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UNITED STATES PATENT APPLICATION  
FOR  
HYDROGEN-CATALYST REACTOR  
BY  
RANDELL L. MILLS

**CROSS-REFERENCES TO RELATED APPLICATIONS**

This application claims the benefit of **(1)** Application No. 60/913,556 filed on April 24, 2007; **(2)** Application No. 60/952,305 filed on July 27, 2007; **(3)** Application No. 60/954,426 filed on August 7, 2007; **(4)** Application No. 60/935,373 filed on August 9, 2007; **(5)** Application No. 60/955,465 filed on August 13, 2007; **(6)** Application No. 60/956,821 filed on August 20, 2007; **(7)** Application No. 60/957,540 filed on August 23, 2007; **(8)** Application No. 60/972,342 filed on September 14, 2007; **(9)** Application No. 60/974,191 filed on September 21, 2007; **(10)** Application No. 60/975,330 filed on September 26, 2007; **(11)** Application No. 60/976,004 filed on September 28, 2007; **(12)** Application No. 60/978,435 filed on October 9, 2007; **(13)** Application No. 60/987,552 filed on November 13, 2007; **(14)** Application No. 60/987,946 filed on November 14, 2007; **(15)** Application No. 60/989,677 filed on November 21, 2007; **(16)** Application No. 60/991,434 filed on November 30, 2007; **(17)** Application No. 60/991,974 filed on December 3, 2007; **(18)** Application No. 60/992,601 filed on December 5, 2007; **(19)** Application No. 61/012,717 filed on December 10, 2007; **(20)** Application No. 61/014,860 filed on December 19, 2007; **(21)** Application No. 61/016,790 filed on December 26, 2007; **(22)** Application No. 61/020,023 filed on January 9, 2008; **(23)** Application No. 61/021,205 filed on January 15, 2008; **(24)** Application No. 61/021,808 filed on January 17, 2008; **(25)** Application No. 61/022,112 filed on January 18, 2008; **(26)** Application No. 61/022,949 filed on January 23, 2008; **(27)** Application No. 61/023,297 filed on January 24, 2008; **(28)** Application No. 61/023,687 filed on January 25, 2008; **(29)** Application No. 61/024,730 filed on January 30, 2008; **(30)** Application No. 61/025,520 filed on February 1, 2008; **(31)** Application No. 61/028,605 filed on February 14, 2008; **(32)** Application No. 61/030,468 filed on February 21, 2008; **(33)**

Application No. 61/064,453 filed on March 6, 2008; **(34)** Application No. 61/xxx,xxx filed on March 21, 2008, and **(35)** Application No. 61/xxx,xxx filed on April 17, 2008, all of which are herein incorporated by reference in their entirety.

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## DESCRIPTION OF THE INVENTION

### 1. Field of the Invention:

As disclosed in the paper R. Mills, J. He, Z. Chang, W. Good, Y. Lu, B. Dhandapani, "Catalysis of Atomic Hydrogen to Novel Hydrogen Species  $H^-(1/4)$  and  $H_2(1/4)$  as a New Power Source", Int. J. Hydrogen Energy, Vol. 32, No. 12, 10 (2007), pp. 2573-2584 which is herein incorporated by reference, the data from a broad spectrum of investigational techniques strongly and consistently indicates that hydrogen can exist in lower-energy states than previously thought possible. The predicted reaction involves a resonant, nonradiative energy transfer from otherwise stable atomic hydrogen to a catalyst capable of accepting the energy. The product is 15  $H(1/p)$ , fractional Rydberg states of atomic hydrogen wherein  $n = \frac{1}{2}, \frac{1}{3}, \frac{1}{4}, \dots, \frac{1}{p}$ ; ( $p \leq 137$  is an integer) replaces the well known parameter  $n = \text{integer}$  in the Rydberg equation for hydrogen excited states.  $He^+$ ,  $Ar^+$ , and  $K$  are predicted to serve as catalysts since they meet the catalyst criterion—a chemical or physical process with an enthalpy change equal to an integer multiple of the potential energy of atomic 20 hydrogen, 27.2 eV. Specific predictions based on closed-form equations for energy levels were tested. For example, two  $H(1/p)$  may react to form  $H_2(1/p)$  that have vibrational and rotational energies that are  $p^2$  times those of  $H_2$  comprising uncatalyzed atomic hydrogen. Rotational lines were observed in the 145-300 nm region from atmospheric pressure electron-beam excited argon-hydrogen plasmas.

The unprecedented energy spacing of  $4^2$  times that of hydrogen established the internuclear distance as  $1/4$  that of  $H_2$  and identified  $H_2(1/4)$ .

The predicted products of alkali catalyst  $K$  are  $H^-(1/4)$  which form  $KH * X$ , a novel alkali halide ( $X$ ) hydride compound, and  $H_2(1/4)$  which may be trapped in the crystal. The  $^1H$  MAS NMR spectrum of novel compound  $KH * Cl$  relative to external tetramethylsilane (TMS) showed a large distinct upfield resonance at  $-4.4$  ppm corresponding to an absolute resonance shift of  $-35.9$  ppm that matched the theoretical prediction of  $H^-(1/p)$  with  $p = 4$ . The predicted frequencies of ortho and para- $H_2(1/4)$  were observed at  $1943\text{ cm}^{-1}$  and  $2012\text{ cm}^{-1}$  in the high resolution FTIR spectrum of  $KH * I$  having a  $-4.6$  ppm NMR peak assigned to  $H^-(1/4)$ . The  $1943/2012\text{ cm}^{-1}$ -intensity ratio matched the characteristic ortho-to-para-peak-intensity ratio of 3:1, and the ortho-para splitting of  $69\text{ cm}^{-1}$  matched that predicted.  $KH * Cl$  having  $H^-(1/4)$  by NMR was incident to the 12.5 keV electron-beam which excited similar emission of interstitial  $H_2(1/4)$  as observed in the argon-hydrogen plasma.  $KNO_3$  and Raney nickel were used as a source of  $K$  catalyst and atomic hydrogen, respectively, to produce the corresponding exothermic reaction. The energy balance was  $\Delta H = -17,925\text{ kcal / mole } KNO_3$ , about 300 times that expected for the most energetic known chemistry of  $KNO_3$ , and  $-3585\text{ kcal / mole } H_2$ , over 60 times the hypothetical maximum enthalpy of  $-57.8\text{ kcal / mole } H_2$  due to combustion of hydrogen with atmospheric oxygen, assuming the maximum possible  $H_2$  inventory. The reduction of  $KNO_3$  to water, potassium metal, and  $NH_3$  calculated from the heats of formation only releases  $-14.2\text{ kcal / mole } H_2$  which cannot account for the observed heat; nor can hydrogen combustion. But, the results are consistent

with the formation of  $H^-(1/4)$  and  $H_2(1/4)$  having enthalpies of formation of over 100 times that of combustion.

In embodiments, the invention comprises a power source and a reactor to form lower-energy-hydrogen species and compounds. The invention further  
 5 comprises catalyst reaction mixtures to provide catalyst and atomic hydrogen. Preferred atomic catalysts are lithium, potassium, and cesium atoms. A preferred molecular catalyst is NaH.

### Hydrinos

10 A hydrogen atom having a binding energy given by

$$\text{Binding Energy} = \frac{13.6 \text{ eV}}{(1/p)^2} \quad (1)$$

where  $p$  is an integer greater than 1, preferably from 2 to 137, is disclosed in R. L. Mills, "The Grand Unified Theory of Classical Quantum Mechanics", October 2007 Edition, (posted at <http://www.blacklightpower.com/theory/book.shtml>); R. Mills, *The*  
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The binding energy of an atom, ion, or molecule, also known as the ionization energy, is the energy required to remove one electron from the atom, ion or molecule. A hydrogen atom having the binding energy given in Eq. (1) is hereafter referred to as a hydrino atom or hydrino. The designation for a hydrino of radius  $\frac{a_H}{p}$ , where  $a_H$  is the radius of an ordinary hydrogen atom and  $p$  is an integer, is  $H\left[\frac{a_H}{p}\right]$ . A hydrogen atom with a radius  $a_H$  is hereinafter referred to as "ordinary hydrogen atom" or "normal hydrogen atom." Ordinary atomic hydrogen is characterized by its binding energy of 13.6 eV.

Hydrinos are formed by reacting an ordinary hydrogen atom with a catalyst having a net enthalpy of reaction of about

$$m \cdot 27.2 \text{ eV} \quad (2)$$

where  $m$  is an integer. This catalyst has also been referred to as an energy hole or source of energy hole in Mills earlier filed Patent Applications. It is believed that the rate of catalysis is increased as the net enthalpy of reaction is more closely matched to  $m \cdot 27.2 \text{ eV}$ . It has been found that catalysts having a net enthalpy of reaction within  $\pm 10\%$ , preferably  $\pm 5\%$ , of  $m \cdot 27.2 \text{ eV}$  are suitable for most applications.

This catalysis releases energy from the hydrogen atom with a commensurate decrease in size of the hydrogen atom,  $r_n = na_H$ . For example, the catalysis of  $H(n=1)$  to  $H(n=1/2)$  releases  $40.8 \text{ eV}$ , and the hydrogen radius decreases from  $a_H$  to  $\frac{1}{2}a_H$ . A catalytic system is provided by the ionization of  $t$  electrons from an atom each to a continuum energy level such that the sum of the ionization energies of the  $t$  electrons is approximately  $m \cdot 27.2 \text{ eV}$  where  $m$  is an integer.

One such catalytic system involves lithium metal. The first and second ionization energies of lithium are  $5.39172 \text{ eV}$  and  $75.64018 \text{ eV}$ , respectively [1]. The double ionization ( $t=2$ ) reaction of  $Li$  to  $Li^{2+}$ , then, has a net enthalpy of reaction of  $81.0319 \text{ eV}$ , which is equivalent to  $m=3$  in Eq. (2).

$$81.0319 \text{ eV} + Li(m) + H\left[\frac{a_H}{p}\right] \rightarrow Li^{2+} + 2e^- + H\left[\frac{a_H}{(p+3)}\right] + [(p+3)^2 - p^2] \cdot 13.6 \text{ eV} \quad (3)$$

$$Li^{2+} + 2e^- \rightarrow Li(m) + 81.0319 \text{ eV} \quad (4)$$

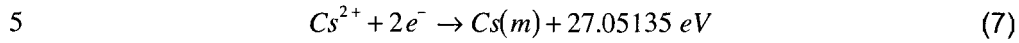
And, the overall reaction is

$$H\left[\frac{a_H}{p}\right] \rightarrow H\left[\frac{a_H}{(p+3)}\right] + [(p+3)^2 - p^2] \cdot 13.6 \text{ eV} \quad (5)$$

In another embodiment, the catalytic system involves cesium. The first and second ionization energies of cesium are  $3.89390 \text{ eV}$  and  $23.15745 \text{ eV}$ , respectively.

The double ionization ( $t = 2$ ) reaction of  $Cs$  to  $Cs^{2+}$ , then, has a net enthalpy of reaction of  $27.05135 \text{ eV}$ , which is equivalent to  $m = 1$  in Eq. (2).

$$27.05135 \text{ eV} + Cs(m) + H\left[\frac{a_H}{p}\right] \rightarrow Cs^{2+} + 2e^- + H\left[\frac{a_H}{(p+1)}\right] + [(p+1)^2 - p^2] \cdot 13.6 \text{ eV} \quad (6)$$



And, the overall reaction is

$$H\left[\frac{a_H}{p}\right] \rightarrow H\left[\frac{a_H}{(p+1)}\right] + [(p+1)^2 - p^2] \cdot 13.6 \text{ eV} \quad (8)$$

An additional catalytic system involves potassium metal. The first, second, and third ionization energies of potassium are  $4.34066 \text{ eV}$ ,  $31.63 \text{ eV}$ ,  $45.806 \text{ eV}$ , respectively [1]. The triple ionization ( $t = 3$ ) reaction of  $K$  to  $K^{3+}$ , then, has a net enthalpy of reaction of  $81.7767 \text{ eV}$ , which is equivalent to  $m = 3$  in Eq. (2).

$$81.7767 \text{ eV} + K(m) + H\left[\frac{a_H}{p}\right] \rightarrow K^{3+} + 3e^- + H\left[\frac{a_H}{(p+3)}\right] + [(p+3)^2 - p^2] \cdot 13.6 \text{ eV} \quad (9)$$

$$K^{3+} + 3e^- \rightarrow K(m) + 81.7426 \text{ eV} \quad (10)$$

And, the overall reaction is

$$H\left[\frac{a_H}{p}\right] \rightarrow H\left[\frac{a_H}{(p+3)}\right] + [(p+3)^2 - p^2] \cdot 13.6 \text{ eV} \quad (11)$$

As a power source, the energy given off during catalysis is much greater than the energy lost to the catalyst. The energy released is large as compared to conventional chemical reactions. For example, when hydrogen and oxygen gases undergo combustion to form water



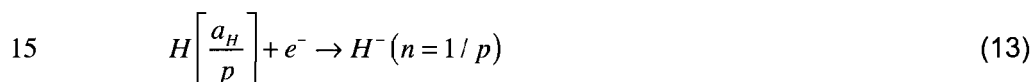
the known enthalpy of formation of water is  $\Delta H_f = -286 \text{ kJ / mole}$  or  $1.48 \text{ eV}$  per hydrogen atom. By contrast, each ( $n = 1$ ) ordinary hydrogen atom undergoing catalysis releases a net of  $40.8 \text{ eV}$ . Moreover, further catalytic transitions may occur:  $n = \frac{1}{2} \rightarrow \frac{1}{3}, \frac{1}{3} \rightarrow \frac{1}{4}, \frac{1}{4} \rightarrow \frac{1}{5}$ , and so on. Once catalysis begins, hydrinos

- 5 autocatalyze further in a process called disproportionation. This mechanism is similar to that of an inorganic ion catalysis. But, hydrino catalysis should have a higher reaction rate than that of the inorganic ion catalyst due to the better match of the enthalpy to  $m \cdot 27.2 \text{ eV}$ .

#### 10 Further Catalysis Products of the Present Invention

The hydrino hydride ion of the present invention can be formed by the reaction of an electron source with a hydrino, that is, a hydrogen atom having a binding energy of about  $\frac{13.6 \text{ eV}}{n^2}$ , where  $n = \frac{1}{p}$  and  $p$  is an integer greater than 1.

The hydrino hydride ion is represented by  $H^-(n = 1/p)$  or  $H^-(1/p)$ :



- The hydrino hydride ion is distinguished from an ordinary hydride ion comprising an ordinary hydrogen nucleus and two electrons having a binding energy of about  $0.8 \text{ eV}$ . The latter is hereafter referred to as "ordinary hydride ion" or
- 20 "normal hydride ion" The hydrino hydride ion comprises a hydrogen nucleus including protium, deuterium, or tritium, and two indistinguishable electrons at a binding energy according to Eq. (15).

The binding energy of a novel hydrino hydride ion can be represented by the following formula:

$$\text{Binding Energy} = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi\mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right) \quad (15)$$

- 5 where  $p$  is an integer greater than one,  $s = 1/2$ ,  $\pi$  is pi,  $\hbar$  is Planck's constant bar,  $\mu_o$  is the permeability of vacuum,  $m_e$  is the mass of the electron,  $\mu_e$  is the reduced electron mass given by  $\mu_e = \frac{m_e m_p}{\frac{m_e}{\sqrt{3}} + m_p}$  where  $m_p$  is the mass of the proton,  $a_H$  is the

radius of the hydrogen atom,  $a_o$  is the Bohr radius, and  $e$  is the elementary charge.

The radii are given by

$$10 \quad r_2 = r_1 = a_0 \left( 1 + \sqrt{s(s+1)} \right); s = \frac{1}{2} \quad (16)$$

The binding energies of the hydrino hydride ion,  $H^-(n = 1/p)$  as a function of  $p$ , where  $p$  is an integer, are shown in TABLE 1.

TABLE 1. The representative binding energy of the hydrino hydride ion  $H^-(n = 1/p)$  as a function of  $p$ , Eq. (15).

|    | Hydride Ion     | $r_1$     | Binding                  | Wavelength |
|----|-----------------|-----------|--------------------------|------------|
| 5  |                 | $(a_o)^a$ | Energy (eV) <sup>b</sup> | (nm)       |
|    | $H^-(n = 1)$    | 1.8660    | 0.7542                   | 1644       |
|    | $H^-(n = 1/2)$  | 0.9330    | 3.047                    | 406.9      |
|    | $H^-(n = 1/3)$  | 0.6220    | 6.610                    | 187.6      |
| 10 | $H^-(n = 1/4)$  | 0.4665    | 11.23                    | 110.4      |
|    | $H^-(n = 1/5)$  | 0.3732    | 16.70                    | 74.23      |
|    | $H^-(n = 1/6)$  | 0.3110    | 22.81                    | 54.35      |
|    | $H^-(n = 1/7)$  | 0.2666    | 29.34                    | 42.25      |
|    | $H^-(n = 1/8)$  | 0.2333    | 36.09                    | 34.46      |
| 15 | $H^-(n = 1/9)$  | 0.2073    | 42.84                    | 28.94      |
|    | $H^-(n = 1/10)$ | 0.1866    | 49.38                    | 25.11      |
|    | $H^-(n = 1/11)$ | 0.1696    | 55.50                    | 22.34      |
|    | $H^-(n = 1/12)$ | 0.1555    | 60.98                    | 20.33      |
|    | $H^-(n = 1/13)$ | 0.1435    | 65.63                    | 18.89      |
| 20 | $H^-(n = 1/14)$ | 0.1333    | 69.22                    | 17.91      |
|    | $H^-(n = 1/15)$ | 0.1244    | 71.55                    | 17.33      |
|    | $H^-(n = 1/16)$ | 0.1166    | 72.40                    | 17.12      |
|    | $H^-(n = 1/17)$ | 0.1098    | 71.56                    | 17.33      |

20

|   |                   |        |        |       |
|---|-------------------|--------|--------|-------|
|   | $H^-(n = 1 / 18)$ | 0.1037 | 68.83  | 18.01 |
|   | $H^-(n = 1 / 19)$ | 0.0982 | 63.98  | 19.38 |
|   | $H^-(n = 1 / 20)$ | 0.0933 | 56.81  | 21.82 |
|   | $H^-(n = 1 / 21)$ | 0.0889 | 47.11  | 26.32 |
| 5 | $H^-(n = 1 / 22)$ | 0.0848 | 34.66  | 35.76 |
|   | $H^-(n = 1 / 23)$ | 0.0811 | 19.26  | 64.36 |
|   | $H^-(n = 1 / 24)$ | 0.0778 | 0.6945 | 1785  |

a Eq. (16)

b Eq. (15)

10

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According to the present invention, a hydrino hydride ion ( $H^-$ ) having a binding energy according to Eqs. (15-16) that is greater than the binding of ordinary hydride ion (about 0.8 eV) for  $p = 2$  up to 23, and less for  $p = 24$  ( $H^-$ ) is provided. For  $p = 2$  to  $p = 24$  of Eqs. (15-16), the hydride ion binding energies are respectively 3, 6.6, 11.2, 16.7, 22.8, 29.3, 36.1, 42.8, 49.4, 55.5, 61.0, 65.6, 69.2, 71.6, 72.4, 71.6, 68.8, 64.0, 56.8, 47.1, 34.7, 19.3, and 0.69 eV. Compositions comprising the novel hydride ion are also provided.

The hydrino hydride ion is distinguished from an ordinary hydride ion comprising an ordinary hydrogen nucleus and two electrons having a binding energy of about 0.8 eV. The latter is hereafter referred to as "ordinary hydride ion" or "normal hydride ion". The hydrino hydride ion comprises a hydrogen nucleus including protium, deuterium, or tritium, and two indistinguishable electrons at a binding energy according to Eqs. (15-16).

Novel compounds are provided comprising one or more hydrino hydride ions and one or more other elements. Such a compound is referred to as a hydrino hydride compound.

Ordinary hydrogen species are characterized by the following binding  
 5 energies (a) hydride ion, 0.754 eV ("ordinary hydride ion"); (b) hydrogen atom ("ordinary hydrogen atom"), 13.6 eV; (c) diatomic hydrogen molecule, 15.3 eV ("ordinary hydrogen molecule"); (d) hydrogen molecular ion, 16.3 eV ("ordinary hydrogen molecular ion"); and (e)  $H_3^+$ , 22.6 eV ("ordinary trihydrogen molecular ion"). Herein, with reference to forms of hydrogen, "normal" and "ordinary" are  
 10 synonymous.

According to a further embodiment of the invention, a compound is provided comprising at least one increased binding energy hydrogen species such as (a) a hydrogen atom having a binding energy of about  $\frac{13.6 \text{ eV}}{\left(\frac{1}{p}\right)^2}$ , preferably within  $\pm 10\%$ ,

more preferably  $\pm 5\%$ , where p is an integer, preferably an integer from 2 to 137; (b)  
 15 a hydride ion ( $H^-$ ) having a binding energy of about

$$\text{Binding Energy} = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi\mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right), \text{ preferably}$$

within  $\pm 10\%$ , more preferably  $\pm 5\%$ , where p is an integer, preferably an integer from 2 to 24; (c)  $H_4^+(1/p)$ ; (d) a trihydrino molecular ion,  $H_3^+(1/p)$ , having a binding  
 energy of about  $\frac{22.6}{\left(\frac{1}{p}\right)^2} \text{ eV}$  preferably within  $\pm 10\%$ , more preferably  $\pm 5\%$ , where p is

20 an integer, preferably an integer from 2 to 137; (e) a dihydrino having a binding

energy of about  $\frac{15.3}{\left(\frac{1}{p}\right)^2} eV$  preferably within  $\pm 10\%$ , more preferably  $\pm 5\%$ , where  $p$  is

an integer, preferably an integer from 2 to 137; (f) a dihydrino molecular ion with a

binding energy of about  $\frac{16.3}{\left(\frac{1}{p}\right)^2} eV$  preferably within  $\pm 10\%$ , more preferably  $\pm 5\%$ ,

where  $p$  is an integer, preferably an integer from 2 to 137.

- 5 According to a further preferred embodiment of the invention, a compound is provided comprising at least one increased binding energy hydrogen species such as (a) a dihydrino molecular ion having a total energy of

$$E_T = -p^2 \left\{ \frac{e^2}{8\pi\epsilon_o a_H} (4 \ln 3 - 1 - 2 \ln 3) \right\} \left[ 1 + p \sqrt{\frac{2\hbar \sqrt{\frac{2e^2}{4\pi\epsilon_o (2a_H)^3}}}{m_e c^2}} \right] - \frac{1}{2} \hbar \sqrt{\frac{k}{\mu}} \quad (17)$$

$$= -p^2 16.13392 eV - p^3 0.118755 eV$$

preferably within  $\pm 10\%$ , more preferably  $\pm 5\%$ , where  $p$  is an integer,  $\hbar$  is Planck's

- 10 constant bar,  $m_e$  is the mass of the electron,  $c$  is the speed of light in vacuum,  $\mu$  is the reduced nuclear mass, and  $k$  is the harmonic force constant solved previously [2] and (b) a dihydrino molecule having a total energy of

$$E_T = -p^2 \left\{ \frac{e^2}{8\pi\epsilon_o a_0} \left[ \left( 2\sqrt{2} - \sqrt{2} + \frac{\sqrt{2}}{2} \right) \ln \frac{\sqrt{2}+1}{\sqrt{2}-1} - \sqrt{2} \right] \right\} \left[ 1 + p \sqrt{\frac{2\hbar \sqrt{\frac{e^2}{4\pi\epsilon_o a_0^3}}}{m_e c^2}} \right] - \frac{1}{2} \hbar \sqrt{\frac{k}{\mu}}$$

$$= -p^2 31.351 eV - p^3 0.326469 eV$$

(18)

preferably within  $\pm 10\%$ , more preferably  $\pm 5\%$ , where  $p$  is an integer and  $a_o$  is the Bohr radius.

According to one embodiment of the invention wherein the compound  
 5 comprises a negatively charged increased binding energy hydrogen species, the compound further comprises one or more cations, such as a proton, ordinary  $H_2^+$ , or ordinary  $H_3^+$ .

A method is provided for preparing compounds comprising at least one increased binding energy hydride ion. Such compounds are hereinafter referred to  
 10 as "hydrino hydride compounds". The method comprises reacting atomic hydrogen with a catalyst having a net enthalpy of reaction of about  $\frac{m}{2} \cdot 27 \text{ eV}$ , where  $m$  is an integer greater than 1, preferably an integer less than 400, to produce an increased binding energy hydrogen atom having a binding energy of about  $\frac{13.6 \text{ eV}}{\left(\frac{1}{p}\right)^2}$  where  $p$  is  
 an integer, preferably an integer from 2 to 137. A further product of the catalysis is  
 15 energy. The increased binding energy hydrogen atom can be reacted with an electron source, to produce an increased binding energy hydride ion. The increased binding energy hydride ion can be reacted with one or more cations to produce a compound comprising at least one increased binding energy hydride ion.

Novel hydrogen species and compositions of matter comprising new forms of  
 20 hydrogen formed by the catalysis of atomic hydrogen are disclosed in "Mills Prior Publications". The novel hydrogen compositions of matter comprise:

(a) at least one neutral, positive, or negative hydrogen species (hereinafter "increased binding energy hydrogen species") having a binding energy

(i) greater than the binding energy of the corresponding ordinary hydrogen species, or

(ii) greater than the binding energy of any hydrogen species for which the corresponding ordinary hydrogen species is unstable or is not observed because  
5 the ordinary hydrogen species' binding energy is less than thermal energies at ambient conditions (standard temperature and pressure, STP), or is negative; and

(b) at least one other element. The compounds of the invention are hereinafter referred to as "increased binding energy hydrogen compounds".

By "other element" in this context is meant an element other than an  
10 increased binding energy hydrogen species. Thus, the other element can be an ordinary hydrogen species, or any element other than hydrogen. In one group of compounds, the other element and the increased binding energy hydrogen species are neutral. In another group of compounds, the other element and increased binding energy hydrogen species are charged such that the other element provides  
15 the balancing charge to form a neutral compound. The former group of compounds is characterized by molecular and coordinate bonding; the latter group is characterized by ionic bonding.

Also provided are novel compounds and molecular ions comprising

(a) at least one neutral, positive, or negative hydrogen species (hereinafter  
20 "increased binding energy hydrogen species") having a total energy  
(i) greater than the total energy of the corresponding ordinary hydrogen species, or

(ii) greater than the total energy of any hydrogen species for which the corresponding ordinary hydrogen species is unstable or is not observed because the  
25 ordinary hydrogen species' total energy is less than thermal energies at ambient

conditions, or is negative; and

(b) at least one other element.

The total energy of the hydrogen species is the sum of the energies to remove all of the electrons from the hydrogen species. The hydrogen species according to the present invention has a total energy greater than the total energy of the corresponding ordinary hydrogen species. The hydrogen species having an increased total energy according to the present invention is also referred to as an "increased binding energy hydrogen species" even though some embodiments of the hydrogen species having an increased total energy may have a first electron binding energy less than the first electron binding energy of the corresponding ordinary hydrogen species. For example, the hydride ion of Eqs. (15-16) for  $p = 24$  has a first binding energy that is less than the first binding energy of ordinary hydride ion, while the total energy of the hydride ion of Eqs. (15-16) for  $p = 24$  is much greater than the total energy of the corresponding ordinary hydride ion.

Also provided are novel compounds and molecular ions comprising

(a) a plurality of neutral, positive, or negative hydrogen species (hereinafter "increased binding energy hydrogen species") having a binding energy

(i) greater than the binding energy of the corresponding ordinary hydrogen species, or

(ii) greater than the binding energy of any hydrogen species for which the corresponding ordinary hydrogen species is unstable or is not observed because the ordinary hydrogen species' binding energy is less than thermal energies at ambient conditions or is negative; and

(b) optionally one other element. The compounds of the invention are hereinafter referred to as "increased binding energy hydrogen compounds".

The increased binding energy hydrogen species can be formed by reacting one or more hydrino atoms with one or more of an electron, hydrino atom, a compound containing at least one of said increased binding energy hydrogen species, and at least one other atom, molecule, or ion other than an increased binding energy hydrogen species.

Also provided are novel compounds and molecular ions comprising

(a) a plurality of neutral, positive, or negative hydrogen species (hereinafter "increased binding energy hydrogen species") having a total energy

(i) greater than the total energy of ordinary molecular hydrogen, or

(ii) greater than the total energy of any hydrogen species for which the corresponding ordinary hydrogen species is unstable or is not observed because the ordinary hydrogen species' total energy is less than thermal energies at ambient conditions or is negative; and

(b) optionally one other element. The compounds of the invention are hereinafter referred to as "increased binding energy hydrogen compounds".

In an embodiment, a compound is provided, comprising at least one increased binding energy hydrogen species selected from the group consisting of (a) hydride ion having a binding energy according to Eqs. (15-16) that is greater than the binding of ordinary hydride ion (about 0.8 eV) for  $p = 2$  up to 23, and less for  $p = 24$  ("increased binding energy hydride ion" or "hydrino hydride ion"); (b) hydrogen atom having a binding energy greater than the binding energy of ordinary hydrogen atom (about 13.6 eV) ("increased binding energy hydrogen atom" or "hydrino"); (c) hydrogen molecule having a first binding energy greater than about 15.3 eV ("increased binding energy hydrogen molecule" or "dihyrino"); and (d) molecular hydrogen ion having a binding energy greater than about 16.3 eV

("increased binding energy molecular hydrogen ion" or "dihydrino molecular ion").

#### Characteristics and Identification of Increased Binding Energy Species

A new chemically generated or assisted plasma source based on a resonant energy transfer mechanism (rt-plasma) between atomic hydrogen and certain catalysts has been developed that may be a new power source. The products are more stable hydride and molecular hydrogen species such as  $H^-(1/4)$  and  $H_2(1/4)$ . One such source operates by incandescently heating a hydrogen dissociator and a catalyst to provide atomic hydrogen and gaseous catalyst, respectively, such that the catalyst reacts with the atomic hydrogen to produce a plasma. It was extraordinary that intense extreme ultraviolet (EUV) emission was observed by Mills et al. [3-10] at low temperatures (e.g.  $\approx 10^3 K$ ) and an extraordinary low field strength of about 1-2 V/cm from atomic hydrogen and certain atomized elements or certain gaseous ions which singly or multiply ionize at integer multiples of the potential energy of atomic hydrogen, 27.2 eV. A number of independent experimental observations confirm that the rt-plasma is due to a novel reaction of atomic hydrogen which produces as chemical intermediates, hydrogen in fractional quantum states that are at lower energies than the traditional "ground" ( $n=1$ ) state. Power is released [3, 9, 11-13], and the final reaction products are novel hydride compounds [3, 14-16] or lower-energy molecular hydrogen [17]. The supporting data include EUV spectroscopy [3-10, 13, 17-22, 25, 27-28], characteristic emission from catalysts and the hydride ion products [3, 5, 7, 21-22, 27-28], lower-energy hydrogen emission [12-13, 18-20], chemically formed plasmas [3-10, 21-22, 27-28], extraordinary ( $>100$  eV) Balmer  $\alpha$  line broadening [3-5, 7, 9-10, 12, 18-19, 21, 23-28], population inversion of H lines [3, 21, 27-29], elevated

electron temperature [19, 23-25], anomalous plasma afterglow duration [3, 8], power generation [3, 9, 11-13], and analysis of novel chemical compounds [3, 14-16].

The theory given previously [6, 18-20, 30] is based on Maxwell's equations to solving the structure of the electron. The familiar Rydberg equation (Eq. (19)) arises

5 for the hydrogen excited states for  $n > 1$  of Eq. (20).

$$E_n = -\frac{e^2}{n^2 8\pi\epsilon_0 a_H} = -\frac{13.598 \text{ eV}}{n^2} \quad (19)$$

$$n = 1, 2, 3, \dots \quad (20)$$

An additional result is that atomic hydrogen may undergo a catalytic reaction with certain atoms, excimers, and ions which provide a reaction with a net enthalpy of an  
 10 integer multiple of the potential energy of atomic hydrogen,  $m \cdot 27.2 \text{ eV}$  wherein  $m$  is an integer. The reaction involves a nonradiative energy transfer to form a hydrogen atom called a hydrino atom that is lower in energy than unreacted atomic hydrogen that corresponds to a fractional principal quantum number. That is

$$n = \frac{1}{2}, \frac{1}{3}, \frac{1}{4}, \dots, \frac{1}{p}; \quad p \text{ is an integer} \quad (21)$$

15 replaces the well known parameter  $n = \text{integer}$  in the Rydberg equation for hydrogen excited states. The  $n = 1$  state of hydrogen and the  $n = \frac{1}{\text{integer}}$  states of hydrogen

are nonradiative, but a transition between two nonradiative states, say  $n = 1$  to  $n = 1/2$ , is possible via a nonradiative energy transfer. Thus, a catalyst provides a net positive enthalpy of reaction of  $m \cdot 27.2 \text{ eV}$  (i.e. it resonantly accepts the

20 nonradiative energy transfer from hydrogen atoms and releases the energy to the surroundings to affect electronic transitions to fractional quantum energy levels). As a consequence of the nonradiative energy transfer, the hydrogen atom becomes unstable and emits further energy until it achieves a lower-energy nonradiative state

having a principal energy level given by Eqs. (19) and (21). Processes such as hydrogen molecular bond formation that occur without photons and that require collisions are common [31]. Also, some commercial phosphors are based on resonant nonradiative energy transfer involving multipole coupling [32].

- 5 Two  $H(1/p)$  may react to form  $H_2(1/p)$ . The hydrogen molecular ion and molecular charge and current density functions, bond distances, and energies were exactly solved previously with remarkable accuracy [30, 33]. Using the Laplacian in ellipsoidal coordinates with the constraint of nonradiation, the total energy of the hydrogen molecule having a central field of  $+pe$  at each focus of the prolate
- 10 spheroid molecular orbital is

$$E_T = -p^2 \left\{ \frac{e^2}{8\pi\epsilon_0 a_0} \left[ \left( 2\sqrt{2} - \sqrt{2} + \frac{\sqrt{2}}{2} \right) \ln \frac{\sqrt{2}+1}{\sqrt{2}-1} - \sqrt{2} \right] \left[ 1 + p \sqrt{\frac{2\hbar \sqrt{\frac{e^2}{4\pi\epsilon_0 a_0^3}}}{m_e c^2}} \right] - \frac{1}{2} \hbar \sqrt{\frac{k}{\mu}} \right\}$$

$$= -p^2 31.351 \text{ eV} - p^3 0.326469 \text{ eV}$$

(22)

where  $p$  is an integer,  $\hbar$  is Planck's constant bar,  $m_e$  is the mass of the electron,  $c$  is the speed of light in vacuum,  $\mu$  is the reduced nuclear mass,  $k$  is the harmonic

15 force constant solved previously in a closed-form equation with fundamental constants only [30, 33] and  $a_0$  is the Bohr radius. The vibrational and rotational energies of fractional-Rydberg-state molecular hydrogen  $H_2(1/p)$  are  $p^2$  those of  $H_2$ . Thus, the vibrational energies,  $E_{vib}$ , for the  $v=0$  to  $v=1$  transition of hydrogen-type molecules  $H_2(1/p)$  are [30, 33]

$$20 \quad E_{vib} = p^2 0.515902 \text{ eV} \quad (23)$$

where the experimental vibrational energy for the  $v = 0$  to  $v = 1$  transition of  $H_2$ ,  $E_{H_2(v=0 \rightarrow v=1)}$ , is given by Beutler [34] and Herzberg [35]. The rotational energies,  $E_{rot}$ , for the  $J$  to  $J + 1$  transition of hydrogen-type molecules  $H_2(1/p)$  are [30, 33]

$$E_{rot} = E_{J+1} - E_J = \frac{\hbar^2}{I} [J + 1] = p^2 (J + 1) 0.01509 \text{ eV} \quad (24)$$

5 where  $I$  is the moment of inertia, and the experimental rotational energy for the  $J = 0$  to  $J = 1$  transition of  $H_2$  is given by Atkins [36]. The  $p^2$  dependence of the rotational energies results from an inverse  $p$  dependence of the internuclear distance and the corresponding impact on  $I$ . The predicted internuclear distance  $2c'$  for  $H_2(1/p)$  is

$$10 \quad 2c' = \frac{a_o \sqrt{2}}{p} \quad (25)$$

The rotational energies provide a very precise measure of  $I$  and the internuclear distance using well established theory [37].

$Ar^+$  may serve as a catalyst since its ionization energy is about 27.2 eV. The catalyst reaction of  $Ar^+$  to  $Ar^{2+}$  forms  $H(1/2)$  which may further serve as both a

15 catalyst and a reactant to form  $H(1/4)$  [19-20, 30]. Thus, the observation of  $H(1/4)$  is predicted to be flow dependent since the formation of  $H_2(1/4)$  requires the buildup of intermediates. The mechanism was tested by experiments with flowing plasma gases. Neutral molecular emission was anticipated for high pressure argon-hydrogen plasmas excited by a 12.5 keV electron beam. Rotational lines for

20  $H_2(1/4)$  were anticipated and sought in the 150-250 nm region. The spectral lines were compared to those predicted by Eqs. (23-24) corresponding to the internuclear

distance of 1/4 that of  $H_2$  given by Eq. (25). For  $p = 4$  in Eqs. (23-24), the predicted energies for the  $v = 1 \rightarrow v = 0$  vibration-rotational series of  $H_2(1/4)$  are

$$\begin{aligned} E_{vib-rot} &= p^2 E_{vib\ H_2(v=0 \rightarrow v=1)} \pm p^2 (J+1) E_{rot\ H_2}, J = 0, 1, 2, 3... \\ &= 8.254432\ eV \pm (J+1) 0.24144\ eV \end{aligned} \quad (26)$$

$He^+$  also fulfills the catalyst criterion—a chemical or physical process with an enthalpy change equal to an integer multiple of 27.2 eV since it ionizes at 54.417 eV which is  $2 \cdot 27.2\ eV$ . The product of the catalysis reaction of  $He^+$ ,  $H(1/3)$ , may further serve as a catalyst to form  $H(1/4)$  and  $H(1/2)$  [19-20, 30] which can lead to transitions to other states  $H(1/p)$ . Novel emission lines with energies of  $q \cdot 13.6\ eV$  where  $q = 1, 2, 3, 4, 6, 7, 8, 9$ , or 11 were previously observed by extreme ultraviolet (EUV) spectroscopy recorded on microwave discharges of helium with 2% hydrogen [18-20]. These lines matched  $H(1/p)$ , fractional Rydberg states of atomic hydrogen given by Eqs. (19) and (21).

Rotational lines were observed in the 145-300 nm region from atmospheric pressure electron-beam excited argon-hydrogen plasmas. The unprecedented energy spacing of  $4^2$  times that of hydrogen established the internuclear distance as 1/4 that of  $H_2$  and identified  $H_2(1/4)$  (Eqs. (23-26)).  $H_2(1/p)$  gas was isolated by liquefaction of helium-hydrogen plasma gas using an high-vacuum ( $10^{-6}$  Torr) capable, liquid nitrogen cryotrap and was characterized by mass spectroscopy (MS). The condensable gas had a higher ionization energy than  $H_2$  by MS [17].  $H_2(1/4)$  gas from chemical decomposition of hydrides containing the corresponding hydride ion  $H^-(1/4)$  as well from liquefaction of the catalysis-plasma gas was also identified by  $^1H$  NMR as an upfield-shifted singlet peak at 2.18 ppm relative to  $H_2$  at 4.63 that

matched theoretical predictions [13, 17].  $H_2(1/4)$  was further characterized by studies on the vibration-rotational emission from electron-beam maintained argon-hydrogen plasmas and from Fourier-transform infrared (FTIR) spectroscopy of solid samples containing  $H^-(1/4)$  with interstitial  $H_2(1/4)$ .

5 Water bath calorimetry was used to determine that measurable power was developed in rt-plasmas due to the reaction to form states given by Eqs. (19) and (21). Specifically,  $He/H_2(10\%)$  (500 mTorr),  $Ar/H_2(10\%)$  (500 mTorr), and  $H_2O(g)$  (500 and 200 mTorr) plasmas generated with an Evenson microwave cavity consistently yielded on the order of 50% more heat than non rt-plasma (controls)

10 such as  $He$ ,  $Kr$ ,  $Kr/H_2(10\%)$ , under identical conditions of gas flow, pressure, and microwave operating conditions. The excess power density of rt-plasmas was of the order  $10\text{ W} \cdot \text{cm}^{-3}$ . In addition to unique vacuum ultraviolet (VUV) lines, earlier studies with these same rt-plasmas demonstrated that other unusual features were present including dramatic broadening of the hydrogen Balmer series lines [3-5, 7, 9-

15 10, 12, 18-19, 21, 23-28], and in the case of water plasmas, population inversion of the hydrogen excited states [3, 21, 27-29]. Both the current results and the earlier results are completely consistent with the existence of a hitherto unknown predicted exothermic chemical reaction occurring in rt-plasmas.

Since the ionization energy of  $Sr^+$  to  $Sr^{3+}$  has a net enthalpy of reaction of

20  $2 \cdot 27.2\text{ eV}$ ,  $Sr^+$  may serve as catalyst alone or with  $Ar^+$  catalyst. It was reported previously that an rt-plasma formed with a low field (1V/cm), at low temperatures (e.g.  $\approx 10^3\text{ K}$ ), from atomic hydrogen generated at a tungsten filament and strontium which was vaporized by heating the metal [4-5, 7, 9-10]. Strong VUV emission was observed that increased with the addition of argon, but not when sodium,

magnesium, or barium replaced strontium or with hydrogen, argon, or strontium alone. Characteristic emission was observed from a continuum state of  $Ar^{2+}$  at 45.6 nm without the typical Rydberg series of Ar I and Ar II lines which confirmed the resonant nonradiative energy transfer of 27.2 eV from atomic hydrogen to  $Ar^+$  [5, 7, 22]. Predicted  $Sr^{3+}$  emission lines were also observed from strontium-hydrogen plasmas [5, 7] that supported the rt-plasma mechanism. Time-dependent line broadening of the H Balmer  $\alpha$  line was observed corresponding to extraordinarily fast H (25 eV). An excess power of  $20 \text{ mW} \cdot \text{cm}^{-3}$  was measured calorimetrically on rt-plasmas formed when  $Ar^+$  was added to  $Sr^+$  as an additional catalyst.

Significant Balmer  $\alpha$  line broadening corresponding to an average hydrogen atom temperature of 14, 24 eV, and 23-45 eV was observed for strontium and argon-strontium rt-plasmas and discharges of strontium-hydrogen, helium-hydrogen, argon-hydrogen, strontium-helium-hydrogen, and strontium-argon-hydrogen, respectively, compared to  $\approx 3 \text{ eV}$  for pure hydrogen, xenon-hydrogen, and magnesium-hydrogen. To achieve that same optically measured light output power, hydrogen-sodium, hydrogen-magnesium, and hydrogen-barium mixtures required 4000, 7000, and 6500 times the power of the hydrogen-strontium mixture, respectively, and the addition of argon increased these ratios by a factor of about two. A glow discharge plasma formed for hydrogen-strontium mixtures at an extremely low voltage of about 2 V compared to 250 V for hydrogen alone and sodium-hydrogen mixtures, and 140-150 V for hydrogen-magnesium and hydrogen-barium mixtures [4-5, 7]. These voltages are too low to be explicable by conventional mechanisms involving accelerated ions with a high applied field. A low-voltage EUV and visible light source is feasible [10].

In general, the energy transfer of  $m \cdot 27.2 \text{ eV}$  from the hydrogen atom to the catalyst causes the central-field interaction of the H atom to increase by  $m$  and its electron to drop  $m$  levels lower from the radius of the hydrogen atom,  $a_H$ , to a radius of  $\frac{a_H}{1+m}$  [19-20]. Since  $K$  to  $K^{3+}$  provides a reaction with a net enthalpy

- 5 equal to three times the potential energy of atomic hydrogen,  $3 \cdot 27.2 \text{ eV}$ , it may serve as a catalyst such that each ordinary hydrogen atom undergoing catalysis releases a net of  $204 \text{ eV}$  [3].  $K$  may then react with the product  $H(1/4)$  to form a yet lower-state  $H(1/7)$  or further catalytic transitions may occur:

$\frac{1}{4} \rightarrow \frac{1}{5}$ ,  $\frac{1}{5} \rightarrow \frac{1}{6}$ ,  $\frac{1}{6} \rightarrow \frac{1}{7}$ , and so, involving only hydrinos in a process called

- 10 *disproportionation*. Since the ionization energies and metastable resonant states of hydrinos due corresponding to the multipole expansion of the potential energy are  $m \cdot 27.2 \text{ eV}$  (Eqs. (19) and (21)) as given previously [19-20, 30] once catalysis begins, hydrinos autocatalyze further transitions to lower states. This mechanism is similar to that of an inorganic ion catalysis. An energy transfer of  $m \cdot 27.2 \text{ eV}$  from
- 15 a first hydrino atom to the second hydrino atom causes the central field of the first atom to increase by  $m$  and its electron to drop  $m$  levels lower from a radius of  $\frac{a_H}{p}$

to a radius of  $\frac{a_H}{p+m}$ .

The catalyst product,  $H(1/p)$ , may also react with an electron to form a novel hydride ion  $H^-(1/p)$  with a binding energy  $E_B$  [3, 14, 16, 21, 30]:

$$E_B = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi\mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right) \quad (27)$$

where  $p$  is an integer greater than one,  $s = 1/2$ ,  $\hbar$  is Planck's constant bar,  $\mu_0$  is the permeability of vacuum,  $m_e$  is the mass of the electron,  $\mu_e$  is the reduced

- 5 electron mass given by  $\mu_e = \frac{m_e m_p}{\frac{m_e}{\sqrt{3}} + m_p}$  where  $m_p$  is the mass of the proton,  $a_H$  is the

radius of the hydrogen atom,  $a_0$  is the Bohr radius, and  $e$  is the elementary charge.

The ionic radius is  $r_i = \frac{a_0}{p} (1 + \sqrt{s(s+1)})$ ;  $s = \frac{1}{2}$ . From Eq. (27), the calculated

ionization energy of the hydride ion is  $0.75418 \text{ eV}$ , and the experimental value given by Lykke [38] is  $6082.99 \pm 0.15 \text{ cm}^{-1}$  ( $0.75418 \text{ eV}$ ).

- 10 Substantial evidence of an energetic catalytic reaction was previously reported [3] involving a resonant energy transfer between hydrogen atoms and  $K$  to form very stable novel hydride ions  $H^-(1/p)$  called hydrino hydrides having a predicted fractional principal quantum number  $p = 4$ . Characteristic emission was observed from  $K^{3-}$  that confirmed the resonant nonradiative energy transfer of
- 15  $3 \cdot 27.2 \text{ eV}$  from atomic hydrogen to  $K$ . From Eq. (27), the binding energy  $E_B$  of  $H^-(1/4)$  is

$$E_B = 11.232 \text{ eV} \quad (\lambda_{vac} = 1103.8 \text{ Å}) \quad (28)$$

The product hydride ion  $H^-(1/4)$  was observed spectroscopically at  $110 \text{ nm}$  corresponding to its predicted binding energy of  $11.2 \text{ eV}$  [3, 21].

