid phase of the fuel while at the same time keeping the chemical aggressiveness of the fuel at acceptable levels.

[0057] The use of the above concepts also facilitates the preparation of fuel pastes and/or concentrates. Regarding corresponding products, reference is made to co-pending U.S. patent application No. 10/757,849, filed January 16, 2004 (U.S. Patent Application Publication No. 2005/0155279 A1), the entire disclosure whereof is expressly incorporated by reference herein. Regarding a method of producing corresponding products, reference is made to the U.S. provisional application having the title "Method of Producing Fuel Dispersion for a Fuel Cell" (Attorney Docket No. P28866), filed concurrently herewith, the entire disclosure whereof is expressly incorporated by reference herein.

[0058] As set forth above, the "efficiency" of a fuel may be assessed by using various parameters, e.g., the initial current that is obtainable or the electric output per volume unit, each determined in a fuel cell.

[0059] The hydride compounds for use in the present invention preferably are compounds which can be oxidized as such at the anode of a fuel cell to provide electrons. It is to be understood that the term "hydride compound" as used in the present specification and the appended claims is used in a broad sense and encompasses, in particular, compounds which are "simple" hydrides, such as, e.g., NaH, KH, etc. as well as compounds which comprise a hydride complex ion such as, e.g., borohydride, aluminum hydride and the like. Non-limiting examples of metal hydride compounds for use in the present invention include hydrides, borohydrides, including cyanoborohydrides and polyborohydrides, and aluminum hydrides of alkali metals such as, e.g., Li, Na, K, Rb and Cs, and alkaline earth metals such as, e.g., Be, Mg, Ca, Sr and Ba, but also of other metals such as Al and Zn, ammonium and complexes of BH_3 and mono-, di-, trialkylamines. Corresponding specific compounds include, but are not limited to, $LiBH_4$, $NaBH_4$, KBH_4 , NH_4BH_4 , $Be(BH_4)_2$, $Ca(BH_4)_2$, $Mg(BH_4)_2$, $(CH_3)_3NHBH_3$, $NaCNBH_3$, LiH, NaH, KH, CaH_2 , BeH_2 , MgH_2 , $NaAlH_4$, $LiAlH_4$ and $KAlH_4$. Polyborohydrides may be used as well. Non-limiting examples of polyborohydrides are those of formulae MB_3H_8 , $M_2B_{10}H_{10}$, $MB_{10}H_{13}$, $M_2B_{12}H_{12}$ and $M_2B_{20}H_{18}$ wherein M may be Li, Na, K, NH_4 , $Be_{1/2}$, $Ca_{1/2}$, $Mg_{1/2}$, $Zn_{1/2}$ or $Al_{1/3}$ (the fractions associated with Ca, Mg, Zn and Al take into account that these metals are bi- or trivalent). Further examples of polyborohydride compounds which are suitable for use in the present invention are disclosed in, e.g., U.S. Patent Application Publication 2005/0132640 A1, the entire disclosure whereof is incorporated by reference herein. Borohydrides and, in particular, $NaBH_4$ and KBH_4 are examples of preferred hydrides for the purposes of the present invention.

[0060] The liquid phase of the compositions of the present invention preferably comprises one or more polar (protic and/or aprotic) solvent components. If the solvent is a pure solvent, i.e., there is only one solvent, the solvent is preferably polar. If the solvent is a solvent mixture, i.e., comprises one or more (e.g., two, three, four, or even more) individual solvents, at least one of the components of the mixture is preferably polar. For example, all or at least substantially all of the solvent components may be polar. Solvents and solvent mixtures for use in the present invention preferably are liquid at room temperature and are preferably present in an amount which is sufficient to dissolve the hydride compound(s) and hydroxide ion providing compound(s). Non-

limiting examples of suitable solvent components include water, mono- and polyhydric alcohols (e.g., methanol, ethanol, propanol, isopropanol, butanol, glycerol, 1,2,4-butanetriol, trimethylolpropane and pentaerythritol) and mono- and polyalkylene glycols (such as, e.g., ethylene glycol, diethylene glycol, propylene glycol, and dipropylene glycol), aliphatic esters of mono- and polycarboxylic acids (e.g., ethyl acetate, methyl acetate, ethyl formiate, and diethyloxalate), aliphatic ketones (such as, e.g., acetone, methyl ethyl ketone, and diethylketone), aliphatic aldehydes (such as, acetaldehyde and propionaldehyde) and (cyclo)aliphatic ethers (such as tetrahydrofuran, dioxane and partial or complete alkyl esters of mono- and polyhydric alcohols and mono- and polyalkylene glycols). A preferred solvent component is water. Examples of other preferred solvent components include monohydric and polyhydric aliphatic and cycloaliphatic alcohols such as methanol and ethanol. If water is present, the concentration thereof will often be at least about 10 % by volume, e.g., at least about 30 % by volume, at least about 50 % by volume, or at least about 70 % by volume, based on the total volume of the combined solvents.

[0061] The hydroxide ion providing compounds for use in the compositions of the present invention may be any compounds which are capable of providing hydroxide ions in the composition, e.g., by dissociation, decomposition, or by (*in situ*) reaction or interaction with any other compound that may be present in the composition. Of course, these compounds must not interfere to any significant extent with the operation of the fuel cell and, in particular, the electrochemical reactions that take place therein. Usually, the hydroxide ion providing compound will include at least one alkali or alkaline earth metal hydroxide and/or ammonium hydroxide. Non-limiting specific examples of suitable compounds are LiOH, NaOH, KOH, RbOH, CsOH, Ca(OH)₂, Mg(OH)₂, Ba(OH)₂, Zn(OH)₂, Al(OH)₃ and NH₄OH. The corresponding oxides, carbonates and bicarbonates are non-limiting examples of further compounds which may serve as hydroxide ion providing compounds. Often, NaOH and/or KOH will be employed. The amount of the hydroxide ion providing compound(s) is apparently dependent on the desired hydroxide ion concentration in the concentrate.

[0062] If two different hydride compounds are used in the present invention, the molar ratio of these hydride compounds will usually be in the range of from about 99 : 1 to about 1 : 99, particularly in the range of from about 95 : 5 to about 5 : 95. Often the molar ratio will be not higher than about 90 : 10, e.g. not higher than about 80 : 20, not higher than about 75 : 25, or not higher than about 60 : 40, and not lower than about 10 : 90, e.g., not lower than about 20 : 80, not lower than about 25 : 75, or not lower than

about 40 : 60. By way of non-limiting example, in the case of, e.g., a caustic aqueous solution, a molar ratio of NaBH_4 and KBH_4 of about 25 : 75 may afford particularly advantageous results.

[0063] One of skill in the art will appreciate that when two different hydride compounds such as, e.g., NaBH_4 and KBH_4 , are employed in a certain molar ratio, this molar ratio is not necessarily the molar ratio in which these two compounds are present in the liquid phase of the fuel, even if both compounds are present in completely dissolved form. This is due to the fact that in the liquid phase these compounds will usually be present in dissociated form. For example, if at the same time a hydroxide ion providing compound such as NaOH is employed, this compound will also be present in dissociated form in the liquid phase, thereby increasing the concentration of sodium cations relative to potassium cations in the liquid phase and, consequently, increasing the concentration of NaBH_4 relative to that of KBH_4 . By the same token, if NaBH_4 is employed as the only borohydride compound and KOH is employed as the only hydroxide ion providing compound, a corresponding solution would contain not only NaBH_4 , but also KBH_4 . In other words, if only one type of hydride compounds is employed (e.g., only two or more borohydrides), the properties of a corresponding solution (fuel) are determined, *inter alia*, by the relative proportions of all (metal) cations which are present in the solution, i.e., not only the relative proportions in which these cations are present in the employed hydride compounds.

[0064] Preferably, a fuel composition of the present invention comprises one or more hydride compounds in a total concentration of at least about 0.5 mole per liter of composition (the composition including a liquid phase and an optionally present solid phase of undissolved material), e.g., in a total concentration of at least about 1 mole per liter, at least about 2 moles per liter, at least about 3 moles per liter, at least about 4 moles per liter, or at least about 5 moles per liter of liquid phase. The concentration may even be higher, especially in the case of fuel concentrates.

[0065] Also, the liquid phase of a fuel composition of the present invention preferably comprises hydroxide ions in a concentration of at least about 0.01 mole per liter, e.g., about 0.05 mole per liter, about 0.1 mole per liter, about 0.5 mole per liter, at least about 1 mole per liter, at least about 1.5 moles per liter, at least about 2 moles per liter, at least about 3 moles per liter, or even at least about 6 moles per liter. On the other hand, the hydroxide ion concentration is preferably not higher than about 8 moles per liter, e.g., not higher than about 7 moles per liter. Especially in the case of fuel concentrates

the hydroxide ion concentration will often be higher than about 7 moles per liter, e.g., up to about 14 moles per liter, or up to about 12 moles per liter.

[0066] One of skill in the art will recognize that the compositions of the present invention may optionally comprise various other components whose presence in the fuel may be desirable, at least as long as these other components do not significantly interfere with the intended use of the fuel composition. By way of non-limiting example, the compositions may comprise additives such as, e.g., stabilizers. Preferred stabilizers include aliphatic and aromatic amines.

BRIEF DESCRIPTION OF THE DRAWINGS

[0067] The present invention is further described in the detailed description which follows, in reference to the noted plurality of drawings by way of non-limiting examples of exemplary embodiments of the present invention, wherein:

Fig. 1 shows a discharge curve in a direct borohydride – air fuel cell of the fuel prepared according to Example 1;

Fig. 2 shows a discharge curve in a direct borohydride – air fuel cell of the fuel prepared according to Example 2;

Fig. 3 shows a discharge curve in a direct borohydride – air fuel cell of the fuel prepared according to Example 3; and

Fig. 4 shows a discharge curve in a direct borohydride – air fuel cell of the fuel prepared according to Example 4.