Upfield-shifted NMR peaks are direct evidence of the existence of lower-energy state hydrogen with a reduced radius relative to ordinary hydride ion and having an increase in diamagnetic shielding of the proton. The total theoretical shift  $\frac{\Delta B_r}{B}$  for  $H^-(1/p)$  is given by the sum of the shift of  $H^-(1/1)$  plus the contribution

5 due to the lower-electronic energy state:

$$\frac{\Delta B_r}{B} = -\mu_0 \frac{e^2}{12m_e a_0 \left(1 + \sqrt{s(s+1)}\right)} (1 + \alpha 2\pi p) = -(29.9 + 1.37p) \text{ ppm} \quad (29)$$

where  $p = \text{integer} > 1$ . Corresponding alkali hydrides and alkali hydrino hydrides (containing  $H^-(1/p)$ ) were characterized by  $^1H$  MAS NMR and compared to the  
10 theoretical values. A match of the predicted and observed peaks with no alternative represents a definite test.

The  $^1H$  MAS NMR spectrum of novel compound  $KH * Cl$  relative to external tetramethylsilane (TMS) showed a large distinct upfield resonance at -4.4 ppm corresponding to an absolute resonance shift of -35.9 ppm that matched the  
15 theoretical prediction of  $p = 4$  [3, 14-16]. This result confirmed the previous observations from the rt-plasmas of intense hydrogen Lyman emission, a stationary inverted Lyman population, excessive afterglow duration, highly energetic hydrogen atoms, characteristic alkali-ion emission due to catalysis, predicted novel spectral lines, and the measurement of a power beyond any conventional chemistry [3] that  
20 matched predictions for a catalytic reaction of atomic hydrogen to form more stable hydride ions designated  $H^-(1/p)$ . Since the comparison of theory and experimental shifts of  $KH * Cl$  is direct evidence of lower-energy hydrogen with an implicit large exotherm during its formation, the NMR results were repeated with the

further analysis by infrared (FTIR) spectroscopy to eliminate any known explanation [39].

Elemental analysis identified [14, 16] these compounds as only containing the alkaline metal, halogen, and hydrogen, and no known hydride compound of this composition could be found in the literature which has an upfield-shifted hydride NMR peak. Ordinary alkali hydrides alone or mixed with alkali halides show down-field shifted peaks [3, 14-16]. From the literature, the list of alternatives to  $H^-(1/p)$  as a possible source of the upfield NMR peaks was limited to U centered H. The intense and characteristic infrared vibration band at  $503\text{ cm}^{-1}$  due to the substitution of  $H^-$  for  $Cl^-$  in  $KCl$  enabled the elimination of U centered H as the source of the upfield-shifted NMR peaks [39].

As further characterizations, the X-ray photoelectron spectrum (XPS) of the hydrino hydride  $KH * I$  was performed to determine if the predicted  $H^-(1/4)$  binding energy given by Eq. (28) was observed, and FTIR analysis of these crystals with  $H^-(1/4)$  was performed before and after storage in argon for 90 days to search for interstitial  $H_2(1/4)$  having a predicted rotational energy given by Eq. (24). The identification of single rotational peaks at this energy with ortho-para splitting due to free rotation of a very small hydrogen molecule would represent definite proof of its existence since there is no other possible assignment [39].

Since the rotational emission of  $H_2(1/4)$  was observed in crystals of  $KH * I$  having a peak assigned to  $H^-(1/4)$  and the vibration-rotational emission of  $H_2(1/4)$  was observed from 12.5 keV-electron-beam-maintained plasmas of argon with 1% hydrogen due to collisional excitation of  $H_2(1/4)$ ,  $H_2(1/4)$  trapped in the lattice of  $KH * Cl$ , or  $H_2(1/4)$  formed from  $H^-(1/4)$  or formed insitu from  $K$

catalysis of H via electron bombardment was investigated by windowless EUV spectroscopy on electron-beam excitation of the crystals using the 12.5 keV electron gun at pressures below which any gas could produce detectable emission ( $<10^{-5}$  Torr) [39]. The rotational energy of  $H_2(1/4)$  was confirmed by this technique as

5 well. Consistent results from the broad spectrum of investigational techniques provide definitive evidence that hydrogen can exist in lower-energy states than previously thought possible in the form of  $H^-(1/4)$  and  $H_2(1/4)$ . In an embodiment, the products of the Li catalyst reaction and NaH catalyst reaction are both  $H^-(1/4)$  and  $H_2(1/4)$  and additionally  $H^-(1/3)$  and  $H_2(1/3)$  for NaH. The

10 present invention provides for their identification and the corresponding energetic exothermic reaction by EUV spectroscopy, characteristic emission from catalysts and the hydride ion products, lower-energy hydrogen emission, chemically formed plasmas, extraordinary Balmer  $\alpha$  line broadening, population inversion of H lines, elevated electron temperature, anomalous plasma afterglow duration, power

15 generation, and analysis of novel chemical compounds. Preferred identification techniques for the species  $H^-(1/p)$  and  $H_2(1/p)$  are NMR of  $H^-(1/p)$  and  $H_2(1/p)$ , FTIR of  $H_2(1/p)$  trapped in a crystal, XPS of  $H^-(1/p)$ , ToF-SIMS of  $H^-(1/p)$ , electron-beam excitation emission spectroscopy of  $H_2(1/p)$ , electron beam emission spectroscopy of  $H_2(1/p)$  trapped in a crystalline lattice, and TOF-

20 SIMS identification of novel compounds comprising  $H^-(1/p)$ . Preferred characterization techniques for the energetic catalysis reaction and the power balance are line broadening, plasma formation, and calorimetry. Preferably,  $H^-(1/p)$  and  $H_2(1/p)$  are  $H^-(1/4)$  and  $H_2(1/4)$ , respectively.

**BRIEF DESCRIPTION OF THE DRAWINGS**

FIGURE 1A is a schematic drawing of an energy reactor and power plant in accordance with the present invention.

5      FIGURE 2A is a schematic drawing of an energy reactor and power plant for recycling or regenerating the fuel in accordance with the present invention.

FIGURE 3A is a schematic drawing of a power reactor in accordance with the present invention.

FIGURE 4A is a schematic drawing of a discharge power and plasma cell and  
10      reactor in accordance with the present invention.

Figure 1 is the experimental set up comprising a filament gas cell to form lithium-argon-hydrogen and lithium-hydrogen rt-plasmas.

Figure 2 is a schematic of the reaction cell and the cross sectional view of the water flow calorimeter used to measure the energy balance of the *NaH* catalyst  
15      reaction to form hydridos. The components were: 1—inlet and outlet thermistors; 2—high-temperature valve; 3—ceramic fiber heater; 4—copper water-coolant coil; 5—reactor; 6—insulation; 7—cell thermocouple, and 8—water flow chamber.

Figure 3 is a schematic of the water flow calorimeter used to measure the energy balance of the *NaH* catalyst reaction to form hydridos.

20      Figure 4 is a schematic of the stainless steel gas cell to synthesize *LiH \* Br*, *LiH \* I*, *NaH \* Cl* and *NaH \* Br* comprising the reaction mixture (i) R-Ni, *Li*, *LiNH<sub>2</sub>*, and *LiBr* or *LiI* or (ii) Pt/Ti dissociator, *Na*, *NaH*, and *NaCl* or *NaBr* as the reactants. The components were: 101—stainless steel cell; 117—internal cavity of cell; 118—high vacuum conflat flange; 119—mating blank conflat flange;  
25      102—stainless steel tube vacuum line and gas supply line; 103—lid to the kiln or top

insulation, 104—surrounding heaters coverer by high temperature insulation;  
108—Pt/Ti dissociator; 109—reactants; 110—high vacuum turbo pump;  
112—pressure gauge; 111—vacuum pump valve; 113—valve; 114—valve;  
115—regulator, and 116—hydrogen tank.

5        Figure 5 shows the 656.3 nm Balmer  $\alpha$  line width recorded with a high-resolution visible spectrometer on (A) the initial emission of a lithium-argon-hydrogen rt-plasma and (B) the emission at 70 hours of operation. Lithium lines and significant broadening of only the  $H$  lines was observed over time corresponding to an average hydrogen atom temperature of  $>40$  eV and fractional population over 90%.

10        Figure 6 shows the 656.3 nm Balmer  $\alpha$  line width recorded with a high-resolution ( $\pm 0.006$  nm) visible spectrometer on (A) the initial emission of a lithium-hydrogen rt-plasma and (B) the emission at 70 hours of operation. Lithium lines and broadening of only the  $H$  lines was observed over time, but diminished relative to the case having the argon-hydrogen gas (95/5%). The Balmer width corresponded to an  
15        average hydrogen atom temperature of 6 eV and a 27% fractional population.

Figure 7 shows the results of the DSC (100-750 °C) of  $NaH$  at a scan rate of 0.1 degree/minute. A broad endothermic peak was observed at 350°C to 420 °C which corresponds to  $47$  kJ / mole and matches sodium hydride decomposition in this temperature range with a corresponding enthalpy of  $57$  kJ / mole . A large exotherm  
20        was observed under conditions that form  $NaH$  catalyst in the region 640°C to 825 °C which corresponds to at least  $-354$  kJ / mole  $H_2$  , greater than that of the most exothermic reaction possible for  $H$  , the  $-241.8$  kJ / mole  $H_2$  enthalpy of combustion of hydrogen.

Figure 8 shows the results of the DSC (100-750 °C) of  $MgH_2$  at a scan rate of 0.1  
25        degree/minute. Two sharp endothermic peaks were observed. A first peak centered

at 351.75 °C corresponding to 68.61 kJ / mole  $MgH_2$  matches the 74.4 kJ / mole  $MgH_2$  decomposition energy. The second peak at 647.66 °C corresponding to 6.65 kJ / mole  $MgH_2$  matches the known melting point of  $Mg(m)$  is 650 °C and enthalpy of fusion of 8.48 kJ / mole  $Mg(m)$ . Thus, the expected behavior was

5 observed for the decomposition of a control, noncatalyst hydride.

Figure 9 shows the temperature versus time for the calibration run with an evacuated test cell and resistive heating only.

Figure 10 shows the power versus time for the calibration run with an evacuated test cell and resistive heating only. The numerical integration of the input and output

10 power curves yielded an output energy of 292.2 kJ and an input energy of 303.1 kJ corresponding to a coupling of flow of 96.4% of the resistive input to the output coolant.

Figure 11 shows the cell temperature with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 1g  $Li$ , 0.5g  $LiNH_2$ , 10g

15  $LiBr$ , and 15g  $Pd / Al_2O_3$ . The reaction liberated 19.1 kJ of energy in less than 120 s to develop a system-response-corrected peak power in excess of 160 W.

Figure 12 shows the coolant power with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 1g  $Li$ , 0.5g  $LiNH_2$ , 10g  $LiBr$ , and 15g  $Pd / Al_2O_3$ . The numerical integration of the input and output power

20 curves with the calibration correction applied yielded an output energy of 227.2 kJ and an input energy of 208.1 kJ corresponding to an excess energy of 19.1 kJ.

Figure 13 shows the cell temperature with time for the R-Ni control power test with the cell containing the reagents comprising the starting material for R-Ni, 15g R-Ni/Al alloy powder, and 3.28g of Na.

Figure 14 shows the coolant power with time for the control power test with the cell containing the reagents comprising the starting material for R-Ni, 15g R-Ni/Al alloy powder, and 3.28g of Na. Energy balance was obtained with the calibration-corrected numerical integration of the input and output power curves yielding an  
5 output energy of 384 kJ and an input energy of 385 kJ.

Figure 15 shows the cell temperature with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 15g *NaOH* -doped R-Ni 2800, and 3.28g of Na. The reaction liberated 36 kJ of energy in less than 90 s to develop a system-response-corrected peak power in excess of 0.5 kW.

10 Figure 16 shows the coolant power with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 15g *NaOH* -doped R-Ni 2800, and 3.28g of Na. The numerical integration of the input and output power curves with the calibration correction applied yielded an output energy of 185.1 kJ and an input energy of 149.1 kJ corresponding to an excess energy of 36 kJ.

15 Figure 17 shows the cell temperature with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 15g *NaOH* -doped R-Ni 2400. The cell temperature jumped from 60°C to 205°C in 60 s wherein the reaction liberated 11.7 kJ of energy in less time to develop a system-response-corrected peak power in excess of 0.25 kW.

20 Figure 18 shows the coolant power with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 15g *NaOH* -doped R-Ni 2400. The numerical integration of the input and output power curves with the calibration correction applied yielded an output energy of 195.7 kJ and an input energy of 184.0 kJ corresponding to an excess energy of 11.7 kJ.

25 Figure 19 shows the positive ToF-SIMS spectrum ( $m/e = 0 - 100$ ) of *LiBr*.

Figure 20 shows the positive ToF-SIMS spectrum ( $m/e = 0 - 100$ ) of the  $LiH * Br$  crystals.

Figure 21 shows the negative ToF-SIMS spectrum ( $m/e = 0 - 100$ ) of  $LiBr$ .

Figure 22 shows the negative ToF-SIMS spectrum ( $m/e = 0 - 100$ ) of the  
 5  $LiH * Br$  crystals. A dominant hydride,  $LiHBr^-$ , and  $Li_2H_2Br^-$  peaks were uniquely observed.

Figure 23 shows the positive ToF-SIMS spectrum ( $m/e = 0 - 200$ ) of  $LiI$ .

Figure 24 shows the positive ToF-SIMS spectrum ( $m/e = 0 - 200$ ) of the  $LiH * I$   
 crystals.  $LiHI^+$ ,  $Li_2H_2I^+$ ,  $Li_4H_2I^+$ , and  $Li_6H_2I^+$  were only observed in the positive  
 10 ion spectrum of the  $LiH * I$  crystals.

Figure 25 shows the negative ToF-SIMS spectrum ( $m/e = 0 - 180$ ) of  $LiI$ .

Figure 26 shows the negative ToF-SIMS spectrum ( $m/e = 0 - 180$ ) of the  $LiH * I$   
 crystals. A dominant hydride,  $LiHI^-$ ,  $Li_2H_2I^-$ , and  $NaHI^-$  peaks were uniquely  
 observed.

15 Figure 27 shows the negative ToF-SIMS spectrum ( $m/e = 20 - 30$ ) of  $NaH * -$   
 coated  $Pt / Ti$  following the production of 15 kJ of excess heat. Hydrino hydride  
 compounds  $NaH_x^-$  were observed.

Figure 28 shows the positive ToF-SIMS spectrum ( $m/e = 0 - 100$ ) of R-Ni reacted  
 over a 48 hour period at 50°C. The dominant ion on the surface was  $Na^+$  consistent  
 20 with  $NaOH$  doping of the surface. The ions of the other major elements of R-Ni  
 2400 such as  $Al^+$ ,  $Ni^+$ ,  $Cr^+$ , and  $Fe^+$  were also observed.

Figure 29 shows the negative ToF-SIMS spectrum ( $m/e = 0 - 180$ ) of R-Ni  
 reacted over a 48 hour period at 50°C. A dominant hydride,  $NaH_3^-$  and  $NaH_3NaOH^-$   
 assigned to sodium hydrino hydride and this ion in combination with  $NaOH$ , as well

as other unique ions assignable to sodium hydrido hydrides  $NaH_x^-$  in combinations with  $NaOH$ ,  $NaO$ ,  $OH^-$  and  $O^-$  were observed.

Figures 30A-B show  $^1H$  MAS NMR spectra relative to external TMS. (A)  $LiH * Br$  showing a broad  $-2.5$  ppm upfield-shifted peak and a peak at  $1.13$  ppm assigned to  $H^-(1/4)$  and  $H_2(1/4)$ , respectively. (B)  $LiH * I$  showing a broad  $-2.09$  ppm upfield-shifted peak assigned to  $H^-(1/4)$  and peaks at  $1.06$  ppm and  $4.38$  ppm assigned to  $H_2(1/4)$  and  $H_2$ , respectively.

Figures 31A-B show  $^1H$  MAS NMR spectra relative to external TMS. (A)  $KH * Cl$  showing a very sharp  $-4.46$  ppm upfield-shifted peak corresponding to an environment that is essentially that of a free ion. (B)  $KH * I$  showing a broad  $-2.31$  ppm upfield-shifted peak similar to the case of  $LiH * Br$  and  $LiH * I$ . Both spectra also had a  $1.13$  ppm peak assigned to  $H_2(1/4)$ .

Figures 32A-B show  $^1H$  MAS NMR spectra relative to external TMS showing an H-content selectivity of  $LiH * X$  for molecular species alone based on the nonpolarizability of the halide and the corresponding nonreactivity towards  $H^-(1/4)$ . (A)  $LiH * F$  comprising a nonpolarizable fluorine showing peaks at  $4.31$  ppm assigned to  $H_2$  and  $1.16$  ppm assigned to  $H_2(1/4)$  and the absence of the  $H^-(1/4)$  ion peak. (B)  $LiH * Cl$  comprising a nonpolarizable chlorine showing peaks at  $4.28$  ppm assigned to  $H_2$  and  $1.2$  ppm assigned to  $H_2(1/4)$  and the absence of the  $H^-(1/4)$  ion peak.

Figure 33 shows the  $^1H$  MAS NMR spectra of  $NaH * Br$  relative to external TMS showing a  $-3.58$  ppm upfield-shifted peak, a peak at  $1.13$  ppm, and a peak at  $4.3$  ppm assigned to  $H^-(1/4)$ ,  $H_2(1/4)$ , and  $H_2$ , respectively.

Figures 34A-B show the  $NaH * Cl$   $^1H$  MAS NMR spectra relative to external TMS showing the effect of hydrogen addition on the relative intensities of  $H_2$ ,

$H_2(1/4)$ , and  $H^-(1/4)$ . The addition of hydrogen increased the  $H^-(1/4)$  peak and decreased the  $H_2(1/4)$  while the  $H_2$  increased. (A)  $NaH * Cl$  synthesized with

5 hydrogen addition showing a  $-4$  ppm upfield-shifted peak assigned to  $H^-(1/4)$ , a  $1.1$  ppm peak assigned to  $H_2(1/4)$ , and a dominant  $4$  ppm peak assigned to  $H_2$ .

(B)  $NaH * Cl$  synthesized without hydrogen addition showing a  $-4$  ppm upfield-shifted peak assigned to  $H^-(1/4)$ , a dominant  $1.0$  ppm peak assigned to  $H_2(1/4)$ , and a small  $4.1$  ppm assigned to  $H_2$ .

10 Figure 35 shows the  $^1H$  MAS NMR spectrum relative to external TMS of  $NaH * Cl$  from reaction of  $NaCl$  and the solid acid  $KHSO_4$  as the only source of hydrogen showing both the  $H^-(1/4)$  peak at  $-3.97$  ppm and an upfield-shifted peak at  $-3.15$  ppm assigned to  $H^-(1/3)$ . The corresponding  $H_2(1/4)$  and  $H_2(1/3)$  peaks are shown at  $1.15$  ppm and  $1.7$  ppm, respectively. Both fractional hydrogen

15 states were present and the  $H_2$  peak was absent at  $4.3$  ppm due to the synthesis of  $NaH * Cl$  using a solid acid as the  $H$  source rather than addition of hydrogen gas and a dissociator. (SB=side band).

Figures 36A-B show XPS survey spectra ( $E_b = 0$  eV to  $1200$  eV). (A)  $LiBr$ . (B)  $LiH * Br$ .

20 Figure 37 shows the  $0-85$  eV binding energy region of a high resolution XPS spectrum of  $LiH * Br$  and the control  $LiBr$  (dashed). The XPS spectrum of  $LiH * Br$  differs from that of  $LiBr$  by having additional peaks at  $9.5$  eV and  $12.3$  eV that could

not be assigned to known elements and do not correspond to any other primary element peak. The peaks match  $H^-(1/4)$  in two different chemical environments.

Figures 38A-B show the XPS survey spectra ( $E_b = 0 \text{ eV}$  to  $1200 \text{ eV}$ ). (A)  $NaBr$ . (B)  $NaH^*Br$ .

5 Figure 39 shows the 0-40 eV binding energy region of a high resolution XPS spectrum of  $NaH^*Br$  and the control  $NaBr$  (dashed). The XPS spectrum of  $NaH^*Br$  differs from that of  $NaBr$  by having additional peaks at 9.5 eV and 12.3 eV that could not be assigned to known elements and do not correspond to any other primary element peak. The peaks match  $H^-(1/4)$  in two different chemical  
10 environments.

Figures 40A-B show XPS survey spectra ( $E_b = 0 \text{ eV}$  to  $1200 \text{ eV}$ ). (A)  $Pt/Ti$ . (B)  $NaH^*$ -coated  $Pt/Ti$  following the production of 15 kJ of excess heat.

Figures 41A-B show high resolution XPS spectra ( $E_b = 0 \text{ eV}$  to  $100 \text{ eV}$ ). (A)  $Pt/Ti$ . (B)  $NaH^*$ -coated  $Pt/Ti$  following the production of 15 kJ of excess heat.

15 The  $Pt\ 4f_{7/2}$ ,  $Pt\ 4f_{5/2}$ , and  $O\ 2s$  peaks were observed at 70.7 eV, 74 eV, and 23 eV, respectively. The  $Na\ 2p$  and  $Na\ 2s$  peaks were observed at 31 eV and 64 eV on  $NaH^*$ -coated  $Pt/Ti$ , and a valance band was only observed for  $Pt/Ti$ .

Figures 42A-B show high resolution XPS spectra ( $E_b = 0 \text{ eV}$  to  $50 \text{ eV}$ ). (A)  $Pt/Ti$ . (B)  $NaH^*$ -coated  $Pt/Ti$  following the production of 15 kJ of excess heat.

20 The XPS spectrum of  $NaH^*$ -coated  $Pt/Ti$  differs from that of  $Pt/Ti$  by having additional peaks at 6 eV, 10.8 eV, and 12.8 eV that could not be assigned to known elements and do not correspond to any other primary element peak. The 10.8 eV, and 12.8 eV peaks match  $H^-(1/4)$  in two different chemical environments, and the

6 eV peak matched and was assigned to  $H^-(1/3)$ . Thus, both fractional hydrogen states, 1/3 and 1/4, were present as predicted by Eq. (27).

Figure 43 shows XPS survey spectrum ( $E_b = 0 \text{ eV}$  to  $120 \text{ eV}$ ) of  $NaH^*$ -coated  $Si$  with the primary-element peaks identified.

5 Figure 44 shows high resolution XPS spectrum ( $E_b = 0 \text{ eV}$  to  $120 \text{ eV}$ ) of  $NaH^*$ -coated  $Si$  having peaks at 6 eV, 10.8 eV, and 12.8 eV that could not be assigned to known elements and do not correspond to any other primary element peak. The 10.8 eV, and 12.8 eV peaks match  $H^-(1/4)$  in two different chemical environments, and the 6 eV peak matched and was assigned to  $H^-(1/3)$ . Thus, both fractional  
10 hydrogen states, 1/3 and 1/4, were present as predicted by Eq. (27) matching the results of  $NaH^*$ -coated  $Pt/Ti$  shown in Figure 42B.

Figures 45A-B show high resolution ( $0.5 \text{ cm}^{-1}$ ) FTIR spectra ( $490\text{--}4000 \text{ cm}^{-1}$ ). (A)  $LiBr$ . (B)  $LiH^*Br$  sample having a NMR peak assigned to  $H^-(1/4)$  that was heated to  $>600^\circ\text{C}$  under dynamic vacuum that retained the  $-2.5 \text{ ppm}$  NMR peak.  
15 The amide peaks at  $3314$ ,  $3259$ ,  $2079(\text{broad})$ ,  $1567$ , and  $1541 \text{ cm}^{-1}$  and the imide peaks at  $3172$  (broad),  $1953$ , and  $1578 \text{ cm}^{-1}$  were eliminated; thus, they were not the source of the  $-2.5 \text{ ppm}$  NMR peak that remained. The  $-2.5 \text{ ppm}$  peak in  $^1H$  NMR spectrum was assigned to the  $H^-(1/4)$  ion. In addition, the  $1989 \text{ cm}^{-1}$  FTIR peak could not be assigned to any known compound, but matched the predicted  
20 frequency of para  $H_2(1/4)$ .

Figure 46 shows the  $150\text{--}350 \text{ nm}$  spectrum of electron-beam excited  $CsCl$  crystals having trapped  $H_2(1/4)$ . A series of evenly spaced lines was observed in the  $220\text{--}300 \text{ nm}$  region that matched the spacing and intensity profile of the P branch of  $H_2(1/4)$ .

Figure 47 shows the 100-550 nm spectrum of an electron-beam excited silicon wafer coated with  $\text{NaH} \cdot \text{Cl}$  having trapped  $\text{H}_2(1/4)$ . A series of evenly spaced lines was observed in the 220-300 nm region that matched the spacing and intensity profile of the P branch of  $\text{H}_2(1/4)$

5

### **DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS**

#### **Hydrogen Catalyst Reactor**

A hydrogen catalyst reactor 50 for producing energy and lower-energy hydrogen species, in accordance with the invention, is shown in FIGURE 1A and  
10 comprises a vessel 52 which contains an energy reaction mixture 54, a heat exchanger 60, and a power converter such as a steam generator 62 and turbine 70. In an embodiment, the catalysis involves reacting atomic hydrogen from the source 56 with the catalyst 58 to form lower-energy hydrogen "hydrinos" and produce power. The heat exchanger 60 absorbs heat released by the catalysis reaction,  
15 when the reaction mixture, comprised of hydrogen and a catalyst, reacts to form lower-energy hydrogen. The heat exchanger exchanges heat with the steam generator 62 which absorbs heat from the exchanger 60 and produces steam. The energy reactor 50 further comprises a turbine 70 which receives steam from the steam generator 62 and supplies mechanical power to a power generator 80 which  
20 converts the steam energy into electrical energy, which can be received by a load 90 to produce work or for dissipation.

In an embodiment, the energy reaction mixture 54 comprises an energy releasing material 56 such as a solid fuel supplied through supply passage 42. The reaction mixture may comprise a source of hydrogen isotope atoms or a source of  
25 molecular hydrogen isotope, and a source of catalyst 58 which resonantly remove

approximately  $m \cdot 27.2 \text{ eV}$  to form lower-energy atomic hydrogen where  $m$  is an integer, preferably an integer less than 400 wherein the reaction to lower energy states of hydrogen occurs by contact of the hydrogen with the catalyst. The catalyst may be in the molten, liquid, gaseous, or solid state. The catalysis releases energy in a form such as heat and forms at least one of lower-energy hydrogen isotope atoms, molecules, hydride ions, and lower-energy hydrogen compounds. Thus, the power cell also comprises a lower-energy hydrogen chemical reactor.

The source of hydrogen can be hydrogen gas, dissociation of water including thermal dissociation, electrolysis of water, hydrogen from hydrides, or hydrogen from metal-hydrogen solutions. In another embodiment, molecular hydrogen of the energy releasing material 56 is dissociated into atomic hydrogen by a molecular hydrogen dissociating catalyst of the mixture 54. Such dissociating catalysts may also absorb hydrogen, deuterium, or tritium atoms and/or molecules and include, for example, an element, compound, alloy, or mixture of noble metals such as palladium and platinum, refractory metals such as molybdenum and tungsten, transition metals such as nickel and titanium, inner transition metals such as niobium and zirconium, and other such materials listed in the Prior Mills Publications. Preferably, the dissociator has a high surface area such as a noble metal such as Pt, Pd, Ru, Ir, Re, or Rh, or Ni on  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ , or combinations thereof.

In an embodiment, a catalyst is provided by the ionization of  $t$  electrons from an atom or ion to a continuum energy level such that the sum of the ionization energies of the  $t$  electrons is approximately  $m \cdot 27.2 \text{ eV}$  where  $t$  and  $m$  are each an integer. A catalyst may also be provided by the transfer of  $t$  electrons between participating ions. The transfer of  $t$  electrons from one ion to another ion provides a net enthalpy of reaction whereby the sum of the  $t$  ionization energies of the electron-

donating ion minus the ionization energies of  $t$  electrons of the electron-accepting ion equals approximately  $m \cdot 27.2 \text{ eV}$  where  $t$  and  $m$  are each an integer. In another preferred embodiment, the catalyst comprises  $MH$  such as  $NaH$  having an atom  $M$  bound to hydrogen, and the enthalpy of  $m \cdot 27.2 \text{ eV}$  is provided by the sum of the

5  $M - H$  bond energy and the ionization energies of the  $t$  electrons.

In a preferred embodiment, a source of catalyst comprises a catalytic material

58 supplied through catalyst supply passage 41, that typically provides a net enthalpy of approximately  $\frac{m}{2} \cdot 27.2 \text{ eV}$  plus or minus  $1 \text{ eV}$ . The catalysts include those given herein and the atoms, ions, molecules, and hydrinos described in Mills

10 Prior Publications (e.g. TABLE 4 of PCT/US90/01998 and pages 25-46, 80-108 of PCT/US94/02219) which are incorporated herein by reference. In embodiments, the catalyst may comprise at least one species selected from the group of molecules of  $AlH$ ,  $BiH$ ,  $ClH$ ,  $CoH$ ,  $GeH$ ,  $InH$ ,  $NaH$ ,  $RuH$ ,  $SbH$ ,  $SeH$ ,  $SiH$ ,  $SnH$ ,  $C_2$ ,  $N_2$ ,  $O_2$ ,  $CO_2$ ,  $NO_2$ , and  $NO_3$  and atoms or ions of  $Li$ ,  $Be$ ,  $K$ ,  $Ca$ ,  $Ti$ ,  $V$ ,  $Cr$ ,  $Mn$ ,  $Fe$ ,  $Co$ ,  $Ni$ ,  $Cu$ ,  $Zn$ ,

15  $As$ ,  $Se$ ,  $Kr$ ,  $Rb$ ,  $Sr$ ,  $Nb$ ,  $Mo$ ,  $Pd$ ,  $Sn$ ,  $Te$ ,  $Cs$ ,  $Ce$ ,  $Pr$ ,  $Sm$ ,  $Gd$ ,  $Dy$ ,  $Pb$ ,  $Pt$ ,  $Kr$ ,  $2K^+$ ,  $He^+$ ,  $Na^+$ ,  $Rb^+$ ,  $Sr^+$ ,  $Fe^{3+}$ ,  $Mo^{2+}$ ,  $Mo^{4+}$ ,  $In^{3+}$ ,  $He^+$ ,  $Ar^+$ ,  $Xe^+$ ,  $Ar^{2+}$  and  $H^+$ , and  $Ne^+$  and  $H^+$ .

#### Hydrogen Catalyst Reactor and Electrical Power System

20 In an embodiment of a power system, the heat is removed by a heat exchanger having a heat exchange medium. The heat exchanger may be a water wall and the medium may be water. The heat may be transferred directly for space and process heating. Alternatively, the heat exchanger medium such as water undergoes a phase change such as conversion to steam. This conversion may

occur in a steam generator. The steam may be used to generate electricity in a heat engine such as a steam turbine and a generator.

An embodiment of an hydrogen catalyst energy and lower-energy-hydrogen species-producing reactor 5, for recycling or regenerating the fuel in accordance with the Invention, is shown in FIGURE 2A and comprises a boiler 10 which contains a solid fuel reaction mixture 11, a hydrogen source 12, steam pipes and steam generator 13, a power converter such as a turbine 14, a water condenser 16, a water-make-up source 17, a solid-fuel recycler 18, and a hydrogen-dihydrino gas separator 19. At Step 1, the solid fuel comprising a source of catalyst and a source of hydrogen reacts to form hydrinos and lower-energy hydrogen products. At Step 2, the spent fuel is reprocessed to re-supply the boiler 10 to maintain thermal power generation. The heat generated in the boiler 10 forms steam in the pipes and steam generator 13 that is delivered to the turbine 14 that in turn generates electricity by powering a generator. At Step 3, the water is condensed by the water condensor 16. Any water loss may be made up by the water source 17 to complete the cycle to maintain thermal to electric power conversion. At Step 4, lower-energy hydrogen products such as hydrino hydride compounds and dihydrino gas may be removed, and unreacted hydrogen may be returned to the fuel recycler 18 or hydrogen source 12 to be added back to spent fuel to make-up recycled fuel. The gas products and unreacted hydrogen may be separated by hydrogen-dihydrino gas separator 19. Any product hydrino hydride compounds may be separated and removed using solid-fuel recycler 18. The processing may be performed in the boiler or externally to the boiler with the solid fuel returned. Thus, the system may further comprise at least one of gas and mass transporters to move the reactants and products to achieve the spent fuel removal, regeneration, and re-supply. Hydrogen make-up for

that spent in the formation of hydrinos is added from the source 12 during fuel reprocessing and may involve recycled, unconsumed hydrogen. The recycled fuel maintains the production of thermal power to drive the power plant to generate electricity.

5 In a preferred embodiment, the reaction mixture comprises species that can generate the reactants of atomic or molecular catalyst and atomic hydrogen that further react to form hydrinos, and the product species formed by the generation of catalyst and atomic hydrogen can be regenerated by at least the step of reacting the products with hydrogen. In an embodiment, the reactor comprises a moving bed  
10 reactor that may further comprise a fluidized-reactor section wherein the reactants are continuously supplied and side products are removed and regenerated and returned to the reactor. In an embodiment, the lower-energy hydrogen products such as hydrino hydride compounds or dihydrino molecules are collected as the reactants are regenerated. Furthermore, the hydrino hydride ions may be formed  
15 into other compounds or converted into dihydrino molecules during the regeneration of the reactants.

The power system may further comprise a catalyst condensor means to maintain the catalyst vapor pressure by a temperature control means which controls the temperature of a surface at a lower value than that of the reaction cell. The  
20 surface temperature is maintained at a desired value which provides the desired vapor pressure of the catalyst. In an embodiment, the catalyst condensor means is a tube grid in the cell. In an embodiment with a heat exchanger, the flow rate of the heat transfer medium may be controlled at a rate that maintains the condensor at the desired lower temperature than the main heat exchanger. In an embodiment, the  
25 working medium is water, and the flow rate is higher at the condensor than the water

wall such that the condensor is the lower, desired temperature. The separate streams of working media may be recombined to be transferred for space and process heating or for conversion to steam.

The present energy invention is further described in Mills Prior Publications which are incorporated herein by reference. The cells of the present invention include those described previously and further comprise the catalysts, reaction mixtures, methods, and systems disclosed herein. The electrolytic cell energy reactor, plasma electrolysis reactor, barrier electrode reactor, RF plasma reactor, pressurized gas energy reactor, gas discharge energy reactor, microwave cell energy reactor, and a combination of a glow discharge cell and a microwave and or RF plasma reactor of the present invention comprises: a source of hydrogen; one of a solid, molten, liquid, and gaseous source of catalyst; a vessel containing hydrogen and the catalyst wherein the reaction to form lower-energy hydrogen occurs by contact of the hydrogen with the catalyst or by reaction of *MH* catalyst; and a means for removing the lower-energy hydrogen product. For power conversion, each cell type may be interfaced with any of the converters of thermal energy or plasma to mechanical or electrical power described in Mills Prior Publications as well as converters known to those skilled in the Art such as a heat engine, steam or gas turbine system, Sterling engine, or thermionic or thermoelectric converter. Further plasma converters comprise the magnetic mirror magnetohydrodynamic power converter, plasmadynamic power converter, gyrotron, photon bunching microwave power converter, charge drift power, or photoelectric converter disclosed in Mills Prior Publications. In an embodiment, the cell comprises at least one cylinder of an internal combustion engine as given in Mills Prior Publications.

### Hydrogen Gas Cell and Solid Fuel Reactor

According to an embodiment of the invention, a reactor for producing hydrinos and power may take the form of a hydrogen gas cell. A gas cell hydrogen reactor of the present invention is shown in FIGURE 3A. Reactant hydrinos are provided by a  
5 catalytic reaction with catalyst. Catalysis may occur in the gas phase or in solid or liquid state.

The reactor of FIGURE 3A comprises a reaction vessel 207 having a chamber 200 capable of containing a vacuum or pressures greater than atmospheric. A source of hydrogen 221 communicating with chamber 200 delivers  
10 hydrogen to the chamber through hydrogen supply passage 242. A controller 222 is positioned to control the pressure and flow of hydrogen into the vessel through hydrogen supply passage 242. A pressure sensor 223 monitors pressure in the vessel. A vacuum pump 256 is used to evacuate the chamber through a vacuum line 257.

15 In an embodiment, the catalysis occurs in the gas phase. The catalyst may be made gaseous by maintaining the cell temperature at an elevated temperature that, in turn, determines the vapor pressure of the catalyst. The atomic and/or molecular hydrogen reactant is also maintained at a desired pressure that may be in any pressure range. In an embodiment, the pressure is less than atmospheric,  
20 preferably in the range about 10 millitorr to about 100 Torr. In another embodiment, the pressure is determined by maintaining a mixture of source of catalyst such as a metal source and the corresponding hydride such as a metal hydride in the cell maintained at the desired operating temperature.

A source of catalyst 250 for generating hydrino atoms can be placed in a  
25 catalyst reservoir 295, and gaseous catalyst can be formed by heating. The reaction

vessel 207 has a catalyst supply passage 241 for the passage of gaseous catalyst from the catalyst reservoir 295 to the reaction chamber 200. Alternatively, the catalyst may be placed in a chemically resistant open container, such as a boat, inside the reaction vessel.

- 5           The source of hydrogen can be hydrogen gas and the molecular hydrogen. Hydrogen may be dissociated into atomic hydrogen by a molecular hydrogen dissociating catalyst. Such dissociating catalysts or dissociators include, for example, Raney nickel (R-Ni), precious or noble metals, and a precious or noble metal on a support. The precious or noble metal may be Pt, Pd, Ru, Ir, and Rh, and
- 10   the support may be at least one of Ti, Nb, Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and combinations thereof. Further dissociators are Pt or Pd on carbon that may comprise a hydrogen spillover catalyst, nickel fiber mat, Pd sheet, Ti sponge, Pt or Pd electroplated on Ti or Ni sponge or mat, TiH, Pt black, and Pd black, refractory metals such as molybdenum and tungsten, transition metals such as nickel and titanium, inner transition metals
- 15   such as niobium and zirconium, and other such materials listed in the Prior Mills Publications. In a preferred embodiment, hydrogen is dissociated on Pt or Pd. The Pt or Pd may be coated on a support material such as titanium or Al<sub>2</sub>O<sub>3</sub>. In another embodiment, the dissociator is a refractory metal such as tungsten or molybdenum, and the dissociating material may be maintained at elevated temperature by
- 20   temperature control means 230, which may take the form of a heating coil as shown in cross section in FIGURE 3A. The heating coil is powered by a power supply 225. Preferably, the dissociating material is maintained at the operating temperature of the cell. The dissociator may further be operated at a temperature above the cell temperature to more effectively dissociate, and the elevated temperature may

prevent the catalyst from condensing on the dissociator. Hydrogen dissociator can also be provided by a hot filament such as 280 powered by supply 285.

In an embodiment, the hydrogen dissociation occurs such that the dissociated hydrogen atoms contact gaseous catalyst to produce hydrino atoms. The catalyst vapor pressure is maintained at the desired pressure by controlling the temperature of the catalyst reservoir 295 with a catalyst reservoir heater 298 powered by a power supply 272. When the catalyst is contained in a boat inside the reactor, the catalyst vapor pressure is maintained at the desired value by controlling the temperature of the catalyst boat, by adjusting the boat's power supply. The cell temperature can be controlled at the desired operating temperature by the heating coil 230 that is powered by power supply 225. The cell (called a permeation cell) may further comprise an inner reaction chamber 200 and an outer hydrogen reservoir 290 such that hydrogen may be supplied to the cell by diffusion of hydrogen through the wall 291 separating the two chambers. The temperature of the wall may be controlled with a heater to control the rate of diffusion. The rate of diffusion may be further controlled by controlling the hydrogen pressure in the hydrogen reservoir.

To maintain the catalyst pressure at the desire level, the cell having permeation as the hydrogen source may be sealed. Alternatively, the cell further comprises high temperature valves at each inlet or outlet such that the valve contacting the reaction gas mixture is maintained at the desired temperature. The cell may further comprise a getter or trap 255 to selectively collect the lower-energy-hydrogen species and/or the increased-binding-energy hydrogen compounds and may further comprise a selective valve 206 for releasing dihydrino gas product.

The catalyst may be at least one of the group of atomic lithium, potassium, or cesium, NaH molecule and hydrino atoms wherein catalysis comprises a

disproportionation reaction. Lithium catalyst may be made gaseous by maintaining the cell temperature in the 500-1000 °C range. Preferably, the cell is maintained in the 500-750 °C range. The cell pressure may be maintained at less than atmospheric, preferably in the range about 10 millitorr to about 100 Torr. Most preferably, at least one of the catalyst and hydrogen pressure is determined by maintaining a mixture of catalyst metal and the corresponding hydride such as lithium and lithium hydride, potassium and potassium hydride, sodium and sodium hydride, and cesium and cesium hydride in the cell maintained at the desired operating temperature. The catalyst in the gas phase may comprise lithium atoms from the metal or a source of lithium metal. Preferably, the lithium catalyst is maintained at the pressure determined by a mixture of lithium metal and lithium hydride at the operating temperature range of 500-1000 °C and most preferably, the pressure with the cell at the operating temperature range of 500-750 °C. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

In an embodiment of the gas cell reactor comprising a catalyst reservoir or boat, gaseous Na, NaH catalyst, or the gaseous catalyst such as Li, K, and Cs vapor is maintained in a super-heated condition in the cell relative to the vapor in the reservoir or boat which is the source of the cell vapor. In one embodiment, the superheated vapor reduces the condensation of catalyst on the hydrogen dissociator or the dissociator of at least one of metal and metal hydride molecules disclosed *infra*. In an embodiment comprising Li as the catalyst from a reservoir or boat, the reservoir or boat is maintained at a temperature at which Li vaporizes. H<sub>2</sub> may be maintained at a pressure that is lower than that which forms a significant mole fraction of LiH at the reservoir temperature. The pressures and temperatures that

achieve this condition can be determined from the data plots of Mueller et al. such as Figure 6.1 [40] of  $H_2$  pressure versus LiH mole fraction at given isotherms. In an embodiment, the cell reaction chamber containing a dissociator is operated at a higher temperature such that the Li does not condense on the walls or the

5 dissociator. The  $H_2$  may flow from the reservoir to the cell to increase the catalyst transport rate. Flow such as from the catalyst reservoir to the cell and then out of the cell is a means to remove hydride product to prevent hydride product inhibition of the reaction. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

10 Hydrogen is supplied to the reaction from a source of hydrogen. Preferably the hydrogen is supplied by permeation from a hydrogen reservoir. The pressure of the hydrogen reservoir may be in the range of 10 Torr to 10,000 Torr, preferably 100 Torr to 1000 Torr, and most preferably about atmospheric pressure. The cell may be operated in the temperature of about 100 °C to 3000 °C, preferably in the  
15 temperature of about 100 °C to 1500 °C, and most preferably in the temperature of about 500 °C to 800 °C.

The source of hydrogen may be from decomposition of an added hydride. A cell design that supplies  $H_2$  by permeation is one comprising an internal metal hydride placed in a sealed vessel wherein atomic H permeates out at high  
20 temperature. The vessel may comprise Pd, Ni, Ti, or Nb. In an embodiment, the hydride is placed in a sealed tube such as a Nb tube containing a hydride and sealed at both ends with seals such as Swagelocks. In the sealed case, the hydride could be an alkaline or alkaline earth hydride. Or, in this as well as the internal-hydride-reagent case, the hydride could be at least one of the group of saline  
25 hydrides, titanium hydride, vanadium, niobium, and tantalum hydrides, zirconium and

hafnium hydrides, rare earth hydrides, yttrium and scandium hydrides, transition element hydrides, intermetallic hydrides, and their alloys given by W. M. Mueller et al. [40].

In an embodiment the hydride and operating temperature  $\pm 200$  °C, based on  
5 each hydride decomposition temperature is at least one of the list of:

a rare earth hydride with an operating temperature of about 800 °C;  
lanthanum hydride with an operating temperature of about 700 °C; gadolinium  
hydride with an operating temperature of about 750 °C; neodymium hydride with an  
operating temperature of about 750 °C; yttrium hydride with an operating  
10 temperature of about 800 °C; scandium hydride with an operating temperature of  
about 800 °C; ytterbium hydride with an operating temperature of about 850-900 °C;  
titanium hydride with an operating temperature of about 450 °C; cerium hydride with  
an operating temperature of about 950 °C; praseodymium hydride with an operating  
temperature of about 700 °C; zirconium-titanium (50%/50%) hydride with an  
15 operating temperature of about 600 °C; an alkali metal/alkali metal hydride mixture  
such as Rb/RbH or K/KH with an operating temperature of about 450 °C, and an  
alkaline earth metal/alkaline earth hydride mixture such as Ba/BaH<sub>2</sub> with an  
operating temperature of about 900-1000 °C.

Metals in the gas state comprise diatomic covalent molecules. An objective of  
20 the present invention is to provide atomic catalyst such as Li as well as K and Cs.  
Thus, the reactor may further comprise a dissociator of at least one of metal  
molecules ("MM") and metal hydride molecules ("MH"). Preferably, the source of  
catalyst, the source of H<sub>2</sub>, and the dissociator of MM, MH, and HH, wherein M is the  
atomic catalyst are matched to operate at the desired cell conditions of temperature  
25 and reactant concentrations for example. In the case that a hydride source of H<sub>2</sub> is

used, in an embodiment, its decomposition temperature is in the range of the temperature that produces the desired vapor pressure of the catalyst. In the case of that the source of hydrogen is permeation from a hydrogen reservoir to the reaction chamber, preferable sources of catalysts for continuous operation are Sr and Li  
5 metals since each of their vapor pressures may be in the desired range of 0.01 to 100 Torr at the temperatures for which permeation occurs. In other embodiments of the permeation cell, the cell is operated at a high temperature permissive of permeation, then the cell temperature is lowered to a temperature which maintains the vapor pressure of the volatile catalyst at the desired pressure.

10 In an embodiment of a gas cell, a dissociator comprises a means to generate catalyst and H from sources. Surface catalysts such as Pt on Ti or Pd, iridium, or rhodium alone or on a substrate such as Ti may also serve the role as a dissociator of molecules of combinations of catalyst and hydrogen atoms. Preferably, the dissociator has a high surface area such as Pt/Al<sub>2</sub>O<sub>3</sub> or Pd/Al<sub>2</sub>O<sub>3</sub>.

15 The H<sub>2</sub> source can also be H<sub>2</sub> gas. In this case, the pressure can be monitored and controlled. This is possible with catalyst and catalyst sources such as K or Cs metal and LiNH<sub>2</sub>, respectively, since they are volatile at low temperature which is permissive of using a high-temperature valve. LiNH<sub>2</sub> also lowers the necessary operating temperature of the Li cell and is less corrosive which is  
20 permissive of long-duration operation using a feed through in the case of plasma and filament cells wherein a filament serves as a hydrogen dissociator.