DETAILED DESCRIPTION OF THE PRESENT INVENTION

[0068] The particulars shown herein are by way of example and for purposes of illustrative discussion of the embodiments of the present invention only and are presented in the cause of providing what is believed to be the most useful and readily understood description of the principles and conceptual aspects of the present invention. In this regard, no attempt is made to show details of the present invention in more detail than is necessary for the fundamental understanding of the present invention, the description making apparent to those skilled in the art how the several forms of the present invention may be embodied in practice.

Example 1 (comparative)

[0069] A fuel of the following composition (in % by weight) is prepared:

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Water	58 %
KOH	23 %
KBH ₄	19 %

The discharge curve of this fuel at a constant voltage of 0.6 V in a direct borohydride – air fuel cell (anode area = 17 cm²; fuel volume 55 cc) is shown in Fig. 1. The discharge energy of the fuel is 13.8 Wh/24h.

Example 2

[0070] A fuel of the following composition (in % by weight) is prepared:

Water	71 %
KOH	10 %
NaOH	7 %
KBH ₄	7 %
NaBH ₄	5 %

[0071] The discharge curve of this fuel at a constant voltage of 0.6 V in a direct borohydride – air fuel cell (anode area = 17 cm²; fuel volume 55 cc) is shown in Fig. 2. The discharge energy of the fuel is 16.2 Wh/24h.

Example 3

[0072] A fuel of the following composition (in % by weight) is prepared:

Water	67 %
KOH	10 %
NaOH	7 %
KBH ₄	9.5 %
NaBH ₄	6.5 %

[0073] The discharge curve of this fuel at a constant voltage of 0.6 V in a direct borohydride – air fuel cell (anode area = 17 cm²; fuel volume 55 cc) is shown in Fig. 3. The discharge energy of the fuel is 18 Wh/24h.

Example 4

[0074] A fuel of the following composition (in % by weight) is prepared:

Water	64.9 %
KOH	14.6 %
NaOH	3.5 %
KBH ₄	14 %

NaBH_4

3 %

[0075] The discharge curve of this fuel at a constant voltage of 0.6 V in a direct borohydride – air fuel cell (anode area = 17 cm²; fuel volume 55 cc) is shown in Fig. 4. The discharge energy of the fuel is 19.2 Wh/24h.

[0076] It is noted that the foregoing examples have been provided merely for the purpose of explanation and are in no way to be construed as limiting of the present invention. While the present invention has been described with reference to exemplary embodiments, it is understood that the words which have been used herein are words of description and illustration, rather than words of limitation. Changes may be made, within the purview of the appended claims, as presently stated and as amended, without departing from the scope and spirit of the present invention in its aspects. Although the present invention has been described herein with reference to particular means, materials and embodiments, the present invention is not intended to be limited to the particulars disclosed herein; rather, the present invention extends to all functionally equivalent structures, methods and uses, such as are within the scope of the appended claims.

WHAT IS CLAIMED IS:

1. A hydride containing fuel composition for a liquid fuel cell, wherein the composition comprises an alkaline liquid phase and at least a first hydride compound and a second hydride compound and wherein a solubility of the first hydride compound in the liquid phase is higher than a solubility of the second hydride compound in the liquid phase and a solubility of an anodic oxidation product of the first hydride compound in the liquid phase is lower than a solubility of an anodic oxidation product of the second hydride compound in the liquid phase.
2. The fuel composition of claim 1, wherein the first hydride compound and the second hydride compound are independently selected from hydrides, borohydrides and aluminum hydrides of alkali and alkaline earth metals, ammonium, Zn and Al.
3. The fuel composition of any one of claims 1 and 2, wherein the first hydride compound and the second hydride compound are independently selected from NaBH_4 , KBH_4 , LiBH_4 , NH_4BH_4 , $\text{Be}(\text{BH}_4)_2$, $\text{Ca}(\text{BH}_4)_2$, $\text{Mg}(\text{BH}_4)_2$, $\text{Zn}(\text{BH}_4)_2$, $\text{Al}(\text{BH}_4)_3$, polyborohydrides, $(\text{CH}_3)_3\text{NBH}_3$, NaCNBH_3 , LiH , NaH , KH , CaH_2 , BeH_2 , MgH_2 , NaAlH_4 , LiAlH_4 and KAlH_4 .
4. The fuel composition of claim 3, wherein the first hydride compound and the second hydride compound are independently selected from NaBH_4 , KBH_4 , LiBH_4 , NH_4BH_4 and polyborohydrides of formula MB_3H_8 , $\text{M}_2\text{B}_{10}\text{H}_{10}$, $\text{MB}_{10}\text{H}_{13}$, $\text{M}_2\text{B}_{12}\text{H}_{12}$ or $\text{M}_2\text{B}_{20}\text{H}_{18}$ wherein $\text{M} = \text{Li}$, Na , K , NH_4 , $\text{Be}_{1/2}$, $\text{Ca}_{1/2}$, $\text{Mg}_{1/2}$, $\text{Zn}_{1/2}$ or $\text{Al}_{1/3}$.
5. The fuel composition of any one of claims 1 to 4, wherein at least one of the first hydride compound and the second hydride compound is selected from borohydrides and polyborohydrides.
6. The fuel composition of claim 5, wherein the first hydride compound and the second hydride compound are selected from borohydrides and polyborohydrides of alkali and alkaline earth metals.
7. The fuel composition of claim 5, wherein at least one of the first hydride compound and the second hydride compound is NaBH_4 or KBH_4 .

8. The fuel composition of any one of claims 1 to 7, wherein a molar ratio of the first hydride compound and the second hydride compound is from about 95 : 5 to about 5 : 95.
9. The fuel composition of claim 8, wherein the molar ratio is from about 60 : 40 to about 40 : 60.
10. The fuel composition of any one of claims 1 to 9, wherein the composition comprises hydride compounds in a total concentration of at least about 0.5 mole per liter of composition.
11. The fuel composition of claim 10, wherein the total concentration is at least about 3 moles per liter of composition.
12. The fuel composition of any one of claims 1 to 11, wherein the liquid phase comprises hydroxide ions.
13. The fuel composition of claim 12, wherein a hydroxide ion concentration in the liquid phase is at least about 0.01 mole per liter.
14. The fuel composition of claim 13, wherein the hydroxide ion concentration is at least about 0.1 mole per liter.
15. The fuel composition of any one of claims 1 to 14, wherein the liquid phase comprises at least one hydroxide ion providing compound dissolved therein, said hydroxide ion providing compound being selected from alkali and alkaline earth metal hydroxides and ammonium hydroxide.
16. The fuel composition of any one of claims 1 to 15, wherein the liquid phase comprises dissolved therein one or more of LiOH, NaOH, KOH, RbOH, CsOH, Ca(OH)₂, Mg(OH)₂, Ba(OH)₂, Zn(OH)₂, Al(OH)₃, and NH₄OH.
17. The fuel composition of claim 16, wherein the liquid phase comprises dissolved therein at least one of NaOH and KOH.

18. The fuel composition of any one of claims 1 to 17, wherein the liquid phase comprises at least one of water, a (cyclo)aliphatic alcohol having up to about 6 carbon atoms and up to about 6 hydroxy groups, a C₂₋₄ alkylene glycol, a di(C₂₋₄ alkylene glycol), a poly(C₂₋₄ alkylene glycol), a mono-C₁₋₄-alkyl ether of a C₂₋₄ alkylene glycol, di(C₂₋₄ alkylene glycol) or poly(C₂₋₄ alkylene glycol), a di-C₁₋₄-alkyl ether of a C₂₋₄ alkylene glycol, di(C₂₋₄ alkylene glycol) or poly(C₂₋₄ alkylene glycol), an ethylene oxide/propylene oxide block copolymer, an ethoxylated aliphatic polyol, a propoxylated aliphatic polyol, an ethoxylated and propoxylated aliphatic polyol, an aliphatic ether having up to about 6 carbon atoms, an aliphatic ketone having up to about 6 carbon atoms, an aliphatic aldehyde having up to about 6 carbon atoms, a C₁₋₄-alkyl ester of a C₁₋₄ alkanolic (aliphatic) acid and a primary, secondary or tertiary aliphatic amine having a total of up to about 10 carbon atoms.

19. The fuel composition of claim 18, wherein the liquid phase comprises water and at least one of an aliphatic alcohol having up to about 6 carbon atoms and up to about 6 hydroxy groups, a C₂₋₄ alkylene glycol, a di(C₂₋₄ alkylene glycol), a poly(C₂₋₄ alkylene glycol), a mono-C₁₋₄-alkyl ether of a C₂₋₄ alkylene glycol, a di(C₂₋₄ alkylene glycol) or a poly(C₂₋₄ alkylene glycol), a di-C₁₋₄-alkyl ether of a C₂₋₄ alkylene glycol, a di(C₂₋₄ alkylene glycol) or a poly(C₂₋₄ alkylene glycol), an ethylene oxide/propylene oxide block copolymer, an ethoxylated and/or propoxylated aliphatic polyol, an aliphatic ether having up to about 6 carbon atoms, an aliphatic ketone having up to about 6 carbon atoms, and a primary, secondary or tertiary aliphatic amine having a total of up to about 10 carbon atoms.

20. The fuel composition of any one of claims 1 to 19, wherein the liquid phase comprises water and at least one of methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol, glycerol, acetone, methyl ethyl ketone, diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran, diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine.

21. The fuel composition of any one of claims 1 to 20, wherein the liquid phase comprises water.

22. The fuel composition of claim 21, wherein the composition comprises two hydride compounds.

23. A hydride containing fuel composition for a liquid fuel cell, wherein the composition comprises an alkaline liquid phase and at least a first hydride compound and a second hydride compound and wherein a solubility of the first hydride compound in the liquid phase is higher than a solubility of the second hydride compound in the liquid phase and a solubility of an anodic oxidation product of the first hydride compound in the liquid phase is lower than a solubility of an anodic oxidation product of the second hydride compound in the liquid phase, said alkaline liquid phase having a hydroxide ion concentration of at least about 0.5 mole per liter and comprising dissolved therein one or more of LiOH, NaOH, KOH, RbOH, CsOH, Ca(OH)₂, Mg(OH)₂, Ba(OH)₂, Zn(OH)₂, Al(OH)₃ and NH₄OH, and said first hydride compound and said second hydride compound being independently selected from NaBH₄, KBH₄, LiBH₄, NH₄BH₄, Be(BH₄)₂, Ca(BH₄)₂, Mg(BH₄)₂, Zn(BH₄)₂, Al(BH₄)₃, polyborohydrides, (CH₃)₃NBH₃, NaCNBH₃, LiH, NaH, KH, CaH₂, BeH₂, MgH₂, NaAlH₄, LiAlH₄ and KAlH₄.