Further embodiments of the gas cell hydrogen reactor having NaH as the catalyst comprise a filament with a dissociator in the reactor cell and Na in the reservoir. H<sub>2</sub> may be flowed through the reservoir to main chamber. The power may  
25 be controlled by controlling the gas flow rate, H<sub>2</sub> pressure, and Na vapor pressure.

The latter may be controlled by controlling the reservoir temperature. In another embodiment, the hydrino reaction is initiated by heating with the external heater and an atomic H is provided by a dissociator.

The invention is also directed to other reactors for producing increased  
5 binding energy hydrogen compounds of the invention, such as dihydrino molecules and hydrino hydride compounds. A further products of the catalysis is plasma, light, and power. Such a reactor is hereinafter referred to as a "hydrogen reactor" or "hydrogen cell". The hydrogen reactor comprises a cell for making hydrinos. The cell for making hydrinos may take the form of a gas cell, a gas discharge cell, a  
10 plasma torch cell, or microwave power cell, for example. These exemplary cells which are not meant to be exhaustive are disclosed in Mills Prior Publications and are incorporated by reference. Each of these cells comprises: a source of atomic hydrogen; at least one of a solid, molten, liquid, or gaseous catalyst for making hydrinos; and a vessel for reacting hydrogen and the catalyst for making hydrinos.  
15 As used herein and as contemplated by the subject invention, the term "hydrogen", unless specified otherwise, includes not only proteum ( $^1H$ ), but also deuterium ( $^2H$ ) and tritium ( $^3H$ ).

#### Hydrogen Gas Discharge Power and Plasma Cell and Reactor

20 A hydrogen gas discharge power and plasma cell and reactor of the present invention is shown in FIGURE 4A. The hydrogen gas discharge power and plasma cell and reactor of FIGURE 4A, includes a gas discharge cell 307 comprising a hydrogen gas-filled glow discharge vacuum vessel 315 having a chamber 300. A hydrogen source 322 supplies hydrogen to the chamber 300 through control valve  
25 325 via a hydrogen supply passage 342. A catalyst is contained in the cell chamber

300. A voltage and current source 330 causes current to pass between a cathode 305 and an anode 320. The current may be reversible.

In an embodiment, the material of cathode 305 may be a source of catalyst such as Fe, Dy, Be, or Pd. In another embodiment of the hydrogen gas discharge power and plasma cell and reactor, the wall of vessel 313 is conducting and serves as the cathode which replaces electrode 305, and the anode 320 may be hollow such as a stainless steel hollow anode. The discharge may vaporize the catalyst source to catalyst. Molecular hydrogen may be dissociated by the discharge to form hydrogen atoms for generation of hydrinos and energy. Additional dissociation may be provided by a hydrogen dissociator in the chamber.

Another embodiment of the hydrogen gas discharge power and plasma cell and reactor where catalysis occurs in the gas phase utilizes a controllable gaseous catalyst. The gaseous hydrogen atoms for conversion to hydrinos are provided by a discharge of molecular hydrogen gas. The gas discharge cell 307 has a catalyst supply passage 341 for the passage of the gaseous catalyst 350 from catalyst reservoir 395 to the reaction chamber 300. The catalyst reservoir 395 is heated by a catalyst reservoir heater 392 having a power supply 372 to provide the gaseous catalyst to the reaction chamber 300. The catalyst vapor pressure is controlled by controlling the temperature of the catalyst reservoir 395, by adjusting the heater 392 by means of its power supply 372. The reactor further comprises a selective venting valve 301. A chemically resistant open container, such as a stainless steel, tungsten or ceramic boat, positioned inside the gas discharge cell may contain the catalyst. The catalyst in the catalyst boat may be heated with a boat heater using an associated power supply to provide the gaseous catalyst to the reaction chamber. Alternatively, the glow gas discharge cell is operated at an elevated temperature

such that the catalyst in the boat is sublimed, boiled, or volatilized into the gas phase. The catalyst vapor pressure is controlled by controlling the temperature of the boat or the discharge cell by adjusting the heater with its power supply. To prevent the catalyst from condensing in the cell, the temperature is maintained

5 above the temperature of the catalyst source, catalyst reservoir 395 or catalyst boat.

In a preferred embodiment, the catalysis occurs in the gas phase, lithium is the catalyst, and a source of atomic lithium such as lithium metal or a lithium compound such as  $\text{LiNH}_2$  is made gaseous by maintaining the cell temperature in the range of about 300-1000 °C. Most preferably, the cell is maintained in the range

10 of about 500-750 °C. The atomic and/or molecular hydrogen reactant may be maintained at a pressure less than atmospheric, preferably in the range of about 10 millitorr to about 100 Torr. Most preferably, the pressure is determined by maintaining a mixture of lithium metal and lithium hydride in the cell maintained at the desired operating temperature. The operating temperature range is preferably in

15 the range of about 300-1000 °C and most preferably, the pressure is that achieved with the cell at the operating temperature range of about 300-750 °C. The cell can be controlled at the desired operating temperature by the heating coil such as 380 of FIGURE 4A that is powered by power supply 385. The cell may further comprise an inner reaction chamber 300 and an outer hydrogen reservoir 390 such that hydrogen

20 may be supplied to the cell by diffusion of hydrogen through the wall 313 separating the two chambers. The temperature of the wall may be controlled with a heater to control the rate of diffusion. The rate of diffusion may be further controlled by controlling the hydrogen pressure in the hydrogen reservoir.

An embodiment of the plasma cell of the present invention regenerates the

25 reactants such as Li and  $\text{LiNH}_2$ . In an embodiment, the reaction given by Eqs. (32)

and (37) occurs to generate the hydrino reactants Li and H with a large excess of energy released due to hydrino production. The products are then hydrogenated by a hydrogen source. In the case that LiH is formed, one reaction to regenerate the lower-energy-hydrogen-catalysis reactants is given by Eq. (66). This may be

5 achieved with the reactants placed in a reactive region in the plasma cell such as at the cathode region in a hydrogen plasma cell. The reaction may be



and then the reaction



10 may occur to some extent to maintain a steady-state level of Li + LiNH<sub>2</sub>. The H<sub>2</sub> pressure, electron density, and energy may be controlled to achieve the maximum or desired extent of the reaction to regenerate hydrino reactants Li + LiNH<sub>2</sub>.

In an embodiment, the mixture is stirred or mixed during the plasma reaction.

In a further embodiment of the plasma regeneration system and method of the

15 present invention, the cell comprises a heated flat-bottom stainless steel plasma chamber. LiH and Li<sub>2</sub>NH comprise a mixture in molten Li. Since stainless steel is not magnetic, the liquid mixture may be stirred with a stainless-steel-coated stirring bar driven by a stirring motor upon which the flat-bottom plasma reactor sits. The Li-metal mixture may serve as a cathode. The reduction of LiH to Li and H<sup>-</sup> and the

20 further reaction of H<sup>-</sup> + Li<sub>2</sub>NH to Li and LiNH<sub>2</sub> can be monitored by XRD and FTIR of the product.

In another embodiment of a system having a reaction mixture comprising species of the group of Li, LiNH<sub>2</sub>, Li<sub>2</sub>NH, Li<sub>3</sub>N, LiNO<sub>3</sub>, LiX, NH<sub>4</sub>X (X is a halide), NH<sub>3</sub>, and H<sub>2</sub>, at least one of the reactants is regenerated by adding one or more of the

25 reagents and by a plasma regeneration. The plasma may be one of the gases such

as  $\text{NH}_3$  and  $\text{H}_2$ . The plasma may be maintained in situ (in the reaction cell) or in an external cell in communication with the reaction cell. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

To maintain the catalyst pressure at the desire level, the cell having  
5 permeation as the hydrogen source may be sealed. Alternatively, the cell further comprises high temperature valves at each inlet or outlet such that the valve contacting the reaction gas mixture is maintained at the desired temperature.

The plasma cell temperature can be controlled independently over a broad range by insulating the cell and by applying supplemental heater power with heater  
10 380. Thus, the catalyst vapor pressure can be controlled independently of the plasma power.

The discharge voltage may be in the range of about 100 to 10,000 volts. The current may be in any desired range at the desired voltage. Furthermore, the plasma may be pulsed as disclosed in Mills Prior Publications such as  
15 PCT/US04/10608 entitled "Pulsed Plasma Power Cell and Novel Spectral Lines" which is herein incorporated by reference in its entirety.

Boron nitride may comprise the feed-throughs of the plasma cell since this material is stable to Li vapor. Crystalline or transparent alumina are other stable feed-through materials of the present invention.

20

#### Solid Fuels and Hydrogen Catalyst Reactor

Metals in the gas state comprise diatomic covalent molecules. An objective of the present Invention is to provide atomic catalyst such as Li as well as K and Cs and molecular catalyst NaH. Thus, in a solid-fuels embodiment, the reactants  
25 comprise alloys, complexes, or sources of complexes that reversibly form with a

metal catalyst M and decompose or react to provide gaseous catalyst such as Li. In another embodiment, at least one of the catalyst source and atomic hydrogen source further comprises at least one reactant which reacts to form at least one of the catalyst and atomic hydrogen. In an embodiment, the source or sources comprise at least one of amides such as  $\text{LiNH}_2$ , imides such as  $\text{Li}_2\text{NH}$ , nitrides such as  $\text{Li}_3\text{N}$ , and catalyst metal with  $\text{NH}_3$ . Reactions of these species provide both Li atoms and atomic hydrogen. These and other embodiments are given *infra.*, wherein, additionally, K, Cs, and Na may replace Li and the catalyst is atomic K, atomic Cs, and molecular NaH.

10       The present invention comprises an energy reactor comprising a reaction vessel constructed and arranged to contain pressures lower, equal to, and higher than atmospheric pressure, a source of atomic hydrogen for chemically producing atomic hydrogen in communication with the vessel, a source of catalyst comprising at least one of atomic lithium, atomic cesium, atomic potassium, and molecular NaH  
15   in communication with the vessel, and may further comprise a getter such as source of an ionic compound for binding or reacting with a lower-energy hydride. The source of catalyst and reactant atomic hydrogen may comprise a solid fuel that may be continuously or batch-wise regenerated inside or outside of the cell wherein a physical process or chemical reaction generates the catalyst and H from a source  
20   such that H catalysis occurs and hydrinos are formed. Thus, embodiments of the present invention of hydrino reactants comprise solid fuels, and preferable embodiments comprise those solid fuels that can be regenerated. Solid fuels can be used in many applications ranging from space and process heating, electricity generation, motive applications, propellants, and others applicants well known to  
25   those skilled in the Art.

A gas cell or plasma cell of the present invention such as those shown in FIGURES 3A and 4A comprises a means for the formation of catalyst and H atoms from sources. In solid-fuels embodiments, the cell further comprises reactants to provide catalyst and H upon initiation of a chemical or physical process. The  
5 initiation may be by means such as heating or plasma reaction. Preferably the external power requirement to maintain the production of hydrinos is low or zero based on the large power of the H catalysis reaction to form hydrinos. With a large energy gain, the reactants can be regenerated with a net release of energy for each cycle of reaction and regeneration.

10 In other embodiments, the reactor shown in FIGURE 3A comprises a solid-fuels reactor wherein a reaction mixture comprises a source of catalyst and a source of hydrogen. The reaction mixture can be regenerated by supplying a flow of reactants and by removing products from the corresponding product mixture. In an embodiment, the reaction vessel 207 has a chamber 200 capable of containing a  
15 vacuum or pressures equal to or greater than atmospheric. At least one source of reagent such as a gaseous reagent 221 is in communication with chamber 200 and delivers reagent to the chamber through at least one reagent supply passage 242. A controller 222 is positioned to control the pressure and flow of reagent into the vessel through reagent supply passage 242. A pressure sensor 223 monitors  
20 pressure in the vessel. A vacuum pump 256 is used to evacuate the chamber through a vacuum line 257. Alternatively, line 257 represents at least one output path such as a product passage line to remove material from the reactor. The reactor further comprises a source of heat such as a heater 230 to bring the reactants up to a desired temperature that initiates the solids fuel chemistry and the  
25 hydrino-forming catalysis reaction. In an embodiment, the temperature is in the

range of about 50 to 1000 °C; preferably it is in the range of about 100-600 °C, and for reactants comprising at least the Li/N-alloy system, the desired temperature is in the range of about 100-500 °C.

The cell may further comprise a source of hydrogen gas and dissociator to  
5 form atomic hydrogen. The vessel may further comprise a source of hydrogen 221  
in communication with the vessel for regenerating at least one of the source of  
atomic catalyst such as atomic lithium and the source of atomic hydrogen. The  
hydrogen source may be hydrogen gas. The H<sub>2</sub> gas may be supplied by a hydrogen  
line 242 or by permeation from a hydrogen reservoir 290. In exemplary regeneration  
10 reactions, the source of atomic lithium and atomic hydrogen may be generated by  
hydrogen addition according to Eqs. (66-71). The first step of an alternative  
regeneration reaction may given by Eq. (69).

In an embodiment, the cell size and materials are such that a high operating  
temperature is archived. The cell may be appropriately sized to the power output to  
15 achieve the desired operating temperature. High-temperature materials for the cell  
construction are niobium and a high-temperature stainless steel such as Hastalloy.  
The source of H<sub>2</sub> may be an internal metal hydride that does not react with LiNH<sub>2</sub>,  
but releases H only at very high temperature. Also, even in the cases that the  
hydride does react with LiNH<sub>2</sub>, it can be separated from the reagents such as Li and  
20 LiNH<sub>2</sub> by placing it in an open or closed vessel in the cell. A cell design that supplies  
H<sub>2</sub> by permeation is one comprising an internal metal hydride placed in a sealed  
vessel wherein atomic H permeates out at high temperature.

The reactor may further comprise means to separate components of a  
product mixture such as sieves for mechanically separating by differences in  
25 physical properties such as size. The reactor may further comprise means to

separate one or more components based on a differential phase change or reaction.

In an embodiment, the phase change comprises melting using a heater, and the liquid is separated from the solid by means known in the Art such as gravity filtration, filtration using a pressurized gas assist, and centrifugation. The reaction may

- 5   comprise decomposition such as hydride decomposition or reaction to form a hydride, and the separations may be achieved by melting the corresponding metal followed by its separation and by mechanically separating the hydride, respectively. The latter may be achieved by sieving. In an embodiment, the phase change or reaction may produce a desired reactant or intermediate. In embodiments, the
- 10   regeneration including any desired separation steps may occur inside or outside of the reactor.

#### Chemical Reactor

- A chemical reactor of the present invention further comprises a source of
- 15   inorganic compound such as MX wherein M is an alkali metal and X is a halide. Additionally to halides, the inorganic compound may be an alkali or alkaline earth salt such as hydroxide, oxide, carbonate, sulfate, phosphate, borate, and silicate (other suitable inorganic compounds are given in D. R. Lide, *CRC Handbook of Chemistry and Physics*, 86th Edition, CRC Press, Taylor & Francis, Boca Raton,
- 20   (2005-6), pp. 4-45 to 4-97 which is herein incorporated by reference). The inorganic compound may further serve as a getter in the generation of power by preventing product accumulation and a consequent back reaction or other product inhibition. A preferred Li chemical-type power cell comprises Li, LiNH<sub>2</sub>, LiBr or LiI, and R-Ni in a hydrogen cell run at about 760 Torr H<sub>2</sub> and about 700+ °C. A preferred NaH
- 25   chemical-type power cell comprises Na, NaX (X is a halide, preferably Br or I) and R-

Ni in a hydrogen cell run at about 760 Torr  $H_2$  and about 700+ °C. The cell may further comprise at least one of NaH and  $NaNH_2$ . A preferred K chemical-type power cell comprises K, KI, and Ni screen or R-Ni dissociator in a hydrogen cell run at about 760 Torr  $H_2$  and about 700+ °C. In an embodiment, the  $H_2$  pressure range is about 1 Torr to  $10^5$  Torr. Preferably, the H pressure is maintained in the range of about 760-1000 Torr. LiHX such as LiHBr and LiHI is typically synthesized in the temperature range of about 450-550 °C, but can be run at lower temp (~350 °C) with LiH present. NaHX such as NaHBr and NaHI is typically synthesized in the temperature range of about 450-550 °C. KHX such as KHI is preferably synthesized in the temperature range of about 450-550 °C. In embodiments of the NaHX and KHX reactors, NaH and K are supplied from a source such as catalyst reservoir wherein the cell temperature is maintained at a higher level than that of the catalyst reservoir. Preferably, the cell is maintained at the temperature range of about 300-550 °C and the reservoir is maintained in a temperature range of about 50 to 200 °C lower.

Another embodiment of the hydrogen reactor having NaH as the catalyst comprises a plasma torch for the production of power and increased-binding-energy hydrogen compounds such as NaHX wherein H is increased-binding-energy hydrogen and X is a halide. At least one of NaF, NaCl, NaBr, NaI may be aerosolized in the plasma gas such as  $H_2$  or a noble gas/hydrogen mixture such as He/ $H_2$  or Ar/ $H_2$ .

#### General Solid Fuels Chemistry

A reaction mixture of the present invention comprises a catalyst or a source of catalyst and atomic hydrogen or a source of atomic hydrogen (H) wherein at least

one of the catalyst and atomic hydrogen is released by a chemical reaction of at least one species of the reaction mixture or between two or more reaction-mixture species. Preferably, the reaction is reversible. Preferably, the energy released is greater than the enthalpy of reaction of the formation of catalyst and reactant hydrogen, and in the case that the reactants of the reaction mixture are regenerated and recycled, preferably, net energy is given off over the cycle of reaction and regeneration due to the large energy of formation of product H states given by Eq. (1). The species may be at least one of an element, alloy, or a compound such as a molecular or inorganic compound wherein each may be at least one of a reagent or product in the reactor. In an embodiment, the species may form an alloy or compound such as a molecular or inorganic compound with at least one of hydrogen and the catalyst. One or more of the reaction-mixture species may form one or more reaction product species such that the energy to release H or free catalyst is lowered relative to the case in the absence of the formation of the reaction product species.

In embodiments of the reactants to provide a catalyst and atomic hydrogen to form states with energy levels given by Eq. (1), the reactants comprise at least one of solid, liquid (including molten), and gaseous reactants. The reactions to form the catalyst and atomic hydrogen to form states with energy levels given by Eq. (1) occurs in one or more of the solid, liquid (including molten), and gaseous phase.

Exemplary solid-fuels reactions are given herein that are certainly not meant to be limiting in that other reactions comprising additional reagents are within the scope of the Invention.

In an embodiment, the reaction product species is an alloy or compound of at least one of the catalyst and hydrogen or sources thereof. In an embodiment, the reaction-mixture species is a catalyst hydride and the reaction product species is a

catalyst alloy or compound that has a lower hydrogen content. The energy to release H from a hydride of the catalyst may be lowered by the formation of an alloy or second compound with the at least one another species such as an element or first compound. In an embodiment, the catalyst is one of Li, K, Cs, and NaH molecule and the hydride is one of LiH, KH, CsH, NaH(s) and the at least one other element is selected from the group of M (catalyst), Al, B, Si, C, N, Sn, Te, P, S, Ni, Ta, Pt, and Pd. The first and the second compound may be one of the group of H<sub>2</sub>, H<sub>2</sub>O, NH<sub>3</sub>, NH<sub>4</sub>X, (X is a couterion such as halide (other anions are given in D. R. Lide, *CRC Handbook of Chemistry and Physics*, 86th Edition, CRC Press, Taylor & Francis, Boca Raton, (2005-6), pp. 4-45 to 4-97 which is herein incorporated by reference) MX, MNO<sub>3</sub>, MAIH<sub>4</sub>, M<sub>3</sub>AlH<sub>6</sub>, MBH<sub>4</sub>, M<sub>3</sub>N, M<sub>2</sub>NH, and MNH<sub>2</sub> wherein M is an alkali metal that may be the catalyst. In another embodiment, a hydride comprising at least one other element than the catalyst element releases H by reversible decomposition.

One or more of the reaction-mixture species may form one or more reaction product species such that the energy to release free catalyst is lowered relative to the case in the absence of the formation of the reaction product species. A reaction species such as an alloy or compound may release free catalyst by a reversible reaction or decomposition. Also, the free catalyst may be formed by a reversible reaction of a source of catalyst with at least one other species such as an element or first compound to form a species such as an alloy or second compound. The element or alloy may comprise at least one of M (catalyst atom), H, Al, B, Si, C, N, Sn, Te, P, S, Ni, Ta, Pt, and Pd. The first and the second compound may be one of the group of H<sub>2</sub>, NH<sub>3</sub>, NH<sub>4</sub>X where X is a couterion such as halide, MMX, MNO<sub>3</sub>, MAIH<sub>4</sub>, M<sub>3</sub>AlH<sub>6</sub>, MBH<sub>4</sub>, M<sub>3</sub>N, M<sub>2</sub>NH, and MNH<sub>2</sub>, wherein M is an alkali metal that may

be the catalyst. The catalyst may be one of Li, K, and Cs, and NaH molecule. The source of catalyst may be M-M such as LiLi, KK, CsCs, and NaNa. The source of H may be MH such as LiH, KH, CsH, or NaH(s).

Li catalyst may be alloyed or react to form a compound with at least one other  
5 element or compound such that the energy barrier for the release of H from LiH or Li from LiH and LiLi molecules is lowered. The alloy or compound may also release H or Li by decomposition or reaction with further reaction species. The alloy or compound may be one or more of  $\text{LiAlH}_4$ ,  $\text{Li}_3\text{AlH}_6$ ,  $\text{LiBH}_4$ ,  $\text{Li}_3\text{N}$ ,  $\text{Li}_2\text{NH}$ ,  $\text{LiNH}_2$ ,  $\text{LiX}$ , and  $\text{LiNO}_3$ . The alloy or a compound may be one or more of Li/Ni, Li/Ta, Li/Pd,  
10 Li/Te, Li/C, Li/Si, and Li/Sn wherein the stoichiometry of Li and any other element of the alloy or compound is varied to achieve the optimal release of Li and H which subsequently react during the catalysis reaction to form lower energy states of hydrogen. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

15 In an embodiment, the alloy or compound has the formula  $\text{M}_x\text{E}_y$  wherein M is the catalyst such as Li, K, or Cs, or it is Na, E is the other element, and x and y designate the stoichiometry. M and  $\text{E}_y$  may be in any desired molar ratio. In an embodiment x is in the range of 1 to 50 and y is in the range of 1 to 50, and preferably x is in the range of 1 to 10 and y is in the range of 1 to 10.

20 In another embodiment, the alloy or compound has the formula  $\text{M}_x\text{E}_y\text{E}_z$  wherein M is the catalyst such as Li, K, or Cs, or it is Na,  $\text{E}_y$  is a first other element,  $\text{E}_z$  is a second other element, and x, y, and z designate the stoichiometry. M,  $\text{E}_y$ , and  $\text{E}_z$  may be in any desired molar ratio. In an embodiment, x is in the range of 1 to 50, y is in the range of 1 to 50, and z is in the range of 1 to 50, and preferably x is in  
25 the range of 1 to 10, y is in the range of 1 to 10, and z is in the range of 1 to 10. In

preferred embodiments,  $E_y$  and  $E_z$  are selected from the group of H, N, C, Si, and Sn. The alloy or compound may be at least one of  $Li_xC_ySi_z$ ,  $Li_xSn_ySi_z$ ,  $Li_xN_ySi_z$ ,  $Li_xSn_yC_z$ ,  $Li_xN_ySn_z$ ,  $Li_xC_yN_z$ ,  $Li_xC_yH_z$ ,  $Li_xSn_yH_z$ ,  $Li_xN_yH_z$ , and  $Li_xSi_yH_z$ . In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

In another embodiment, the alloy or compound has the formula  $M_xE_wE_yE_z$  wherein M is the catalyst such as Li, K, or Cs, or it is Na,  $E_w$  is a first other element,  $E_y$  is a second other element,  $E_z$  is a third other element, and x, w, y, and z designate the stoichiometry. M,  $E_w$ ,  $E_y$ , and  $E_z$  may be in any desired molar ratio. In an embodiment, x is in the range of 1 to 50, w is in the range of 1 to 50, y is in the range of 1 to 50, and z is in the range of 1 to 50, and preferably x is in the range of 1 to 10, w is in the range of 1 to 10, y is in the range of 1 to 10, and z is in the range of 1 to 10. In preferred embodiments,  $E_w$ ,  $E_y$ , and  $E_z$  are selected from the group of H, N, C, Si, and Sn. The alloy or compound may be at least one of  $Li_xH_wC_ySi_z$ ,  $Li_xH_wSn_ySi_z$ ,  $Li_xH_wN_ySi_z$ ,  $Li_xH_wSn_yC_z$ ,  $Li_xH_wN_ySn_z$ , and  $Li_xH_wC_yN_z$ . In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH. Species such as  $M_xE_wE_yE_z$  are exemplary and are certainly not meant to be limiting in that other species comprising additional elements are within the scope of the invention.

In an embodiment, the reaction contains a source of atomic hydrogen and a source of Li catalyst. The reaction contains one or more species from the group of a hydrogen dissociator,  $H_2$ , a source of atomic hydrogen, Li, LiH,  $LiNO_3$ ,  $LiNH_2$ ,  $Li_2NH$ ,  $Li_3N$ , LiX,  $NH_3$ ,  $LiBH_4$ ,  $LiAlH_4$ ,  $Li_3AlH_6$ ,  $NH_3$ , and  $NH_4X$  wherein X is a counterion such halide and those given in the CRC [41]. The weight % of the reactants may be in any desired molar range. The reagents may be well mixed using a ball mill.

In an embodiment, the reaction mixture comprises a source of catalyst and a source of H. In an embodiment, the reaction mixture further comprises reactants which undergo reaction to form Li catalyst and atomic hydrogen. The reactants may comprise one or more of the group of H<sub>2</sub>, hydrido catalyst, MNH<sub>2</sub>, M<sub>2</sub>NH, M<sub>3</sub>N, NH<sub>3</sub>,  
 5 LiX, NH<sub>4</sub>X (X is a counterion such as a halide), MNO<sub>3</sub>, MAIH<sub>4</sub>, M<sub>3</sub>AlH<sub>6</sub>, and MBH<sub>4</sub>, wherein M is an alkali metal that may be the catalyst. The reaction mixture may comprise reagents selected from the group of Li, LiH, LiNO<sub>3</sub>, LiNO, LiNO<sub>2</sub>, Li<sub>3</sub>N, Li<sub>2</sub>NH, LiNH<sub>2</sub>, LiX, NH<sub>3</sub>, LiBH<sub>4</sub>, LiAlH<sub>4</sub>, Li<sub>3</sub>AlH<sub>6</sub>, LiOH, Li<sub>2</sub>S, LiHS, LiFeSi, Li<sub>2</sub>CO<sub>3</sub>, LiHCO<sub>3</sub>, Li<sub>2</sub>SO<sub>4</sub>, LiHSO<sub>4</sub>, Li<sub>3</sub>PO<sub>4</sub>, Li<sub>2</sub>HPO<sub>4</sub>, LiH<sub>2</sub>PO<sub>4</sub>, Li<sub>2</sub>MoO<sub>4</sub>, LiNbO<sub>3</sub>, Li<sub>2</sub>B<sub>4</sub>O<sub>7</sub>  
 10 (lithium tetraborate), LiBO<sub>2</sub>, Li<sub>2</sub>WO<sub>4</sub>, LiAlCl<sub>4</sub>, LiGaCl<sub>4</sub>, Li<sub>2</sub>CrO<sub>4</sub>, Li<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>, Li<sub>2</sub>TiO<sub>3</sub>, LiZrO<sub>3</sub>, LiAlO<sub>2</sub>, LiCoO<sub>2</sub>, LiGaO<sub>2</sub>, Li<sub>2</sub>GeO<sub>3</sub>, LiMn<sub>2</sub>O<sub>4</sub>, Li<sub>4</sub>SiO<sub>4</sub>, Li<sub>2</sub>SiO<sub>3</sub>, LiTaO<sub>3</sub>, LiCuCl<sub>4</sub>, LiPdCl<sub>4</sub>, LiVO<sub>3</sub>, LiIO<sub>3</sub>, LiFeO<sub>2</sub>, LiIO<sub>4</sub>, LiClO<sub>4</sub>, LiScO<sub>n</sub>, LiTiO<sub>n</sub>, LiVO<sub>n</sub>, LiCrO<sub>n</sub>, LiCr<sub>2</sub>O<sub>n</sub>, LiMn<sub>2</sub>O<sub>n</sub>, LiFeO<sub>n</sub>, LiCoO<sub>n</sub>, LiNiO<sub>n</sub>, LiNi<sub>2</sub>O<sub>n</sub>, LiCuO<sub>n</sub>, and LiZnO<sub>n</sub>, where n=1, 2, 3, or 4, an oxyanion, an oxyanion of a strong acid, an oxidant, a molecular oxidant  
 15 such as V<sub>2</sub>O<sub>3</sub>, I<sub>2</sub>O<sub>5</sub>, MnO<sub>2</sub>, Re<sub>2</sub>O<sub>7</sub>, CrO<sub>3</sub>, RuO<sub>2</sub>, AgO, PdO, PdO<sub>2</sub>, PtO, PtO<sub>2</sub>, and NH<sub>4</sub>X wherein X is a nitrate or other suitable anion given in the CRC [41], and a reductant. In each case, the mixture further comprises hydrogen or a source of hydrogen. In other embodiments, other dissociators are used or one may not be used wherein atomic hydrogen, and, optionally, atomic catalyst, are generated  
 20 chemically by reaction of the species of the mixture. In a further embodiment, the reactant catalyst may be added to the reaction mixture.

The reaction mixture may further comprise an acid such as H<sub>2</sub>SO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, H<sub>2</sub>CO<sub>3</sub>, HNO<sub>2</sub>, HNO<sub>3</sub>, HClO<sub>4</sub>, H<sub>3</sub>PO<sub>3</sub>, and H<sub>3</sub>PO<sub>4</sub> or a source of an acid such as an anhydrous acid. The latter may comprise at least one of the list of SO<sub>2</sub>, SO<sub>3</sub>, CO<sub>2</sub>,  
 25 NO<sub>2</sub>, N<sub>2</sub>O<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, Cl<sub>2</sub>O<sub>7</sub>, PO<sub>2</sub>, P<sub>2</sub>O<sub>3</sub>, and P<sub>2</sub>O<sub>5</sub>.

In an embodiment, the reaction mixture further comprises a reactant catalyst to generate the reactants that serve as a lower-energy-hydrogen catalyst or a source of lower-energy-hydrogen catalyst and atomic hydrogen or a source of atomic hydrogen. Suitable reactant catalysts comprise at one of the group of acids, bases, halide ions, metal ions and free radical sources. The reactant catalyst may be at least one of the group of a weak-base-catalysts such as  $\text{Li}_2\text{SO}_4$ , a weak-acid catalyst such as a solid acid such as  $\text{LiHSO}_4$ , a metal ion source such as  $\text{TiCl}_3$  or  $\text{AlCl}_3$  which provide  $\text{Ti}^{3+}$  and  $\text{Al}^{3+}$  ions, respectively, a free radical source such a  $\text{CoX}_2$  wherein X is a halide such as Cl wherein  $\text{Co}^{2+}$  may react with  $\text{O}_2$  to form the  $\text{O}_2^-$  radical, metals such as Ni, Fe, Co preferably at a concentration of about 1 mol%, a source of  $\text{X}^-$  ion (X is halide) such as  $\text{Cl}^-$  or  $\text{F}^-$  from  $\text{LiX}$ , a source of free radical initiators/propagators such as peroxides, azo-group compounds, and UV light.

In an embodiment, the reactant mixture to form lower-energy hydrogen comprises a source of hydrogen, a source of catalyst, and at least one of a getter for hydrino and a getter for electrons from the catalyst as it is ionized to resonantly accept energy from atomic hydrogen to form hydrinos having energies given by Eq. (1). The hydrino getter may bind to lower-energy hydrogen to prevent the reverse reaction to ordinary hydrogen. In an embodiment, the reaction mixture comprises a getter for hydrino such as  $\text{LiX}$  or  $\text{Li}_2\text{X}$  (X is halide or other anion such anions from the CRC [41]). The electron getter may perform at least one of accepting electrons from the catalyst and stabilizing the catalyst-ion intermediate such as a  $\text{Li}^{2+}$  intermediate to allow the catalysis reaction to occur with fast kinetics. The getter may be an inorganic compound comprising at least one cation and one anion. The cation may be  $\text{Li}^+$ . The anion may be a halide or other anion given in the CRC [41] such as one of the group comprising  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{I}^-$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{HSO}_4^-$ ,  $\text{CoO}_2^-$ ,  $\text{IO}_3^-$ ,  $\text{IO}_4^-$ ,

$\text{TiO}_3^-$ ,  $\text{CrO}_4^-$ ,  $\text{FeO}_2^-$ ,  $\text{PO}_4^{3-}$ ,  $\text{HPO}_4^{2-}$ ,  $\text{H}_2\text{PO}_4^-$ ,  $\text{VO}_3^-$ ,  $\text{ClO}_4^-$  and  $\text{Cr}_2\text{O}_7^{2-}$  and other anions of the reactants. The hydride binder and/or stabilizer may be at least one of the group of  $\text{LiX}$  ( $\text{X}$  = halide) and the other compounds comprising the reactants.

In an embodiment of the reaction mixture such as  $\text{Li}$ ,  $\text{LiNH}_2$ , and  $\text{X}$  wherein  $\text{X}$  is the hydride binding compound,  $\text{X}$  is at least one of  $\text{LiHBr}$ ,  $\text{LiHI}$ , a hydrino hydride compound, and a lower-energy hydrogen compound. In an embodiment, the catalyst reaction mixture is regenerated by addition of hydrogen from a source of hydrogen.

In an embodiment, the hydrino product may bind to form a stable hydrino hydride compound. The hydride binder may be  $\text{LiX}$  wherein  $\text{X}$  is a halide or other anion. The hydride binder may react with a hydride that has an NMR upfield shift greater than that of TMS. The binder may be an alkali halide, and the product of hydride binding may be an alkali hydride halide having an NMR upfield shift greater than that of TMS. The hydride may have a binding energy determined by XPS of 11 to 12 eV. In an embodiment, the product of the catalysis reaction is the hydrogen molecule  $\text{H}_2(1/4)$  having an solid NMR peak at about 1 ppm relative to TMS and a binding energy of about 250 eV that is trapped in a crystalline ionic lattice. In an embodiment, the product  $\text{H}_2(1/4)$  is trapped in the crystalline lattice of an ionic compound of the reactor such that the selection rules for infrared absorption are such that the molecule becomes IR active and a FTIR peak is observed at about  $1990\text{ cm}^{-1}$ .

Additional sources of atomic  $\text{Li}$  of the present invention comprise additional alloys of  $\text{Li}$  such as these comprising  $\text{Li}$  and at least one of alkali, alkaline earth metals, transitions, metals, rare earth metals, noble metals, tin, aluminum, other Group III and Group IV metal, actinides, and lanthanides. Some representative

alloys comprise one or more members of the group of LiBi, LiAg, LiIn, LiMg, LiAl, LiMgSi, LiFeSi, LiZr, LiAlCu, LiAlZr, LiAlMg, LiB, LiCa, LiZn, LiBSi, LiNa, LiCu, LiPt, LiCaNa, LiAlCuMgZr, LiPb, LiCaK, LiV, LiSn, and LiNi. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

In another embodiment, an anion can form a hydrogen-type bond with a Li atom of a covalently bound Li-Li molecule. This hydrogen-type bond can weaken the Li-Li bond to the point that a Li atom is at vacuum energy (equivalent to free a atom) such that it can serve as a catalyst atom to form hydrides. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

In an embodiment, the function of the hydrogen dissociator is provided by a chemical reaction. Atomic H is generated by the reaction of at least two species of the reaction mixture or by the decomposition of at least one species. In an embodiment, Li-Li reacts with  $\text{LiNH}_2$  to form atomic Li, atomic H, and  $\text{Li}_2\text{NH}$ . Atomic Li may also form by the decomposition or reaction of  $\text{LiNO}_3$ . In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

In further embodiments, in addition to a catalyst or source of catalyst to form lower-energy hydrogen, the reaction mixture comprises heterogeneous catalysts to dissociate MM and MH such as LiLi and LiH as to provide M and H atoms. The heterogeneous catalyst may comprise at least one element from the group of transition elements, precious metals, rare earth and other metals and elements such as Mo, W, Ta, Ni, Pt, Pd, Ti, Al, Fe, Ag, Cr, Cu, Zn, Co, and Sn.

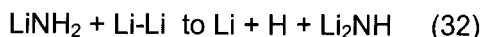
In an embodiment of the Li carbon alloy, the reaction mixture comprises an

excess of Li over the Li-carbon intercalation limit. The excess may be in the range of 1% and 1000% and preferably in the range of 1% to 10%. The carbon may further comprise a hydrogen spillover catalyst having a hydrogen dissociator such as Pd or Pt on activated carbon. In a further embodiment, the cell temperature exceeds that at which Li is completely intercalated into the carbon. The cell temperature may be in the range of about 100 to 2000 °C, preferably in the range of about 200 to 800 °C, and most preferably in the range of about 300 to 700 °C. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

In an embodiment of the Li silicon alloy, the cell temperature is in the range over which the silicon alloy further comprising H releases atomic hydrogen. The range may be about 50-1500 °C, preferably about 100 to 800 °C, and most preferably in the range of about 100 to 500 °C. The hydrogen pressure may be in range of about 0.01 to  $10^5$  Torr, preferably in the range of about 10 to 5000 Torr, and most preferably in the range of about 0.1 to 760 Torr. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

The reaction mixture, alloys, and compounds may be formed by mixing the catalyst such as Li or a source of catalyst such as catalyst hydride with the other element(s) or compound(s) or a source of the other element(s) or compound(s) such as a hydride of the other element(s). The catalyst hydride may be LiH, KH, CsH, or NaH. The reagents may be mixed by ball milling. An alloy of the catalyst may also be formed from a source of alloy comprising the catalyst and at least one other element or compound.

In an embodiment, the reaction mechanism for the Li/N system to form hydrido reactants of atomic Li and H is



In embodiments of the other Li-alloy systems, the reaction mechanism is analogous to that of the Li/N system with the other alloy element(s) replacing N. Exemplary reaction mechanisms to carry out the reaction to form hydride reactants, atomic Li and H, involving the reaction mixtures comprising Li with at least one of S, Sn, Si, and C are



Preferred embodiments of the Li/S alloy-catalyst system comprises Li with  $\text{Li}_2\text{S}$  and Li with LiHS. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

#### 15 Primary Li/Nitrogen Alloy Reactions

Lithium in the solid and liquid states is a metal, and the gas comprises covalent  $\text{Li}_2$  molecules. In order to generate atomic lithium, the reaction mixture of the solid fuel comprises Li/N alloy reactants. The reaction mixture may comprise at least one of the group of Li, LiH,  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ ,  $\text{Li}_3\text{N}$ ,  $\text{NH}_3$ , a dissociator, a hydrogen source such as  $\text{H}_2$  gas or a hydride, a support, and a getter such as LiX (X is a halide). The dissociator is preferably Pt or Pd on a high surface area support inert to Li. It may comprise Pt or Pd on carbon or  $\text{Pd/Al}_2\text{O}_3$ . The latter support may comprise a protective surface coating of a material such as  $\text{LiAlO}_2$ . Preferred dissociators for a reagent mixture comprising a Li/N alloy or Na/N alloy are Pt or Pd on  $\text{Al}_2\text{O}_3$ , Raney nickel (R-Ni), and Pt or Pd on carbon. In the case that the

dissociator support is  $\text{Al}_2\text{O}_3$ , the reactor temperature may be maintained below that which results in its substantial reaction with Li. The temperature may be below the range of about  $250^\circ\text{C}$  to  $600^\circ\text{C}$ . In another embodiment, Li is in the form of LiH and the reaction mixture comprises one or more of  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ ,  $\text{Li}_3\text{N}$ ,  $\text{NH}_3$ , a  
 5 dissociator, a hydrogen source such as  $\text{H}_2$  gas or a hydride, a support, and a getter such as  $\text{LiX}$  (X is a halide) wherein the reaction of LiH with  $\text{Al}_2\text{O}_3$  is substantially endothermic. In other embodiments, the dissociator may be separate from the balance of the reaction mixture wherein the separator passes H atoms.

Two preferred embodiments comprise the first reaction mixture of LiH,  $\text{LiNH}_2$ ,  
 10 and Pd on  $\text{Al}_2\text{O}_3$  powder and a second reaction mixture of Li,  $\text{Li}_3\text{N}$ , and hydrided Pd on  $\text{Al}_2\text{O}_3$  powder that may further comprise  $\text{H}_2$  gas. The first reaction mixture can be regenerated by addition of  $\text{H}_2$ , and the second mixture can be regenerated by removing  $\text{H}_2$  and hydriding the dissociator or by reintroducing  $\text{H}_2$ . The reactions to generate catalyst and H as well as the regeneration reactions are given *infra*.

15 In an embodiment,  $\text{LiNH}_2$  is added to the reaction mixture.  $\text{LiNH}_2$  generates atomic hydrogen as well as atomic Li according to the reversible reactions



and



20 In an embodiment, the reaction mixture comprises about 2:1 Li and  $\text{LiNH}_2$ . In the hydrino reaction cycle, Li-Li and  $\text{LiNH}_2$  react to form atomic Li, atomic H, and  $\text{Li}_2\text{NH}$ , and the cycle continues according to Eq. (38). The reactants may be present in any wt%.

The mechanism of the formation of  $\text{Li}_2\text{NH}$  from  $\text{LiNH}_2$  involves a first step that  
 25 forms ammonia [42]:



With LiH present, the ammonia reacts to release H<sub>2</sub>



and the net reaction is the consumption of LiNH<sub>2</sub> with the formation of H<sub>2</sub>:



With Li present, the amide is not consumed due to the energetically much more favorable back reaction of Li with ammonia:



Thus, in an embodiment, the reactants comprise a mixture of Li and LiNH<sub>2</sub> to form  
10 atomic Li and atomic H according to Eqs. (37-38).

The reaction mixture of Li and LiNH<sub>2</sub> that serves as a source of Li catalyst and atomic hydrogen may be regenerated. During the regeneration cycle, the reaction product mixture comprising species such as Li, Li<sub>2</sub>NH, and Li<sub>3</sub>N can be reacted with H to form LiH and LiNH<sub>2</sub>. LiH has a melting point of 688 °C; whereas, LiNH<sub>2</sub> melts at  
15 380 °C, and Li melts at 180 °C. LiNH<sub>2</sub> liquid and any Li liquid that forms can be physically removed from the LiH solid at about 380 °C, and then LiH solid can be heated separately to form Li and H<sub>2</sub>. The Li and LiNH<sub>2</sub> can be recombined to regenerate the reaction mixture. And, the excess H<sub>2</sub> from LiH thermal decomposition can be reused in the next regeneration cycle with some make-up H<sub>2</sub>  
20 to replace any H<sub>2</sub> consumed in hydride formation.

In a preferred embodiment, the competing kinetics of the hydriding or dehydriding of one reactant over another is exploited to achieve a desired reaction mixture comprising hydrided and non-hydrided compounds. For example, hydrogen can be added under appropriate temperature and pressure conditions such that the  
25 reverse of reactions of Eqs. (37) and (38) occur over the competing reaction of the

formation of LiH such that the hydrogenated products are predominantly Li and LiNH<sub>2</sub>. Alternatively, a reaction mixture comprising compounds of the group of Li, Li<sub>2</sub>NH, and Li<sub>3</sub>N may be hydrogenated to form the hydrides and the LiH can be selectively dehydrided by pumping at the temperature and pressure ranges and  
 5 duration which achieves the selectivity based on differential kinetics.

In an embodiment, Li is deposited as a thin film over a large area and a mixture of LiH and LiNH<sub>2</sub> is formed by addition of ammonia. The reaction mixture may further comprise excess Li. Atomic Li and H are formed according to Eqs. (37-38) with the subsequent reaction to form states with energies given by Eq. (1).  
 10 Then, the mixture can be regenerated by H<sub>2</sub> addition followed by heating and pumping with selective pumping and removal of H<sub>2</sub>.

A reversible system of the present invention to generate atomic lithium catalyst is the Li<sub>3</sub>N + H system which can be regenerated by pumping. The reaction mixture comprises at least one of Li<sub>3</sub>N and a source of Li<sub>3</sub>N such as Li and N<sub>2</sub>, and  
 15 a source of H such as at least one of H<sub>2</sub> and a hydrogen dissociator, LiNH<sub>2</sub>, Li<sub>2</sub>NH, LiH, Li, NH<sub>3</sub>, and a metal hydride. The reaction of H<sub>2</sub> with Li<sub>3</sub>N gives LiH and Li<sub>2</sub>NH; whereas, the reaction of Li<sub>3</sub>N and H from an atomic hydrogen source such as H<sub>2</sub> and a dissociator or from a hydride undergoing decomposition gives



20 The atomic Li catalyst can then react with additional atomic H to form hydrides. The side products such as LiH, Li<sub>2</sub>NH, and LiNH<sub>2</sub> can be converted to Li<sub>3</sub>N by evacuating the reaction vessel of H<sub>2</sub>. Representative Li/N alloy reactions are as follows:





$Li_3N$ , a source of H, and a hydrogen dissociator are in any desired molar ratio.

- 5 Each are in molar ratios of greater than 0 and less than 100%. Preferably the molar ratios are similar. In an embodiment, the ratios of  $Li_3N$ , at least one of  $LiNH_2$ ,  $Li_2NH$ ,  $LiH$ ,  $Li$ , and  $NH_3$ , and a H source such as a metal hydride are similar. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

- 10 In an embodiment, lithium amide and hydrogen is reacted to form ammonia and lithium:



The reaction can be driven to form  $Li$  by increasing the  $H_2$  concentration.

Alternatively, the forward reaction can be driven via the formation of atomic H using

- 15 a dissociator. The reaction with atomic H is given by



In an embodiment of the reaction mixture that comprises one or more compounds that react with a source of Li to form Li catalyst, the reaction mix comprises at least one species from the group of  $LiNH_2$ ,  $Li_2NH$ ,  $Li_3N$ ,  $Li$ ,  $LiH$ ,  $NH_3$ ,  $H_2$  and a dissociator.

- 20 In an embodiment, Li catalyst is generated from a reaction of  $LiNH_2$  and hydrogen, preferably atomic hydrogen as given in reaction Eq. (50). The ratios of reactants may be any desired amount. Preferably the ratios are about stoichiometric to those of Eqs. (49-50). The reactions to form catalyst are reversible with the addition of a source of H such as  $H_2$  gas to replace that reacted to form hydrides wherein the

catalyst reactions are given by Eqs. (3-5), and lithium amide forms by the reaction of ammonia with Li:



In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic  
5 K, atomic Cs, and molecular NaH. In a preferred embodiment, the reaction mixture comprises a hydrogen dissociator, a source of atomic hydrogen, and Na or K and NH<sub>3</sub>. In an embodiment, ammonia reacts with Na or K to form NaNH<sub>2</sub> or KNH<sub>2</sub> that serves as a source of catalyst. Another embodiment comprises a source of K catalyst such as K metal, a hydrogen source such as at least one of NH<sub>3</sub>, H<sub>2</sub>, and a  
10 hydride such as a metal hydride, and a dissociator. A preferred hydride is one comprising R-Ni that also may serve as a dissociator. Additionally, a hydride getter such as KX may be present wherein X is preferably a halide such as Cl, Br, or I. The cell may be run continuously with the replacement of the hydrogen source. The NH<sub>3</sub> may act as a source of atomic K by the reversible formation of KN alloy compounds  
15 from K-K such as at least one of amide, imide, or nitride or by formation of KH with the release of atomic K.

In a further embodiment, the reactants comprise the catalyst such as Li and an atomic hydrogen source such H<sub>2</sub> and a dissociator or a hydride such as hydrided R-Ni. H can react with Li-Li to form LiH and Li which can further serve as the  
20 catalyst to react with additional H to form hydrinos. Then, Li can be regenerated by evacuating H<sub>2</sub> released from LiH. The plateau temperature at 1 Torr for LiH decomposition is about 560 °C. LiH can be decomposed at about 0.5 Torr and about 500 °C, below the alloy-formation and sintering temperatures of R-Ni. The molted Li can be separated from R-Ni, the R-Ni may be rehydrided, and Li and  
25 hydrided R-Ni can be returned to another reaction cycle.

In an embodiment, Li atoms are vapor deposited on a surface. The surface may support or be a source of H atoms. The surface may comprise at least one of a hydride and hydrogen dissociator. The surface may be R-Ni which may be hydrided. The vapor deposition may be from a reservoir containing a source of Li atoms. The

5 Li source may be controlled by heating. One source that provides Li atoms when heated is Li metal. The surface may be maintained at a low temperature such as room temperature during the vapor deposition. The Li-coated surface may be heated to cause the reaction of Li and H to form H states given by Eq. (1). Other thin-film deposition techniques that are well known in the ART comprise further

10 embodiments of the Invention. Such embodiments comprise physical spray, electro-spray, aerosol, electro-arching, Knudsen cell controlled release, dispenser-cathode injection, plasma-deposition, sputtering, and further coating methods and systems such as melting a fine dispersion of Li, electroplating Li, and chemical deposition of Li. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K,

15 atomic Cs, and molecular NaH.

In the case of vapor-deposited Li on a hydride surface, regeneration can be achieved by heating with pumping to remove LiH and Li, the hydride can be rehydrided by introducing H<sub>2</sub>, and Li atoms can be redeposited onto the regenerated hydride after the cell is evacuated in an embodiment. In other embodiments, K, Cs,

20 and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

Li and R-Ni are in any desired molar ratio. Each of Li and R-Ni are in molar ratios of greater than 0 and less than 100%. Preferably the molar ratio of Li and R-Ni are similar.

In a preferred embodiment, the competing kinetics of the hydriding or

25 dehydriding of one reactant over another is exploited to achieve a reaction mixture

comprising hydrided and non-hydrided compounds. For example, the formation of LiH is thermodynamically favored over the formation of R-Ni hydride. However, the rate of LiH formation at low temperature such as the range of about 25 °C-100 °C is very low; whereas, the formation of R-Ni hydride proceeds at a high rate in this

5 temperature range at modest pressures such as the range of about 100 Torr to 3000 Torr. Thus, the reaction mixture of Li and hydrided R-Ni can be regenerated from LiH R-Ni by pumping at about 400-500 °C to dehydride LiH, cooling the vessel to about 25-100 °C, adding hydrogen to preferentially hydride R-Ni for a duration that achieves the desired selectivity, and then removing the excess hydrogen by

10 evacuating the cell. While excess Li is present or is added to be in excess, the R-Ni can be used in repeated cycles by selectively hydriding alone. This can be achieved by adding hydrogen in the temperature and pressure ranges that achieve the selective hydriding of R-Ni and then by removing the excess hydrogen before the vessel is heated to initiate the reactions that form atomic H and atomic Li and the

15 subsequent hydriding reaction. Further hydrides and sources of catalysts can be used in place of Li and R-Ni in this procedure. In a further embodiment, the R-Ni is hydrided to a great extent in a separate preparation step using elevated temperature and high-pressure hydrogen or by using electrolysis. The electrolysis may be in basic aqueous solution. The base may be a hydroxide. The counter electrode may

20 be nickel. In this case, R-Ni can provide atomic H for a long duration with the appropriate temperature, pressure, and temperature ramp rate.

LiH has a high melting point of 688 °C which may be above that which sinters the dissociator or causes the dissociator metal to form an alloy with the catalyst metal. For example, an alloy of LiNi may form at temperatures in excess of about

25 550 °C in the case that the dissociator is R-Ni and the catalyst is Li. Thus, in another

embodiment, LiH is converted to LiNH<sub>2</sub> that can be removed at its lower melting point such that the reaction mixture can be regenerated. The reaction to form lithium amide from lithium hydride and ammonia is given by



- 5 Then, molten LiNH<sub>2</sub> can be recovered at the melting point of 380 °C. LiNH<sub>2</sub> may be converted to Li by decomposition.

In an embodiment comprising the recovery of molten LiNH<sub>2</sub>, gas pressure is applied to the mixture comprising LiNH<sub>2</sub> to increase the rate of its separation from solid components. A screen separator or semi-permeable membrane may retain the  
10 solid components. The gas may be an inert gas such as a noble gas or a decomposition product such as nitrogen to limit the decomposition of LiNH<sub>2</sub>. Molten Li can be separated using gas pressure as well. To clean any residue from a dissociator, gas flow can be used. An inert gas such as a noble gas is preferable. In the case that residual Li adheres to the dissociator such as R-Ni, the residue can be  
15 removed by washing with a basic solution such as a basic aqueous solution which may also regenerate the R-Ni. Alternatively, the Li may be hydrided and the solids of LiH and R-Ni and any additional solid compounds present may be separated mechanically by methods such as sieving. In another embodiment, the dissociator such as R-Ni and the other reactants may be physically separated but maintained in  
20 close proximity to permit diffusion of atomic hydrogen to the balance of reactant mixture. The balance of reaction mixture and dissociator may be placed in open juxtaposed boats, for example. In other embodiments, the reactor further comprises multiple compartments independently containing the dissociator and balance of the reaction mixture. The separator of each compartment allows for atomic hydrogen  
25 formed in a dissociator compartment to flow to the balance-of-reaction-mixture

compartment while maintaining the chemical separation. The separator may be a metallic screen or semipermeable, inert membrane which may be metallic. The contents may be mechanically mixed during the operation of the reactor. The separated balance of the reaction mixture and its products can be removed and  
 5 reprocessed outside of the reaction vessel and returned independently of the dissociator, or either may be independently reprocessed within the reactor.

Other embodiments of systems to generate atomic catalyst Li and atomic H involve Li, ammonia, and LiH. Atomic Li catalyst and atomic H can be generated by reaction of  $\text{Li}_2$  and  $\text{NH}_3$ :



$\text{LiNH}_2$  is a source of  $\text{NH}_3$  by the reaction:



In a preferred embodiment, the Li is dispersed on a support having a large surface area to react with ammonia. Ammonia can also react with LiH to generate  $\text{LiNH}_2$ :



And,  $\text{H}_2$  can react with  $\text{Li}_2\text{NH}$  to regenerate  $\text{LiNH}_2$ :



In another embodiment, the reactants comprise a mixture of  $\text{LiNH}_2$  and a dissociator. The reaction to form atomic lithium is:



The Li can then react with additional H to form hydride.

Other embodiments of systems to generate atomic catalyst Li and atomic H involve Li and  $\text{LiBH}_4$  or  $\text{NH}_4\text{X}$  (X is an anion such as halide). Atomic Li catalyst and atomic H can be generated by reaction of  $\text{Li}_2$  and  $\text{LiBH}_4$ :



$\text{NH}_4\text{X}$  can generate  $\text{LiNH}_2$  and  $\text{H}_2$



Then, atomic Li can be generated according to the reaction of Eqs. (32) and (37). In another embodiment, the reaction mechanism for the Li/N system to form

5 hydride reactants of atomic Li and H is



where X is a counterion, preferably a halide.

Atomic Li catalyst can be generated by reaction of  $\text{Li}_2\text{NH}$  or  $\text{Li}_3\text{N}$  with atomic H formed by the dissociation of  $\text{H}_2$ :



In a further embodiment, the reaction mixture comprises nitrides of metals in addition to Li such as those of Mg Ca Sr Ba Zn and Th. The reaction mixture may comprise metals that exchange with Li or form mixed-metal compounds with Li. The  
15 metals may be from the group of alkali, alkaline earth, and transition metals. The compounds may further comprise N such as amides, imides, and nitrides.

In an embodiment, the catalyst Li is generated chemically by an anion exchange reaction such as a halide (X) exchange reaction. For example, at least one of Li metal and Li-Li molecules are reacted with a halide compound to form  
20 atomic Li and LiX. Alternatively, LiX is reacted with a metal M to form atomic Li and MX. In an embodiment, lithium metal is reacted with a lanthanide halide to form Li and the LiX where X is halide. An example is the reaction of  $\text{CeBr}_3$  with  $\text{Li}_2$  to form Li and LiBr. In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

25 In another embodiment, the reaction mixture further comprises the reactants

and products of the Haber process [43]. The products may be  $\text{NH}_x$   $x=0,1,2,3,4$ .

These products may react with Li or compounds comprising Li to form atomic Li and atomic H. For example, Li-Li may react with  $\text{NH}_x$  to form Li and possibly H:



In other embodiments, K, Cs, and Na replace Li wherein the catalyst is atomic K, atomic Cs, and molecular NaH.

A mixture of compounds may be used which melts at a lower temperature  
10 than that of one or more of compounds individually. Preferably, a eutectic mixture may form that is a molten salt that mixes the reactants such as Li and  $\text{LiNH}_2$ .

The chemistry of the reaction mixture can change very substantially based on the physical state of the reactants and the presence or absence of a solvent or added solute or alloy species. Objectives of the present invention for changing the  
15 physical state are to control the rate of reaction and to alter the thermodynamics to achieve a sustainable lower-energy hydrogen reaction with the addition of H from a source of H. For the Li/N alloy system comprising reactants such as Li and  $\text{LiNH}_2$ , alkali metals, alkaline earth metals, and their mixtures may serve as the solvent. For example, excess Li can serve as a molten solvent for  $\text{LiNH}_2$  to comprise solvated Li  
20 and  $\text{LiNH}_2$  reactants that will have different kinetics and thermodynamics of reaction relative to those of the solid-state mixture. The former effect, control of the kinetics of the lower-energy hydrogen reaction, can be adjusted by controlling the properties of the solute and solvent such as temperature, concentration, and molar ratios. Following the reaction to generate atomic catalyst and atomic hydrogen, the latter  
25 effect can be used to regenerate the initial reactants. This is a route when the

products cannot be directly regenerated by hydrogenation.

One embodiment where the regeneration of the reactants is facilitated by a solvent or added solute or alloy species involves lithium metal wherein the hydriding of Li is not to completion so that Li remains a solvent and a reactant. In Li solvent,  
5 the following regeneration reaction may occur with the addition of H from a source to form LiH:



For the Li/N alloy system comprising reactants such as Li and LiNH<sub>2</sub>, alkali metals, alkaline earth metals, and their mixtures may serve as the solvent. In an  
10 embodiment, the solvent is selected such that it can reduce LiH to Li and form an unstable solvent hydride with the release of H. Preferably, the solvent may be one or more of the group of Li (excess), Na, K, Rb, Cs, and Ba that have the ability to reduce Li<sup>+</sup> and a corresponding hydride having a low thermal stability. In a case that the melting point of the solvent is higher than desired such as in the case of Ba with  
15 a high melting point of 727 °C, the solvent can be mixed with other solvents such as metals to form a solvent with a lower melting point such as one comprising a eutectic mixture. In an embodiment, one or more alkaline earth metals can be mixed with one or more alkali metals to lower the melting point, add the capability to reduce Li<sup>+</sup>, and decrease the stability of the corresponding solvent hydride.

20 Another embodiment where the regeneration of the reactants is facilitated by a solvent or added solute or alloy species involves potassium metal. Potassium metal in a mixture of LiH and LiNH<sub>2</sub> may reduce LiH to Li and form KH. Since KH is thermally unstable at intermediate temperatures such as 300 °C, it may facilitate the further hydrogenation of Li<sub>2</sub>NH to Li and LiNH<sub>2</sub>.

25 Thus, K may catalyze the reaction given by Eq. (66). The reaction steps are



wherein H is added at the rate at which it is consumed by lower-energy hydrogen production. Alternatively, K catalytically generates Li and H from LiH wherein LiNH<sub>2</sub>

5 is formed directly from hydrogenation of Li<sub>2</sub>NH. The reactions steps are



In addition to the favorable condition of the instability of the hydride (KH), the amide  
10 (KNH<sub>2</sub>) is also unstable so that the exchange of lithium amide with potassium amide is not thermodynamically favorable. In addition to K, Na is a preferred metal solvent since it can reduce LiH and has a lower vapor pressure. Other examples of suitable metal solvents are Rb, Cs, Mg, Ca, Sr, Ba, and Sn. The solvent may comprise a mixture of metals such as a mixture of two or more alkaline or alkaline earth metals.  
15 Preferable solvents are Li (excess) and Na above 380 °C since Li is miscible in Na above this temperature.

In another embodiment, an alkali or alkaline earth metal serves as a regeneration catalyst according to Eqs. (70-71). In an embodiment, LiNH<sub>2</sub> is first removed from the LiH/LiNH<sub>2</sub> mixture by melting the LiNH<sub>2</sub>. Then, the metal M may  
20 be added to catalyze the LiH to Li conversion. M can be selectively removed by distillation. Na, K, Rb, and Cs form hydrides that decompose at relatively low temperatures and form amides that thermally decompose; thus, in another embodiment, at least one can serve as a reactant for the catalytic conversion of LiH to Li and H according to the corresponding reaction for K given by Eqs. (67-71). In  
25 addition, some alkaline earths such as Sr can form very stable hydrides which can

serve to convert LiH to Li by reaction of LiH and an alkaline earth metal to form the stable alkaline earth hydride. By operating at an elevated temperature, hydrogen may be supplied from the alkaline earth hydride via decomposition with the lithium inventory being primarily as Li. The reaction mixture may comprise Li, LiNH<sub>2</sub>, X, and  
 5 a dissociator wherein X may be a lithium compound such as LiH, Li<sub>2</sub>NH, Li<sub>3</sub>N with a small amount of an alkaline earth metal that forms a stable hydride to generate Li from LiH. The source of hydrogen may be H<sub>2</sub> gas. The operating temperature may be sufficient such that H is available.

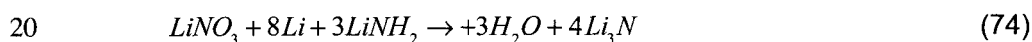
In an embodiment, LiNO<sub>3</sub> can serve to generate the LiNH<sub>2</sub> source of Li and  
 10 H in a set of coupled reactions. Consider an embodiment of the catalysis reaction mixture comprising Li, LiNH<sub>2</sub>, and LiNO<sub>3</sub>. The reaction of Li and LiNH<sub>2</sub> to Li<sub>3</sub>N and release H<sub>2</sub> is



The balanced H<sub>2</sub> reduction reaction of the released H<sub>2</sub> (Eq. (72)) with LiNO<sub>3</sub> to  
 15 form water and lithium amide is



Then, reaction Eq. (72) can proceed with the generated LiNH<sub>2</sub> and the balance of Li, and the coupled reactions given by Eqs. (72) and (73) can occur until the Li is completely consumed. The overall reaction is given by



The water may be dynamically removed by methods such as condensation or reacted with a getter to prevent its reaction with species such as Li, LiNH<sub>2</sub>, Li<sub>2</sub>NH, and Li<sub>3</sub>N.

### Exemplary Regeneration of Li Catalyst Reactants

The present invention further comprises methods and systems to generate, or regenerate the reaction mixture to form states given by Eq. (1) from any side products that form during said reaction. For example, in an embodiment of the energy reactor, the catalysis reaction mixture such as  $Li$ ,  $LiNH_2$ , and  $LiNO_3$  is regenerated from any side products such as  $LiOH$  and  $Li_2O$  by methods known to those skilled in the Art such as given in Cotton and Wilkinson [43]. Components of the reaction mixture including side products may be liquid or solids. The mixture is heated or cooled to a desired temperature, and the products are separated physically by means known by those skilled in the Art. In an embodiment,  $LiOH$  and  $Li_2O$  are solid,  $Li$ ,  $LiNH_2$ , and  $LiNO_3$  are liquid, and the solid components are separated from the liquid ones. The  $LiOH$  and  $Li_2O$  may be converted to lithium metal by reduction with  $H_2$  at high temperature or by electrolysis of the molten compounds or a mixture containing them. The electrolysis cell may comprise a eutectic molten salt comprising at least one of  $LiOH$ ,  $Li_2O$ ,  $LiCl$ ,  $KCl$ ,  $CaCl_2$  and  $NaCl$ . The electrolysis cell is comprised of a material resistant to attack by Li such as a BeO or BN vessel. The Li product may be purified by distillation.  $LiNH_2$  is formed by means known in the Art such as reaction of  $Li$  with nitrogen followed by hydrogen reduction. Alternatively,  $LiNH_2$  can be formed directly by reaction of  $Li$  with  $NH_3$ .

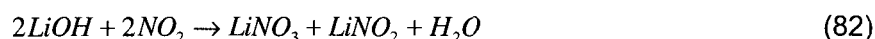
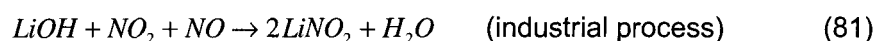
In the case that the initial reaction mixture comprises at least one of Li,  $LiNH_2$ , and  $LiNO_3$ , Li metal may be regenerated by methods such as electrolysis,  $LiNO_3$  can be generated from Li metal. One key step that eliminates the difficult nitrogen

fixation step is the reaction of Li metal with  $N_2$  to form  $Li_3N$  even at room temperature.  $Li_3N$  can be reacted with  $H_2$  to form  $Li_2NH$  and  $LiNH_2$ .  $Li_3N$  can be reacted with an oxygen source to form  $LiNO_3$ . In an embodiment,  $Li_3N$  is used in the synthesis of lithium nitrate ( $LiNO_3$ ) involving reactants or intermediates of at least one or more of lithium (Li), lithium nitride ( $Li_3N$ ), oxygen ( $O_2$ ), an oxygen source, lithium imide ( $Li_2NH$ ), and lithium amide ( $LiNH_2$ ).

In an embodiments, the oxidation reactions are



Lithium nitrate can be regenerated from  $Li_2O$  and  $LiOH$  using at least one of  $NO_2$ ,  $NO$ , and  $O_2$  by the following reactions



Lithium oxide can be converted to lithium hydroxide by reaction with steam:



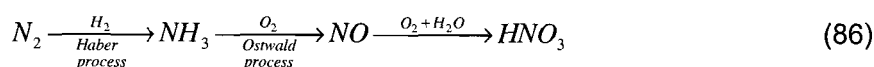
In an embodiment,  $Li_2O$  is converted to  $LiOH$  followed by reaction with  $NO_2$  and  $NO$  according to Eq. (81).

Both lithium oxide and lithium hydroxide can be converted to lithium nitrate by treatment with nitric acid followed by drying:





LiNO<sub>3</sub> can be made by treatment of lithium oxide or lithium hydroxide with nitric acid. Nitric acid, in turn, can be generated by known industrial methods such as by the Haber process followed by the Ostwald process and then by hydration and  
 5 oxidation of NO as given in Cotton and Wilkinson [43]. In one embodiment, the exemplary sequence of steps are:



Specifically, the Haber process may be used to produce NH<sub>3</sub> from N<sub>2</sub> and H<sub>2</sub> at  
 10 elevated temperature and pressure using a catalyst such as  $\alpha$ -iron containing some oxide. The ammonia may be used to form LiNH<sub>2</sub> from Li. The Ostwald process may be used to oxidize the ammonia to NO at a catalyst such as a hot platinum or platinum-rhodium catalyst. The NO may be further reacted with oxygen and water to form nitric acid which can be reacted with lithium oxide or lithium hydroxide to form  
 15 lithium nitrate. The crystalline lithium nitrate reactant is then obtained by drying. In another embodiment, NO and NO<sub>2</sub> are reacted directly with the one or more of lithium oxide and lithium hydroxide to form lithium nitrate. The regenerated Li, LiNH<sub>2</sub>, and LiNO<sub>3</sub> are then returned to the reactor in desired molar ratios.

In further exemplary regeneration reactions, an embodiment of the reactor  
 20 comprises the reactants of Li, LiNH<sub>2</sub>, and LiCoO<sub>2</sub>. LiOH, Li<sub>2</sub>O, and Co and its lower oxides are the side products. The reactants can be regenerated by electrolysis of LiOH and Li<sub>2</sub>O to Li. LiNH<sub>2</sub> can be regenerated by reaction of Li with NH<sub>3</sub> or N<sub>2</sub> and then H<sub>2</sub>. The CoO<sub>2</sub> and its lower oxides can be regenerated by reaction with oxygen. The LiCoO<sub>2</sub> can be formed by reaction of Li with CoO<sub>2</sub>. Li, LiNH<sub>2</sub>, and

LiCoO<sub>2</sub> are then returned to the cell in a batch or continuous regeneration process.

In the case that LiIO<sub>3</sub> or LiIO<sub>4</sub> is a reagent of the mixture, IO<sub>3</sub><sup>-</sup> and or IO<sub>4</sub><sup>-</sup> may be regenerated by reaction of iodine or iodide ion with base and may further undergo electrolysis to the desired anion which may be precipitated out as LiIO<sub>3</sub> or LiIO<sub>4</sub>,

5 dried, and dehydrated.

### NaH Molecular Catalyst

In a further embodiment, a compound comprising hydrogen such as MH where H is hydrogen and M is another element serves as a source of hydrogen and  
10 a source of catalyst. In an embodiment, a catalytic system is provided by the breakage of the M-H bond plus the ionization of  $t$  electrons from an atom M each to a continuum energy level such that the sum of the bond energy and ionization energies of the  $t$  electrons is approximately one of  $m \cdot 27.2 \text{ eV}$  and  $m \cdot \frac{27.2}{2} \text{ eV}$  where  $m$  is an integer.

15 One such catalytic system involves sodium. The bond energy of NaH is 1.9245 eV [44]. The first and second ionization energies of Na are 5.13908 eV and 47.2864 eV, respectively [1]. Based on these energies NaH molecule can serve as a catalyst and H source since the bond energy of NaH plus the double ionization ( $t=2$ ) of Na to Na<sup>2+</sup>, is 54.35 eV (2X27.2 eV) which is equivalent to  $m=2$  in Eq.

20 (2). The catalyst reactions are given by

$$54.35 \text{ eV} + \text{NaH} \rightarrow \text{Na}^{2+} + 2e^- + H \left[ \frac{a_H}{(3)} \right] + [(3)^2 - 1^2] \cdot 13.6 \text{ eV} \quad (88)$$

$$\text{Na}^{2+} + 2e^- + H \rightarrow \text{NaH} + 54.35 \text{ eV} \quad (89)$$

And, the overall reaction is

$$H \rightarrow H \left[ \frac{a_H}{(3)} \right] + [(3)^2 - 1^2] \cdot 13.6 \text{ eV} \quad (90)$$

As given in Chp. 5 of Ref [30], and Ref. [20], hydrogen atoms

$H(1/p)$   $p = 1, 2, 3, \dots, 137$  can undergo further transitions to lower-energy states given

by Eq. (1) wherein the transition of one atom is catalyzed by a second that

5 resonantly and nonradiatively accepts  $m \cdot 27.2 \text{ eV}$  with a concomitant opposite

change in its potential energy. The overall general equation for the transition of

$H(1/p)$  to  $H(1/(p+m))$  induced by a resonance transfer of  $m \cdot 27.2 \text{ eV}$  to  $H(1/p')$

is represented by

$$H(1/p') + H(1/p) \rightarrow H^+ + e^- + H(1/(p+m)) + [2pm + m^2 - p'^2] \cdot 13.6 \text{ eV}$$

$$10 \quad (91)$$

In the case of high hydrogen concentrations, the transition of  $H(1/3)$  ( $p = 3$ ) to

$H(1/4)$  ( $p+m = 4$ ) with  $H$  as the catalyst ( $p = 1$ ;  $m = 1$ ) can be fast:

$$H(1/3) \xrightarrow{H} H(1/4) + 81.6 \text{ eV} \quad (92)$$

Due to the stable binding of  $H^-(1/4)$  in halides and its stability to ionization relative

15 to other reaction species, it and the corresponding molecule formed by the reactions

$2H(1/4) \rightarrow H_2(1/4)$  and  $H^-(1/4) + H^+ \rightarrow H_2(1/4)$  are favored products of the

catalysis of hydrogen.

The  $NaH$  catalyst reaction may be concerted since the sum of the bond

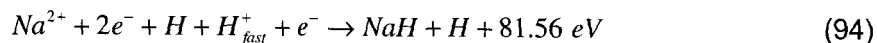
energy of  $NaH$ , the double ionization ( $t = 2$ ) of  $Na$  to  $Na^{2+}$ , and the potential energy

20 of  $H$  is  $81.56 \text{ eV}$  ( $3 \cdot 27.2 \text{ eV}$ ) which is equivalent to  $m = 3$  in Eq. (2). The catalyst

reactions are given by

$$81.56 \text{ eV} + NaH + H \rightarrow Na^{2+} + 2e^- + H_{fast}^+ + e^- + H \left[ \frac{a_H}{(4)} \right] + [(4)^2 - 1^2] \cdot 13.6 \text{ eV}$$

(93)



And, the overall reaction is

$$H \rightarrow H \left[ \frac{a_H}{(4)} \right] + [(4)^2 - 1^2] \cdot 13.6 \text{ eV} \quad (95)$$

- 5 where  $H_{fast}^{+}$  is a fast hydrogen atom having at least 13.6 eV of kinetic energy

In an embodiment, the reaction mixture comprises at least one of a source of NaH molecules and hydrogen. The NaH molecules may serve as the catalyst to form H states given by Eq. (1). A source of NaH molecules may comprise at least one of Na metal, a source of hydrogen, preferably atomic hydrogen, and NaH(s).

- 10 The source of hydrogen may be at least one of H<sub>2</sub> gas and a dissociator and a hydride. Preferably, the dissociator and hydride may be R-Ni. Preferably, the dissociator may also be Pt/Ti, Pt/Al<sub>2</sub>O<sub>3</sub>, and Pd/Al<sub>2</sub>O<sub>3</sub> powder. Solid NaH may be a source of at least one of NaH molecules, H atoms, and Na atoms.

- In a preferred embodiment, one of atomic sodium and molecular NaH is
- 15 provided by a reaction between a metallic, ionic, or molecular form of Na and at least one other compound or element. The source of Na or NaH may be at least one of metallic Na, an inorganic compound comprising Na such as NaOH, and other suitable Na compounds such as NaNH<sub>2</sub>, Na<sub>2</sub>CO<sub>3</sub>, and Na<sub>2</sub>O which are given in the CRC [41], NaX (X is a halide), and NaH(s). The other element may be H, a
- 20 displacing agent, or a reducing agent. The reaction mixture may comprise at least one of (1) a source of sodium such as at least one of Na(m), NaH, NaNH<sub>2</sub>, Na<sub>2</sub>CO<sub>3</sub>, Na<sub>2</sub>O, NaOH, NaOH doped-R-Ni, NaX (X is a halide), and NaX doped R-Ni, (2) a source of hydrogen such as H<sub>2</sub> gas and a dissociator and a hydride, (3) a displacing agent such as an alkali or alkaline earth metal, preferably Li, and (4) a reducing

agent such as at least one of a metal such as an alkaline metal, alkaline earth metal, a lanthanide, a transition metal such as Ti, aluminum, B, a metal alloy such as AlHg, NaPb, NaAl, LiAl, and a source of a metal alone or in combination with reducing agent such as an alkaline earth halide, a transition metal halide, a lanthanide halide, and aluminum halide. Preferably, the alkali metal reductant is Na. Other suitable reductants comprise metal hydrides such as  $\text{LiBH}_4$ ,  $\text{NaBH}_4$ ,  $\text{LiAlH}_4$ , or  $\text{NaAlH}_4$ . Preferably, the reducing agent reacts with NaOH to form a NaH molecules and a Na product such as Na, NaH(s), and  $\text{Na}_2\text{O}$ . The source of NaH may be R-Ni comprising NaOH and a reactant such as a reductant to form NaH catalyst such as an alkali or alkaline earth metal or the Al intermetallic of R-Ni. Further exemplary reagents are an alkaline or alkaline earth metal and an oxidant such as  $\text{AlX}_3$ ,  $\text{MgX}_2$ ,  $\text{LaX}_3$ ,  $\text{CeX}_3$ , and  $\text{TiX}_n$  where X is a halide, preferably Br or I. Additionally, the reaction mixture may comprise another compound comprising a getter or a dispersant such as at least one of  $\text{Na}_2\text{CO}_3$ ,  $\text{Na}_3\text{SO}_4$ , and  $\text{Na}_3\text{PO}_4$  that may be doped into the dissociator such as R-Ni. The reaction mixture may further comprise a support wherein the support may be doped with at least one reactant of the mixture. The support may have preferably a large surface area that favors the production of NaH catalyst from the reaction mixture. The support may comprise at least one of the group of R-Ni, Al, Sn,  $\text{Al}_2\text{O}_3$  such as gamma, beta, or alpha alumina, sodium aluminate (according to Cotton [45] beta-aluminas have other ions present such as  $\text{Na}^+$  and possess the idealized composition  $\text{Na}_2\text{O} \cdot 11\text{Al}_2\text{O}_3$ ), lanthanide oxides such as  $\text{M}_2\text{O}_3$  (preferably M= La, Sm, Dy, Pr, Tb, Gd, and Er), Si, silica, silicates, zeolites, lanthanides, transition metals, metal alloys such as alkali and alkali earth alloys with Na, rare earth metals,  $\text{SiO}_2\text{-Al}_2\text{O}_3$  or  $\text{SiO}_2$  supported Ni, and other supported metals such as at least one of alumina supported platinum, palladium, or ruthenium. The support

may have a high surface area and comprise a high-surface-area (HSA) materials such as R-Ni, zeolites, silicates, aluminates, aluminas, alumina nanoparticles, porous  $\text{Al}_2\text{O}_3$ , Pt, Ru, or Pd/ $\text{Al}_2\text{O}_3$ , carbon, Pt or Pd/C, inorganic compounds such as  $\text{Na}_2\text{CO}_3$ , silica and zeolite materials, preferably Y zeolite powder. In an embodiment, the support such as  $\text{Al}_2\text{O}_3$  (and the  $\text{Al}_2\text{O}_3$  support of the dissociator if present) reacts with the reductant such as a lanthanide to form a surface-modified support. In an embodiment, the surface Al exchanges with the lanthanide to form a lanthanide-substituted support. This support may be doped with a source of NaH molecules such as NaOH and reacted with a reductant such as a lanthanide. The subsequent reaction of the lanthanide-substituted support with the lanthanide will not significantly change it, and the doped NaOH on the surface can be reduced to NaH catalyst by reaction with the reductant lanthanide.

In an embodiment, wherein the reaction mixture comprises a source of NaH catalyst, the source of NaH may be an alloy of Na and a source of hydrogen. The alloy may comprise at least one of those known in the Art such as an alloy of sodium metal and one or more other alkaline or alkaline earth metals, transition metals, Al, Sn, Bi, Ag, In, Pb, Hg, Si, Zr, B, Pt, Pd, or other metals and the H source may be  $\text{H}_2$  or a hydride.

The reagents such as the source of NaH molecules, the source of sodium, the source of NaH, the source of hydrogen, the displacing agent, and the reducing agent are in any desired molar ratio. Each is in a molar ratio of greater than 0 and less than 100%. Preferably, the molar ratios are similar.

A preferred embodiment comprises the reaction mixture of NaH and Pd on  $\text{Al}_2\text{O}_3$  powder wherein the reaction mixture may be regenerated by addition of  $\text{H}_2$ .

In an embodiment, Na atoms are vapor deposited on a surface. The surface

may support or be a source of H atoms to form NaH molecules. The surface may comprise at least one of a hydride and hydrogen dissociator such as Pt, Ru, or Pd/Al<sub>2</sub>O<sub>3</sub> which may be hydrided. Preferably, the surface area is large. The vapor deposition may be from a reservoir containing a source of Na atoms. The Na source  
5 may be controlled via heating. One source that provides Na atoms when heated is Na metal. The surface may be maintained at a low temperature such as room temperature during the vapor deposition. The Na-coated surface may be heated to cause the reaction of Na and H to form NaH and may further cause the NaH molecules to react to form H states given by Eq. (1). Other thin-film deposition  
10 techniques that are well known in the ART comprise further embodiments of the Invention. Such embodiments comprise physical spray, electro-spray, aerosol, electro-arching, Knudsen cell controlled release, dispenser-cathode injection, plasma-deposition, sputtering, and further coating methods and systems such as melting a fine dispersion of Na, electroplating Na, and chemical deposition of Na.  
15 Na metal may be dispersed on a high-surface area material, preferably Na<sub>2</sub>CO<sub>3</sub>, carbon, silica, alumina, R-Ni, and Pt, Ru, or Pd/Al<sub>2</sub>O<sub>3</sub>, to increase the activity to form NaH when reacted with another reagent such as H or a source of H. Other dispersion materials are known in the Art such as those given in Cotton et al. [46].

In an embodiment, at least one reactant comprising the reductant or source of  
20 NaH such as Na and NaOH undergoes aerosolization to create a corresponding reactant vapor to react to form NaH catalyst. Na and NaOH may react in the cell to form NaH catalyst wherein at least one species undergoes aerosolization. The aerosolized species may be transported into the cell to react to form NaH catalyst. The means to carry the aerosolized species may be a carrier gas. The  
25 aerosolization of the reactant may be achieved using a mechanical agitator and a

carrier gas such as a noble gas to carry the reactant into the cell to form NaH catalyst. In an embodiment, Na which may serve as a source of NaH and a reductant is aerosolized by becoming charged and electrically dispensed. The reactants such as at least one of Na and NaOH may be aerosolized mechanically in  
 5 a carrier gas or they may undergo ultrasonic aerosolization. The reactant may be forced through an orifice to form a vapor. Alternatively, the reactant may be heated locally to very high temperature to be vaporized or sublimed to form a vapor. The reactants may further comprise a source of hydrogen. The hydrogen may react with Na to form NaH catalyst. The Na may be in the form of a vapor. The cell may  
 10 comprise a dissociator to form atomic hydrogen from H<sub>2</sub>. Other means of achieving aerosolization that are known to those skilled in the Art are part of the Invention.

In an embodiment, the reaction mixture comprises at least one species of the group comprising Na or a source of Na, NaH or a source of NaH, a metal hydride or source of a metal hydride, a reactant or source of a reactant to form a metal hydride,  
 15 a hydrogen dissociator, and a source of hydrogen. The reaction mixture may further comprise a support. A reactant to form a metal hydride may comprise a lanthanide, preferably La or Gd. In an embodiment, La may reversibly react with NaH to form LaH<sub>n</sub> (n=1,2,3). In an embodiment, the hydride exchange reaction forms NaH catalyst. The reversible general reaction may be given by



The reaction given by Eq. (96) applies to other *MH*-type catalysts given in TABLE 2. The reaction may proceed with the formation of hydrogen that may be dissociated to form atomic hydrogen that reacts with Na to form NaH catalyst. The dissociator is preferably at least one of Pt, Pd, or Ru/Al<sub>2</sub>O<sub>3</sub> powder, Pt/Ti, and R-Ni. Preferentially,  
 25 the dissociator support such as Al<sub>2</sub>O<sub>3</sub> comprises at least surface La substitution for

Al or comprises Pt, Pd, or Ru/M<sub>2</sub>O<sub>3</sub> powder wherein M is a lanthanide. The dissociator may be separated from the rest of the reaction mixture wherein the separator passes atomic H.

A preferred embodiment comprises the reaction mixture of NaH, La, and Pd  
 5 on Al<sub>2</sub>O<sub>3</sub> powder wherein the reaction mixture may be regenerated in an embodiment, by adding H<sub>2</sub>, separating NaH and lanthanum hydride by sieving, heating lanthanum hydride to form La, and mixing La and NaH. Alternatively, the regeneration involves the steps of separating Na and lanthanum hydride by melting Na and removing the liquid, heating lanthanum hydride to form La, hydriding Na to  
 10 NaH, and mixing La and NaH. The mixing may be by ball milling.

In an embodiment, a high-surface-area material such as R-Ni is doped with NaX (X=F, Cl, Br, I). The doped R-Ni is reacted with a reagent that will displace the halide to form at least one of Na and NaH. In an embodiment, the reactant is at least an alkali or alkaline earth metal, preferably at least one of K, Rb, Cs. In  
 15 another embodiment, the reactant is an alkaline or alkaline earth hydride, preferably at least one of KH, RbH, CsH, MgH<sub>2</sub> and CaH<sub>2</sub>. The reactant may be both an alkali metal and an alkaline earth hydride. The reversible general reaction may be given by



20

#### NaOH Catalyst Reactions to Form NaH Catalyst

The reaction of NaOH and Na to Na<sub>2</sub>O and NaH is



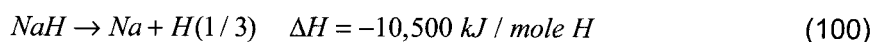
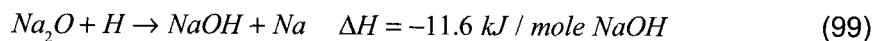
The exothermic reaction can drive the formation of NaH(g). Thus, Na metal can  
 25 serve as a reductant to form catalyst NaH(g). Other examples of suitable reductants

that have a similar highly exothermic reduction reaction with the NaH source are alkali metals, alkaline earth metals such as at least one of Mg and Ca, metal hydrides such as  $\text{LiBH}_4$ ,  $\text{NaBH}_4$ ,  $\text{LiAlH}_4$ , or  $\text{NaAlH}_4$ , B, Al, transition metals such as Ti, lanthanides such as at least one of La, Sm, Dy, Pr, Tb, Gd, and Er, preferably La, Tb, and Sm. Preferably, the reaction mixture comprises a high-surface-area material (HSA material) having a dopant such as NaOH comprising a source of NaH catalyst. Preferably, conversion of the dopant on the material with a high surface area to the catalyst is achieved. The conversion may occur by a reduction reaction. The reductant may be provided as a gas stream. Preferably, Na is flowed into the reactor as a gas stream. In addition to the preferred reductant, Na, other preferred reductants are other alkali metals, Ti, a lanthanide, or Al. Preferably, the reaction mixture comprises NaOH doped into a HSA material preferably R-Ni wherein the reductant is Na or the intermetallic Al. The reaction mixture may further comprise a source of H such as a hydride or  $\text{H}_2$  gas and a dissociator. Preferably the H source is hydrided R-Ni.

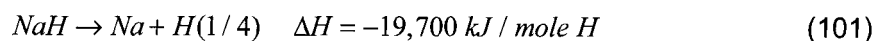
In an embodiment, the reaction temperature is maintained below that at which the reductant such as a lanthanide forms an alloy with the source of catalyst such as R-Ni. In the case of lanthanum, preferably the reaction temperature does not exceed  $532^\circ\text{C}$  which is the alloy temperature of Ni and La as shown by Gasser and Kefif [47]. Additionally, the reaction temperature is maintained below that at which the reaction with the  $\text{Al}_2\text{O}_3$  of R-Ni occurs to a significant extent such as in the range of  $100^\circ\text{C}$  to  $450^\circ\text{C}$ .

In an embodiment,  $\text{Na}_2\text{O}$  formed as a product of a reaction to generate NaH catalyst such as that given by Eq. (98), is reacted with a source of hydrogen to form

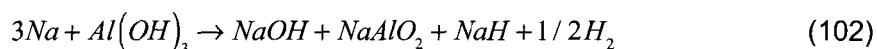
NaOH that can further serve as a source of NaH catalyst. In an embodiment, a regenerative reaction of NaOH from Eq. (98) in the presence of atomic hydrogen is



5 and



Thus, a small amount of NaOH and Na with a source of atomic hydrogen or atomic hydrogen serves as a catalytic source of the NaH catalyst, that in turn forms a large yield of hydrinos via multiple cycles of regenerative reactions such as those given by  
 10 Eqs. (98-101). In an embodiment, from the reaction given by Eq. (102),  $Al(OH)_3$  can serve as a source of NaOH and NaH wherein with Na and H, the reactions given by Eqs. (98-101) proceed to form hydrinos.



In an embodiment, the Al of the intermetallic serves as the reductant to form NaH  
 15 catalyst The balanced reaction is given by



This exothermic reaction can drive the formation of NaH(g) to drive the very exothermic reaction given by Eqs. (88-92) wherein the regeneration of NaH occurs from Na in the presence of atomic hydrogen.

20 Two preferred embodiments comprise the first reaction mixture of Na and R-Ni comprising about 0.5 wt% NaOH wherein Na serves as the reductant and a second reaction mixture of R-Ni comprising about 0.5 wt% NaOH wherein intermetallic Al serves as the reductant. The reaction mixture may be regenerated by adding NaOH and NaH that may serve as an H source and a reductant.

In an embodiment, of the energy reactor, the source of NaH such as NaOH is regenerated by addition of a source of hydrogen such as at least one of a hydride and hydrogen gas and a dissociator. The hydride and dissociator may be hydrided R-Ni. In another embodiment, the source of NaH such as NaOH-doped R-Ni is  
 5 regenerated by at least one of rehydriding, addition of NaH, and addition of NaOH wherein the addition may be by physical mixing. The mixing may be performed mechanically by means such as by ball milling.

In an embodiment, the reaction mixture further comprises oxide-forming reactants that react with NaOH or Na<sub>2</sub>O to form a very stable oxide and NaH. Such  
 10 reactants comprises a cerium, magnesium, lanthanide, titanium, or aluminum or their compounds such as AlX<sub>3</sub>, MgX<sub>2</sub>, LaX<sub>3</sub>, CeX<sub>3</sub>, and TiX<sub>n</sub> where X is a halide, preferably Br or I and a reducing compound such as an alkali or alkaline earth metal. In an embodiment, the source of NaH catalyst comprises R-Ni comprising a sodium compound such as NaOH on its surface. Then, the reaction of NaOH with the oxide-  
 15 forming reactants such as AlX<sub>3</sub>, MgX<sub>2</sub>, LaX<sub>3</sub>, CeX<sub>3</sub>, and TiX<sub>n</sub>, and alkali metal M forms NaH, MX, and Al<sub>2</sub>O<sub>3</sub>, MgO, La<sub>2</sub>O<sub>3</sub>, Ce<sub>2</sub>O<sub>3</sub>, and Ti<sub>2</sub>O<sub>3</sub>, respectively.

In an embodiment, the reaction mixture comprises NaOH doped R-Ni and an alkaline or alkaline earth metal added to form at least one of Na and NaH molecules. The Na may further react with H from a source such as H<sub>2</sub> gas or a hydride such as  
 20 R-Ni to form NaH catalyst. The subsequent catalysis reaction of NaH forms H states given by Eq. (1). The addition of an alkali or alkaline earth metal M may reduce Na<sup>+</sup> to Na by the reactions:



25 M may also react with NaOH to form H as well as Na



Then, the catalyst NaH may be formed by the reaction



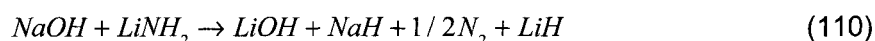
- 5 by reacting with H from reactions such as that given by Eq. (106) as well as from R-Ni and any added source of H. Na is a preferred reductant since it is a further source of NaH.

Hydrogen may be added to reduce NaOH and form NaH catalyst:

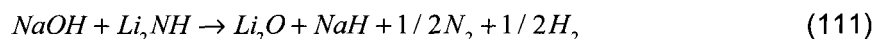


- 10 The H in R-Ni may reduce NaOH to Na metal, and water that may be removed by pumping.

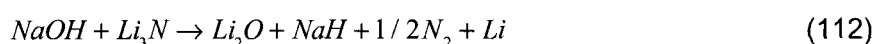
In an embodiment, the reaction mixture comprises one or more compounds that react with a source of NaH to form NaH catalyst. The source may be NaOH. The compounds may comprise at least one of a  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ , and  $\text{Li}_3\text{N}$ . The reaction mixture may further comprise a source of hydrogen such as  $\text{H}_2$ . In embodiments, the reaction of sodium hydroxide and lithium amide to form NaH and lithium hydroxide is



- The reaction of sodium hydroxide and lithium imide to form NaH and lithium hydroxide is
- 20



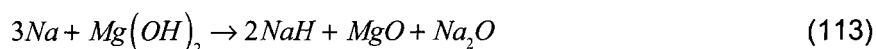
And, the reaction of sodium hydroxide and lithium nitride to form NaH and lithium oxide is



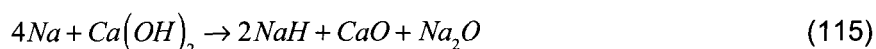
Alkaline Earth Hydroxide Catalyst Reactions to Form NaH Catalyst

In an embodiment, a source of H is provided to a source of Na to form the catalyst NaH. The Na source may be the metal. The source of H may be a hydroxide. The hydroxide may be at least one of alkali, alkaline earth hydroxide, a transition metal hydroxide, and  $\text{Al}(\text{OH})_3$ . In an embodiment, Na reacts with a hydroxide to form the corresponding oxide and NaH catalyst. In an embodiment wherein the hydroxide is  $\text{Mg}(\text{OH})_2$ , the product is  $\text{MgO}$ . In an embodiment wherein the hydroxide is  $\text{Ca}(\text{OH})_2$ , the product is  $\text{CaO}$ . Alkaline earth oxides may be reacted with water to regenerate the hydroxide as given in Cotton [48]. The hydroxide can be collected as a precipitate by means such as filtration and centrifugation.

For example, in an embodiment, the reaction to form NaH catalyst and regeneration cycle for  $\text{Mg}(\text{OH})_2$ , are given by the reactions:



In an embodiment, the reaction to form NaH catalyst and regeneration cycle for  $\text{Ca}(\text{OH})_2$ , are given by the reactions:

20 Na/N Alloy Reactions to Form NaH Catalyst

Sodium in the solid and liquid states is a metal, and the gas comprises covalent  $\text{Na}_2$  molecules. In order to generate NaH catalyst, the reaction mixture of the solid fuel comprises Na/N alloy reactants. In an embodiment, the reaction mixture, solid-fuel reactions, and regeneration reactions comprise those of the Li/N

system wherein Na replaces Li and the catalyst is molecular NaH except that the solid fuel reaction generates molecular NaH rather than atomic Li and H. In an embodiment, the reaction mixture comprises one or more compounds that react with a source of NaH to form NaH catalyst. The reaction mixture may comprise at least

5 one of the group of Na, NaH, NaNH<sub>2</sub>, Na<sub>2</sub>NH, Na<sub>3</sub>N, NH<sub>3</sub>, a dissociator, a hydrogen source such as H<sub>2</sub> gas or a hydride, a support, and a getter such as NaX (X is a halide). The dissociator is preferably Pt, Ru, or Pd/Al<sub>2</sub>O<sub>3</sub> powder. For high-temperature operation, the dissociator may comprise Pt or Pd on a high surface area support suitably inert to Na. The dissociator may be Pt or Pd on carbon or Pd/Al<sub>2</sub>O<sub>3</sub>.