24. The fuel composition of claim 23, wherein at least one of the first hydride compound and the second hydride compound is selected from NaBH₄ and KBH₄.

25. The fuel composition of any one of claims 23 and 24, wherein a molar ratio of the first hydride compound and the second hydride compound is from about 95 : 5 to about 5 : 95.

26. The fuel composition of any one of claims 23 to 25, wherein the composition comprises hydride compounds in a total concentration of at least about 1 mole per liter of composition.

27. The fuel composition of any one of claims 23 to 26, wherein the hydroxide ion concentration is at least about 1 mole per liter of liquid phase.

28. The fuel composition of any one of claims 23 to 27, wherein the hydroxide ion concentration in the liquid phase is not higher than about 7 moles per liter of liquid phase.

29. The fuel composition of any one of claims 23 to 28, wherein the liquid phase comprises dissolved therein one or more of NaOH and KOH.

30. The fuel composition of any one of claims 23 to 29, wherein the liquid phase comprises water and at least one of methanol, ethanol, propanol, isopropanol, ethylene glycol, diethylene glycol, 1,2,4-butanetriol, trimethylolpropane, pentaerythritol, sorbitol, glycerol, acetone, methyl ethyl ketone, diethyl ketone, methyl acetate, ethyl acetate, dioxan, tetrahydrofuran, diglyme, triglyme, monoethanolamine, diethanolamine, triethanolamine, monopropylamine, dipropylamine and tripropylamine.

31. A hydride containing fuel composition for a direct liquid fuel cell, wherein the composition comprises at least two different hydride compounds, the at least two different hydride compounds being such that the composition provides a higher efficiency than an otherwise identical fuel composition that comprises only one of the these different hydride compounds in a molar amount which is identical with a total molar amount of the at least two different hydride compounds.

32. The composition of claim 31, wherein a first hydride compound of the at least two different hydride compounds has a higher solubility in a liquid phase of a liquid fuel than a second hydride compound of the at least two different hydride compounds and wherein an anodic oxidation product of the first hydride compound has a lower solubility in the liquid phase of the liquid fuel than an anodic oxidation product of the second hydride compound.

33. A liquid fuel cell which comprises the fuel composition of any one of claims 1 to 32.

34. A fuel cartridge for filling a liquid fuel cell, wherein the cartridge comprises the fuel composition of any one of claims 1 to 32.

35. A method of increasing the fuel efficiency of a hydride compound containing liquid fuel composition for a direct liquid fuel cell, wherein the method comprises employing at least two different hydride compounds in the fuel composition, the at least two different hydride compounds being selected such that a fuel composition comprising the at least two different hydride compounds has a higher efficiency than an otherwise

identical fuel composition which comprises only one of the at least two different hydride compounds in a molar amount which is identical with a total molar amount of the at least two different hydride compounds.

36. The method of claim 35, wherein a first hydride compound of the at least two different hydride compounds has a higher solubility in a liquid phase of the liquid fuel composition than a second hydride compound of the at least two different hydride compounds and wherein an anodic oxidation product of the first hydride compound has a lower solubility in the liquid phase of the liquid fuel composition than an anodic oxidation product of the second hydride compound.

37. The method of any one of claims 35 and 36, wherein the at least two hydride compounds are present in the liquid phase in a total concentration of at least about 1 mole per liter.

38. The method of any one of claims 35 to 37, wherein the at least two different hydride compounds comprise at least two borohydride compounds.

39. A method of improving the performance of a liquid fuel cell which comprises a hydride compound containing liquid fuel composition, wherein the method comprises employing at least two different hydride compounds in the liquid fuel composition, the at least two different hydride compounds being selected such that the fuel composition comprising the at least two different hydride compounds is chemically less aggressive toward at least one structural component of the fuel cell than, and shows at least the same efficiency as a liquid fuel composition which comprises only one of these hydride compounds in a molar amount which is identical with a total molar amount of the at least two different hydride compounds.