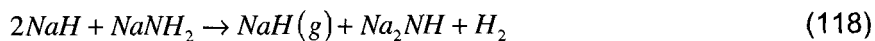
10 The latter support may comprise a protective surface coating of a material such as NaAlO<sub>2</sub>. The reactants may be present in any wt%.

A preferred embodiment comprises the reaction mixture of Na or NaH, NaNH<sub>2</sub>, and Pd on Al<sub>2</sub>O<sub>3</sub> powder wherein the reaction mixture may be regenerated by addition of H<sub>2</sub>.

15 In an embodiment, NaNH<sub>2</sub> is added to the reaction mixture. NaNH<sub>2</sub> generates NaH according to the reversible reactions



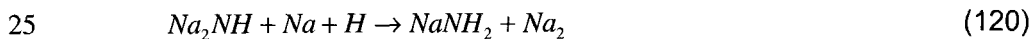
and



20 In the hydrino reaction cycle, Na-Na and NaNH<sub>2</sub> react to form NaH molecule and Na<sub>2</sub>NH, and the NaH forms hydrino and Na. Thus, the reaction is reversible according to the reactions:



and



In an embodiment, NaH of Eq. (119) is molecular such that this reaction is another to generate the catalyst.

The reaction of sodium amide and hydrogen to form ammonia and sodium hydride is



- 5 In an embodiment, this reaction is reversible. The reaction can be driven to form NaH by increasing the  $H_2$  concentration. Alternatively, the forward reaction can be driven via the formation of atomic H using a dissociator. The reaction is given by



The exothermic reaction can drive the formation of NaH(g).

- 10 In an embodiment, NaH catalyst is generated from a reaction of  $NaNH_2$  and hydrogen, preferably atomic hydrogen as given in reaction Eqs. (121-122). The ratios of reactants may be any desired amount. Preferably the ratios are about stoichiometric to those of Eqs. (121-122). The reactions to form catalyst are reversible with the addition of a source of H such as  $H_2$  gas or a hydride to replace  
15 that reacted to form hydrides wherein the catalyst reactions are given by Eqs. (88-95), and sodium amide forms with additional NaH catalyst by the reaction of ammonia with Na:



- In an embodiment, a HSA material is doped with  $NaNH_2$ . The doped HSA  
20 material is reacted with a reagent that will displace the amide group to form at least one of Na and NaH. In an embodiment, the reactant is an alkali or alkaline earth metal, preferably Li. In another embodiment, the reactant is an alkaline or alkaline earth hydride, preferably LiH. The reactant may be both an alkali metal and an alkaline earth hydride. A source of H such as  $H_2$  gas may be further provided in

addition to that provided by any other reagent of the reaction mixture such as a hydride, HSA material, and displacing reagent.

In an embodiment, sodium amide undergoes reaction with lithium to form lithium amide, imide, or nitride and Na or NaH catalyst. The reaction of sodium amide and

5 lithium to form lithium imide and NaH is

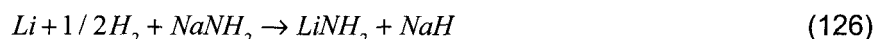


The reaction of sodium amide and lithium hydride to form lithium amide and NaH is



The reaction of sodium amide, lithium, and hydrogen to form lithium amide and NaH

10 is



In an embodiment, the reaction of the mixture forms Na, and the reactants further comprise a source of H that reacts with Na to form catalyst NaH by a reaction such as the following:



and



In an embodiment, the reactants comprise NaNH<sub>2</sub>, a reactant to displace the amide  
20 group of NaNH<sub>2</sub> such as an alkali or alkaline earth metal, preferably Li, and may additionally comprise a source of H such as at least one of MH (M=Li, Na, K, Rb, Cs, Mg, Ca, Sr, and Ba), H<sub>2</sub> and a hydrogen dissociator, and a hydride.

The reagents of the reaction mixture such as M, MH, NaH, NaNH<sub>2</sub>, HSA material, hydride, and the dissociator are in any desired molar ratio. Each of M, MH,

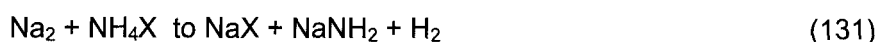
NaNH<sub>2</sub>, and the dissociator are in molar ratios of greater than 0 and less than 100%, preferably the molar ratios are similar.

Other embodiments of systems to generate molecular catalyst NaH involve Na and NaBH<sub>4</sub> or NH<sub>4</sub>X (X is an anion such as halide). Molecular NaH catalyst can

5 be generated by reaction of Na<sub>2</sub> and NaBH<sub>4</sub>:



NH<sub>4</sub>X can generate NaNH<sub>2</sub> and H<sub>2</sub>



Then, NaH catalyst can be generated according to the reaction of Eqs. (117-  
10 129). In another embodiment, the reaction mechanism for the Na/N system to form  
hydrino catalyst NaH is



#### Preparation and Regeneration of NH Catalyst Reactants

15 In an embodiment NaH molecules or Na and hydrided R-Ni can be  
regenerated by systems and methods after those disclosed for the Li-based reactant  
systems. In an embodiment, Na can be regenerated from solid NaH by evacuating  
H<sub>2</sub> released from NaH. The plateau temperature at about 1 Torr for NaH  
decomposition is about 500 °C. NaH can be decomposed at about 1 Torr and 500  
20 °C, below the alloy-formation and sintering temperatures of R-Ni. The molten Na  
can be separated from R-Ni, the R-Ni may be rehydrided, and Na and hydrided R-Ni  
can be returned to another reaction cycle. In the case of vapor-deposited Na on a  
hydride surface, regeneration can be achieved by heating with pumping to remove  
Na, the hydride can be rehydrided by introducing H<sub>2</sub>, and Na atoms can be  
25 redeposited onto the regenerated hydride after the cell is evacuated in an

embodiment.

In a preferred embodiment, the competing kinetics of the hydriding or dehydriding of one reactant over another is exploited to achieve a reaction mixture comprising hydrided and non-hydrided compounds. For example, the formation of

5 NaH solid is thermodynamically favored over the formation of R-Ni hydride.

However, the rate of NaH formation at low temperature such as the range of about 25 °C-100 °C is low; whereas, the formation of R-Ni hydride proceeds at a high rate in this temperature range at modest pressures such as the range of about 100 Torr to 3000 Torr. Thus, the reaction mixture of Na and hydrided R-Ni can be

10 regenerated from NaH solid and R-Ni by pumping at about 400-500 °C to dehydride NaH, cooling the vessel to about 25-100 °C, adding hydrogen to preferentially hydride R-Ni for a duration that achieves the desired selectivity, and then removing the excess hydrogen by evacuating the cell. While excess Na is present or is added to be in excess, the R-Ni can be used in repeated cycles by selectively hydriding

15 alone. This can be achieved by adding hydrogen in the temperature and pressure ranges that achieve the selective hydriding of R-Ni and then by removing the excess hydrogen before the vessel is heated to initiate the reactions that form atomic H and molecular NaH and the subsequent reaction to yield H states given by Eq. (1).

Alternatively, a reaction mixture comprising Na and a hydrogen source such as R-Ni

20 may be hydrogenated to form the hydrides, and the NaH solid can be selectively dehydrided by pumping at the temperature and pressure ranges and durations which achieve the selectivity based on differential kinetics.

In an embodiment having powder reactants such as a powder source of catalyst and a reductant, the reductant powder is mixed with the catalyst-source

25 powder. For example, NaOH-doped R-Ni that provides NaH catalyst is mixed with a

metal or metal hydride powder such as a lanthanide or NaH, respectively. In an embodiment of the reaction mixture having a solid material such as a dissociator, support, or HSA material that is doped or coated with at least one other species of the reaction mixture, the mixing may be achieved by ball milling or the method of incipient wetness. In an embodiment, the surface may be coated by immersing the surface into a solution of the species such as NaOH or NaX (X is a counter anion such as halide) followed by drying. Alternatively, NaOH may be incorporated into Ni/Al alloy or R-Ni by etching with concentrated NaOH (deoxygenated) using the same procedure as used to etch R-Ni as is well known in the Art [49]. In an embodiment, the HSA material such as R-Ni doped with a species such as NaOH is reacted with a reductant such as Na to form NaH catalyst that reacts to form hydrides. Then, the excess reductant such as Na may be removed from the products by evaporation, preferably, under vacuum at elevated temperature. The reductant may be condensed to be recycled. In another embodiment, at least one of the reductant and a product species is removed by using a transporting medium such as a gas or liquid such as a solvent, and the removed species is isolated from the transporting medium. The species can be isolated by means well known in the Art such as precipitation, filtration, or centrifugation. The species may be recycled directly or further reacted to a chemical form suitable for recycling. In addition, the NaOH may be regenerated by H<sub>2</sub> reduction or by reaction with a water-vapor gas stream. In the former case, excess Na may be removed by evaporation, preferably, under vacuum at elevated temperature. Alternatively, the reaction products can be removed by rinsing with a suitable solvent such as water, the HSA material may be dried, and the initial reactants may be added. Separately, the products may be regenerated to the original reactants by methods known to those skilled in the Art.

Or, a reaction product such as NaOH separated by rinsing R-Ni can be used in the process of etching R-Ni to regenerate it. In an embodiment comprising a reactant that reacts with the HSA material, the product such as an oxide may be treated with a solvent such as dilute acid to remove the product. The HSA material may then be  
5 re-doped and reused while the removed product may be regenerated by known methods.

The reductant such as an alkali metal can be regenerated from the product comprising a corresponding compound, preferably NaOH or Na<sub>2</sub>O, using methods and systems known to those skilled in the Art as given in Cotton [48]. One method  
10 comprises electrolysis in a mixture such as a eutectic mixture. In a further embodiment, the reductant product may comprise at least some oxide such as a lanthanide metal oxide (e.g. La<sub>2</sub>O<sub>3</sub>). The hydroxide or oxide may be dissolved in a weak acid such as hydrochloric acid to form the corresponding salt such as NaCl or LaCl<sub>3</sub>. The treatment with acid may be a gas phase reaction. The gases may be  
15 streaming at low pressure. The salt may be treated with a product reductant such as an alkali or alkaline earth metal to form the original reductant. In an embodiment, the second reductant is an alkaline earth metal, preferably Ca wherein NaCl or LaCl<sub>3</sub> is reduced to Na or La metal. Methods known to those skilled in the Art are given in Cotton [48] which is herein incorporated by reference in its entirety. The additional  
20 product of CaCl<sub>3</sub> is recovered and recycled as well. In alternative embodiment, the oxide is reduced with H<sub>2</sub> at high temperature.

In an embodiment wherein NaAlH<sub>4</sub> is the reductant, the product comprises Na and Al that need not be separated from the R-Ni product. The R-Ni is regenerated as a source of catalyst without separation. Regeneration may be by the addition of  
25 NaOH. The NaOH may partially etch Al of R-Ni [49] which is dried [50] for reuse.

Alternatively, Na and Al are reacted insitu or separated from the reaction product mixture and reacted with  $H_2$  to form  $NaAlH_4$  directly as given by Cotton [51] or by reaction of the recovered NaH with Al to form  $NaAlH_4$ .

R-Ni is a preferred HSA material having NaOH as a source of NaH catalyst.

- 5 In an embodiment, the Na content from the manufacturer is in the range of about 0.01 mg to 100 mg per gram of R-Ni, preferably in the range of about 0.1 mg to 10 mg per gram of R-Ni, and most preferably in the range of about 1 mg to 10 mg Na per gram of R-Ni. The R-Ni or an alloy of Ni may further comprise promoters such as at least one of Zn, Mo, Fe, and Cr. The R-Ni or alloy may be at least one of W. R. Grace Davidson Raney 2400, Raney 2800, Raney 2813, Raney 3201, and Raney 4200, preferably 2400, or etched or Na-doped embodiments of these materials. The NaOH content of the R-Ni may be increased by a factor in the range of about 1.01 to 1000 times. Solid NaOH may added by mixing by means such as ball milling, or it may be dissolved in a solution to achieve a desired concentration or pH. The
- 10 solution may be added to R-Ni and the water evaporated to achieve the doping. The doping may be in the range of about 0.1  $\mu g$  to 100 mg per gram of R-Ni, preferably in the range of about 1  $\mu g$  to 100  $\mu g$  per gram of R-Ni, and most preferably in the range of about 5  $\mu g$  to 50  $\mu g$  per gram of R-Ni. In an embodiment, 0.1 g of NaOH is dissolved in 100 ml of distilled water and 10 ml of the NaOH solution is added to
- 20 500g of non-decanted R-Ni from W. R. Grace Chemical Company such lot #2800/05310 having an initial total content of Na of about 0.1 wt%. The mixture is then dried. The drying may be achieved by heating at 50 °C under vacuum for 65 hours. In another embodiment, the doping may be achieved by ball milling NaOH with the R-Ni such as about 1 to 10 mg of NaOH per gram of R-Ni.

- 25 The R-Ni may be dried dry according to the standard R-Ni drying procedure

[50]. The R-Ni may be decanted and dried in the temperature range of about 10-500 °C under vacuum, preferably, it is dried at 50°C. The duration may be in the range of about 1 hr to 200 hours, preferably, the duration is about 65 hours. In an embodiment, the H content of the dried R-Ni is in the range of about 1 ml-100 ml H/g R-Ni, preferably the H content of the dried R-Ni is in the range of about 10-50 ml H/g R-Ni (where ml gas are at STP). The drying temperature, time, vacuum pressure and flow of gases, if any, such as He, Ar, or H<sub>2</sub> during and after drying is controlled to achieve dryness and the desired H content.

In an embodiment of the R-Ni doped with a source of NaH catalyst such as NaOH, the preparation of R-Ni from Ni/Al alloy comprises the step of etching the alloy with aqueous NaOH solution. The concentration of NaOH, etching times, and rinsing exchanges, may be varied to achieve the desired level of incorporation of NaOH. In an embodiment, the NaOH solution is oxygen free. The molarity is in the range of about 1 to 10 M, preferably in the range of about 5 to 8 M, and most preferably about 7 M. In an embodiment, the alloy is reacted with the NaOH for about 2 hours at about 50 °C. The solution is then diluted with water such as deionized water until Al(OH)<sub>3</sub> precipitate forms. In that case, the amphoteric reaction of NaOH with Al(OH)<sub>3</sub> to form water-soluble Na[Al(OH)<sub>4</sub>] is at least partially prevented such that NaOH is incorporated into the R-Ni. The incorporation may be achieved by drying the R-Ni without decanting. The pH of the diluted solution may be in the range of 8 to 14, preferably in the range of 9 to 12, and most preferably about 10-11. Argon may be bubbled through the solution for about 12 hours, and then the solution may be dried.

Following the reaction of the reductant and source of catalyst to form hydrido (H with states given by Eq. (1)), the reductant and catalyst source are regenerated.

In an embodiment, the reaction products are separated. The reductant product may be separated from the product of the source of catalyst. In an embodiment wherein at least one of the reductant and source of catalyst are powders, the products are separated mechanically based on at least one of particle size, shape, weight, density, magnetism, or dielectric constant. Particles having a significant difference in size and shape can be mechanically separated using sieves. Particles with large differences in density can be separated by buoyancy differences. Particles having large differences in magnetic susceptibility can be separated magnetically. Particles with large differences in dielectric constant can be separated electrostatically. In an embodiment, the products are ground to reverse any sintering. The grinding may be with a ball mill.

Methods known by those skilled in the Art that can be applied to the separations of the present Invention by application of routine experimentation. In general, mechanical separations can be divided into four groups: sedimentation, centrifugal separation, filtration, and sieving as described in Earle [52] which is incorporated herein in its entirety by reference. In a preferred embodiment, the separation of the particles is achieved by at least one of sieving and use of classifiers. The size and shape of the particle may be selected in the starting materials to achieve the desired separation of the products.

In a further embodiment, the reductant is a powder or is converted to a powder and mechanically separated from the other components of the product reaction mixture such as a HSA material. In embodiments, Na, NaH, and a lanthanide comprise at least one of the reductant and a source of the reductant, and a HSA material component is R-Ni. The reductant product may be separated from the product mixture by converting any unreacted non-powder reductant metal to the

hydride. The hydride may be formed by the addition of hydrogen. The metal hydride may be ground to form a powder. The powder may then be separated from the other products such as that of the source of the catalyst based on a difference in the size of the particles. The separation may be by agitating the mixture over a series of  
5 sieves that are selective for certain size ranges to cause the separation.

Alternatively, or in combination with sieving, the R-Ni particles are separated from the metal hydride or metal particles based on the large magnetic susceptibility difference between the particles. The reduced R-Ni product may be magnetic. The unreacted lanthanide metal and hydrided metal and any oxide such as  $\text{La}_2\text{O}_3$  are  
10 weakly paramagnetic and diamagnetic, respectively. The product mixture may be agitated over a series of strong magnets alone or in combination with one or more sieves to cause the separation based on at least one of the stronger adherence or attraction of the R-Ni product particles to the magnet and a size difference of the two classes of particles. In an embodiment of the use of sieves and an applied magnetic  
15 field, the latter adds an additional force to that of gravity to draw the smaller R-Ni product particles through the sieve while the weakly paramagnetic or diamagnetic particles of the reductant product are retained on the sieve due to their larger size.

The alkali metal may be recovered from the corresponding hydride by heating and optionally by applying vacuum. The evolved hydrogen can be reacted with alkali  
20 metal in another batch of a repetitive reaction-regeneration cycle. There may be more than one batch in the cycle at various stages. The hydride and any other compound(s) may be separated, and then reacted to form the metal separately from the formation of the metal from the hydride.

In an embodiment, the reaction mixture is regenerated by vapor deposition  
25 techniques, preferably in the case that the reactants are on the surface of a HSA

material such as R-Ni. In further embodiments, having other coated desired reactants comprising at least one of a source of NaH catalyst on a surface and a material that supports the formation of NaH catalyst such as a HSA material, the reactants are provided by reacting gas streams with the HSA material such as R-Ni.

5 The deposited reactants may comprise at least one of the group of Na, NaH, Na<sub>2</sub>O, NaOH, Al, Ni, NiO, NaAl(OH)<sub>4</sub>,  $\beta$ -alumina,  $Na_2O \cdot nAl_2O_3$  (n= integer from 1 to 1000, preferably 11), Al(OH)<sub>3</sub>, and Al<sub>2</sub>O<sub>3</sub> in alpha, beta, and gamma forms. Vapor-deposited elements, compounds, intermediates, and species that are the desired reactants or are converted into the desired reactants as well as the sequence and

10 composition of the gas streams and the chemistry to form the reactants from the gas streams are well by those skilled in the Art of vapor deposition. For example, alkali metals can be directly vapor deposited and any metals with low vapor pressure such as Al can be vapor deposited from the gaseous halide or hydride. Furthermore, oxide products such as Na<sub>2</sub>O may be reacted with a source of hydrogen to form the

15 hydroxide such as NaOH. The source of hydrogen may comprise a water-vapor gas stream to regenerate NaOH. Alternatively, the NaOH can be formed using H<sub>2</sub> or a source of H<sub>2</sub>. In addition, the hydriding of the HSA material such as R-Ni can be achieved by supplying hydrogen gas, and removing excess hydrogen by means such as pumping. The NaOH may be regenerated stoichiometrically by precisely

20 controlling the total moles of reacted H from a source such as water vapor or hydrogen gas. Any additional Na or NaH formed at this stage may be removed by evaporation, and decomposition and evaporation, respectively. Alternative, an oxide or hydroxide product such as Na<sub>2</sub>O or excess NaOH can be removed. This can be achieved by conversion to a halide such as NaI which may be removed by distillation

25 or vaporization. The vaporization can be achieved with heating and by maintaining a

vacuum at elevated temperature. The conversion to a halide may be achieved by reaction with an acid such as HI. The treatment may be by a gas stream comprising the acid gas. In another embodiment, any excess NaOH is removed by sublimation. This occurs under vacuum in the temperature range of 350-400 °C as given by

5 Cotton [53]. Any evaporation, distillation, transport, gas-stream process, or related processes of the reactants may further comprise a carrier gas. The carrier gas may be an inert gas such as a noble gas. Further steps may comprise mechanical mixing or separation. For example, NaOH and NaH can be also be deposited or removed mechanically by methods such as ball milling and sieving, respectively.

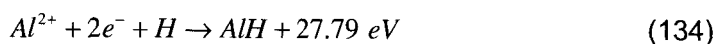
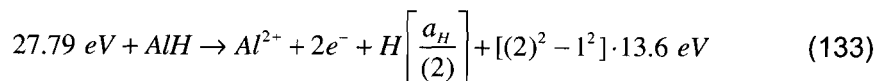
10 In the case that the redundant is an element other than a desired first element such as Na, the other element may be replaced by a second such as Na using methods known in the Art. A step may comprise evaporation of excess reductant. The large surface-area material such as R-Ni may be etched. The etching may be with a base, preferably NaOH. The etched product may be decanted with

15 substantially all of any solvent such as water removed mechanically such as by decanting and possibly centrifugation. The etched R-Ni may be dried under vacuum and recycled.

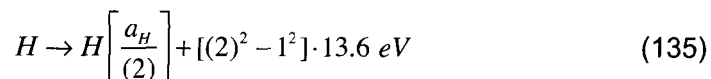
#### Additional MH-Type Catalysts and Reactions

20 Another catalytic system of the type MH involves aluminum. The bond energy of AlH is 2.98 eV [44]. The first and second ionization energies of Al are 5.985768 eV and 18.82855 eV, respectively [1]. Based on these energies AlH molecule can serve as a catalyst and H source since the bond energy of AlH plus the double ionization ( $t = 2$ ) of Al to  $Al^{2+}$ , is 27.79 eV (27.2 eV) which is equivalent to  $m = 1$  in

25 Eq. (2). The catalyst reactions are given by

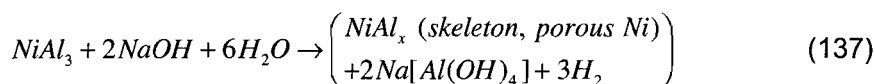


And, the overall reaction is



5 In an embodiment, the reaction mixture comprises at least one of AlH molecules and a source of AlH molecules. A source of AlH molecules may comprise Al metal and a source of hydrogen, preferably atomic hydrogen. The source of hydrogen may be a hydride, preferably R-Ni. In another embodiment, the catalyst AlH is generated by the reaction of an oxide or hydroxide of Al with a reductant. The reductant comprises at least one of the NaOH reductants given previously. In an embodiment, a source of H is provided to a source of Al to form the catalyst AlH. The Al source may be the metal. The source of H may be a hydroxide. The hydroxide may be at least one of alkali, alkaline earth hydroxide, a transition metal hydroxide, and Al(OH)<sub>3</sub>.

15 Raney nickel can be prepared by the following two reaction steps:



Na[Al(OH)<sub>4</sub>] is readily dissolved in concentrated NaOH. It can be washed in de-oxygenated water. The prepared Ni contains Al (~10 wt%, that may vary), is porous, and has a large surface area. It contains large amounts of H, both in the Ni lattice and in the form of Ni-AlH<sub>x</sub> (x=1,2,3).

R-Ni may be reacted with another element to cause the chemical release of AlH molecules which then undergo catalysis according to reactions given by Eqs.

(133-135). In an embodiment, the AlH release is caused by a reduction reaction, etching, or alloy formation. One such other element M is an alkali or alkaline earth metal which reacts with the Ni portion of R-Ni to cause the  $\text{AlH}_x$  component to release AlH molecules that subsequently undergo catalysis. In an embodiment, M may react with Al hydroxides or oxides to form Al metal that may further react with H to form AlH. The reaction can be initiated by heating, and the rate may be controlled by controlling the temperature. M (alkali or alkaline earth metal) and R-Ni are in any desired molar ratio. Each of M and R-Ni are in molar ratios of greater than 0 and less than 100%. Preferably the molar ratio of M and R-Ni are similar.

In an embodiment, Al atoms are vapor deposited on a surface. The surface may support or be a source of H atoms to form AlH molecules. The surface may comprise at least one of a hydride and hydrogen dissociator. The surface may be R-Ni which may be hydrided. The vapor deposition may be from a reservoir containing a source of Al atoms. The Al source may be controlled by heating. One source that provides Al atoms when heated is Al metal. The surface may be maintained at a low temperature such as room temperature during the vapor deposition. The Al-coated surface may be heated to cause the reaction of Al and H to form AlH and may further cause the AlH molecules to react to form H states given by Eq. (1). Other thin-film deposition techniques that are well known in the ART to form layers of at least one of Al and other elements such as metals comprise further embodiments of the Invention. Such embodiments comprise physical spray, electro-spray, aerosol, electro-arching, Knudsen cell controlled release, dispenser-cathode injection, plasma-deposition, sputtering, and further coating methods and systems such as melting a fine dispersion of Al, electroplating Al, and chemical deposition of Al.

In an embodiment, the source of AlH comprises R-Ni and other Raney metals

or alloys of Al known in the Art such as R-Ni or an alloy comprising at least one of Ni, Cu, Si, Fe, Ru, Co, Pd, Pt, and other elements and compounds. The R-Ni or alloy may further comprise promoters such as at least one of Zn, Mo, Fe, and Cr. The R-Ni may be at least one of W. R. Grace Raney 2400, Raney 2800, Raney 2813,

5 Raney 3201, Raney 4200, or an etched or Na doped embodiment of these materials.

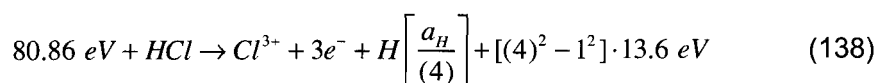
In another embodiment of the AlH catalyst system, the source of catalyst comprises a Ni/Al alloy wherein the Al to Ni ratio is in the range of about 10-90%, preferably about 10-50%, and more preferably about 10-30%. The source of catalyst may comprise palladium or platinum and further comprise Al as a Raney metal.

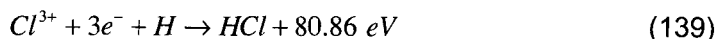
10 The source of AlH may further comprise AlH<sub>3</sub>. The AlH<sub>3</sub> may be deposited on or with Ni to form a NiAlH<sub>x</sub> alloy. The alloy may be activated by the addition of a metal such as an alkali or alkaline earth metal. In an embodiment the reaction mixture comprises AlH<sub>3</sub>, R-Ni, and a metal such as an alkali metal. The metal may be supplied by vaporization from a reservoir or by gravity feed from a source that

15 flows down on the R-Ni at an elevated temperature. In an embodiments, AlH molecules or Al and hydrided R-Ni can be regenerated by systems and methods after those disclosed for the other reactant systems.

Another catalytic system of the type MH involves chlorine. The bond energy of HCl is 4.4703 eV [44]. The first, second, and third ionization energies of Cl are

20 12.96764 eV, 23.814 eV, and 39.61 eV, respectively [1]. Based on these energies HCl can serve as a catalyst and H source since the bond energy of HCl plus the triple ionization ( $t = 3$ ) of Cl to Cl<sup>3+</sup>, is 80.86 eV ( $3 \cdot 27.2 \text{ eV}$ ) which is equivalent to  $m = 3$  in Eq. (2). The catalyst reactions are given by





And, the overall reaction is

$$H \rightarrow H \left[ \frac{a_H}{(4)} \right] + [(4)^2 - 1^2] \cdot 13.6 \text{ eV} \quad (140)$$

In an embodiment, the reaction mixture comprises HCl or a source of HCl. A source may be NH<sub>4</sub>Cl or a solid acid and a chloride such as an alkali or alkaline earth chloride. The solid acid may be at least one of MHSO<sub>4</sub>, MHCO<sub>3</sub>, MH<sub>2</sub>PO<sub>4</sub>, and MHPO<sub>4</sub> wherein M is a cation such as an alkali or alkaline earth cation. Other such solid acids are known to those skilled in the Art. In an embodiment, the reactants comprise HCl catalyst in an ionic lattice such as HCl in an alkali or alkaline earth halide, preferably a chloride. In an embodiment, the reaction mixture comprises a strong acid such as H<sub>2</sub>SO<sub>4</sub> and an ionic compound such as NaCl. The reaction of the acid with the ionic compound such as NaCl generates HCl in the crystalline lattice to serve as a hydrino catalyst and H source.

In general, MH type hydrogen catalysts to produce hydrinos provided by the breakage of the M-H bond plus the ionization of  $t$  electrons from the atom M each to a continuum energy level such that the sum of the bond energy and ionization energies of the  $t$  electrons is approximately  $m \cdot 27.2 \text{ eV}$  where  $m$  is an integer are given in TABLE 2. Each MH catalyst is given in the first column and the corresponding M-H bond energy is given in column two. The atom M of the MH species given in the first column is ionized to provide the net enthalpy of reaction of  $m \cdot 27.2 \text{ eV}$  with the addition of the bond energy in column two. The enthalpy of the catalyst is given in the eighth column where  $m$  is given in the ninth column. The electrons, that participate in ionization are given with the ionization potential (also called ionization energy or binding energy). For example, the bond energy of NaH,

1.9245 eV [44], is given in column two. The ionization potential of the  $n$ th electron of the atom or ion is designated by  $IP_n$  and is given by the CRC [1]. That is for example,  $Na + 5.13908 \text{ eV} \rightarrow Na^+ + e^-$  and  $Na^+ + 47.2864 \text{ eV} \rightarrow Na^{2+} + e^-$ . The first ionization potential,  $IP_1 = 5.13908 \text{ eV}$ , and the second ionization potential,

- 5  $IP_2 = 47.2864 \text{ eV}$ , are given in the second and third columns, respectively. The net enthalpy of reaction for the breakage of the  $NaH$  bond and the double ionization of  $Na$  is 54.35 eV as given in the eighth column, and  $m = 2$  in Eq. (2) as given in the ninth column. Additionally, H can react with each of the MH molecules given in
- TABLE 2 to form a hydrino having a quantum number  $p$  increased by one (Eq. (1))
- 10 relative to the catalyst reaction product of MH alone as given by exemplary Eq. (92).

TABLE 2. MH type hydrogen catalysts capable of providing a net enthalpy of reaction of approximately  $m \cdot 27.2 \text{ eV}$ .

| Catalyst | M-H    | $IP_1$   | $IP_2$   | $IP_3$  | $IP_4$  | $IP_5$ | Enthalpy | m |
|----------|--------|----------|----------|---------|---------|--------|----------|---|
|          | Bond   |          |          |         |         |        |          |   |
|          | Energy |          |          |         |         |        |          |   |
| AlH      | 2.98   | 5.985768 | 18.82855 |         |         |        | 27.79    | 1 |
| BiH      | 2.936  | 7.2855   | 16.703   |         |         |        | 26.92    | 1 |
| ClH      | 4.4703 | 12.96763 | 23.8136  | 39.61   |         |        | 80.86    | 3 |
| CoH      | 2.538  | 7.88101  | 17.084   |         |         |        | 27.50    | 1 |
| GeH      | 2.728  | 7.89943  | 15.93461 |         |         |        | 26.56    | 1 |
| InH      | 2.520  | 5.78636  | 18.8703  |         |         |        | 27.18    | 1 |
| NaH      | 1.925  | 5.139076 | 47.2864  |         |         |        | 54.35    | 2 |
| RuH      | 2.311  | 7.36050  | 16.76    |         |         |        | 26.43    | 1 |
| SbH      | 2.484  | 8.60839  | 16.63    |         |         |        | 27.72    | 1 |
| SeH      | 3.239  | 9.75239  | 21.19    | 30.8204 | 42.9450 |        | 107.95   | 4 |

|     |       |         |          |          |       |   |
|-----|-------|---------|----------|----------|-------|---|
| SiH | 3.040 | 8.15168 | 16.34584 |          | 27.54 | 1 |
| SnH | 2.736 | 7.34392 | 14.6322  | 30.50260 | 55.21 | 2 |

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In other embodiments of the MH type catalyst, the reactants comprise sources of SbH, SiH, SnH, and InH. In embodiments providing the catalyst MH, the sources comprise at least one of M and a source of H<sub>2</sub> and MH<sub>x</sub> such as at least one of Sb, Si, Sn, and In and a source of H<sub>2</sub>, and SbH<sub>3</sub>, SiH<sub>4</sub>, SnH<sub>4</sub>, and InH<sub>3</sub>.

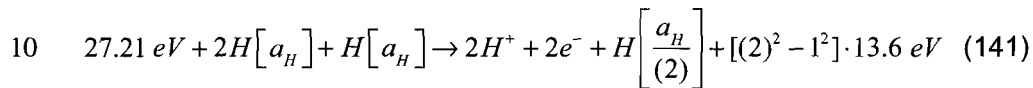
The reaction mixture may further comprise a source of H and a source of catalyst wherein the source of at least one of H and catalyst may be a solid acid or NH<sub>4</sub>X where X is a halide, preferably Cl to form HCl catalyst. Preferably, the reaction mixture may comprise at least one of NH<sub>4</sub>X, a solid acid, NaX, LiX, KX, NaH, LiH, KH, Na, Li, K, a support, a hydrogen dissociator and H<sub>2</sub> where X is a halide, preferably Cl. The solid acid may be NaHSO<sub>4</sub>, KHSO<sub>4</sub>, LiHSO<sub>4</sub>, NaHCO<sub>3</sub>, KHCO<sub>3</sub>, LiHCO<sub>3</sub>, Na<sub>2</sub>HPO<sub>4</sub>, K<sub>2</sub>HPO<sub>4</sub>, Li<sub>2</sub>HPO<sub>4</sub>, NaH<sub>2</sub>PO<sub>4</sub>, KH<sub>2</sub>PO<sub>4</sub>, and LiH<sub>2</sub>PO<sub>4</sub>. The catalyst may be at least one of NaH, Li, K, and HCl. The reaction mixture may further comprise at least one of a dissociator and a support.

Other thin-film deposition techniques that are well known in the ART comprise further embodiments of the Invention. Such embodiments comprise physical spray, electro-spray, aerosol, electro-arching, Knudsen cell controlled release, dispenser-cathode injection, plasma-deposition, sputtering, and further coating methods and systems such as melting a fine dispersion of M, electroplating M, and chemical deposition of M where MH comprises a catalyst.

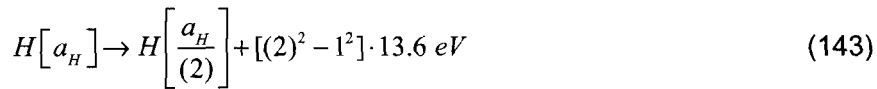
In each case of a source of MH comprising an M alloy such as AlH and Al, respectively, the alloy may be hydrided with a source of H<sub>2</sub> such as H<sub>2</sub> gas. H<sub>2</sub> can be supplied to the alloy during the reaction, or H<sub>2</sub> may be supplied to form the alloy of a desired H content with the H pressure changed during the reaction. In this case,

the initial H<sub>2</sub> pressure may be about zero. The alloy may be activated by the addition of a metal such as an alkali or alkaline earth metal. For MH catalysts and sources of MH, the hydrogen gas may be maintained in the range of about 1 Torr to 100 atm, preferably about 100 Torr to 10 atm, more preferably about 500 Torr to 2 atm. In  
 5 other embodiments, the source of hydrogen is from a hydride such as an alkali or alkaline earth metal hydride or a transition metal hydride.

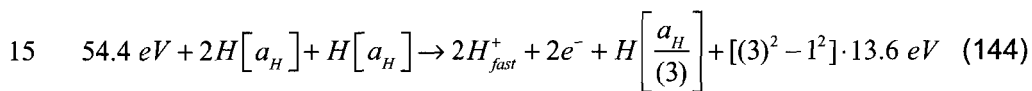
Atomic hydrogen in high density can undergo three-body-collision reactions to form hydrinos wherein one H atom undergoes the transition to form states given by Eq. (1) when two additional H atoms ionize. The reaction are given by



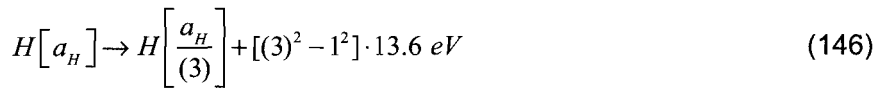
And, the overall reaction is



In another embodiment, the reaction are given by



And, the overall reaction is



In an embodiment, the material that provides H atoms in high density is R-Ni.

20 The atomic H may be from at least one of the decomposition of H within R-Ni and the dissociation of H<sub>2</sub> from an H<sub>2</sub> source such as H<sub>2</sub> gas supplied to the cell. R-Ni may

be reacted with an alkali or alkaline earth metal M to enhance the production of layers of atomic H to cause the catalysis. R-Ni can be regenerated by evaporating the metal M followed by addition of hydrogen to rehydride the R-Ni.

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### **EXPERIMENTAL**

Equation numbers, section numbers, and reference numbers given hereafter in this Experimental section refer to those given in this Experimental section of the Disclosure.

20

#### **Abstract**

The data from a broad spectrum of investigational techniques strongly and consistently indicates that hydrogen can exist in lower-energy states than previously thought possible. The predicted reaction involves a resonant, nonradiative energy transfer from otherwise stable atomic hydrogen to a catalyst capable of accepting

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the energy. The product is  $H(1/p)$ , fractional Rydberg states of atomic hydrogen called "hydrino atoms" wherein  $n = \frac{1}{2}, \frac{1}{3}, \frac{1}{4}, \dots, \frac{1}{p}$  ( $p \leq 137$  is an integer) replaces the well-known parameter  $n = \text{integer}$  in the Rydberg equation for hydrogen excited states. Atomic lithium and molecular  $NaH$  served as catalysts since they meet the catalyst criterion—a chemical or physical process with an enthalpy change equal to an integer multiple  $m$  of the potential energy of atomic hydrogen, 27.2 eV (e.g.  $m = 3$  for  $Li$  and  $m = 2$  for  $NaH$ ). Specific predictions based on closed-form equations for energy levels of the corresponding hydrino hydride ions  $H^-(1/4)$  of novel alkali halido hydrino hydride compounds ( $MH * X$ ;  $M = Li \text{ or } Na$ ,  $X = \text{halide}$ ) and dihydrino molecules  $H_2(1/4)$  were tested using chemically generated catalysis reactants.

First,  $Li$  catalyst was tested.  $Li$  and  $LiNH_2$  were used as a source of atomic lithium and hydrogen atoms. Using water-flow, batch calorimetry, the measured power from 1g  $Li$ , 0.5g  $LiNH_2$ , 10g  $LiBr$ , and 15g  $Pd / Al_2O_3$  was about 160W with an energy balance of  $\Delta H = -19.1 \text{ kJ}$ . The observed energy balance was 4.4 times the maximum theoretical based on known chemistry. Next, Raney nickel (R-Ni) served as a dissociator when the power reaction mixture was used in chemical synthesis wherein  $LiBr$  acted as a getter of the catalysis product  $H(1/4)$  to form  $LiH * X$  as well as to trap  $H_2(1/4)$  in the crystal. The ToF-SIMs showed  $LiH * X$  peaks. The  $^1H$  MAS NMR  $LiH * Br$  and  $LiH * I$  showed a large distinct upfield resonance at about  $-2.5 \text{ ppm}$  that matched  $H^-(1/4)$  in a  $LiX$  matrix. An NMR peak at 1.13 ppm matched interstitial  $H_2(1/4)$ , and the rotation frequency of  $H_2(1/4)$  of  $4^2$  times that of ordinary  $H_2$  was observed at  $1989 \text{ cm}^{-1}$  in the FTIR spectrum. The

XPS spectrum recorded on the  $LiH * Br$  crystals showed peaks at about 9.5 eV and 12.3 eV that could not be assigned to any known elements based on the absence of any other primary element peaks, but matched the binding energy of  $H^-(1/4)$  in two chemical environments. A further signature of the energetic process was the

5 observation of the formation of a plasma called a resonant transfer- or rt-plasma at low temperatures (e.g.  $\approx 10^3 K$ ) and very low field strengths of about 1-2 V/cm when atomic  $Li$  was present with atomic hydrogen. Time-dependent line broadening of the H Balmer  $\alpha$  line was observed corresponding to extraordinarily fast H ( $>40$  eV).

$NaH$  uniquely achieves high kinetics since the catalyst reaction relies on the

10 release of the intrinsic  $H$ , which concomitantly undergoes the transition to form  $H(1/3)$  that further reacts to form  $H(1/4)$ . High-temperature differential scanning calorimetry (DSC) was performed on ionic  $NaH$  under a helium atmosphere at an extremely slow temperature ramp rate ( $0.1^\circ C/min$ ) to increase the amount of molecular  $NaH$  formation. A novel exothermic effect of  $-177 kJ / mole NaH$  was

15 observed in the temperature range of  $640^\circ C$  to  $825^\circ C$ . To achieve high power, R-Ni having a surface area of about  $100 m^2 / g$  was surface-coated with  $NaOH$  and reacted with  $Na$  metal to form  $NaH$ . Using water-flow, batch calorimetry, the measured power from 15g of R-Ni was about 0.5 kW with an energy balance of  $\Delta H = -36 kJ$  compared to  $\Delta H \approx 0 kJ$  from the R-Ni starting material, R-NiAl alloy,

20 when reacted with  $Na$  metal. The observed energy balance of the  $NaH$  reaction was  $-1.6 \times 10^4 kJ / mole H_2$ , over 66 times the  $-241.8 kJ / mole H_2$  enthalpy of combustion.

The ToF-SIMS showed sodium hydride,  $NaH_x$ , peaks. The  $^1H$  MAS NMR spectra of  $NaH * Br$  and  $NaH * Cl$  showed large distinct upfield resonance at

–3.6 ppm and –4 ppm, respectively, that matched  $H^-(1/4)$ , and an NMR peak at 1.1 ppm matched  $H_2(1/4)$ .  $NaH * Cl$  from reaction of  $NaCl$  and the solid acid  $KHSO_4$  as the only source of hydrogen comprised two fractional hydrogen states. The  $H^-(1/4)$  NMR peak was observed at –3.97 ppm, and the  $H^-(1/3)$  peak was also present at –3.15 ppm. The corresponding  $H_2(1/4)$  and  $H_2(1/3)$  peaks were observed at 1.15 ppm and 1.7 ppm, respectively. The XPS spectrum recorded on  $NaH * Br$  showed the  $H^-(1/4)$  peaks at about 9.5 eV and 12.3 eV that matched the results from  $LiH * Br$  and  $KH * I$ ; whereas, sodium hydride showed two fractional hydrogen states additionally having the  $H^-(1/3)$  XPS peak at 6 eV in the absence of a halide peak. The predicted rotational transitions having energies of  $4^2$  times those of ordinary  $H_2$  were also observed from  $H_2(1/4)$  which was excited using a 12.5 keV electron beam.

### I. Introduction

Mills [1-12] solved the structure of the bound electron using classical laws and subsequently developed a unification theory based on those laws called the Grand Unified Theory of Classical Physics (GUTCP) with results that match observations for the basic phenomena of physics and chemistry from the scale of the quarks to cosmos. This paper is the first in a series of two that covers two specific predictions of GUTCP involving the existence of lower-energy states of the hydrogen atom, which represents a powerful new energy source and the transitions of atomic hydrogen to lower-energy states [2].

GUTCP predicts a reaction involving a resonant, nonradiative energy transfer from otherwise stable atomic hydrogen to a catalyst capable of accepting the energy

to form hydrogen in lower-energy states than previously thought possible.

Specifically, the product is  $H(1/p)$ , fractional Rydberg states of atomic hydrogen

wherein  $n = \frac{1}{2}, \frac{1}{3}, \frac{1}{4}, \dots, \frac{1}{p}$  ( $p \leq 137$  is an integer) replaces the well known parameter

$n = \text{integer}$  in the Rydberg equation for hydrogen excited states.  $He^+$ ,  $Ar^+$ ,  $Sr^+$ ,  $Li$ ,

- 5  $K$ , and  $NaH$  are predicted to serve as catalysts since they meet the catalyst criterion—a chemical or physical process with an enthalpy change equal to an integer multiple of the potential energy of atomic hydrogen,  $27.2 \text{ eV}$ . The data from a broad spectrum of investigational techniques strongly and consistently support the existence of these states called hydrino, for “small hydrogen”, and the corresponding
- 10 diatomic molecules called dihydrino molecules. Some of these prior related studies supporting the possibility of a novel reaction of atomic hydrogen, which produces hydrogen in fractional quantum states that are at lower energies than the traditional “ground” ( $n = 1$ ) state, include extreme ultraviolet (EUV) spectroscopy, characteristic emission from catalysts and the hydride ion products, lower-energy hydrogen
- 15 emission, chemically-formed plasmas, Balmer  $\alpha$  line broadening, population inversion of H lines, elevated electron temperature, anomalous plasma afterglow duration, power generation, and analysis of novel chemical compounds [13-40].

- Recently, there has been the announcement of some unexpected astrophysical results that support the existence of hydrinos. In 1995, Mills
- 20 published the GUTCP prediction [41] that the expansion of the universe was accelerating from the same equations that correctly predicted the mass of the top quark before it was measured. To the astonishment of cosmologists, this was confirmed by 2000. Mills made another prediction about the nature of dark matter based on GUTCP that may be close to being confirmed. Based on recent evidence,

Bournaud et al. [42-43] suggest that dark matter is hydrogen in dense molecular form that somehow behaves differently in terms of being unobservable except by its gravitational effects. Theoretical models predict that dwarfs formed from collisional debris of massive galaxies should be free of nonbaryonic dark matter. So, their gravity should tally with the stars and gas within them. By analyzing the observed gas kinematics of such recycled galaxies, Bournaud et al. [42-43] have measured the gravitational masses of a series of dwarf galaxies lying in a ring around a massive galaxy that has recently experienced a collision. Contrary to the predictions of Cold-Dark-Matter (CDM) theories, their results demonstrate that they contain a massive dark component amounting to about twice the visible matter. This baryonic dark matter is argued to be cold molecular hydrogen, but it is distinguished from ordinary molecular hydrogen in that it is not traced at all by traditional methods, such as emission of CO lines. These results match the predictions of the dark matter being dihydrino molecules.

Emission lines recorded on cold interstellar regions containing dark matter matched  $H(1/p)$ , fractional Rydberg states of atomic hydrogen given by Eqs. (2a) and (2c) [29]. Such emission lines with energies of  $q \cdot 13.6 \text{ eV}$ , where  $q = 1, 2, 3, 4, 6, 7, 8, 9, \text{ or } 11$  were also observed by extreme ultraviolet (EUV) spectroscopy recorded on microwave discharges of helium with 2% hydrogen [27-29]. These  $He^+$  fulfills the catalyst criterion—a chemical or physical process with an enthalpy change equal to an integer multiple of  $27.2 \text{ eV}$  since it ionizes at  $54.417 \text{ eV}$ , which is  $2 \cdot 27.2 \text{ eV}$ . The product of the catalysis reaction of  $He^+$ ,  $H(1/3)$ , may further serve as a catalyst to lead to transitions to other states  $H(1/p)$ .

J. R. Rydberg showed that all of the spectral lines of atomic hydrogen were given by a completely empirical relationship:

$$\bar{\nu} = R \left( \frac{1}{n_f^2} - \frac{1}{n_i^2} \right) \quad (1)$$

where  $R = 109,677 \text{ cm}^{-1}$ ,  $n_f = 1, 2, 3, \dots$ ,  $n_i = 2, 3, 4, \dots$  and  $n_i > n_f$ . Bohr, Schrödinger, and Heisenberg, each developed a theory for atomic hydrogen that gave the energy levels in agreement with Rydberg's equation.

$$E_n = -\frac{e^2}{n^2 8\pi\epsilon_0 a_H} = -\frac{13.598 \text{ eV}}{n^2} \quad (2a)$$

$$n = 1, 2, 3, \dots \quad (2b)$$

where  $e$  is the elementary charge,  $\epsilon_0$  is the permittivity of vacuum, and  $a_H$  is the radius of the hydrogen atom. The excited energy states of atomic hydrogen are given by Eq. (2a) for  $n > 1$  in Eq. (2b). The  $n = 1$  state is the "ground" state for "pure" photon transitions (i.e. the  $n = 1$  state can absorb a photon and go to an excited electronic state, but it cannot release a photon and go to a lower-energy electronic state). However, an electron transition from the ground state to a lower-energy state may be possible by a resonant nonradiative energy transfer such as multipole coupling or a resonant collision mechanism. Processes such as hydrogen molecular bond formation that occur without photons and that require collisions are common [44]. Also, some commercial phosphors are based on resonant nonradiative energy transfer involving multipole coupling [45].

The theory reported previously [1, 13-40] predicts that atomic hydrogen may undergo a catalytic reaction with certain atoms, excimers, ions, and diatomic hydrides which provide a reaction with a net enthalpy of an integer multiple of the potential energy of atomic hydrogen,  $E_h = 27.2 \text{ eV}$  where  $E_h$  is one Hartree. Specific species (e.g.  $\text{He}^+$ ,  $\text{Ar}^+$ ,  $\text{Sr}^+$ ,  $\text{K}$ ,  $\text{Li}$ ,  $\text{HCl}$ , and  $\text{NaH}$ ) identifiable on the basis of their known electron energy levels are required to be present with atomic hydrogen

to catalyze the process. The reaction involves a nonradiative energy transfer followed by  $q \cdot 13.6 \text{ eV}$  emission or  $q \cdot 13.6 \text{ eV}$  transfer to H to form extraordinarily hot, excited-state H [13-17, 19-20, 32-39] and a hydrogen atom that is lower in energy than unreacted atomic hydrogen that corresponds to a fractional principal quantum number. That is

$$n = 1, \frac{1}{2}, \frac{1}{3}, \frac{1}{4}, \dots, \frac{1}{p}; \quad p \leq 137 \text{ is an integer} \quad (2c)$$

replaces the well known parameter  $n = \text{integer}$  in the Rydberg equation for hydrogen excited states. The  $n = 1$  state of hydrogen and the  $n = \frac{1}{\text{integer}}$  states of hydrogen are nonradiative, but a transition between two nonradiative states, say  $n = 1$  to  $n = 1/2$ , is possible via a nonradiative energy transfer. Thus, a catalyst provides a net positive enthalpy of reaction of  $m \cdot 27.2 \text{ eV}$  (i.e. it resonantly accepts the nonradiative energy transfer from hydrogen atoms and releases the energy to the surroundings to affect electronic transitions to fractional quantum energy levels). As a consequence of the nonradiative energy transfer, the hydrogen atom becomes unstable and emits further energy until it achieves a lower-energy nonradiative state having a principal energy level given by Eqs. (2a) and (2c).

The catalyst product,  $H(1/p)$ , may also react with an electron to form a novel hydride ion  $H^-(1/p)$  with a binding energy  $E_B$  [1, 13-14, 18, 30]:

$$E_B = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi\mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right) \quad (3)$$

where  $p = \text{integer} > 1$ ,  $s = 1/2$ ,  $\hbar$  is Planck's constant bar,  $\mu_0$  is the permeability of vacuum,  $m_e$  is the mass of the electron,  $\mu_e$  is the reduced electron mass given by

$$\mu_e = \frac{m_e m_p}{\frac{m_e}{\sqrt{\frac{3}{4}}} + m_p} \text{ where } m_p \text{ is the mass of the proton, } a_o \text{ is the Bohr radius, and the}$$

ionic radius is  $r_1 = \frac{a_0}{p} (1 + \sqrt{s(s+1)})$ . From Eq. (3), the calculated ionization energy of the hydride ion is 0.75418 eV, and the experimental value given by Lykke [46] is  $6082.99 \pm 0.15 \text{ cm}^{-1}$  (0.75418 eV).

- 5 Upfield-shifted NMR peaks are a direct evidence of the existence of lower-energy state hydrogen with a reduced radius relative to ordinary hydride ion and having an increase in diamagnetic shielding of the proton. The shift is given by the sum of that of ordinary hydride ion  $H^-$  and a component due to the lower -energy state [1, 15]:

$$10 \quad \frac{\Delta B_T}{B} = -\mu_0 \frac{e^2}{12m_e a_0 (1 + \sqrt{s(s+1)})} (1 + \alpha 2\pi p) = -(29.9 + 1.37p) \text{ ppm} \quad (4)$$

where for  $H^-$   $p=0$  and  $p = \text{integer} > 1$  for  $H^-(1/p)$  and  $\alpha$  is the fine structure constant.

$H(1/p)$  may react with a proton and two  $H(1/p)$  may react to form

$H_2(1/p)^+$  and  $H_2(1/p)$ , respectively. The hydrogen molecular ion and molecular

- 15 charge and current density functions, bond distances, and energies were solved previously [1, 6] from the Laplacian in ellipsoidal coordinates with the constraint of nonradiation.

$$(\eta - \zeta)R_\xi \frac{\partial}{\partial \xi} (R_\xi \frac{\partial \phi}{\partial \xi}) + (\zeta - \xi)R_\eta \frac{\partial}{\partial \eta} (R_\eta \frac{\partial \phi}{\partial \eta}) + (\xi - \eta)R_\zeta \frac{\partial}{\partial \zeta} (R_\zeta \frac{\partial \phi}{\partial \zeta}) = 0 \quad (5)$$

The total energy  $E_T$  of the hydrogen molecular ion having a central field of  $+pe$  at

- 20 each focus of the prolate spheroid molecular orbital is

$$E_T = -p^2 \left\{ \frac{e^2}{8\pi\epsilon_o a_H} (4 \ln 3 - 1 - 2 \ln 3) \left[ 1 + p \sqrt{\frac{2\hbar \sqrt{\frac{2e^2}{4\pi\epsilon_o (2a_H)^3}}}{m_e c^2}} \right] - \frac{1}{2} \hbar \sqrt{\frac{k}{\mu}} \right\} \quad (6)$$

$$= -p^2 16.13392 \text{ eV} - p^3 0.118755 \text{ eV}$$

where  $p$  is an integer,  $c$  is the speed of light in vacuum,  $\mu$  is the reduced nuclear mass, and  $k$  is the harmonic force constant solved previously in a closed-form equation with fundamental constants only [1, 6]. The total energy of the hydrogen molecule having a central field of  $+pe$  at each focus of the prolate spheroid molecular orbital is

$$E_T = -p^2 \left\{ \frac{e^2}{8\pi\epsilon_o a_0} \left[ \left( 2\sqrt{2} - \sqrt{2} + \frac{\sqrt{2}}{2} \right) \ln \frac{\sqrt{2}+1}{\sqrt{2}-1} - \sqrt{2} \right] \left[ 1 + p \sqrt{\frac{2\hbar \sqrt{\frac{e^2}{4\pi\epsilon_o a_0^3}}}{m_e c^2}} \right] - \frac{1}{2} \hbar \sqrt{\frac{k}{\mu}} \right\}$$

$$= -p^2 31.351 \text{ eV} - p^3 0.326469 \text{ eV} \quad (7)$$

The bond dissociation energy,  $E_D$ , of hydrogen molecule  $H_2(1/p)$  is the difference between the total energy of the corresponding hydrogen atoms and  $E_T$

$$E_D = E(2H(1/p)) - E_T \quad (8)$$

where [47]

$$E(2H(1/p)) = -p^2 27.20 \text{ eV} \quad (9)$$

$E_D$  is given by Eqs. (8-9) and (7):

$$E_D = -p^2 27.20 \text{ eV} - E_T$$

$$= -p^2 27.20 \text{ eV} - (-p^2 31.351 \text{ eV} - p^3 0.326469 \text{ eV}) \quad (10)$$

$$= p^2 4.151 \text{ eV} + p^3 0.326469 \text{ eV}$$

The calculated and experimental parameters of  $H_2$ ,  $D_2$ ,  $H_2^+$ , and  $D_2^+$  from Ref. [1, 6] are given in TABLE 3.

TABLE 3. The Maxwellian closed-form calculated and experimental parameters of

5  $H_2$ ,  $D_2$ ,  $H_2^+$  and  $D_2^+$ .

| Parameter                                | Calculated                              | Experimental                            |
|------------------------------------------|-----------------------------------------|-----------------------------------------|
| $H_2$ Bond Energy                        | 4.478 eV                                | 4.478 eV                                |
| $D_2$ Bond Energy                        | 4.556 eV                                | 4.556 eV                                |
| $H_2^+$ Bond Energy                      | 2.654 eV                                | 2.651 eV                                |
| $D_2^+$ Bond Energy                      | 2.696 eV                                | 2.691 eV                                |
| $H_2$ Total Energy                       | 31.677 eV                               | 31.675 eV                               |
| $D_2$ Total Energy                       | 31.760 eV                               | 31.760 eV                               |
| $H_2$ Ionization Energy                  | 15.425 eV                               | 15.426 eV                               |
| $D_2$ Ionization Energy                  | 15.463 eV                               | 15.466 eV                               |
| $H_2^+$ Ionization Energy                | 16.253 eV                               | 16.250 eV                               |
| $D_2^+$ Ionization Energy                | 16.299 eV                               | 16.294 eV                               |
| $H_2^+$ Magnetic Moment                  | $9.274 \times 10^{-24} \text{ JT}^{-1}$ | $9.274 \times 10^{-24} \text{ JT}^{-1}$ |
|                                          | $(\mu_B)$                               | $(\mu_B)$                               |
| Absolute $H_2$ Gas-Phase<br>NMR Shift    | -28.0 ppm                               | -28.0 ppm                               |
| $H_2$ Internuclear Distance <sup>a</sup> | 0.748 Å<br>$\sqrt{2}a_o$                | 0.741 Å                                 |
| $D_2$ Internuclear Distance <sup>a</sup> | 0.748 Å<br>$\sqrt{2}a_o$                | 0.741 Å                                 |
| $H_2^+$ Internuclear Distance            | 1.058 Å<br>$2a_o$                       | 1.06 Å                                  |

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|                                                   |                         |                          |
|---------------------------------------------------|-------------------------|--------------------------|
| $D_2^+$ Internuclear Distance <sup>a</sup>        | 1.058 Å                 | 1.0559 Å                 |
|                                                   | $2a_o$                  |                          |
| $H_2$ Vibrational Energy                          | 0.517 eV                | 0.516 eV                 |
| $D_2$ Vibrational Energy                          | 0.371 eV                | 0.371 eV                 |
| $H_2$ $\omega_e x_e$                              | $120.4 \text{ cm}^{-1}$ | $121.33 \text{ cm}^{-1}$ |
| $D_2$ $\omega_e x_e$                              | $60.93 \text{ cm}^{-1}$ | $61.82 \text{ cm}^{-1}$  |
| $H_2^+$ Vibrational Energy                        | 0.270 eV                | 0.271 eV                 |
| $D_2^+$ Vibrational Energy                        | 0.193 eV                | 0.196 eV                 |
| $H_2$ J=1 to J=0 Rotational Energy <sup>a</sup>   | 0.0148 eV               | 0.01509 eV               |
| $D_2$ J=1 to J=0 Rotational Energy <sup>a</sup>   | 0.00741 eV              | 0.00755 eV               |
| $H_2^+$ J=1 to J=0 Rotational Energy              | 0.00740 eV              | 0.00739 eV               |
| $D_2^+$ J=1 to J=0 Rotational Energy <sup>a</sup> | 0.00370 eV              | 0.003723 eV              |

<sup>a</sup> Not corrected for the slight reduction in internuclear distance due to  $\bar{E}_{osc}$ .

The  $^1H$  NMR resonance of  $H_2(1/p)$  is predicted to be upfield from that of  $H_2$  due to the fractional radius in elliptic coordinates [1, 6] wherein the electrons are

5 significantly closer to the nuclei. The predicted shift,  $\frac{\Delta B_r}{B}$ , for  $H_2(1/p)$  derived previously [1, 6] is given by the sum of that of  $H_2$  and a term that depends on

$p = \text{integer} > 1$  for  $H_2(1/p)$ :

$$\frac{\Delta B_r}{B} = -\mu_0 \left( 4 - \sqrt{2} \ln \frac{\sqrt{2} + 1}{\sqrt{2} - 1} \right) \frac{e^2}{36a_0 m_e} (1 + \pi \phi p) \quad (11)$$

$$\frac{\Delta B_r}{B} = -(28.01 + 0.64 p) \text{ ppm} \quad (12)$$

10 where for  $H_2$   $p = 0$ .

The vibrational energies,  $E_{vib}$ , for the  $v = 0$  to  $v = 1$  transition of hydrogen-type molecules  $H_2(1/p)$  are [1, 6]

$$E_{vib} = p^2 0.515902 \text{ eV} \quad (13)$$

where  $p$  is an integer and the experimental vibrational energy for the  $v = 0$  to  $v = 1$  transition of  $H_2$ ,  $E_{H_2(v=0 \rightarrow v=1)}$ , is given by Beutler [48] and Herzberg [49].

The rotational energies,  $E_{rot}$ , for the  $J$  to  $J + 1$  transition of hydrogen-type molecules  $H_2(1/p)$  are [1, 6]

$$E_{rot} = E_{J+1} - E_J = \frac{\hbar^2}{I} [J + 1] = p^2 (J + 1) 0.01509 \text{ eV} \quad (14)$$

where  $p$  is an integer,  $I$  is the moment of inertia, and the experimental rotational energy for the  $J = 0$  to  $J = 1$  transition of  $H_2$  is given by Atkins [50].

The  $p^2$  dependence of the rotational energies results from an inverse  $p$  dependence of the internuclear distance and the corresponding impact on the moment of inertia  $I$ . The predicted internuclear distance  $2c'$  for  $H_2(1/p)$  is

$$2c' = \frac{a_o \sqrt{2}}{p} \quad (15)$$

The formation of new states of hydrogen is very energetic. A new chemically generated or assisted plasma source based on the resonant energy transfer mechanism (rt-plasma) has been developed that may be a new power source. One such source operates by incandescently heating a hydrogen dissociator and a catalyst to provide atomic hydrogen and gaseous catalyst, respectively, such that the catalyst reacts with the atomic hydrogen to produce a plasma. It was extraordinary that intense EUV emission was observed by Mills et al. [13-21, 38-39] at low temperatures (e.g.  $\approx 10^3 \text{ K}$ ), as well as an extraordinary low field strength of about 1-

2 V/cm from atomic hydrogen and certain atomized elements or certain gaseous ions, which singly or multiply ionize at integer multiples of the potential energy of atomic hydrogen, 27.2 eV.

$K$  to  $K^{3+}$  provides a reaction with a net enthalpy equal to three times the  
5 potential energy of atomic hydrogen. It was reported previously [13-21, 38-39] that the presence of these gaseous atoms with thermally dissociated hydrogen formed an rt-plasma having strong EUV emission with a stationary inverted Lyman population. Other noncatalyst metals such as  $Mg$  produced no plasma. Significant line broadening of the Balmer  $\alpha$ ,  $\beta$ , and  $\gamma$  lines of 18 eV was observed. Emission  
10 from rt-plasmas occurred even when the electric field applied to the plasma was zero. Since a conventional discharge power source was not present, the formation of a plasma would require an energetic reaction. The origin of Doppler broadening is the relative thermal motion of the emitter with respect to the observer. Line broadening is a measure of the atom temperature, and a significant increase was  
15 expected and observed for catalysts,  $K$  as well as  $Sr^+$  or  $Ar^+$  [13-21, 38-39], with hydrogen. The observation of a high hydrogen temperature with no conventional explanation would indicate that an rt-plasma must have a source of free energy. An energetic chemical reaction was further implicated since it was found that the broadening is time dependent [13-14, 20]. Therefore, the thermal power balance  
20 was measured calorimetrically. The reaction was exothermic since excess power of  $20\text{ mW} \cdot \text{cm}^{-3}$  was measured by Calvet calorimetry [20]. In further experiments,  $KNO_3$  and Raney nickel were used as a source of  $K$  catalyst and atomic hydrogen, respectively, to produce the corresponding exothermic reaction. The energy balance was  $\Delta H = -17,925\text{ kcal / mole } KNO_3$ , about 300 times that expected for the most  
25 energetic known chemistry of  $KNO_3$ , and  $-3585\text{ kcal / mole } H_2$ , over 60 times the

hypothetical maximum enthalpy of  $-57.8 \text{ kcal / mole } H_2$  due to combustion of hydrogen with atmospheric oxygen, assuming the maximum possible  $H_2$  inventory [14]. Additional substantial evidence of an energetic catalytic reaction was previously reported [13-15, 24-26, 30-31] involving a resonant energy transfer between hydrogen atoms and  $K$  to form very stable novel hydride ions and molecules  $H^-(1/4)$  and  $H_2(1/4)$ , respectively. Characteristic emission was observed from  $K^{3+}$  that confirmed the resonant nonradiative energy transfer of  $3 \cdot 27.2 \text{ eV}$  from atomic hydrogen to  $K$  that served as a predicted catalyst. From Eq. (3), the binding energy  $E_b$  of  $H^-(1/4)$  is

$$E_b = 11.232 \text{ eV} \quad (\lambda_{vac} = 110.38 \text{ nm}) \quad (16)$$

The product hydride ion  $H^-(1/4)$  was observed by EUV spectroscopy at  $110 \text{ nm}$  corresponding to its predicted binding energy of  $11.2 \text{ eV}$  [13-15, 24-26, 30-31]. The identification of  $H^-(1/4)$  was confirmed previously by the XPS measurement of its binding energy. The XPS spectrum of  $KH^*I$  differed from that of  $KI$  by having additional features at  $8.9 \text{ eV}$  and  $10.8 \text{ eV}$  that did not correspond to any other primary element peaks but did match the  $H^-(1/4) E_b = 11.2 \text{ eV}$  hydride ion (Eq. (3)) in two different chemical environments. The  $^1H$  MAS NMR spectrum of novel compound  $KH^*Cl$  relative to external tetramethylsilane (TMS) showed a large distinct upfield resonance at  $-4.4 \text{ ppm}$  corresponding to an absolute resonance shift of  $-35.9 \text{ ppm}$  that matched the theoretical prediction of  $p = 4$  [13-15, 25-26, 30-31]. Elemental analysis identified [13-15, 25-26, 30-31] these compounds as only containing the alkaline metal, halogen, and hydrogen, and no known hydride compound of this composition could be found in the literature that had an upfield-shifted hydride NMR peak. Ordinary alkali hydrides alone or mixed with alkali

halides show down-field shifted peaks [13-15, 25-26, 30-31]. From the literature, the list of alternatives to  $H^-(1/p)$  as a possible source of the upfield NMR peaks was limited to U centered H. This was eliminated by the absence of the intense and characteristic infrared vibration band at  $503\text{ cm}^{-1}$  due to the substitution of  $H^-$  for  $Cl^-$  in  $KCl$  [51].

As a further characterization, FTIR analysis of  $KH^*I$  crystals with  $H^-(1/4)$  was performed and interstitial  $H_2(1/4)$  having a predicted rotational energy given by Eq. (14) was observed. Rotational lines were observed previously [13-14] in the 145-300 nm region from atmospheric pressure electron beam-excited argon—hydrogen plasmas. The unprecedented energy spacing of  $4^2$  times that of hydrogen established the internuclear distance as  $1/4$  that of  $H_2$  and identified  $H_2(1/4)$  (Eqs. (13-15)). The spectrum was asymmetric with the P branch dominant corresponding to the absence of populated rotational states in the excited  $v = 1$  vibrational state. This was due to the high rotational energy (10 times the thermal energy), the short lifetime of the rotational excited states, and the low cross section for electron-beam rotational excitation; whereas, the vibrational dipole excitation was allowed. Thus, only the  $v = 1, J = 0$  state was populated significantly from e-beam excitation, and transitions occurred with  $\Delta J > 0$  during the  $v = 1$  to  $v = 0$  transition.  $KH^*Cl$  having  $H^-(1/4)$  by NMR was incident to the 12.5 keV electron beam, which excited similar emission of interstitial  $H_2(1/4)$  as observed in the argon—hydrogen plasma [13-14]. Specifically,  $H_2(1/4)$  trapped in the lattice of  $KH^*Cl$  was investigated by windowless EUV spectroscopy on electron-beam excitation of the crystals using the 12.5 keV electron gun at pressures below which any gas could produce detectable emission ( $<10^{-5}$  Torr). The rotational energy of  $H_2(1/4)$  was

confirmed by this technique as well. These results confirmed the previous observations from the plasmas formed by the energetic hydrino-forming reaction having intense hydrogen Lyman emission, a stationary inverted Lyman population, excessive afterglow duration, highly energetic hydrogen atoms, characteristic alkali-ion emission due to catalysis, predicted novel spectral lines, and the measurement of a power beyond any conventional chemistry [13-40] that matched predictions for a catalytic reaction of atomic hydrogen to form more stable hydride ions designated  $H^-(1/p)$ . Since the comparison of theory and experimental energies is direct evidence of lower-energy hydrogen with an implicit large exotherm during its formation, we report in this paper the results when these experiments were repeated with additionally predicted catalysts  $Li$  and  $NaH$ .

A catalytic system used to make and analyzed predicted hydride compounds involves lithium atoms. The first and second ionization energies of lithium are 5.39172 eV and 75.64018 eV, respectively [52]. The double ionization ( $t = 2$ ) reaction of  $Li$  to  $Li^{2+}$  then, has a net enthalpy of reaction of 81.0319 eV, which is equivalent to 3·27.2 eV.

$$81.0319 \text{ eV} + Li(m) + H\left[\frac{a_H}{p}\right] \rightarrow Li^{2+} + 2e^- + H\left[\frac{a_H}{(p+3)}\right] + [(p+3)^2 - p^2] \cdot 13.6 \text{ eV} \quad (17)$$

$$Li^{2+} + 2e^- \rightarrow Li(m) + 81.0319 \text{ eV} \quad (18)$$

And, the overall reaction is

$$H\left[\frac{a_H}{p}\right] \rightarrow H\left[\frac{a_H}{(p+3)}\right] + [(p+3)^2 - p^2] \cdot 13.6 \text{ eV} \quad (19)$$

Lithium is a metal in the solid and liquid states, and the gas comprises covalent  $Li_2$  molecules [53], each having a bond energy of 110.4 kJ/mole [54]. In

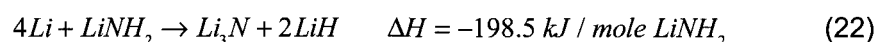
order to generate atomic lithium,  $LiNH_2$  was added to the reaction mixture.  $LiNH_2$  generates atomic hydrogen as well, according to the reversible reactions [55-64]:



and



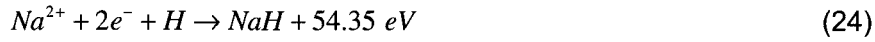
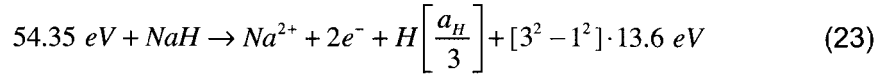
The energy for the reaction of lithium amide to lithium nitride and lithium hydride is exothermic [65-66]:



Thus, it should occur to a significant extent. The specific predictions of the energetic reaction given by Eqs. (17-19) were tested by rt-plasma formation and  $H$  line broadening. The power developed was measured using water-flow, batch calorimetry. Then, the predicted products of  $H^-(1/4)$  and  $H_2(1/4)$  having the energies given by Eqs. (3) and (5-15), respectively, were tested by magic angle solid proton nuclear magnetic resonance spectroscopy (MAS  $^1H$  NMR), X-ray photoelectron spectroscopy (XPS), time of flight secondary ion mass spectroscopy (ToF-SIMS), and Fourier transform infrared spectroscopy (FTIR).

A compound comprising hydrogen such as  $MH$ , where  $M$  is element other than hydrogen, serves as a source of hydrogen and a source of catalyst. A catalytic reaction is provided by the breakage of the  $M-H$  bond plus the ionization of  $t$  electrons from the atom  $M$  each to a continuum energy level such that the sum of the bond energy and ionization energies of the  $t$  electrons is approximately  $m \cdot 27.2 \text{ eV}$ , where  $m$  is an integer. One such catalytic system involves sodium. The bond energy of  $NaH$  is  $1.9245 \text{ eV}$  [54], and the first and second ionization energies of  $Na$  are  $5.13908 \text{ eV}$  and  $47.2864 \text{ eV}$ , respectively [52]. Based on these energies  $NaH$  molecule can serve as a catalyst and  $H$  source, since the bond

energy of  $NaH$  plus the double ionization ( $t=2$ ) of  $Na$  to  $Na^{2+}$  is  $54.35 \text{ eV}$  ( $2 \cdot 27.2 \text{ eV}$ ). The catalyst reactions are given by



5 And, the overall reaction is



As given in Chp. 5 of Ref [1], and Ref. [29], hydrogen atoms

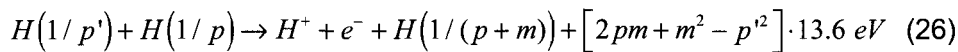
$H(1/p)$   $p=1,2,3,\dots,137$  can undergo further transitions to lower-energy states given

by Eqs. (2a) and (2c) wherein the transition of one atom is catalyzed by a second

10 that resonantly and nonradiatively accepts  $m \cdot 27.2 \text{ eV}$  with a concomitant opposite change in its potential energy. The overall general equation for the transition of

$H(1/p)$  to  $H(1/(p+m))$  induced by a resonance transfer of  $m \cdot 27.2 \text{ eV}$  to  $H(1/p')$

is represented by



15 In the case of a high hydrogen atom concentration, the transition of  $H(1/3)$  ( $p=3$ )

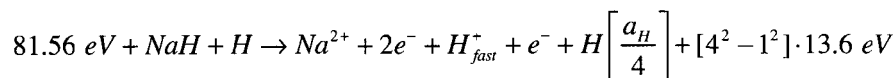
to  $H(1/4)$  ( $p+m=4$ ) with  $H$  as the catalyst ( $p=1$ ;  $m=1$ ) can be fast:



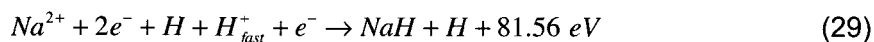
The  $NaH$  catalyst reactions may be concerted since the sum of the bond energy of

$NaH$ , the double ionization ( $t=2$ ) of  $Na$  to  $Na^{2+}$ , and the potential energy of  $H$  is

20  $81.56 \text{ eV}$  ( $3 \cdot 27.2 \text{ eV}$ ). The catalyst reactions are given by



(28)



And, the overall reaction is

$$H \rightarrow H \left[ \frac{a_H}{4} \right] + [4^2 - 1^2] \cdot 13.6 \text{ eV} \quad (30)$$

5 where  $H_{fast}^{+}$  is a fast hydrogen atom having at least 13.6 eV of kinetic energy.

$H^{-}(1/4)$  forms stable halidohydrides and is a favored product together with the

corresponding molecule formed by the reactions  $2H(1/4) \rightarrow H_2(1/4)$  and

$H^{-}(1/4) + H^{+} \rightarrow H_2(1/4)$  [13-15, 24-26, 30-31]. The corresponding hydrino atom

$H(1/4)$  is a preferred final product consistent with observation since the  $p = 4$

10 quantum state has a multipolarity greater than that of a quadrupole giving it a long

theoretical lifetime.  $H(1/4)$  may be formed directly from  $H$  (e.g. Eqs. (36-38)) or

via multiple transitions (e.g. Eqs. (23-27)). In the latter case, the higher-energy

$H(1/p)$  states with quantum numbers  $p = 2; \ell = 0, 1$  and  $p = 3; \ell = 0, 1, 2$

corresponding to dipole and quadrupole transitions, respectively, have theoretically

15 allowed, fast transitions.

Sodium hydride is typically in the form of an ionic crystalline compound

formed by the reaction of gaseous hydrogen with metallic sodium. And, in the

gaseous state, sodium comprises covalent  $Na_2$  molecules [53] with a bond energy of

74.8048 kJ/mole [54]. It was found that when  $NaH(s)$  was heated at a very slow

20 temperature ramp rate (0.1°C/min) under a helium atmosphere to form  $NaH(g)$ , the

predicted exothermic reaction given by Eqs. (23-25) was observed at high

temperature by differential scanning calorimetry (DSC). To achieve high power, a

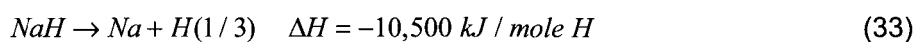
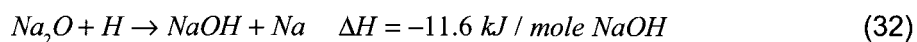
chemical system was designed to greatly increase the amount and rate of formation

of  $NaH(g)$ . The reaction of  $NaOH$  and  $Na$  to  $Na_2O$  and  $NaH(s)$  calculated from the heats of formation [54, 65] releases  $\Delta H = -44.7 \text{ kJ / mole } NaOH$ :

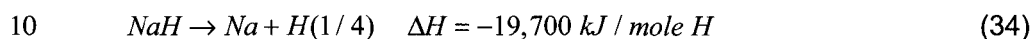


This exothermic reaction can drive the formation of  $NaH(g)$  and was exploited to

- 5 drive the very exothermic reaction given by Eqs. (23-25). The regenerative reaction in the presence of atomic hydrogen is



and



Thus, a small amount of  $NaOH$ ,  $Na$ , and atomic hydrogen serves as a catalytic source of the  $NaH$  catalyst that in turn forms a large yield of hydridos via multiple cycles of regenerative reactions such as those given by Eqs. (31-34). R-Ni having a high surface area of about  $100 \text{ m}^2 / \text{g}$  and containing  $H$  was surface coated with

- 15  $NaOH$  and reacted with  $Na$  metal to form  $NaH(g)$ . Since the energy balance in the formation of  $NaH(g)$  was negligible due to the small amounts involved, the energy and power due to the hydrido reactions given by Eqs. (23-25) were specifically measured using water-flow, batch calorimetry. Next, R-Ni 2400 was prepared such that it comprised about 0.5 wt%  $NaOH$ , and the  $Al$  of the intermetallic served as the
- 20 reductant to form  $NaH$  catalyst during calorimetry measurement. The reaction of

$NaOH + Al$  to  $Al_2O_3 + NaH$  calculated from the heats of formation [65] is

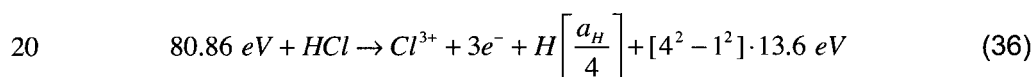
exothermic by  $\Delta H = -189.1 \text{ kJ / mole } NaOH$ . The balanced reaction is given by



This exothermic reaction can drive the formation of  $NaH(g)$  and was exploited to drive the very exothermic reaction given by Eqs. (23-25) wherein the regeneration of  $NaH$  occurred from  $Na$  in the presence of atomic hydrogen. For 0.5wt%  $NaOH$ , the exothermic reaction given by Eq. (35) gave a negligible  $\Delta H = -0.024 \text{ kJ}$  background heat during measurement.

It was reported previously [28-29] that the reaction products  $H(1/p)$  may undergo further reaction to lower-energy states. For example, the catalyst reaction of  $Ar^+$  to  $Ar^{2+}$  forms  $H(1/2)$ , which may further serve as both a catalyst and a reactant to form  $H(1/4)$  [1, 13-14, 28-29] and the corresponding favored molecule  $H_2(1/4)$ , observed using different catalysts [13-14]. Thus, predicted products of  $NaH$  catalyst from Eqs. (23-25) and Table 1 of Ref. [29] are  $H^-(1/3)$  and  $H_2(1/4)$  having the energies given by Eqs. (3) and (5-15), respectively. They were tested by MAS  $^1H$  NMR and ToF-SIMs.

Another catalytic system of the type  $MH$  involves chlorine. The bond energy of  $HCl$  is 4.4703 eV [54]. The first, second, and third ionization energies of  $Cl$  are 12.96764 eV, 23.814 eV, and 39.61 eV, respectively [52]. Based on these energies,  $HCl$  can serve as a catalyst and  $H$  source, since the bond energy of  $HCl$  plus the triple ionization ( $t = 3$ ) of  $Cl$  to  $Cl^{3+}$  is 80.86 eV ( $3 \cdot 27.2 \text{ eV}$ ). The catalyst reactions are given by



And, the overall reaction is



The anticipated product then is  $H_2(1/4)$ .

Alkali chlorides contain both  $Cl$  and  $H$ , typically from  $H_2O$  contamination.

Thus, some  $HCl$  can form interstitially in the crystalline matrix. Since  $H^+$  can most easily substitute for  $Li^+$ , and the substitution is least likely in the case of  $Cs^+$ , it was

5 anticipated that alkali chlorides may form  $HCl$  that undergoes catalysis to form  $H_2(1/4)$  with the trend of the rate of formation increasing in the order of the Group I elements. Due to the difference in lattice structure,  $MgCl_2$  may not form  $HCl$

catalyst; thus, it serves as a chlorine control. This condition applies to other alkaline earth halides and transition metal halides such as those of copper that can serve as

10 controls for the formation of  $H_2(1/4)$ . One exception from this set is  $Mg^{2+}$  in a suitable lattice, since the ionization of  $Mg^{2+}$  to  $Mg^{3+}$  is 80.1437 eV [52] which is close to  $3 \cdot 27.2$  eV. These hypotheses were tested by electron beam-excitation emission spectroscopy on alkali halides,  $MgX_2$  ( $X = F, Cl, Br, I$ ), and  $CuX_2$

( $X = F, Cl, Br$ ) with the goal of determining whether the predicted emission of

15  $H_2(1/4)$  is selectively observed when a catalyst reaction is possible and not otherwise. NMR was recorded on these compounds to search for the corresponding predicted  $H_2(1/4)$  peak to be compared with the emission results.

## II. Experimental Methods

20 Rt-plasma and Line Broadening Measurements.  $LiNH_2$  argon-hydrogen (95/5%) and  $LiNH_2$  hydrogen rt-plasmas was generated in the experimental set up described previously [15-21] (Figure 1) comprising a thermally insulated stainless steel cell with a cap that incorporated ports for gas inlet, and outlet. A titanium filament (55 cm long, 0.5 mm diameter) that served as a heater and hydrogen

dissociator was in the cell. 1 g of  $\text{LiNH}_2$  (Alfa Aesar 99.95%) was placed in the center of the cell under 1 atm of dry argon in a glove box. The cell was sealed and removed from the glove box. The cell was maintained at 50 °C for 4 hours with helium flowing at 30 sccm at a pressure of 1 Torr. The filament power was

5 increased to 200 W in 20 W increments every 20 minutes. At 120 W, the filament temperature was estimated to be in the range 800 to 1000 °C. The external cell wall temperature was about 700 °C. The cell was then operated with and without an argon-hydrogen (95/5%) flow rate of 5.5 sccm maintained at 1 Torr. Additionally, the cell was operated with hydrogen gas flow replacing argon-hydrogen (95/5%). The

10  $\text{LiNH}_2$  was vaporized by the filament heater as evidence the presence of *Li* lines. The presence of an argon-hydrogen or hydrogen plasma was determined by recording the visible spectrum over the Balmer region with a Jobin Yvon Horiba 1250 M spectrometer with a CCD detector described previously [15-21] using entrance/exits slits of 80/80  $\mu\text{m}$  and a 3 second integration time. The width of the

15 656.3 nm Balmer  $\alpha$  line emitted from the argon-hydrogen (95/5%)- $\text{LiNH}_2$  or hydrogen- $\text{LiNH}_2$  rt-plasma having a titanium filament was measured initially and periodically during operation. As further controls, the experiment was run with each of the flowing gases in the absence of  $\text{LiNH}_2$ .

Differential Scanning Calorimetry (DSC) Measurements. Differential scanning

20 calorimeter (DSC) measurements were performed using the DSC mode of a Setaram HT-1000 calorimeter (Setaram, France). Two matched alumina glove fingers were used as the sample compartment and the reference compartment. The fingers permitted the control of the reaction atmosphere. 0.067 g  $\text{NaH}$  was placed in a flat-base Al-23 crucible (Alfa-Aesar, 15 mm high x 10 mm OD x 8 mm ID). The

25 crucible was then placed in the bottom of the sample alumina glove finger cell. As a

reference, an aluminum oxide sample (Alfa-Aesar, -400 Mesh powder, 99.9%) with matching weight of the sample was placed in a matched Al-23 crucible. All samples were handled in a glove box. Each alumina glove finger cell was sealed in the glove box, removed from the glove box, and then quickly attached to the Setaram  
5 calorimeter. The system was immediately evacuated to pressure of 1 mTorr or less. The cell was back filled with 1 atm of helium, evacuated again, and then refilled with helium to 760 Torr. The cells were then inserted into the oven, and secured to their positions in the DSC instrument. The oven temperature was brought to the desired starting temperature of 100 °C. The oven temperature was scanned from 100 °C to  
10 750 °C at a ramp rate of 0.1 degree/minute. As a control,  $MgH_2$  replaced  $NaH$ . A 0.050 g  $MgH_2$  sample (Alfa-Aesar, 90%, reminder  $Mg$ ) was added to the sample cell, while a similar weight of aluminum oxide (Alfa-Aesar) was added to the reference cell. Both samples were also handled in a glove box.

Water-Flow, Batch Calorimetry. The cylindrical stainless steel reactor of  
15 approximately 60  $cm^3$  volume (1.0" outside diameter (OD), 5.0" length, and 0.065" wall thickness) is shown in Figure 2. The cell further comprised a welded-in 2.5" long, cylindrical thermocouple well with a wall thickness of 0.035" along the centerline that held a Type K thermocouple (Omega) read by a meter (DAS). For the cell sealed with a high temperature valve, a 3/8" OD, 0.065" thick SS tube  
20 welded at the end of the cell 1/4" off-center served as a port to introduce combinations of the reagents comprising the group of (i) 1g  $Li$ , 0.5g  $LiNH_2$ , 10g  $LiBr$ , and 15g  $Pd / Al_2O_3$ , (ii) 3.28 g  $Na$ , 15g Raney (R-)  $Ni / Al$  alloy, (iii) 15g R- $Ni$  doped with  $NaOH$ , and (iv) 3 wt%  $Al(OH)_3$  doped  $Ni / Al$  alloy. In the case that this port was spot-weld sealed, the SS tube had a 1/4" OD and a 0.02" wall-thickness.  
25 The reactants were loaded in a glove box, and a valve was attached to the port tube

to seal the cell before it was removed from the glove box and connected to a vacuum pump. The cell was evacuated to a pressure of 10 mTorr and crimped. The cell was then sealed with the valve or hermetically sealed by spot-welding 1/2" from the cell with the remaining tube cut off.

5        The reactor was installed inside a cylindrical calorimeter chamber shown in Figure 3. The stainless steel chamber had 15.2 cm ID, 0.305 cm wall thickness, and 40.4 cm length. The chamber was sealed at both ends by removable stainless steel plates and Viton o-rings. The space between the reactor and the inside surface of the cylindrical chamber was filled with high temperature insulation. The gas  
10       composition and pressure in the chamber was controlled to modulate the thermal conductance between the reactor and the chamber. The interior of the chamber was first filled with 1000 Torr helium to allow the cell to reach ambient temperature, the chamber was then evacuated during the calorimetric run to increase the cell temperature. Afterwards, 1000 Torr helium was added to increase the heat transfer  
15       rate from the hot cell to the coolant and balance any heat associated with P-V work. The relative dimensions of the reactor and the chamber were such that heat flow from the reactor to the chamber was primarily radial. Heat was removed from the chamber by cooling water which flowed turbulently through 6.35 mm OD copper tubing, which was wound tightly (63 turns) onto the outer cylindrical surface of the  
20       chamber. The reactor and chamber system were designed to safely absorb a thermal power pulse of 50 kW with one a minute duration. The absorbed energy was subsequently released to the cooling water stream in a controlled manner for calorimetric measurement. The temperature rise of the cooling water was measured by precision thermistor probes (Omega, OL-703-PP, 0.01°C) at the cooling coil inlet  
25       and exit. The inlet water temperature was controlled by a Cole Parmer (digital

Polystat, model 12101-41) circulating bath with 0.01°C temperature stability and 900 W cooling capacity at 20°C. A well insulated eight-liter damping tank was installed just downstream of the bath in order to reduce temperature fluctuations caused by cycling of the bath. Coolant flow through the system was maintained by an FMI model QD variable flow rate positive displacement lab pump. Cooling water flow rate was set by a variable area flow meter with a high-resolution control valve. The flow meter was calibrated directly by water collection in situ. A secondary flow rate measurement was performed by a turbine flow meter (McMillan Co., G111 Flometer,  $\pm 1\%$ ) which continuously output the flow rate to the data acquisition system. The calorimeter chamber was installed in a covered HDPE tank which was filled with melamine foam insulation to minimize heat loss from the system. Careful measurement of the thermal power release to the coolant and comparison with the measured input power indicated that thermal losses were less than 2-3 %.

The calorimeter was calibrated with a precision heater applied for a set time period to determine the percentage recovery of the total energy applied by the heater. The energy recovery was determined by integrating the total output power  $P_T$  over time. The power was given by

$$P_T = \dot{m}C_p\Delta T \quad (39)$$

where  $\dot{m}$  was the mass flow rate,  $C_p$  was the specific heat of water, and  $\Delta T$  was the absolute change in temperature between the inlet and outlet where the two thermistors were matched to correct any offset using a constant flow with no input power. In first step of the calibration test, an empty reaction cell, that was identical to the latter tested power cell containing the reactants, was evacuated to below 1 Torr and inserted into the calorimeter vacuum chamber. The chamber was evacuated and then filled with helium to 1000 Torr. The unpowered assembly

reached equilibrium over an approximately two-hour period at which time the temperature difference between the thermistors became constant. The system was run another hour to confirm the value of the difference due to absolute calibrations of the two sensors. The magnitude of the correction was 0.036 °C, and it was

5 confirmed to be consistent over all of the tests performed over the reported data set.

To increase the temperature of the cell per input power, ten minutes before the end of the ten-hour equilibration period, helium was evacuated from the chamber by the vacuum pump, and the chamber was maintained under dynamic pumping at a pressure below 1 Torr. 100.00 W of power was supplied to the heater (50.23 V and

10 1.991 A) for a period of 50 minutes. During this period, the cell temperature increased to approximately 650 °C, and the maximum change in water temperature (outlet minus inlet) was approximately 1.2 °C. After 50 minutes, the program directed the power to zero. To increase the rate of heat transfer to the coolant, the chamber was re-pressurized with 1000 Torr of helium and the assembly was allowed

15 to fully reach equilibrium over a 24-hour period as confirmed by the observation of full equilibrium in the flow thermistors.

The hydrino-reaction procedure followed that of the calibration run, but the cell contained the reagents. The equilibration period with 1000 Torr helium in the chamber was 90 minutes. 100.00 W of power was applied to the heater, and after

20 10 minutes, the helium was evacuated from the chamber. The cell heated at a faster rate post evacuation, and the reagents reached a hydrino reaction threshold temperature of 190 °C at 57 minutes. The onset of reaction was confirmed by a rapid rise in cell temperature that reached 378 °C at about 58 minutes. After ten minutes, the power was terminated, and helium was reintroduced into the cell slowly

25 over a period of 1 hour at a rate of 150 sccm.

The reactants 0.1 wt% *NaOH* -doped R-Ni 2800 or 0.5 wt% *NaOH* -doped R-Ni 2400 (elemental analysis was provided by the manufacturer, W. R. Grace Davidson, and the wt% *NaOH* was confirmed by elemental analysis (Galbraith) performed on samples handled in an inert atmosphere) and the products following the reaction of these reactants as well as those of the reaction mixture comprising *Li* (1 g) and *LiNH<sub>2</sub>* (0.5 g) (Alfa Aesar 99%), *LiBr* (10g) (Alfa Aesar ACS grade 99+%), and *Pd / Al<sub>2</sub>O<sub>3</sub>* (15g) (1% Pd, Alfa Aesar) were analyzed by quantitative X-ray diffraction (XRD) using hermetically sealed sample holders (Bruker Model #A100B37) loaded in a glove box under argon and analyzed with a Siemens D5000 diffractometer using *Cu* radiation at 40kV/30mA over the range 10° - 70° with a step size of 0.02° and a counting time of eight hours. In addition, a weighed sample of R-Ni in a 16.5 cc stainless steel cell connected to a vacuum system having a total volume of 291cc was heated with a temperature ramp from 25°C to 550°C to decompose any physically absorbed or chemisorbed gasses and to identify and quantify the released gasses. The hydrogen content was determined by mass spectroscopy, quantitative gas chromatography (HP 5890 Series II with a ShinCarbon ST 100/120 micropacked column (2 m long, 1/16" OD), N<sub>2</sub> carrier gas with a flow rate of 14 ml/min, an oven temperature of 80 °C, an injector temperature of 100 °C, and thermal-conductivity detector temperature of 100 °C), and by using the ideal gas law and the measured pressure, volume, and temperature. Hydrogen dominated each analysis with trace water only detected by mass spectroscopy, and <2% methane was also quantified by gas chromatography. The trace water of the R-Ni and controls was quantified independently of the hydrogen by liquefying the *H<sub>2</sub>O* in a liquid nitrogen trap, pumping off the hydrogen, and allowing all the water to vaporize by using a sample size of 0.5 g which is less than that which gives rise to a

saturated water-vapor pressure at room temperature.