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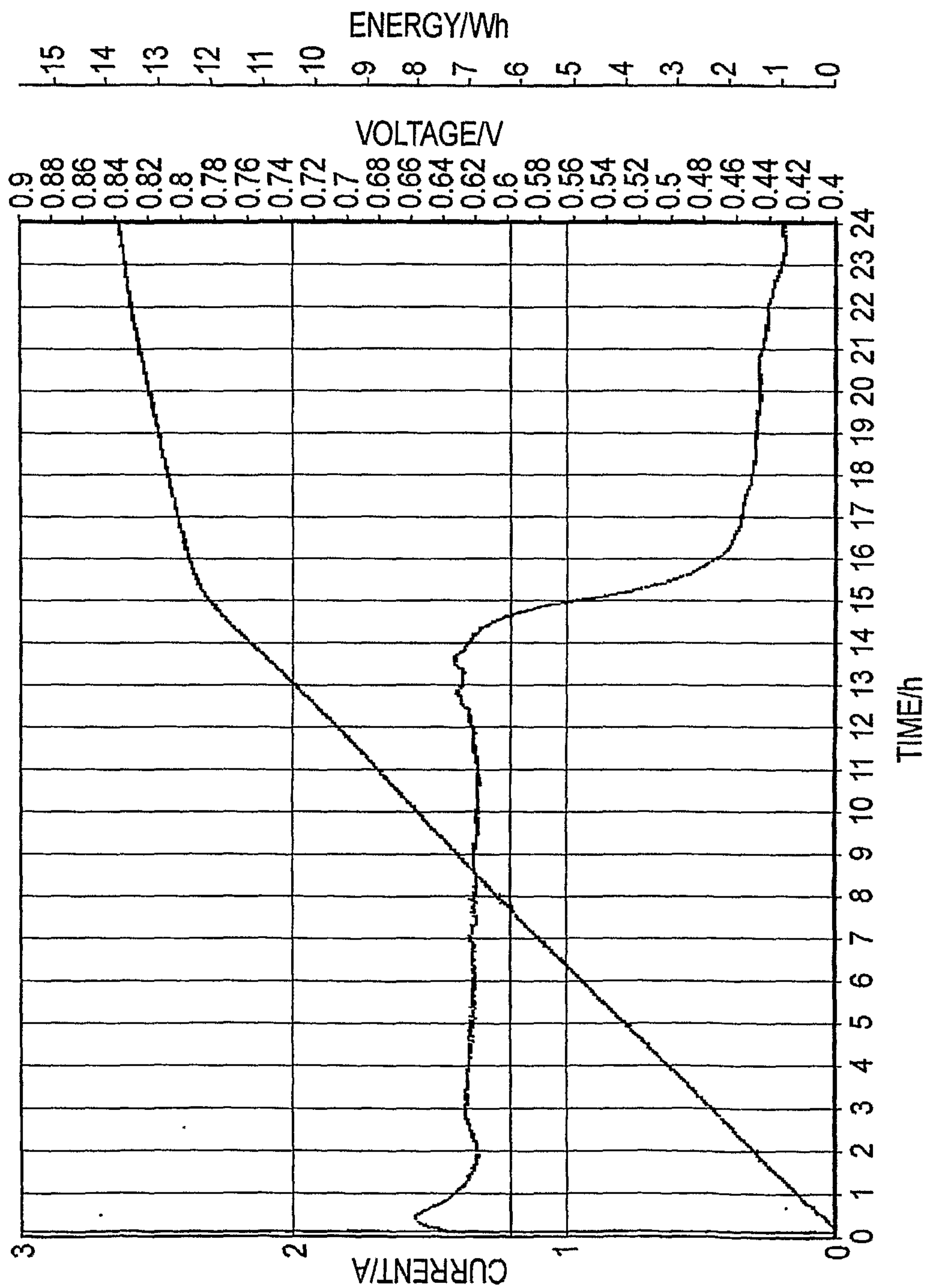