Synthesis and Solid  $^1\text{H}$  MAS NMR of  $\text{LiH} * \text{Br}$ ,  $\text{LiH} * \text{I}$ ,  $\text{NaH} * \text{Cl}$  and  $\text{NaH} * \text{Br}$ . Lithium bromo and iodohydridohydride ( $\text{LiH} * \text{Br}$  and  $\text{LiH} * \text{I}$ ) were synthesized by reaction of hydrogen with  $\text{Li}$  (1 g) and  $\text{LiNH}_2$  (0.5 g) (Alfa Aesar 99%) as a source of atomic catalyst and additional atomic  $\text{H}$  with the corresponding alkali halide (10 g),  $\text{LiBr}$  (Alfa Aesar ACS grade 99+%) or  $\text{LiI}$  (Alfa Aesar 99.9 %), as an additional reactant. The compounds were prepared in a stainless steel gas cell (Figure 4) further containing Raney Ni (15 g) (W. R. Grace Davidson) as the hydrogen dissociator according to the methods described previously [13-14]. The reactor was run at 500 °C in a kiln for 72 hours with make-up hydrogen addition such that the pressure ranged cyclically between 1 Torr to 760 Torr. Then, the reactor was cooled under helium atmosphere. The sealed reactor was then opened in a glove box under an argon atmosphere. NMR samples were placed in glass ampules, sealed with rubber septa, and transferred out of the glove box to be flame sealed.  $^1\text{H}$  MAS NMR was performed on solid samples of  $\text{LiH} * \text{X}$  ( $\text{X}$  is a halide) at Spectral Data Services, Inc., Champaign, Illinois as described previously [13-14]. Chemical shifts were referenced to external TMS. XPS was also performed on crystalline samples that were handled as air-sensitive materials.

Since the synthesis reaction comprised  $\text{LiNH}_2$ , and  $\text{Li}_2\text{NH}$  was a reaction product, both were run as controls alone and in a  $\text{LiBr}$  or  $\text{LiI}$  matrix. The  $\text{LiNH}_2$  was the commercial starting material, and  $\text{Li}_2\text{NH}$  was synthesized by the reaction of  $\text{LiNH}_2$  and  $\text{LiH}$  [67] and by decomposition of  $\text{LiNH}_2$  [68] with the  $\text{Li}_2\text{NH}$  product confirmed by X-ray diffraction (XRD). To eliminate the possibility that the alkali halide influenced the local environment of the protons or that any given known species produced an NMR resonance that was shifted upfield relative to the ordinary

peak, controls comprising  $LiH$  (Aldrich Chemical Company 99%),  $LiNH_2$ , and  $Li_2NH$  with an equimolar mixture of  $LiX$  were run. The controls were prepared by mixing equimolar amounts of compounds in a glove box under argon. To further eliminate F centers as a possible contributor to the local environment of the protons of any given known species to produce an upfield-shifted NMR resonance, electron spin resonance spectroscopy (ESR) was performed on the  $LiH * Br$  and  $LiH * I$  samples. For the ESR studies, the samples were loaded into 4 mm OD Suprasil quartz tubes and evacuated to a final pressure of  $10^{-4}$  Torr. ESR spectra were recorded with a Bruker ESP 300 X-band spectrometer at room temperature and 77 K. The magnetic field was calibrated with a Varian E-500 gauss meter. The microwave frequency was measured by a HP 5342A frequency counter.

Elemental analysis was performed at Galbraith Laboratories to confirm the product composition and to eliminate the possibility of NMR-detectable amounts of any transition metal hydrides or other exotic hydrides that may give rise to upfield-shifted peaks. Specifically, the abundance of all elements present in the product ( $Li, H, X$ ) and the stainless steel reaction vessel and R-Ni ( $Ni, Fe, Cr, Mo, Mn, Al$ ) were determined.

$NaH * Cl$  and  $NaH * Br$  were synthesized by reaction of hydrogen with  $Na$  (3.28 g) and  $NaH$  (1 g) (Aldrich Chemical Company 99%) as a source of  $NaH$  catalyst and intrinsic atomic  $H$  with the corresponding alkali halide (15 g),  $NaCl$  or  $NaBr$  (Alfa Aesar ACS grade 99+%), as an additional reactant. The compounds were prepared in a stainless steel gas cell (Figure 4) further containing  $Pt / Ti$  ( $Pt$  coated  $Ti$  (15 g); Titan Metal Fabricators, platinum plated titanium mini-expanded anode, 0.089 cm x 0.5 cm x 2.5 cm with 2.54  $\mu m$  of platinum) as the hydrogen dissociator. Each synthesis was run according to the methods described for  $Li$

except that the kiln was maintained at 500 °C, and, the  $NaH * Cl$  synthesis was repeated without the addition of hydrogen gas to determine the effect of using  $NaH(s)$  as the sole hydrogen source. XPS was performed on  $NaH * Cl$  since no primary element peaks were possible in the region for  $H^-(1/4)$ , and NMR

5 investigations of both products were preformed.

$NaH * Cl$  was also synthesized from  $NaCl$  (10 g) and the solid acid  $KHSO_4$  (1.6 g) as the only source of hydrogen with the kiln maintained at 580 °C. NMR was performed to test whether  $H^-(1/3)$  formed by the reactions of Eqs. (23-25) could be observed when the rapid reaction to  $H^-(1/4)$  according to Eq. (27) was partially

10 inhibited due to the absence of a high concentration of  $H$  from a dissociator with  $H_2$  or a hydride.

A silicon wafer (2 g, 0.5 x 0.5 x 0.05 cm, Silicon Quest International, silicon (100), boron-doped, cleaned by heating to 700 °C under vacuum) was coated by the product  $NaH * Cl$  and  $NaH *$  by placing it in reactants comprising  $Na$  (1.7 g),  $NaH$  (0.5 g),  $NaCl$  (10 g), and  $Pt / Ti$  (15 g) wherein the  $NaCl$  that was initially heated to 400°C under vacuum to remove any  $H_2(1/4)$ . The reaction was run at 550°C in the kiln for 19 hours with an initial hydrogen pressure of 760 Torr. XPS was performed on a spot comprising only sodium hydride coated silicon wafer ( $NaH *$ -coated  $Si$ ). The  $NaH * Cl$ -coated silicon wafer ( $NaH * Cl$ -coated  $Si$ ) was

15 investigated by electron-beam excitation spectroscopy. An emission spectrum of a pressed pellet of the  $NaH * Cl$  crystals was also recorded.

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ToF-SIMS Spectra. The crystalline samples of  $LiH * Br$ ,  $LiH * I$ ,  $NaH * Cl$ ,  $NaH * Br$ , and the corresponding alkali halide controls were sprinkled onto the surface of a double-sided adhesive tape and characterized using a Physical

Electronics TFS-2000 ToF-SIMS instrument. The primary ion gun utilized a  $^{69}\text{Ga}^+$  liquid metal source. A region on each sample of  $(60\mu\text{m})^2$  was analyzed. In order to remove surface contaminants and expose a fresh surface, the samples were sputter-cleaned for 60 seconds using a  $180\mu\text{m} \times 100\mu\text{m}$  raster. The aperture setting was 3, and the ion current was 600 pA resulting in a total ion dose of  $10^{15} \text{ ions/cm}^2$ .

During acquisition, the ion gun was operated using a bunched (pulse width 4 ns bunched to 1 ns) 15 kV beam [69-70]. The total ion dose was  $10^{12} \text{ ions/cm}^2$ . Charge neutralization was active, and the post accelerating voltage was 8000 V. The positive and negative SIMS spectra were acquired. Representative post sputtering data is reported.

In addition, 0.1g *Na*, 0.5g *NaH*, and 15g *Pt / Ti* were loaded into the water flow calorimetry cell, and water flow calorimetry was performed under the same conditions as described for *Na* and *R-Ni*. The cell generated 15 kJ of excess energy; whereas, the theoretical energy balance from the decomposition of *NaH* is endothermic by +1.2 kJ. Thus, to confirm the presence of hydrino hydrides corresponding to the reactions given by Eqs. (23-25) as the source of the excess heat, a sample of the *Pt / Ti* coated with sodium hydrino hydride (*NaH*\*-coated *Pt / Ti*) was analyzed directly by the same procedure as for the crystalline samples except that the sputtering was for 100s. Unreacted *Pt / Ti* coated with the starting materials served as a control. XPS was also performed.

ToF-SIMS of *R-Ni* 2400 reacted over a 48 hour period at 50°C was also performed by the same procedure as for the crystalline samples. The reactions to form hydrinos are given by Eqs. (32-35). Since the surface was coated with *NaOH*, sodium hydrino hydride compounds with *NaOH* were predicted.

FTIR Spectroscopy. FTIR analysis was performed on solid-sample- *KBr* pellets of *LiH \* Br* using the transmittance mode at the Department of Chemistry, Princeton University, New Jersey using a Nicolet 730 FTIR spectrometer with DTGS detector at resolution of  $4\text{ cm}^{-1}$  as described previously [13-14]. The samples were  
5 handled under an inert atmosphere. The resolution was  $0.5\text{ cm}^{-1}$ . Controls comprised  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ , and  $\text{Li}_3\text{N}$  that were commercially available except  $\text{Li}_2\text{NH}$  that was synthesized by the reaction of  $\text{LiNH}_2$  and  $\text{LiH}$  [67] and by decomposition of  $\text{LiNH}_2$  [68] with the  $\text{Li}_2\text{NH}$  product confirmed by X-ray diffraction (XRD).

XPS Spectra. A series of XPS analyses were made on the crystalline  
10 samples using a Scienta 300 XPS Spectrometer. The fixed analyzer transmission mode and the sweep acquisition mode were used. The step energy in the survey scan was  $0.5\text{ eV}$ , and the step energy in the high-resolution scan was  $0.15\text{ eV}$ . In the survey scan, the time per step was 0.4 seconds, and the number of sweeps was 4. In the high-resolution scan, the time per step was 0.3 seconds, and the number of  
15 sweeps was 30.  $\text{C } 1s$  at  $284.5\text{ eV}$  was used as the internal standard.

UV Spectroscopy of Electron-Beam Excited Interstitial  $\text{H}_2(1/4)$ . Vibration-rotational emission of  $\text{H}_2(1/4)$  trapped in the lattice of alkali halides,  $\text{MgCl}_2$ , and in a silicon wafer was investigated via electron bombardment of the crystals. Windowless UV spectroscopy of the emission from electron-beam excitation of the  
20 crystals was recorded using a 12.5 keV electron gun at a beam current of  $10\text{-}20\text{ }\mu\text{A}$  in the pressure range of  $<10^{-5}$  Torr. The UV spectrum was recorded with a photomultiplier tube (PMT). The wavelength resolution was about  $2\text{ nm}$  (FWHM) with an entrance and exit slit width of  $300\text{ }\mu\text{m}$ . The increment was  $0.5\text{ nm}$  and the dwell time was 1 second.

### III. Results and Discussion

A. RT-plasma Emission and Balmer  $\alpha$  Line Widths. An argon-hydrogen (95/5%)-lithium rt-plasma formed with a low field (1V/cm), at low temperatures (e.g.  $\approx 10^3$  K), from atomic hydrogen generated at a titanium filament and  $LiNH_2$  that was

- 5 vaporized by heating. Lithium and H emission were observed that confirmed  $LiNH_2$  and its decomposition product  $Li$  served as a source of atomic  $Li$  and  $H$ . Argon of the argon-hydrogen mixture increased the amount of atomic H as evidenced by the significantly decreased H emission in the absence of argon.  $H$  Balmer emission corresponding to population of a level with energy  $> 12$  eV was observed, as shown
- 10 in Figures 5 and 6, which also requires that Lyman emission was present.

- No plasma formed with argon/hydrogen alone. No possible chemical reaction of the titanium filament, the vaporized  $LiNH_2$ , and 0.6 Torr argon-hydrogen mixture at a cell temperature of 700°C could be found to account for the Balmer emission. In fact, no known chemical reaction releases enough energy to excite Balmer and
- 15 Lyman emission from hydrogen. In addition to known chemical reactions, electron collisional excitation, resonant photon transfer, and the lowering of the ionization and excitation energies by the state of "non ideality" in dense plasmas were also rejected as the source of ionization or excitation to form the hydrogen plasma [21]. The formation of an energetic reaction of atomic hydrogen was consistent with a source
- 20 of free energy from the catalysis of atomic hydrogen by  $Li$ .

The energetic hydrogen atom energies were calculated from the width of the 656.3 nm Balmer  $\alpha$  line emitted from RF rt-plasmas. The full half-width  $\Delta\lambda_G$  of each Gaussian results from the Doppler ( $\Delta\lambda_D$ ) and instrumental ( $\Delta\lambda_I$ ) half-widths:

$$\Delta\lambda_G = \sqrt{\Delta\lambda_D^2 + \Delta\lambda_I^2} \quad (40)$$

$\Delta\lambda_i$  in our experiments was  $\pm 0.006 \text{ nm}$ . The temperature was calculated from the Doppler half-width using the formula:

$$\Delta\lambda_D = 7.16 \times 10^{-7} \lambda_0 \left( \frac{T}{\mu} \right)^{1/2} \quad (41)$$

where  $\lambda_0$  is the line wavelength,  $T$  is the temperature in K ( $1 \text{ eV} = 11,605 \text{ K}$ ), and  $\mu$  is the molecular weight ( $=1$  for atomic hydrogen). In each case, the average Doppler half-width that was not appreciably changed with pressure, varied by  $\pm 5\%$  corresponding to an error in the energy of  $\pm 10\%$ .

The 656.3 nm Balmer  $\alpha$  line widths recorded on the argon-hydrogen (95/5%)-lithium rt-plasma, initially and after 70 hours of operation, are shown in Figures 5A and 5B, respectively. The Balmer  $\alpha$  line profile of the plasma emission at both time points comprised two distinct Gaussian peaks, an inner, narrower peak corresponding to a slow component of less than 0.5 eV and an outer, significantly broadened peak corresponding to a fast component of  $>40 \text{ eV}$ . The fast component accounted for 90% of the  $n = 3$  excited-state H population initially and increased to 97% at 70 hours. Only the hydrogen lines were broadened. As shown previously, the source of energy of the fast H cannot be attributed to any applied electric field, but is predicted by the mechanism of the catalysis of hydrogen to lower-states [32-37].

A lithium rt-plasma also formed in the case of pure  $H_2$  gas at a pressure of 1 Torr, except that the line broadening and populations were less, about 6 eV with only a 27% population, at the initial and 70-hour time points as shown in Figures 6A and 6B, respectively. This result was expected, since the excess  $H_2$  can react with  $Li$  to form  $LiH$  that catalyzes the destruction of  $LiNH_2$  by the reaction:



Thus, the reactions to produce atomic  $Li$  and  $H$  are diminished. In addition, argon of the argon-hydrogen mixture can increase the amount of atomic H by preventing its recombination, and  $Ar^+$  generated by the plasma can participate as a catalyst as well as  $Li$ .

5 We have assumed that Doppler broadening due to thermal motion was the dominant source to the extent that other sources may be neglected. This assumption was confirmed when each source was considered. In general, the experimental profile is a convolution of two Doppler profiles, an instrumental profile, the natural (lifetime) profile, Stark profiles, van der Waals profiles, a resonance  
10 profile, and fine structure. The contribution from each source was determined to be below the limit of detection [13-21, 38-39].

The formation of fast H can be explained by a resonant energy transfer from hydrogen atoms to  $Li$  atoms, of three times the potential energy of atomic hydrogen, to form a short-lived intermediate  $H^*(1/4)$  having a central field equivalent to four  
15 times that of a proton and a radius of the hydrogen atom. The intermediate spontaneously decays by a collisional or through-space energy transfer as the radius decreases to  $a_0/4$  yielding fast  $H(n=1)$ , as well as the emission of  $q \cdot 13.6 \text{ eV}$  photons reported previously [27-29]. Collisional energy transfer including through-space coupling is common. For example, the exothermic chemical reaction of  
20  $H + H$  to form  $H_2$  does not occur with the emission of a photon. Rather, the reaction requires a collision with a third body,  $M$ , to remove the bond energy-  
 $H + H + M \rightarrow H_2 + M^*$  [44]. The third body distributes the energy from the exothermic reaction, and the end result is the  $H_2$  molecule and an increase in the temperature of the system. In the case of the catalytic reaction with the formation of  
25 states given by Eqs. (2a) and (2c), the temperature of H becomes very high.

B. Differential Scanning Calorimetry (DSC) Measurements. The DSC (100-750 °C) of  $NaH$  is shown in Figure 7. A broad endothermic peak was observed at 350 °C to 420 °C which corresponded to  $47 \text{ kJ / mole}$ . Sodium hydride decomposes in this temperature range with a corresponding enthalpy of  $57 \text{ kJ / mole}$  [71]. A large exotherm was observed in the region 640 °C to 825 °C which corresponded to  $-177 \text{ kJ / mole}$ . The DSC (100-750 °C) of  $MgH_2$  is shown in Figure 8. Two sharp endothermic peaks were observed. A first peak was observed centered at 351.75 °C corresponding to  $68.61 \text{ kJ / mole } MgH_2$ . The decomposition of  $MgH_2$  is observed at 440 °C to 560 °C corresponding to  $74.4 \text{ kJ / mole } MgH_2$  [71]. In Figure 8, a second peak was observed centered at 647.66 °C corresponding to  $6.65 \text{ kJ / mole } MgH_2$ . The known melting point of  $Mg(m)$  is 650 °C corresponding to an enthalpy of fusion of  $8.48 \text{ kJ / mole } Mg(m)$  [72]. Thus, the expected behavior was observed for the decomposition of a control, noncatalyst hydride. In contrast, a novel exothermic effect of  $-177 \text{ kJ / mole } NaH$  or at least  $-354 \text{ kJ / mole } H_2$  was observed under conditions that form  $NaH$  catalyst with some portion of the  $H$  undergoing the catalysis reactions given by Eqs. (23-25). The observed enthalpy was greater than that of the most exothermic reaction possible for  $H$ , the  $-241.8 \text{ kJ / mole } H_2$  enthalpy of combustion of hydrogen.

20

C. Water-Flow Calorimetry Power Measurements. In each test, the energy input and energy output were calculated by integration of the corresponding power. For the input power, the voltage and current measured at the end of each time interval were multiplied by the time interval (typically 10 seconds) to obtain the energy increment in Joules. All energy increments were summed over the entire experiment

25

after the equilibration period to obtain total energy. For output energy, the thermistor offset was calculated after each test assuming that the final readings of inlet and outlet temperature were identical. This offset was calculated to be 0.036 °C. The thermal energy in the coolant flow in each time increment was calculated using Eq. (39) by multiplying volume flow rate of water by the water density at 19 °C (0.998 kg/liter), the specific heat of water (4.181 kJ/kg-°C), the corrected temperature difference, and the time interval. Values were summed over the entire experiment to obtain the total energy output. The total energy from cell  $E_T$  must equal the energy input  $E_{in}$  and any excess energy  $E_{ex}$ :

$$E_T = E_{in} + E_{ex} \quad (43)$$

From the energy balance, any excess heat was determined.

The calibration test results are shown in Figures 9 and 10. In the plot of Figure 10, there is a time point at which the slope of the coolant power changes almost discontinuously. This point at about one hour corresponds to the helium addition enhancing heat transfer from the cell to the chamber wall. The numerical integration of the input and output power curves yielded an output energy of 292.2 kJ and an input energy of 303.1 kJ corresponding to a coupling of flow of 96.4% of the resistive input to the output coolant.

The cell temperature with time and the coolant power with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 1g  $Li$ , 0.5g  $LiNH_2$ , 10g  $LiBr$ , and 15g  $Pd / Al_2O_3$  are shown in Figures 11 and 12, respectively. The numerical integration of the input and output power curves with the calibration correction applied yielded an output energy of 227.2 kJ and an input energy of 208.1 kJ. Thus, from Eq. (43), the excess energy was 19.1 kJ. In the plot of Figure 12, there is a point at which the slope of the temperature changes almost

discontinuously. The slope change occurs just slightly after 1 hour, and this corresponds to the cell temperature rising rapidly with the onset of reaction. Based on the system response to a power pulse, the excess energy of 19.1 kJ occurred in less than 2 minutes which places the power for the reaction at over 160 W.

5           The quantitative XRD of the composition of the products following the reaction showed that the  $LiBr$  and  $Pd / Al_2O_3$  were unchanged. Thus, assuming a 100% yield, the maximum theoretical energy released by known chemistry is 4.3 kJ from the formation lithium nitride and hydride according to Eq. (22); whereas, the observed energy balance was 4.4 times this maximum. The only exothermic  
10   reaction possible to account for the energy balance is that given by Eqs. (17-19). The hydrogen content of the 0.5g  $LiNH_2$  was 22 mmoles  $H_2$ . Thus, the observed energy balance is  $-870 \text{ kJ / mole } H_2$ , over 3.5 times the  $-241.8 \text{ kJ / mole } H_2$  enthalpy of combustion, the most energetic reaction of hydrogen assuming the maximum possible  $H_2$  inventory.

15           The cell temperature with time and the coolant power with time for the R-Ni control power test with the cell containing the reagents comprising the starting material for R-Ni, 15g R-Ni/Al alloy powder, and 3.28g of Na are shown in Figures 13 and 14, respectively. The temperature and coolant power time profiles curves were very similar to the calibration. The numerical integration of the input and output  
20   power curves with the calibration correction applied yielded an output energy of 384 kJ and an input energy of 385 kJ. Energy balance was obtained.

          The cell temperature with time and the coolant power with time for the hydrino reaction with the cell containing the reagents comprising the catalyst material, 15g  $NaOH$ -doped R-Ni, and 3.28g of Na are shown in Figures 15 and 16, respectively.  
25   The numerical integration of the input and output power curves with the calibration

correction applied yielded an output energy of 185.1 kJ and an input energy of 149.1 kJ. Thus, from Eq. (43), the excess energy was 36 kJ. In the plot of Figure 15, there is a point at which the slope of the temperature changes almost discontinuously. The slope change occurs just slightly before 1 hour, and this corresponds to the cell temperature rising rapidly with the onset of reaction. Based on the system response to a power pulse, the excess energy of 36 kJ occurred in less than 1.5 minutes which places the power for the reaction at over 0.5 kW.

The composition of the reactant *NaOH*-doped R-Ni and the product following the reaction with the alkali metal determined by quantitative XRD was Ni with trace Bayerite and Ni with trace alkali hydroxide, respectively. The formation of a sodium-Ni alloy or the reaction of sodium with  $Al_2O_3$  of R-Ni [73-74] is significantly endothermic ( $\Delta H = +138 \text{ kJ / mole Na}$  [75] and  $\Delta H = +72.18 \text{ kJ / mole Na}$  [65], respectively). Using the heat of formations, the reaction of Bayerite with sodium to form *NaOH* ( $\Delta H = -15.6 \text{ kJ / mole Al(OH)}_3$  [65, 76]) contributes negligibly to the energy balance based on the XRD analysis showing trace Bayerite initially and the corresponding *NaOH* product from reaction with *Na*. Consistent with the literature [74], the  $H_2O$  content from Bayerite decomposition was  $47.7 \mu\text{ moles } H_2O/\text{g R-Ni}$  corresponding to a negligible contribution due to the formation of *NaOH* ( $\Delta H = -184.0 \text{ kJ / mole } H_2O$  [65]) from the decomposition of  $Al(OH)_3$  ( $2Al(OH)_3 \rightarrow Al_2O_3 + 3H_2O \quad \Delta H = +92.45 \text{ kJ / mole Al}$ ). The overall reaction is the reaction of Bayerite with sodium to form *NaOH* ( $\Delta H = -15.6 \text{ kJ / mole Al(OH)}_3$ ).

The only exothermic reaction possible to account for the energy balance is that given by Eqs. (23-25). The hydrogen content of the R-Ni determined using quantitative GC and by using the ideal gas law on the measured P, V, and T was

150  $\mu$  moles  $H_2$ /g R-Ni. Thus, the observed energy balance is  
 $-1.6 \times 10^4$  kJ / mole  $H_2$ , over 66 times the  $-241.8$  kJ / mole  $H_2$  enthalpy of combustion,  
the most energetic reaction of hydrogen assuming the maximum possible  $H_2$   
inventory. The conservative theoretical energy yield for the reaction of Eq. (44) is  
5 259 eV /  $H_2$  or 25 MJ / mole  $H_2$  (Eq. (7)).



Among the most energetic known oxidation reactions involving a solid fuel is the  
reaction  $Be + 1/2 O_2 \rightarrow BeO$ , which has a heat of combustion of 24 kJ/g, and there  
are very few known fuel/oxidizer systems producing greater than 10 kJ/g [65]. As a  
10 comparison, even without possibly going to completion, the H content of the  
recyclable catalyst  $NaH$  produced energy of over 300 times that of the best known  
solid fuel per weight.

With increased  $NaOH$  doping and a switch to R-Ni 2400, the catalytic material  
generated high power and energy without requiring the addition of  $Na$ . The cell  
15 temperature with time and the coolant power with time for the hydride reaction with  
the cell containing the catalyst material, 15g  $NaOH$ -doped R-Ni 2400, are shown in  
Figures 17 and 18, respectively. The numerical integration of the input and output  
power curves with the calibration correction applied yielded an output energy of  
195.7 kJ and an input energy of 184.0 kJ corresponding to an excess energy of 11.7  
20 kJ, and the power was over 0.25 kW.

The composition of the reactant  $NaOH$ -doped R-Ni and the product following  
the reaction determined by quantitative XRD was R-Ni with 3.7wt% Bayerite and R-  
Ni, respectively. The measured  $H_2O$  content from Bayerite decomposition of the  
initial R-Ni was 32.8  $\mu$  moles  $H_2O$ /g R-Ni compared to the measured  $H_2O$  content

from Bayerite decomposition of 34.0  $\mu$  moles  $H_2O/g$  for 3 wt%  $Al(OH)_3$  doped  $Ni / Al$  alloy. The most exothermic reaction possible was the reaction of  $Al(OH)_3$  to  $Al_2O_3$ . The balanced reaction is given by [65, 75, 77]:



- 5 For 3.7wt%  $Al(OH)_3$ , the maximum theoretical energy from the reaction given by Eq. (45) is  $\Delta H = -1.88 \text{ kJ}$ . This was confirmed by the heat measurement of 15g of 3 wt%  $Al(OH)_3$  doped  $Ni / Al$  alloy that showed an average energy of  $\Delta H = -1.1 \text{ kJ}$  compared to the theoretical energy of  $\Delta H = -1.7 \text{ kJ}$  ( $\Delta H = -300 \text{ kJ / mole } Al(OH)_3$  using Eq. (45) with  $\Delta H_f(NiAl_{crystal}) = -96 \text{ kJ/mole}$  [75]). Thus, the observed energy
- 10 from the  $NaOH$ -doped R-Ni was 4.4 times the theoretical; thus, it was predominantly attributable to the catalysis reaction given by Eqs. (23-25).

D. ToF-SIMS Spectra. The positive ToF-SIMS spectrum obtained from  $LiBr$  and the  $LiH * Br$  crystals are shown in Figures 19 and 20, respectively. The positive ion spectrum of the  $LiH * Br$  crystals and that of the  $LiBr$  control were dominated by the  $Li^+$  ion.  $Li_2^+$ ,  $Na^+$ ,  $Ga^+$ , and  $Li(LiBr)^+$  were also observed.

The negative ion ToF-SIMS of  $LiBr$  and the  $LiH * Br$  crystals are shown in Figures 21 and 22, respectively. The  $LiH * Br$  spectrum was dominated by  $H^-$  and  $Br^-$  peaks with the intensity of  $H^- > Br^-$ . Bromide alone dominated the negative ion ToF-SIMS of the  $LiBr$  control. For both,  $O^-$ ,  $OH^-$ ,  $Cl^-$ , and  $LiBr^-$  were also observed. In addition to the increased hydride, other unique peaks of the  $LiH * Br$  sample were  $LiHBr^-$  and  $Li_2H_2Br^-$  consistent with the formation of novel lithium bromohydride.

The positive ToF-SIMS spectrum obtained from *LiI* and the *LiH \* I* crystals are shown in Figures 23 and 24, respectively. The positive ion spectrum of the *LiH \* I* crystals and that of the *LiI* control were dominated by the  $Li^+$  ion.  $Li_2^+$ ,  $Na^+$ ,  $Ga^+$ , and a series of positive ions  $Li[LiI]_n^+$  were also observed. Unique peaks

5 of the *LiH \* I* sample were  $LiHI^+$ ,  $Li_2H_2I^+$ ,  $Li_4H_2I^+$ , and  $Li_6H_2I^+$ .

The negative ion ToF-SIMS of *LiI* and the *LiH \* I* crystals are shown in Figure 25 and 26, respectively. The *LiH \* I* spectrum was dominated by  $H^-$  and  $I^-$  peaks with the intensity of  $H^- > I^-$ . Iodide alone dominated the negative ion ToF-SIMS of the *LiI* control. For both,  $O^-$ ,  $OH^-$ ,  $Cl^-$ , and a series of negative ions

10  $I[LiI]_n^-$  were also observed. In addition to the increased hydride, other unique peaks of *LiH \* I* sample were  $LiHI^-$ ,  $Li_2H_2I^-$ , and  $NaHI^-$  consistent with the formation of novel lithium iodohydride.

The negative ToF-SIMS spectrum ( $m/e = 20 - 30$ ) of *NaH \**-coated *Pt / Ti* following the production of 15 kJ of excess heat is shown in Figure 27. Hydride-

15 hydride-compound series  $NaH_x^-$  was observed wherein the mass deficit from the high resolution (10,000) mass determination definitively distinguished this assignment over the  $C_2H_x^-$  series observed in controls. The XPS spectrum showed that *NaH \**-coated *Pt / Ti* comprised two fractional hydrogen states,  $H^-(1/3)$  and  $H^-(1/4)$  (Sec. IIIF).

20  $NaH_x^-$  having the mass-deficit series was also observed in the spectrum of R-Ni from the Na/R-Ni water-flow calorimetric run that produced 36 kJ of excess heat. The positive ToF-SIMS spectrum obtained from R-Ni reacted over a 48 hour period at 50°C is shown in Figure 28. The dominant ion on the surface was  $Na^+$  consistent

with  $\text{NaOH}$  doping of the surface. The ions of the other major elements of R-Ni 2400 such as  $\text{Al}^+$ ,  $\text{Ni}^+$ ,  $\text{Cr}^+$ , and  $\text{Fe}^+$  were also observed.

The negative ion ToF-SIMS of R-Ni reacted over a 48 hour period at  $50^\circ\text{C}$  is shown in Figure 29. The spectrum showed a very large  $\text{H}^-$  peak as well as 5 hydroxide fragments  $\text{OH}^-$  and  $\text{O}^-$ . Two other dominant peaks matched the high resolution mass of  $\text{NaH}_3^-$  and  $\text{NaH}_3\text{NaOH}^-$  to 10,000 and were assigned to sodium hydride and this ion in combination with  $\text{NaOH}$ . Other unique ions assignable to sodium hydride hydrides  $\text{NaH}_x^-$  in combinations with  $\text{NaOH}$ ,  $\text{NaO}$ ,  $\text{OH}^-$  and  $\text{O}^-$  were observed.

10

E. NMR Identification of  $\text{H}^-(1/3)$ ,  $\text{H}^-(1/4)$ ,  $\text{H}_2(1/3)$  and  $\text{H}_2(1/4)$ . The  $^1\text{H}$  MAS NMR spectra of  $\text{LiH} * \text{Br}$  and  $\text{LiH} * \text{I}$  relative to external TMS are shown in Figures 30A and 30B, respectively.  $\text{LiH} * \text{X}$  samples showed a large distinct upfield resonance at  $-2.51$  ppm and  $-2.09$  ppm for  $\text{X} = \text{Br}$  and  $\text{X} = \text{I}$ , respectively. None 15 of the controls comprising  $\text{LiH}$ , equal molar mixtures of  $\text{LiH}$  and  $\text{LiBr}$  or  $\text{LiI}$ ,  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ , and equal molar mixtures of  $\text{LiNH}_2$  or  $\text{Li}_2\text{NH}$  and  $\text{LiBr}$  or  $\text{LiI}$  showed an upfield-shifted peak. Since the upfield peak of  $\text{LiH} * \text{X}$  at about  $-2.2$  ppm was very broad, it is useful to compare these results to those of the prior identification of  $\text{H}^-(1/4)$  of  $\text{KH} * \text{Cl}$  and  $\text{KH} * \text{I}$ .

20

The  $^1\text{H}$  MAS NMR spectra relative to TMS of  $\text{KH} * \text{Cl}$  samples (Figure 31A) from independent syntheses and controls were given previously [13-15, 24-26]. The experimental absolute resonance shift of TMS is  $-31.5$  ppm relative to the proton's gyromagnetic frequency [78-79]. The  $\text{KH}$  experimental shift of  $+1.1$  ppm relative to TMS corresponding to absolute resonance shift of  $-30.4$  ppm matches very well the

predicted shift of  $H^-(1/1)$  of  $-30$  ppm given by Eq. (4) wherein  $p = 0$ . The novel peak at  $-4.46$  ppm relative to TMS corresponding to an absolute resonance shift of  $-35.96$  ppm indicates that  $p = 4$  in Eq. (4).  $H^-(1/4)$  is the hydride ion predicted by using  $K$  as the catalyst [1, 15, 30]. Furthermore, the extraordinarily narrow peak-width is indicative of a small hydride ion that is a free rotator. In contrast,  $KH * I$  (Figure 31B) shows a very broad peak at  $-2.31$  ppm. The predicted product hydride ion  $H^-(1/4)$  of the reaction with  $K$  catalyst to form  $KH * I$  was observed by XPS [13-15, 26, 30] at its predicted binding energy of  $11.2$  eV. Thus, the diamagnetic shift due to the larger halide is  $+2.15$  ppm. The corrected upfield NMR peaks for  $LiH * X$  are each about  $-4.46$  ppm which matches the predicted shift of the free ion given by Eq. (4).

The elemental analysis of  $LiH * Br$  by wt% was Li (8%), H (1.1%), I (90.9%) corresponding stoichiometrically to  $LiHBr$  with the stainless steel and R-Ni components at less than detectable levels. The elemental analysis of  $LiH * I$  by wt% was Li (5.2%), H (0.8%), I (94%) corresponding stoichiometrically to  $LiHI$  with the stainless steel and R-Ni components at less than detectable levels. Thus, no hydrides other than those of Li are possible assignments. U H does not have an upfield-shifted NMR peak as determined previously [13-14]. F centers could not have been the source since no ESR signal was detectable in  $LiH * Br$  or  $LiH * I$  at room temperature or 77 K.  $^1H$  MAS NMR spectra obtained on  $LiNH_2$ ,  $Li_2NH$ , and these compounds in a  $LiBr$  or  $LiI$  matrix also showed that neither of these compounds have an upfield-shifted NMR peak. To further eliminate  $LiNH_2$  and  $Li_2NH$  as the source of the  $-2.5$  ppm peak,  $LiH * Br$  samples with the  $-2.5$  ppm peak were heated to  $>600^\circ C$  under dynamic vacuum to decompose  $LiNH_2$  and  $Li_2NH$ . The heat-treated samples were analyzed by FTIR spectroscopy to confirm

that the amide and imide were eliminated as indicated by the absence of the amide peaks at 3314, 3259, 2079(broad), 1567, and 1541  $\text{cm}^{-1}$  and the imide peaks at 3172 (broad), 1953, and 1578  $\text{cm}^{-1}$  while the  $-2.5$  ppm peak remained upon reanalysis by NMR. The FTIR spectrum shown in Figure 45B shows the elimination of these species while the corresponding NMR showed the  $-2.5$  ppm peak. Since the past and present NMR and FTIR analysis leads to the conclusion that the  $-2.5$  ppm peak in  $^1\text{H}$  NMR spectrum is not associated with the  $\text{U H}$ ,  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ , or any other known species, the  $-2.5$  ppm peak in  $^1\text{H}$  NMR spectrum is assigned to the  $\text{H}^-(1/4)$  ion which matches theoretical prediction and is direct evidence of a lower-energy state hydride ion.

In addition to the  $-2.5$  ppm and  $-2.09$  ppm peaks assigned to  $\text{H}^-(1/4)$ , a  $1.3$  ppm peak was observed in the  $^1\text{H}$  MAS NMR spectra of  $\text{LiH} * \text{Br}$  and  $\text{LiH} * \text{I}$  shown in Figures 30A. and 30B, respectively. None of the controls showed this peak which eliminated any of the starting compounds or their possible known products. However, the peak may be due to the  $\text{H}_2(1/4)$  molecule corresponding to  $\text{H}^-(1/4)$ .

$\text{H}_2$  has been characterized by gas-phase  $^1\text{H}$  NMR. The experimental absolute resonance shift of gas-phase TMS relative to the proton's gyromagnetic frequency is  $-28.5$  ppm [80].  $\text{H}_2$  was observed at  $0.48$  ppm compared to gas phase TMS set at  $0.00$  ppm [81]. Thus, the corresponding absolute  $\text{H}_2$  gas-phase resonance shift of  $-28.0$  ppm ( $-28.5 + 0.48$ ) ppm was in excellent agreement with the predicted absolute gas-phase shift of  $-28.01$  ppm given by Eq. (12).

The absolute  $\text{H}_2$  gas-phase shift can be used to determine the matrix shift for  $\text{H}_2$  in a lithium-compound matrix. The correction for the matrix shift can then be applied to the  $1.3$  ppm peak to determine the gas-phase absolute shift to compare to

Eq. (12). The shifts of all of the peaks were relative to liquid-phase TMS which has an experimental absolute resonance shift of  $-31.5$  ppm relative to the proton's gyromagnetic frequency [78-79]. The experimental shift of  $H_2$  in a lithium-compound matrix of  $4.06$  ppm relative to liquid-phase TMS is shown in Figure 7 of Lu et al. [82] and corresponds to an absolute resonance shift of  $-27.44$  ppm ( $-31.5$  ppm +  $4.06$  ppm). Using the absolute  $H_2$  gas-phase resonance shift of  $-28.0$  ppm corresponding to  $3.5$  ppm ( $-28.0$  ppm –  $31.5$  ppm) relative to liquid TMS, the lithium-compound matrix effect is  $+0.56$  ppm ( $4.06$  ppm –  $3.5$  ppm) requiring a correction of the measured shift of  $-0.56$  ppm. Then, the peak upfield of  $H_2$  at  $1.26$  ppm peak relative to TMS corresponds to a matrix-corrected absolute resonance shift of  $-30.8$  ppm ( $-31.5$  ppm +  $1.26$  ppm –  $0.56$  ppm). Using Eq. (12), the data indicates  $p = 4$  and matches  $H_2(1/4)$ :

$$\begin{aligned}
 \frac{\Delta B_r}{B} &= -(28.01 + 0.64p) \text{ ppm} \\
 &= -(28.01 + 0.64(4)) \text{ ppm} \\
 &= -30.6 \text{ ppm}
 \end{aligned}
 \tag{46}$$

Lu et al. [82] also observed a peak at this position that increased in intensity relative to  $H_2$  with the duration of in situ heating of  $LiH + LiNH_2$  (1.1/1). They were unable to assign the peak labeled unknown in their Figures 6 and 7. The assignment of the peak that matched the theoretical shift of  $H_2(1/4)$  extremely well, was confirmed by FTIR (Sec. IIIG) and electron beam-excitation emission spectroscopy (Sec. IIIH).

The presence of the  $H^-(1/4)$  ion in  $LiH * X$  was found to depend on the polarizability of the halide ion. The  $^1H$  MAS NMR spectra of  $LiH * F$  and  $LiH * Cl$  are shown in Figures 32A and 32B, respectively. Peaks at  $4.3$  ppm and  $1.2$  ppm matched theoretical predictions of molecular hydrogen in two different quantum

states [1, 6]. The 4.3 ppm peak matched the assignment of Lu et al. [82] for  $H_2$ , and the 1.2 ppm peak labeled unknown by Lu et al. [82] matched  $H_2(1/4)$ . The  $H_2(1/4)$  assignment was confirmed by the observation of the predicted rotational transition in the FTIR spectrum (Sec. IIIG) and the predicted rotational spacing by electron beam-excitation emission spectroscopy (Sec. IIIH). The  $H^-(1/4)$  ion peak was absent in  $LiH * F$  comprising a nonpolarizable fluorine as well as in  $LiH * Cl$  comprising a nonpolarizable chlorine; whereas, it was the dominant peak in both  $LiH * Br$  and  $LiH * I$  as shown in Figures 30A and 30B, respectively. These results indicate that a polarizable halide is required for  $LiX$  to react with the  $H^-(1/4)$  ion to form the corresponding lithium halidohydride. Since molecular species are nonspecifically trapped in the crystalline lattice, the H-content selectivity of  $LiH * X$  for molecular species alone or in combination with  $H^-(1/4)$  ions is based on the polarizability of the halide and the corresponding reactivity towards  $H^-(1/4)$ . Potassium catalyst formed  $H_2(1/4)$  as well, but in  $KCl$  and  $KI$  matrices with  $H^-(1/4)$ , as shown in Figures 31A and 31B.

The  $^1H$  MAS NMR spectra of  $NaH * Br$  relative to external TMS is shown in Figure 32.  $NaH * Br$  showed a large distinct upfield resonance at  $-3.58$  ppm. None of the controls comprising  $NaH$  or equal molar mixtures of  $NaH$  and  $NaBr$  showed an upfield-shifted peak. The  $-3.58$  ppm upfield peak of  $NaH * Br$  was broadened, but not significantly as in the case of  $KH * I$ ; thus, the matrix may not have as large an effect as in the prior case of the identification of  $H^-(1/4)$  in  $KH * I$ . Thus, the measured shift is directly compared to theory with the expectation of that it is the peak shifted downfield due to the matrix effect. The experimental absolute resonance shift of TMS is  $-31.5$  ppm relative to the proton's gyromagnetic frequency

[78-79]. The novel peak at  $-3.58$  ppm relative to TMS corresponding to an absolute resonance shift of  $-35.08$  ppm indicates that  $p = 4$  in Eq. (4).  $H^-(1/4)$  is the favored hydride ion predicted by using  $NaH$  as the catalyst (Eqs. (3-4) and (23-27)). Similar to the case of  $LiH * X$ , the  $4.3$  ppm peak shown in Figure 33 is assigned to  $H_2$ , and the  $1.13$  ppm peak is assigned to  $H_2(1/4)$ . The latter is commonly observed as a favored catalysis molecular product [29].

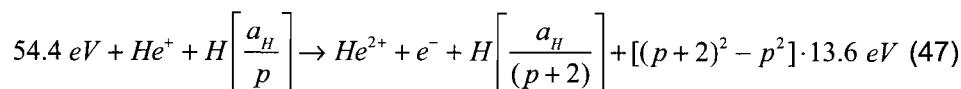
$NaH * Cl$   $^1H$  MAS NMR spectra relative to external TMS showing the effect of hydrogen addition on the relative intensities of  $H_2$ ,  $H_2(1/4)$ , and  $H^-(1/4)$  is shown in Figures 34A-B. The addition of hydrogen increased the  $H^-(1/4)$  peak and decreased the  $H_2(1/4)$  while the  $H_2$  increased. (A)  $NaH * Cl$  synthesized with hydrogen addition showing a  $-4$  ppm upfield-shifted peak assigned to  $H^-(1/4)$ , a  $1.1$  ppm peak assigned to  $H_2(1/4)$ , and a dominant  $4$  ppm peak assigned to  $H_2$ . (B)  $NaH * Cl$  synthesized without hydrogen addition showing a  $-4$  ppm upfield-shifted peak assigned to  $H^-(1/4)$ , a dominant  $1.0$  ppm peak assigned to  $H_2(1/4)$ , and a small  $4.1$  ppm assigned to  $H_2$ .

The effect of hydrogen addition on the relative  $^1H$  MAS NMR intensities of  $H_2$ ,  $H_2(1/4)$ , and  $H^-(1/4)$  in  $NaH * Cl$  is shown in Figures 34A-B. The dominant peak switched from being  $H_2$  to  $H_2(1/4)$  with the addition of external hydrogen indicating that  $H_2$  may occupy sites in the lattice that are filled by  $H_2(1/4)$  when  $H_2$  is less abundant. However, the addition of hydrogen increased the relative intensity of the  $H^-(1/4)$  peak, mostly likely by increasing the hydride reactant concentration.

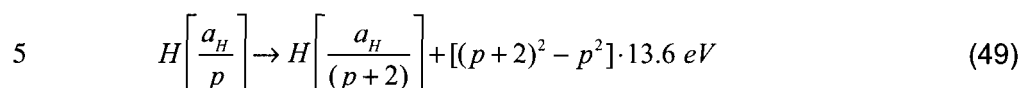
NMR was performed on  $NaH * Cl$  synthesized from  $NaCl$  and the solid acid  $KHSO_4$  as the only source of hydrogen to test whether  $H^-(1/3)$  formed by the reactions of Eqs. (23-25) could be observed when the rapid reaction to  $H^-(1/4)$  according to Eq. (27) was partially inhibited due to the absence of a high concentration of  $H$  from a dissociator with  $H_2$  or a hydride. The  $^1H$  MAS NMR spectrum of  $NaH * Cl$  formed using the solid acid relative to external TMS is shown in Figure 35. Peaks at  $-3.97$  ppm and  $1.15$  ppm matched the  $-4$  ppm and  $1.1$  ppm peaks of Figures 34A-B that were assigned to  $H^-(1/4)$  and  $H_2(1/4)$ , respectively, of  $NaH * Cl$  synthesized using  $H$  from a dissociator with  $H_2$  or a hydride. The close match was expected since the  $KHSO_4$  was only 6.5 mole% of the mixture with  $NaCl$  such that the matrix effect was essentially constant between samples. Uniquely, another set of peaks at  $-3.15$  ppm and  $1.7$  ppm was observed for the solid-acid product. Using Eqs. (4) and (12) with the matrix shift given previously for  $NaH * Cl$ , these peaks matched and were assigned to  $H^-(1/3)$  and  $H_2(1/3)$ , respectively. Curve fitting of two peaks put the peaks at about  $-3$  ppm and  $-4$  ppm, the theoretical values with experimental error. Thus, both fractional hydrogen states were present, and the  $H_2$  peak was absent at  $4.3$  ppm due to the synthesis of  $NaH * Cl$  using a solid acid as the only  $H$  source which confirms the reactions given by Eqs. (23-30). The presence of  $H^-(1/4)$  and  $H_2(1/4)$  in  $NaH * Cl$  from reaction of  $NaCl$  and the solid acid  $KHSO_4$  was confirmed by XPS and electron beam-excitation emission spectroscopy.