FIG. 1

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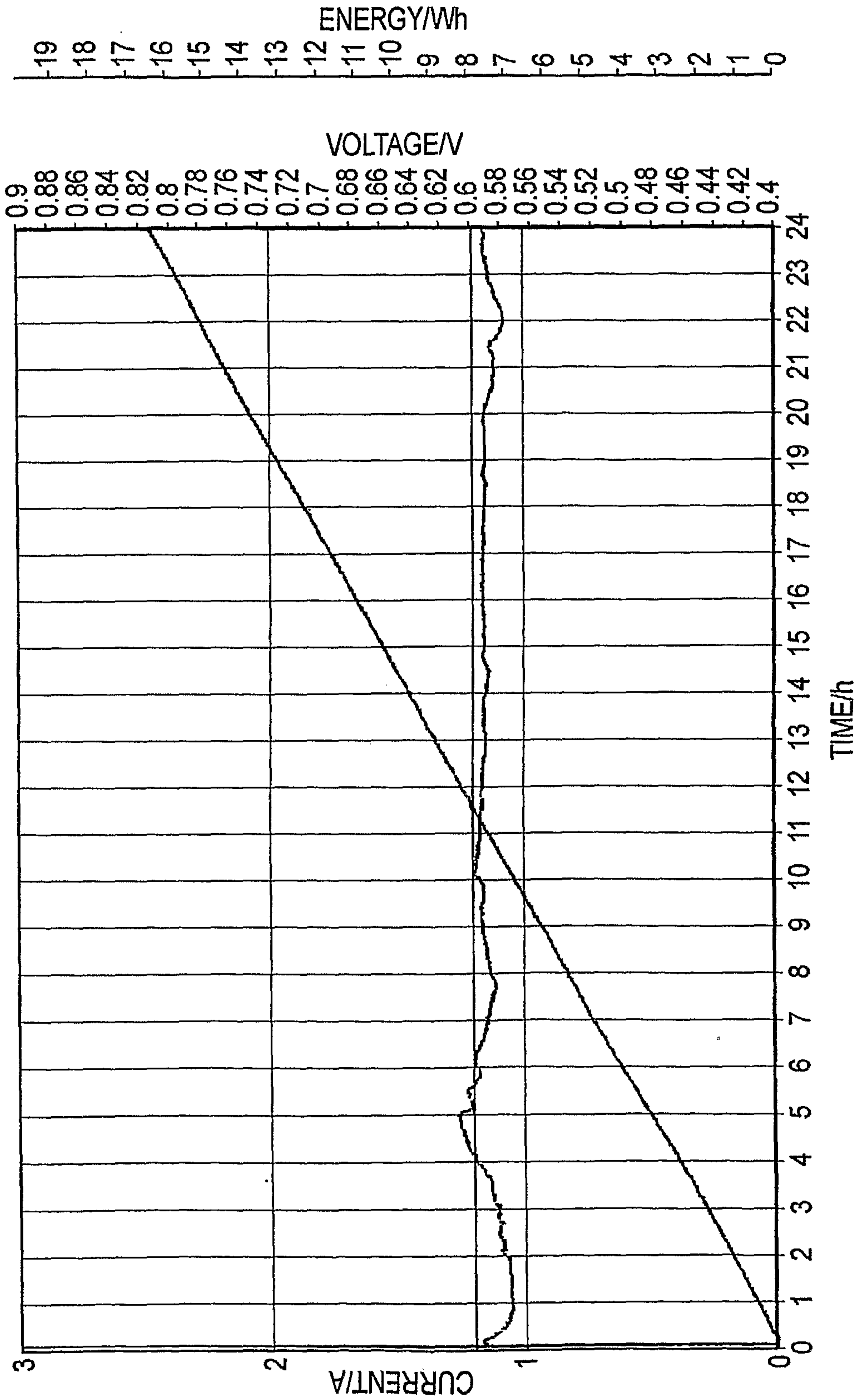


FIG. 2

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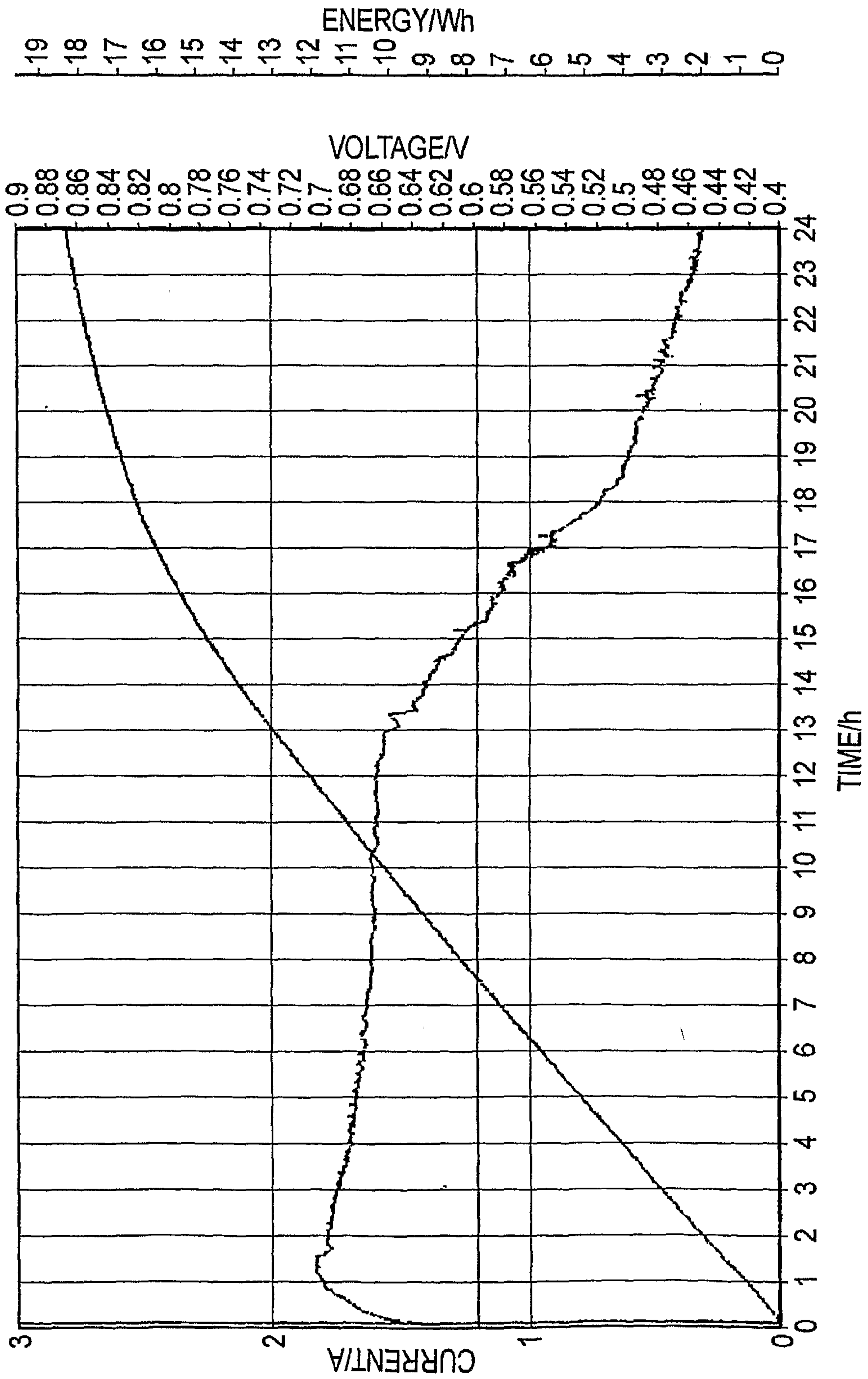


FIG. 3

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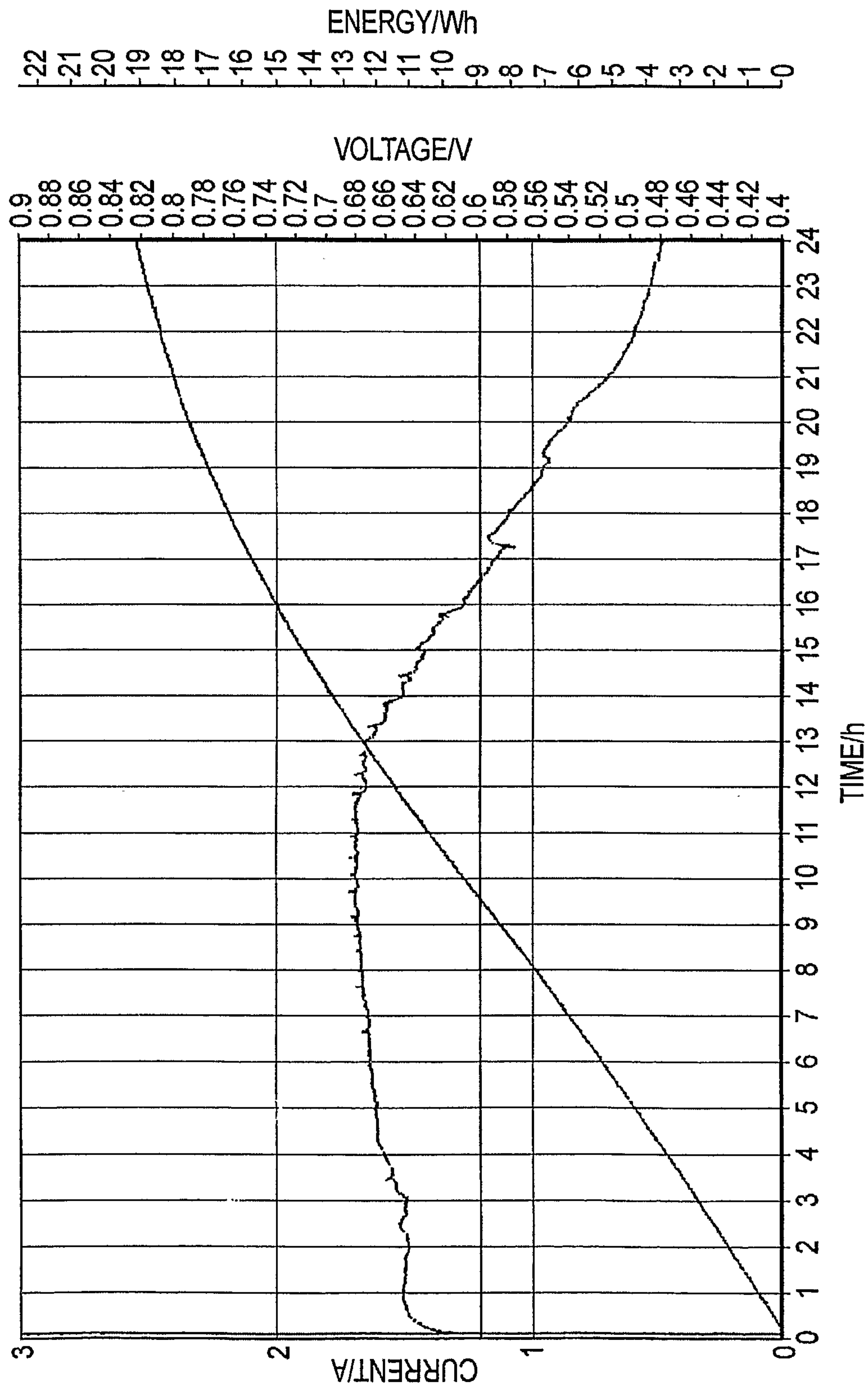


FIG. 4