Helium is another catalyst that can cause a transition reaction to  $\left[ \frac{a_H}{3} \right]$  because the second ionization energy is  $54.4 \text{ eV}$ ,  $(2 \cdot 27.2 \text{ eV})$ . The catalyst

reactions are given by



And, the overall reaction is



As in the case of the *NaH* catalyst reaction, the subsequently rapid transition of the

*He*<sup>+</sup> catalysis product  $\left[ \frac{a_H}{3} \right]$  to  $\left[ \frac{a_H}{4} \right]$  may occur via further catalysis by atomic

hydrogen that first accepts 27.2 eV from  $\left[ \frac{a_H}{3} \right]$  as given by Eq. (27). Characteristic

broad emission starting at 46.5 nm and continuing to shorter wavelengths is

10 predicted for this transition reaction as the energetic H catalyst decays. The

emission has been observed by EUV spectroscopy recorded on microwave

discharges of helium with 2% hydrogen [27-29]. The spectroscopic and NMR data

provide strong support for the catalyst mechanism of the formation of  $\left[ \frac{a_H}{3} \right]$  with the

subsequent transition to  $\left[ \frac{a_H}{4} \right]$ . Additional evidence is the observation of both

15  $\text{H}^-(1/3)$  and  $\text{H}^-(1/4)$  in *NaH \* Cl* as given in Sec. IIIF.

F. XPS Identification of  $\text{H}^-(1/4)$  and  $\text{H}^-(1/3)$ . A survey spectrum was obtained

on each of *LiBr* and *LiH \* Br* over the region  $E_b = 0 \text{ eV}$  to 1200 eV (Figures 36A-B).

The primary element peaks allowed for the determination of all of the elements

20 present in the *LiH \* Br* crystals and the control *LiBr*. No elements were present in

the survey scan which could be assigned to peaks in the low binding energy region (Figure 37) with the exception of the *Li* 1*s* peak at 55 eV (shifted 1 eV lower compared to *LiBr*), the *O* 2*s* at 23 eV, the *Br* 3*d*<sub>5/2</sub> and *Br* 3*d*<sub>3/2</sub> peaks at 69 eV and 70 eV, respectively, the *Br* 4*s* at 15 eV, and the *Br* 4*d* at 5 eV. Accordingly, any other peaks in this region must be due to novel species. As shown in Figure 37, the XPS spectrum of *LiH* \* *Br* differs from that of *LiBr* by having additional peaks at 9.5 eV and 12.3 eV that do not correspond to any other primary element peaks but do match the  $H^-(1/4)$   $E_b = 11.2$  eV hydride ion (Eqs. (4) and (16)). The literature was searched for elements having a peak in the valence-band region that could be assigned to these peaks. Given the primary element peaks present, there was no known alternative assignment. Thus, the 9.5 eV and 12.3 eV peaks that could not be assigned to known elements and do not correspond to any other primary element peak were assigned to the  $H^-(1/4)$  in two different chemical environments. These features closely matched those for  $H^-(1/4)$  of *KH* \* *I* reported previously [13-15, 26, 30].

A survey spectrum was obtained on each of *NaBr* and *NaH* \* *Br* over the region  $E_b = 0$  eV to 1200 eV (Figures 38A-B). The primary element peaks allowed for the determination of all of the elements present in the *NaH* \* *Br* crystals and the control *NaBr*. No elements were present in the survey scan which could be assigned to peaks in the low binding energy region (Figure 39) with the exception of the *Na* 2*p* and *Na* 2*s* peaks at 30 eV and 63 eV (shifted 1 eV lower compared to *NaBr*), the *O* 2*s* at 23 eV, the *Br* 3*d*<sub>5/2</sub> and *Br* 3*d*<sub>3/2</sub> peaks at 69 eV and 70 eV, respectively, the *Br* 4*s* at 15.2 eV, and the *Br* 4*d* at 5 eV. Accordingly, any other peaks in this region must be due to novel species. As shown in Figure 39, the XPS

spectrum of  $NaH^*Br$  differs from that of  $NaBr$  by having additional peaks at 9.5 eV and 12.3 eV that do not correspond to any other primary element peaks but do match the  $H^-(1/4) E_b = 11.2 \text{ eV}$  hydride ion (Eqs. (4) and (16)). The literature was searched for elements having a peak in the valance-band region that could be assigned to these peaks. Given the primary element peaks present, there was no known alternative assignment. Thus, the 9.5 eV and 12.3 eV peaks that could not be assigned to known elements and do not correspond to any other primary element peak were assigned to the  $H^-(1/4)$  in two different chemical environments.

Survey spectra over the region  $E_b = 0 \text{ eV}$  to  $1200 \text{ eV}$  were obtained on each of  $Pt/Ti$  and  $NaH^*$ -coated  $Pt/Ti$  following the production of 15 kJ of excess heat (Figures 40A-B). The primary element peaks allowed for the determination of all of the elements present in the  $NaH^*$ -coated  $Pt/Ti$  and the control  $Pt/Ti$ . No elements were present in the survey scan which could be assigned to peaks in the low binding energy region (Figures 41A-B) with the exception of the  $Pt 4f_{7/2}$  and  $Pt 4f_{5/2}$  peaks at 70.7 eV and 74 eV, respectively, and the  $O 2s$  at 23 eV. The  $Na 2p$  and  $Na 2s$  peaks were observed at 31 eV and 64 eV on  $NaH^*$ -coated  $Pt/Ti$ , and a valance band was only observed for  $Pt/Ti$ . Accordingly, any other peaks in this region must be due to novel species. As shown in Figures 42A-B, the XPS spectrum of  $NaH^*$ -coated  $Pt/Ti$  differs from that of  $Pt/Ti$  by having additional peaks at 6 eV, 10.8 eV, and 12.8 eV that do not correspond to any other primary element peaks but do match the  $H^-(1/3) E_b = 6.6 \text{ eV}$  and  $H^-(1/4) E_b = 11.2 \text{ eV}$  hydride ions (Eqs. (4) and (16)). The literature was searched for elements having a peak in the valance-band region that could be assigned to these peaks. Given the primary element peaks present, there was no known

alternative assignment. Thus, the 10.8 eV, and 12.8 eV peaks that could not be assigned to known elements and do not correspond to any other primary element peak were assigned to the  $H^-(1/4)$  in two different chemical environments. The 6 eV peak matched and was assigned to  $H^-(1/3)$ . Thus, in the absence of a halide peak in this region, both fractional hydrogen states, 1/3 and 1/4, were observed as predicted by Eq. (27). The absence of a valance band due to the high-binding energies was also consistent with the hydrido hydride assignments of  $NaH^*$ -coated  $Pt/Ti$ .

The results of the  $NaH^*$ -coated  $Pt/Ti$  shown in Figure 42B were replicated with  $NaH^*$ -coated  $Si$ . As shown in Figures 43 and 44, the XPS spectra of  $NaH^*$ -coated  $Si$  showed peaks at 6 eV, 10.8 eV, and 12.8 eV that could not be assigned to known elements and do not correspond to any other primary element peak, but matched  $H^-(1/3)$  and  $H^-(1/4)$ . Thus, both fractional hydrogen states, 1/3 as  $H^-(1/3)$  at the 6 eV and 1/4 as  $H^-(1/4)$  at 10.8 eV and 12.8 eV, were present as predicted by Eq. (27).

G. FTIR Identification of  $H_2(1/4)$ . Samples of  $LiH^*Br$  having an upfield-shifted  $^1H$  NMR peak at  $-2.5$  ppm assigned to  $H^-(1/4)$  and an NMR peak at 1.3 ppm assigned to the corresponding molecule  $H_2(1/4)$  were analyzed by high resolution FTIR spectroscopy. As shown in Figure 45B, a single narrow peak was observed at  $1989\text{ cm}^{-1}$ . The compounds,  $LiNH_2$ ,  $Li_2NH$ , and  $Li_3N$  are possible, based on the starting materials and predicted reactions, but none of these compounds showed peaks in the region of  $1989\text{ cm}^{-1}$ . No additional peaks other than those easily assignable to  $LiBr$  were observed (Figure 45A). An exhaustive list of species that

have features in this region were considered, including exotic species such as azide, metal carbonyls, and metaborate ion. The former were eliminated based on their known spectra, which have very broad bands. Metaborate ion was eliminated by ToF-SIMS analysis, which showed a total boron content that was not detectable at the ppb level which is orders of magnitude below its FTIR detection limit and the absence of two peaks corresponding to the boron isotopes  $^{10}\text{B}$  (20% N.A.) and  $^{11}\text{B}$  (80% N.A.).

Considering a possible matrix effect, the peak at  $1989\text{ cm}^{-1}$  ( $0.24\text{ eV}$ ) matched the theoretical prediction of  $1947\text{ cm}^{-1}$  for  $\text{H}_2(1/4)$ . From Eqs. (14-15), the unprecedented rotational energy of  $4^2$  times that of ordinary hydrogen establishes the internuclear distance of  $\text{H}_2(1/4)$  as  $1/4$  that of  $\text{H}_2$ . Interstitial  $\text{H}_2$  in silicon and *GaAs* is a nearly free rotator showing single rovibrational transitions [83-87].  $\text{H}_2$  is FTIR active as well as Raman active due to the induced dipole from interactions with the crystalline lattice [83]. The crystalline lattice may also influence the selection rules to permit an otherwise forbidden transition in  $\text{H}_2(1/4)$ . Considering a matrix effect, the match to the predicted  $1943\text{ cm}^{-1}$  peak and the relatively narrow peak width, indicates that  $\text{H}_2(1/4)$  can rotate essentially freely inside of the crystal and confirms its small size corresponding to  $1/4$  the dimensions of ordinary hydrogen. Ordinary hydrogen shows a 3:1 ortho-para ratio at non-cryogenic temperatures; whereas, a single peak of  $\text{H}_2(1/4)$  formed under the synthesis conditions is assigned to the para form only due to the 64 times increase in stability due to the  $1/4$  relative internuclear separation. Given the frequency match of the  $1989\text{ cm}^{-1}$  peak and the absence of any known alternative, wherein hydrogen is the only known species that exhibits single rovibrational transitions in a solid matrix, the  $1989\text{ cm}^{-1}$  peak is assigned to the  $J = 0$  to  $J = 1$  rotational transitions of para  $\text{H}_2(1/4)$ .

H.  $H_2(1/4)$  Rotational UV Spectrum by Electron Beam Excitation.  $H_2(1/4)$  trapped in the lattice of alkali halides,  $MgX_2$  ( $X = F, Cl, Br, I$ ), and  $CuX_2$  ( $X = F, Cl, Br$ ) was investigated by windowless UV spectroscopy on electron beam

5 excitation of the crystals using the 12.5 keV electron gun at a beam current of 10-20  $\mu A$  in the pressure range of  $<10^{-5}$  Torr. Of the alkali metals, it was found that only alkali chlorides showed the peaks predicted by Eq. (14), and the intensity roughly matched the order predicted, increasing intensity down the column of the Group I elements. In all cases, the peaks could be eliminated by heating with the loss of the

10 Lyman  $\alpha$  peak, and no other peaks were observed in the UV. The on-line mass spectrometer recorded hydrogen only. Of the compounds of the series  $MgX_2$  ( $X = F, Cl, Br, I$ ) and  $CuX_2$  ( $X = F, Cl, Br$ ), the predicted band was just detectable only for  $MgI_2$  which, in this case, can be attributed to  $Mg^{2+}$  as the catalyst. NMR on these crystals showed the  $H_2(1/4)$  peak at 1.13 ppm only in  $MgX_2$  with relative

15 intensities  $F, Cl, Br, \ll I$  that matched the detection of the band by electron beam-excitation emission for  $MgI_2$  only.

The 100-350 nm spectrum of electron beam-excited  $CsCl$  crystals having trapped  $H_2(1/4)$  is shown in Figure 46. A series of evenly spaced lines was observed in the 220-300 nm region as shown in Figure 46. The series matched the

20 spacing and intensity profile of the P branch of  $H_2(1/4)$  given by Eq. (14). P(1), P(2), P(3), P(4), P(5), and P(6) were observed at 226.0 nm, 237.0 nm, 249.5 nm, 262.5 nm, 277.0 nm, and 292.5 nm, respectively. The slope of the linear curve-fit of the energies of the peaks shown in Figure 46 is 0.25 eV with an intercept of 5.73 eV and a sum of residual errors  $r^2 < 0.0000$ . The slope matches the predicted rotational

energy spacing of 0.241 eV (Eq. (14);  $p = 4$ ) with  $\Delta J = +1$ ;  $J = 1, 2, 3, 4, 5, 6$  where  $J$  is the rotational quantum number of the final state.  $H_2(1/4)$  is a free rotator, but is not a free vibrator which is similar to the case of interstitial hydrogen in silicon discussed previously [83-87]. The observed intercept of 5.73 eV is shifted from the predicted

5  $v=1 \rightarrow v=0$  vibrational energy of  $H_2(1/4)$  of 8.25 eV (Eq. (13)) by about twice the percentage as that of interstitial  $H_2$  in silicon [83-87]. In the latter case, vibrational energy of free  $H_2$  is  $4161 \text{ cm}^{-1}$ , whereas the vibrational peaks in silicon are observed at 3618 and  $3627 \text{ cm}^{-1}$  corresponding to ortho and para- $H_2$ , respectively [83]. In the former case the shift is about 30% lower, possibly due to an increase in

10 the effective mass from coupling of the molecular vibrational mode with the crystal lattice.

Using Eqs. (14) and (15) with the measured rotational energy spacing of 0.25 eV establishes an internuclear distance of 1/4 that of the ordinary  $H_2$  for  $H_2(1/4)$ . A corresponding weak band was observed from  $NaH * Br$ , and a more intense band

15 was observed from  $NaH * Cl$ . Regarding the latter case, the intensity of the emission was significantly increased by trapping  $H_2(1/4)$  in a silicon matrix. The 100-550 nm spectrum of an electron beam-excited silicon wafer coated with  $NaH * Cl$  having trapped  $H_2(1/4)$  is shown in Figure 47. The series matching the spacing and intensity profile of the P branch of  $H_2(1/4)$  given by Eq. (14) was

20 observed. P(1), P(2), P(3), P(4), P(5), and P(6) were observed at 222.5 nm, 233.4 nm, 245.2 nm, 258.2 nm, 272.2 nm, and 287.4 nm, respectively. The slope of the linear curve-fit of the energies of the peaks shown in Figure 47 is 0.25 eV with an intercept of 5.82 eV and a sum of residual errors  $r^2 < 0.0000$ . The linearity is characteristic of rotation, and the results again match  $H_2(1/4)$ . This technique

confirms the solid NMR and FTIR results given in Secs. IIIE and IIIG, respectively. It was reported previously [13-14] that when  $KH^*Cl$  having  $H^-(1/4)$  by NMR was incident to the 12.5 keV electron beam, similar excited emission of interstitial  $H_2(1/4)$  was observed as that from electron-beam excited alkali chlorides,  $NaH^*Cl$ -coated  $Si$ , and argon-hydrogen plasmas [13-14]. It was further observed that the band assigned to  $H_2(1/4)$  was eliminated from the  $KCl$  starting material by heating to high temperature.  $KH^*Cl$  was then synthesized from the heat-treated  $KCl$ , and  $H_2(1/4)$  trapped in the lattice of  $KH^*Cl$  was then observed in addition to  $H^-(1/4)$  demonstrating that multiple catalysts,  $HCl$ ,  $NaH$ ,  $K$ , and  $Ar^+$ , can give rise to  $H_2(1/4)$ .

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**What Is Claimed Is:**

1. A power source and hydride reactor, comprising:
  - a reaction cell for the catalysis of atomic hydrogen to form novel hydrogen species and compositions of matter comprising new forms of hydrogen;
  - 5 a reaction vessel constructed and arranged to contain a pressure in the range of lower, equal to, or greater than atmospheric pressure;
  - a vacuum pump;
  - a source of atomic hydrogen from a source in communication with the reaction vessel;
  - 10 a source of a hydrogen catalyst in communication with the reaction vessel comprising a reaction mixture of at least one reactant comprising the element or elements that form the catalyst and at least one other element, whereby the catalyst is formed from the source; and
  - a heater to heat the vessel to initiate the formation the catalyst in the reaction
  - 15 vessel if the reaction is not spontaneous at ambient temperature, whereby the catalysis of atomic hydrogen releases energy in an amount greater than about 300 kJ per mole of hydrogen during the catalysis of the hydrogen atom.
2. A power source and hydride reactor of claim 1, comprising an energy cell for
  - 20 the catalysis of atomic hydrogen to form novel hydrogen species and compositions of matter comprising new forms of hydrogen, a source of hydrogen catalyst, and a source of atomic hydrogen whereby the source of hydrogen catalyst comprises at least one reactant having hydrogen and at least one other element, and
  - the at least one reactant undergoes reaction such that the energy released is
  - 25 greater than the difference between the standard enthalpy of formation of

compounds having the stoichiometry or elemental composition of the products and the energy of formation of the at least one reactant.

3. The power source and hydride reactor of claim 1, whereby the source of  
5 hydrogen catalyst comprises at least one reactant having hydrogen and at least one other element, and

the at least one reactant undergoes reaction such that the energy released is greater than the theoretical standard enthalpy required to regenerate the at least one reactant from the products wherein the energy to replace any reacted hydrogen is  
10 the standard value.

4. The power source and hydride reactor of claim 1, for generating power comprising reactants of hydrogen and at least one other element that undergo reaction such that the energy released is greater than the difference between the  
15 standard enthalpy of formation of compounds having the stoichiometry or elemental composition of the products and the energy of formation of the reactants.

5. The power source and hydride reactor of claim 1, for generating power comprising reactants of hydrogen and at least one other element that undergo  
20 reaction such that the energy released is greater than the theoretical standard enthalpy required to regenerate the reactants from the products wherein the energy to replace any reacted hydrogen is the standard value for the combustion of the hydrogen.

6. The power source and hydride reactor of claim 1, wherein the catalyst is capable of accepting energy from atomic hydrogen in integer units of one of about  $27.2 \text{ eV} \pm 0.5 \text{ eV}$  and  $\frac{27.2}{2} \text{ eV} \pm 0.5 \text{ eV}$ .
- 5 7. The power source and hydride reactor of claim 1, wherein the catalyst comprises an atom or ion M wherein the ionization of  $t$  electrons from the atom or ion M each to a continuum energy level is such that the sum of ionization energies of the  $t$  electrons is approximately one of  $m \cdot 27.2 \text{ eV}$  and  $m \cdot \frac{27.2}{2} \text{ eV}$  where  $m$  is an integer.
- 10 8. A power source and hydride reactor of claim 7 wherein the catalyst atom M is at least one of the group of atomic Li, K, and Cs.
9. A power source and hydride reactor of claim 8 wherein the source of catalyst
- 15 comprises a diatomic molecule of catalyst atoms.
10. A power source and hydride reactor of claim 8 wherein the reaction mixture comprises at least a first reactant as the source of atomic catalyst and atomic hydrogen comprising one of the group of Li, K, Cs, and H;
- 20 the reaction mixture further comprising at least one other reactant wherein the atomic hydrogen and atomic catalyst are formed by reaction of at least one first and at least one other reactant.

11. A power source and hydride reactor of claim 10 wherein the source of catalyst comprises MH wherein M is the catalyst atom whereby atomic catalyst is formed from the source by reaction with a species comprising at least one other element.

5 12. The catalyst of claim 1 comprising a diatomic molecule MH wherein the breakage of the M-H bond plus the ionization of  $t$  electrons from the atom M each to a continuum energy level is such that the sum of the bond energy and ionization energies of the  $t$  electrons is approximately one of  $m \times 27.2 \text{ eV}$  and  $m \cdot \frac{27.2}{2} \text{ eV}$  where  $m$  is an integer.

10

13. A power source and hydride reactor of claim 12 wherein the source of catalyst comprises a reaction that generates a diatomic molecule comprising hydrogen and another element.

15 14. A power source and hydride reactor of claim 13 wherein the catalyst comprises hydrogen and an element other than hydrogen.

15. A power source and hydride reactor of claim 14 wherein the catalyst and source of reactant atomic hydrogen comprises a diatomic molecule of hydrogen and  
20 another element.

16. A power source and hydride reactor of claim 15 wherein the catalyst comprises at least one of molecular AlH, BiH, ClH, CoH, GeH, InH, NaH, RuH, SbH, SeH, SiH, and SnH.

25

17. A power source and hydride reactor of claim 1 comprising at least one of the element or elements of the catalyst, another element, and a composition of matter of the same composition as that of the catalyst, but in a different physical state from that of the catalyst.

5

18. A power source and hydride reactor of claim 1 wherein the source of catalyst comprises hydrogen and another element other than hydrogen.

19. A power source and hydride reactor of claim 1 wherein the reaction mixture  
10 comprises a catalyst or a source of catalyst and atomic hydrogen or a source of atomic hydrogen (H) wherein at least one of the catalyst and atomic hydrogen is released by a chemical reaction of at least one species of the reaction mixture or between two or more reaction-mixture species.

15 20. A power source and hydride reactor of claim 19 wherein the species may be at least one of an element, complex, alloy, or a compound such as a molecular or inorganic compound wherein each may be at least one of a reagent or product in the reactor.

20 21. A power source and hydride reactor of claim 20 wherein the species may form a complex, alloy, or compound with at least one of hydrogen and the catalyst.

22. A power source and hydride reactor of claim 21 wherein the element or alloy  
comprises at least one of M (catalyst atom), H, Al, B, Si, C, N, Sn, Te, P, S, Ni, Ta,  
25 Pt, and Pd.

23. A power source and hydride reactor of claim 22 wherein the catalyst atom M is at least one of the group of Li, K, Cs, and Na and the catalyst is atomic Li, K, and Cs, and molecular NaH.

5

24. A power source and hydride reactor of claim 23 wherein one or more of the reaction-mixture species may form one or more reaction product species such that the energy to release H or free catalyst is lowered relative to the case in the absence of the formation of the reaction product species.

10

25. A power source and hydride reactor of claim 24 wherein the reaction to generate at least one of atomic H and catalyst is reversible.

15

26. A power source and hydride reactor of claim 25 wherein the complex, alloy, or compound comprises a lithium alloy or compound.

20

27. A power source and hydride reactor of claim 26 wherein the reaction mixture comprises at least one of alloy or compound of the group of  $\text{LiAlH}_4$ ,  $\text{Li}_3\text{AlH}_6$ ,  $\text{LiBH}_4$ ,  $\text{Li}_3\text{N}$ ,  $\text{Li}_2\text{NH}$ ,  $\text{LiNH}_2$ ,  $\text{NH}_3$ ,  $\text{H}_2$ ,  $\text{LiNO}_3$ ,  $\text{Li/Ni}$ ,  $\text{Li/Ta}$ ,  $\text{Li/Pd}$ ,  $\text{Li/Te}$ ,  $\text{Li/C}$ ,  $\text{Li/Si}$ , and  $\text{Li/Sn}$ .

25

28. A power source and hydride reactor of claim 27 wherein the reaction mixture comprises one or more compounds that react with a source of Li to form Li catalyst, the reaction mixture comprises at least one species from the group of  $\text{LiNH}_2$ ,  $\text{Li}_2\text{NH}$ ,  $\text{Li}_3\text{N}$ ,  $\text{Li}$ ,  $\text{LiH}$ ,  $\text{NH}_3$ ,  $\text{H}_2$ , and a dissociator.

29. A power source and hydride reactor of claim 28 wherein the reaction mixture comprises LiH, LiNH<sub>2</sub>, and Pd on Al<sub>2</sub>O<sub>3</sub> powder.
- 5 30. A power source and hydride reactor of claim 29 wherein the reaction mixture comprises Li, Li<sub>3</sub>N, and hydrided Pd on Al<sub>2</sub>O<sub>3</sub> powder and optionally H<sub>2</sub> gas.
31. A power source and hydride reactor of claim 25 wherein the source of catalyst comprises a source of NaH catalyst, wherein the source of NaH is an alloy of Na and  
10 a source of hydrogen.
32. A power source and hydride reactor of claim 31 wherein the alloy source of catalyst comprises at least one of sodium metal and one or more other nitrogen-based compounds, alkaline or alkaline earth metals, transition metals, Al, Sn, Bi, Ag,  
15 In, Pb, Hg, Si, Zr, B, Pt, Pd, or other metals and the H source comprises H<sub>2</sub> or a hydride.
33. A power source and hydride reactor of claim 32 wherein the source of hydrogen catalyst comprises an inorganic compound comprising Na.  
20
34. A power source and hydride reactor of claim 33 wherein the reaction mixture comprises one or more compounds that react with a source of NaH to form NaH catalyst;  
at least one of the source of NaH catalyst and the reaction mixture comprising  
25 at least one of Na, NaH, an alkaline or alkaline earth hydroxide, aluminum hydroxide,

an alkali metal, an alkaline earth metal, NaOH-doped R-Ni, NaOH, Na<sub>2</sub>O, and Na<sub>2</sub>CO<sub>3</sub>, and at least one species from the group of NaNH<sub>2</sub>, Na<sub>2</sub>NH, Na<sub>3</sub>N, Na, NaH, NH<sub>3</sub>, H<sub>2</sub> and a dissociator.

- 5 35. A power source and hydride reactor of claim 33 wherein the reaction mixture comprises one or more compounds that react with a source of NaH to form NaH catalyst;

the reaction mix comprises at least one species from the group of NaNH<sub>2</sub>, Na<sub>2</sub>NH, Na<sub>3</sub>N, Na, NaH, NH<sub>3</sub>, H<sub>2</sub> and a dissociator.

10

36. A power source and hydride reactor of claim 35 wherein the reaction mixture comprises one or more compounds that react with a source of NaH to form NaH catalyst;

the reaction mix comprises at least one species from the group of NaH, Na, a  
15 metal, a metal hydride, a lanthanide metal, a lanthanide metal hydride, lanthanum, lanthanum hydride, H<sub>2</sub>, and a dissociator.

37. A power source and hydride reactor of claim 34 wherein the reaction mixture comprises at least one of NaH molecules and a source of NaH molecules whereby  
20 the NaH molecules serve as the catalyst to form H states given by

$$\text{Binding Energy} = \frac{13.6 \text{ eV}}{(1/p)^2} \text{ where } p \text{ is an integer greater than 1, preferably from 2 to}$$

137;

the source of NaH molecules comprise at least one of:

- (a) Na metal, atomic Na, a source of hydrogen, atomic hydrogen, and

NaH(s);

(b) R-Ni comprising NaOH and a reactant to form NaH comprising a reductant, and  
a source of hydrogen.

5

38. A power source and hydride reactor of claim 34 wherein the reaction mixture comprises at least one of

reactant comprising a reductant to form NaH from NaOH;

the source of hydrogen comprising at least one of NaH, H<sub>2</sub> gas and a

10 dissociator, and a hydride.

39. A power source and hydride reactor of claim 34 whereby one of atomic sodium and molecular NaH is provided by a reaction between a metallic, ionic, or molecular form of Na and at least one other compound or element;

15 the source of Na or NaH is at least one of metallic Na, NaNH<sub>2</sub>, NaOH, NaX (X is a halide), and NaH(s);

the other element is H, a displacing agent, or a reducing agent.

40. A power source and hydride reactor of claim 34 wherein the reaction mixture  
20 comprises at least one of;

(1) a source of sodium;

(2) a support material;

(3) a source of hydrogen;

(4) a displacing agent, and

25 (5) a reductant or reducing agent.

41. A power source and hydride reactor of claim 40 wherein the source of sodium comprises Na, NaH, NaNH<sub>2</sub>, NaOH, NaOH coated R-Ni, NaX (X is a halide), and NaX coated R-Ni.

5

42. A power source and hydride reactor of claim 41 wherein the reductant or reducing agent comprises at least one of a metal such as an alkaline metal, alkaline earth metal, a lanthanide, a transition metal such as Ti, aluminum, B, a metal alloy such as AlHg, NaPb, NaAl, LiAl, and a source of a metal alone or in combination  
10 with reducing agent such as an alkaline earth halide, a transition metal halide, a lanthanide halide, an aluminum halide, metal hydrides such as LiBH<sub>4</sub>, NaBH<sub>4</sub>, LiAlH<sub>4</sub>, or NaAlH<sub>4</sub>, and an alkaline or alkaline earth metal and an oxidant such as AlX<sub>3</sub>, MgX<sub>2</sub>, LaX<sub>3</sub>, CeX<sub>3</sub>, and TiX<sub>n</sub> where X is a halide, preferably Br or I.

15 43. A power source and hydride reactor of claim 42 wherein the source of hydrogen comprises H<sub>2</sub> gas and a dissociator and a hydride.

44. A power source and hydride reactor of claim 43 wherein the displacing agent comprises at least one of an alkali metal, alkaline earth metal, alkali metal hydride,  
20 and alkaline earth metal hydride.

45. A power source and hydride reactor of claim 44 wherein the support comprises at least one of R-Ni, Al, Sn, Al<sub>2</sub>O<sub>3</sub> such as gamma, beta, or alpha alumina, aluminates, sodium aluminate, alumina nanoparticles, porous Al<sub>2</sub>O<sub>3</sub>, Pt, Ru,  
25 or Pd/Al<sub>2</sub>O<sub>3</sub>, carbon, Pt or Pd/C, inorganic compounds such as Na<sub>2</sub>CO<sub>3</sub>, lanthanide

oxides such as  $M_2O_3$  (preferably  $M = \text{La, Sm, Dy, Pr, Tb, Gd, and Er}$ ), Si, silica, silicates, zeolites, Y zeolite powder, lanthanides, transition metals, metal alloys such as alkali and alkali earth alloys with Na, rare earth metals,  $\text{SiO}_2\text{-Al}_2\text{O}_3$  or  $\text{SiO}_2$  supported Ni, and other supported metals such as at least one of alumina supported  
5 platinum, palladium, and ruthenium.

46. A power source and hydride reactor of claim 45 wherein the dissociator comprises at least one of Raney nickel (R-Ni), a precious or noble metal, and a precious or noble metal on a support where in the precious or noble metal may be  
10 Pt, Pd, Ru, Ir, and Rh, and the support may be at least one of Ti, Nb,  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$  and combinations thereof;

Pt or Pd on carbon, a hydrogen spillover catalyst, nickel fiber mat, Pd sheet, Ti sponge, Pt or Pd electroplated on Ti or Ni sponge or mat, TiH, Pt black, and Pd black, refractory metals such as molybdenum and tungsten, transition metals such  
15 as nickel and titanium, inner transition metals such as niobium and zirconium, and a refractory metal such as tungsten or molybdenum, and the dissociating material may be maintained at elevated temperature.

47. A power source and hydride reactor of claim 40 wherein the source of NaH  
20 may be R-Ni comprising NaOH and a reactant to form NaH, and the reactant is a reductant comprising at least one of an alkali metal, alkaline earth metal, and the Al intermetallic of R-Ni.

48. A power source and hydride reactor, comprising:

a reaction cell for the catalysis of atomic hydrogen to form novel hydrogen species and compositions of matter comprising new forms of hydrogen;

a reaction vessel constructed and arranged to contain a pressure in the range of lower, equal to, or greater than atmospheric pressure;

5 a vacuum pump;

a source of atomic hydrogen from a source in communication with the reaction vessel;

a source of a hydrogen catalyst M in communication with the reaction vessel, whereby the ionization of  $t$  electrons from the catalyst each to a continuum energy level is such that the sum of the ionization energies of the  $t$  electrons is

approximately one of  $m \cdot 27.2 \text{ eV}$  and  $m \cdot \frac{27.2}{2} \text{ eV}$  where  $m$  is an integer ;

a reaction mixture that forms catalyst from the source of catalyst, if the catalyst is not already present; and

a heater to heat the vessel to initiate at least one of the reaction for the formation of the catalyst and the hydride reaction in the reaction vessel if the reaction is not spontaneous at ambient temperature, whereby the catalyzed atomic H releases energy in an amount greater than about 300 kJ per mole of hydrogen during the catalysis of the hydrogen atom.

20 49. A power source and hydride reactor of claim 48 wherein the catalyst atom M is at least one of the group of atomic Li, K, and Cs.

50. A power source and hydride reactor, comprising:

a reaction cell for the catalysis of atomic hydrogen to form novel hydrogen species and compositions of matter comprising new forms of hydrogen;

a reaction vessel constructed and arranged to contain a pressure in the range of lower, equal to, or greater than atmospheric pressure;

a vacuum pump;

5 a source of atomic hydrogen from a source in communication with the reaction vessel;

a source of at least one of the group of atomic Li, K, and Cs catalyst in communication with the reaction vessel;

a reaction mixture that forms atomic catalyst from the source of atomic catalyst, if the catalyst is not already present; and

10 a heater to heat the vessel to initiate the formation of at least one of atomic Li, K, and Cs catalyst in the reaction vessel if the reaction is not spontaneous at ambient temperature, whereby the catalyst reaction with H releases energy in an amount greater than about 300 kJ per mole of hydrogen during the catalysis of the hydrogen atom.

15

51. A power source and hydride reactor of claim 50 wherein the reaction mixture comprises LiH, LiNH<sub>2</sub>, and Pd on Al<sub>2</sub>O<sub>3</sub> powder.

52. A power source and hydride reactor of claim 50 wherein the reaction mixture  
20 comprises Li, Li<sub>3</sub>N, and hydrided Pd on Al<sub>2</sub>O<sub>3</sub> powder and optionally H<sub>2</sub> gas.

53. A power source and hydride reactor, comprising:

a reaction cell for the catalysis of atomic hydrogen to form novel hydrogen species and compositions of matter comprising new forms of hydrogen;

a reaction vessel constructed and arranged to contain a pressure in the range of lower, equal to, or greater than atmospheric pressure;

a vacuum pump;

a source of a hydrogen catalyst in communication with the reaction vessel

- 5 comprising MH, whereby the breakage of the M-H bond plus the ionization of  $t$  electrons from the atom M each to a continuum energy level is such that the sum of the bond energy and ionization energies of the  $t$  electrons is approximately one of  $m \cdot 27.2 \text{ eV}$  and  $m \cdot \frac{27.2}{2} \text{ eV}$  where  $m$  is an integer ;

- a reaction mixture that forms molecular MH from the source of molecular MH,  
10 if the molecular MH is not already present; and

- a heater to heat the vessel to initiate the formation of molecular MH in the reaction vessel if the reaction is not spontaneous at ambient temperature, whereby the molecular MH serves as a hydrogen catalyst and a source of H reactant with the release of energy in an amount greater than about 300 kJ per mole of hydrogen  
15 during the catalysis of the hydrogen atom.

54. A power source and hydride reactor or claim 53, further comprising a source of atomic hydrogen from a source in communication with the reaction vessel.

- 20 55. A power source and hydride reactor of claim 53 wherein MH comprises at least one from the group of AlH, BiH, ClH, CoH, GeH, InH, NaH, RuH, SbH, SeH, SiH, and SnH.

56. A power source and hydride reactor, comprising:

a reaction cell for the catalysis of atomic hydrogen to form novel hydrogen species and compositions of matter comprising new forms of hydrogen;

a reaction vessel constructed and arranged to contain a pressure in the range of lower, equal to, or greater than atmospheric pressure;

5 a vacuum pump;

a source of molecular NaH catalyst in communication with the reaction vessel;

a reaction mixture that forms molecular NaH from the source of molecular NaH, if the molecular NaH is not already present; and

10 a heater to heat the vessel to initiate the formation of molecular NaH in the reaction vessel if the reaction is not spontaneous at ambient temperature, whereby the molecular NaH serves as a hydrogen catalyst and a source of H reactant with the release of energy in an amount greater than about 300 kJ per mole of hydrogen during the catalysis of the hydrogen atom.

15 57. A power source and hydride reactor or claim 56, further comprising a source of atomic hydrogen from a source in communication with the reaction vessel.

58. A power source and hydride reactor of claim 56 wherein the reaction mixture comprises NaH and Pd on  $\text{Al}_2\text{O}_3$  powder.

20

59. A power source and hydride reactor of claim 56 wherein the reaction mixture comprises Na and R-Ni comprising about 0.5 wt% NaOH wherein Na serves as the reductant.

60. A power source and hydride reactor of claim 56 wherein the reaction mixture comprises R-Ni comprising about 0.5 wt% NaOH wherein intermetallic Al serves as the reductant.
- 5 61. A power source and hydride reactor of claim 56 wherein the reaction mixture comprises NaH, La, and Pd on Al<sub>2</sub>O<sub>3</sub> powder.
62. A power source and hydride reactor of claim 56 wherein the reaction mixture comprises NaH, NaNH<sub>2</sub>, and Pd on Al<sub>2</sub>O<sub>3</sub> powder.
- 10 63. A power plant comprising:
- at least one reaction vessel constructed and arranged to contain a pressure in the range of lower, equal to, or greater than atmospheric pressure;
- a vacuum pump in communication with the reaction vessel;
- 15 a reaction mixture comprising
- a first source of hydrogen atoms in communication with the reaction vessel;
- a source of catalyst in communication with the reaction vessel;
- a heater for intimating the catalysis reaction;
- a means to regenerate the reaction mixture, and
- 20 a power converter.
64. The power plant according to claim 63, wherein the converter comprises a steam generator in communication with the reaction vessel, a steam turbine in communication with the steam generator, and an electrical generator in
- 25 communication with the steam turbine.

65. A power source and hydride reactor of claim 1 wherein the novel hydrogen species and compositions of matter comprising new forms of hydrogen comprises:

(a) at least one neutral, positive, or negative increased binding energy

5 hydrogen species having a binding energy

(i) greater than the binding energy of the corresponding ordinary

hydrogen species, or

(ii) greater than the binding energy of any hydrogen species for which

the corresponding ordinary hydrogen species is unstable or is not observed because

10 the ordinary hydrogen species' binding energy is less than thermal energies at

ambient conditions, or is negative; and

(b) at least one other element.

66. A power source and hydride reactor of claim 66 wherein the compound is

15 characterized in that the increased binding energy hydrogen species is selected from

the group consisting of  $H_n$ ,  $H_n^-$ , and  $H_n^+$  where n is a positive integer, with the

proviso that n is greater than 1 when H has a positive charge.

67. A power source and hydride reactor of claim 66 wherein the compound is

20 characterized in that the increased binding energy hydrogen species is selected from

the group consisting of (a) hydride ion having a binding energy that is greater than

the binding of ordinary hydride ion (about 0.8 eV) for  $p = 2$  up to 23 in which the

binding energy is represented by

$$\text{Binding Energy} = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi \mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right)$$

where  $p$  is an integer greater than one,  $s = 1/2$ ,  $\pi$  is pi,  $\hbar$  is Planck's constant bar,  $\mu_0$  is the permeability of vacuum,  $m_e$  is the mass of the electron,  $\mu_e$  is the reduced

- 5 electron mass given by  $\mu_e = \frac{m_e m_p}{\frac{m_e}{\sqrt{\frac{3}{4}}} + m_p}$  where  $m_p$  is the mass of the proton,  $a_H$  is the

radius of the hydrogen atom,  $a_0$  is the Bohr radius, and  $e$  is the elementary charge;

(b) hydrogen atom having a binding energy greater than about 13.6 eV; (c) hydrogen molecule having a first binding energy greater than about 15.3 eV; and (d) molecular hydrogen ion having a binding energy greater than about 16.3 eV.

10

68. A power source and hydride reactor of claim 67 wherein the compound is characterized in that the increased binding energy hydrogen species is a hydride ion having a binding energy of about 3, 6.6, 11.2, 16.7, 22.8, 29.3, 36.1, 42.8, 49.4, 55.5, 61.0, 65.6, 69.2, 71.6, 72.4, 71.6, 68.8, 64.0, 56.8, 47.1, 34.7, 19.3, and 0.69

15 eV.

69. A power source and hydride reactor of claim 68 wherein the compound is characterized in that the increased binding energy hydrogen species is a hydride ion having the binding energy:

$$\text{Binding Energy} = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi \mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right)$$

where  $p$  is an integer greater than one,  $s = 1/2$ ,  $\pi$  is pi,  $\hbar$  is Planck's constant bar,

$\mu_o$  is the permeability of vacuum,  $m_e$  is the mass of the electron,  $\mu_e$  is the reduced

electron mass given by  $\mu_e = \frac{m_e m_p}{\frac{m_e}{\sqrt{\frac{3}{4}}} + m_p}$  where  $m_p$  is the mass of the proton,  $a_H$  is the

5 radius of the hydrogen atom,  $a_o$  is the Bohr radius, and  $e$  is the elementary charge.

70. A power source and hydride reactor of claim 69 wherein the compound is characterized in that the increased binding energy hydrogen species is selected from the group consisting of

10 (a) a hydrogen atom having a binding energy of about  $\frac{13.6 \text{ eV}}{\left(\frac{1}{p}\right)^2}$  where  $p$  is an

integer,

(b) an increased binding energy hydride ion ( $H^-$ ) having a binding energy of

$$\text{about Binding Energy} = \frac{\hbar^2 \sqrt{s(s+1)}}{8\mu_e a_0^2 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^2} - \frac{\pi \mu_0 e^2 \hbar^2}{m_e^2} \left( \frac{1}{a_H^3} + \frac{2^2}{a_0^3 \left[ \frac{1 + \sqrt{s(s+1)}}{p} \right]^3} \right)$$

where  $p$  is an integer greater than one,  $s = 1/2$ ,  $\pi$  is pi,  $\hbar$  is Planck's constant bar,

15  $\mu_o$  is the permeability of vacuum,  $m_e$  is the mass of the electron,  $\mu_e$  is the reduced

electron mass given by  $\mu_e = \frac{m_e m_p}{\frac{m_e}{\sqrt{\frac{3}{4}}} + m_p}$  where  $m_p$  is the mass of the proton,  $a_H$  is the

radius of the hydrogen atom,  $a_o$  is the Bohr radius, and  $e$  is the elementary charge;

(c) an increased binding energy hydrogen species  $H_4^+(1/p)$ ;

(d) an increased binding energy hydrogen species trihydrino molecular ion,

5  $H_3^+(1/p)$ , having a binding energy of about  $\frac{22.6}{\left(\frac{1}{p}\right)^2} eV$  where  $p$  is an integer,

(e) an increased binding energy hydrogen molecule having a binding energy

of about  $\frac{15.3}{\left(\frac{1}{p}\right)^2} eV$ ; and

(f) an increased binding energy hydrogen molecular ion with a binding energy

of about  $\frac{16.3}{\left(\frac{1}{p}\right)^2} eV$ .

10

71. A power source and hydride reactor of claim 1 wherein the catalyst comprises a chemical or physical process that provides a net enthalpy of  $m \cdot 27.2 \pm 0.5 eV$  where  $m$  is an integer or  $m/2 \cdot 27.2 \pm 0.5 eV$  where  $m$  is an integer greater than one.

15

72. A power source and hydride reactor of claim 1 wherein the catalytic system is provided by the ionization of  $t$  electrons from a participating species such as an atom, an ion, a molecule, and an ionic or molecular compound to a continuum energy level such that the sum of the ionization energies of the  $t$  electrons is

approximately  $m \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer or  $m/2 \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer greater than one and  $t$  is an integer.

73. A power source and hydride reactor of claim 1 wherein the catalyst is
- 5 provided by the transfer of  $t$  electrons between participating ions;
- the transfer of  $t$  electrons from one ion to another ion provides a net enthalpy of reaction whereby the sum of the ionization energy of the electron donating ion minus the ionization energy of the electron accepting ion equals approximately  $m \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer or  $m/2 \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer
- 10 greater than one and  $t$  is an integer.

74. A power source and hydride reactor of claims 71, 72, and 73 wherein  $m$  is an integer less than 400.

- 15 75. A power source and hydride reactor of claim 74 wherein catalyst is selected from the group of Li, Be, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Kr, Rb, Sr, Nb, Mo, Pd, Sn, Te, Cs, Ce, Pr, Sm, Gd, Dy, Pb, Pt,  $2K^+$ ,  $He^+$ ,  $Na^+$ ,  $Rb^+$ ,  $Sr^+$ ,  $Fe^{3+}$ ,  $Mo^{2+}$ ,  $Mo^{4+}$ , and  $In^{3+}$ ,  $Ar^+$ ,  $Xe^+$ ,  $Ar^{2+}$  and  $H^+$ , and  $Ne^+$  and  $H^+$ .

- 20 76. A power source and hydride reactor of claim 75 wherein the catalyst of atomic hydrogen capable of providing a net enthalpy of  $m \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer or  $m/2 \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer greater than one and capable of forming a hydrogen atom having a binding energy of about  $\frac{13.6 \text{ eV}}{\left(\frac{1}{p}\right)^2}$  where  $p$  is an

integer wherein the net enthalpy is provided by the breaking of a molecular bond of

the catalyst and the ionization of  $t$  electrons from an atom of the broken molecule each to a continuum energy level such that the sum of the bond energy and the ionization energies of the  $t$  electrons is approximately  $m \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer or  $m/2 \cdot 27.2 \pm 0.5 \text{ eV}$  where  $m$  is an integer greater than one.

5

77. A power source and hydride reactor of claim 76 wherein the catalyst comprises at least one of AlH, BiH, ClH, CoH, GeH, InH, NaH, RuH, SbH, SeH, SiH, SnH,  $C_2$ ,  $N_2$ ,  $O_2$ ,  $CO_2$ ,  $NO_2$ , and  $NO_3$ .

10 78. A power source and hydride reactor of claim 77 wherein the catalyst comprises a molecule in combination with an ion or atom catalyst.

79. A power source and hydride reactor of claim 78 wherein the catalyst combination comprises at least one molecule selected from the group of AlH, BiH, ClH, CoH, GeH, InH, NaH, RuH, SbH, SeH, SiH, SnH,  $C_2$ ,  $N_2$ ,  $O_2$ ,  $CO_2$ ,  $NO_2$ , and  $NO_3$  in combination with at least one atom or ion selected from the group of Li, Be, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Kr, Rb, Sr, Nb, Mo, Pd, Sn, Te, Cs, Ce, Pr, Sm, Gd, Dy, Pb, Pt, Kr,  $2K^+$ ,  $He^+$ ,  $Na^+$ ,  $Rb^+$ ,  $Sr^+$ ,  $Fe^{3+}$ ,  $Mo^{2+}$ ,  $Mo^{4+}$ ,  $In^{3+}$ ,  $He^+$ ,  $Ar^+$ ,  $Xe^+$ ,  $Ar^{2+}$  and  $H^+$ , and  $Ne^+$  and  $H^+$ .

20

80. A power source and hydride reactor of claim 79 wherein the catalyst comprises two hydrogen atoms which absorbs at least one of  $27.21 \text{ eV}$  and  $54.4 \text{ eV}$  and is ionized to  $2H^+$ , to catalyze the transition of atomic hydrogen from the ( $p$ ) energy level to at least one of the ( $p+1$ ) and ( $p+2$ ) energy levels given by at least

25 one of

$$27.21 \text{ eV} + 2H\left[\frac{a_H}{1}\right] + H\left[\frac{a_H}{p}\right] \rightarrow 2H^+ + 2e^- + H\left[\frac{a_H}{(p+1)}\right] + [(p+1)^2 - p^2] \cdot 13.6 \text{ eV}$$

$$2H^+ + 2e^- \rightarrow 2H\left[\frac{a_H}{1}\right] + 27.21 \text{ eV}$$

wherein, the overall reaction is

$$5 \quad H\left[\frac{a_H}{p}\right] \rightarrow H\left[\frac{a_H}{(p+1)}\right] + [(p+1)^2 - p^2] \cdot 13.6 \text{ eV}$$

and

$$54.4 \text{ eV} + 2H[a_H] + H[a_H] \rightarrow 2H_{fast}^+ + 2e^- + H\left[\frac{a_H}{(3)}\right] + [(3)^2 - 1^2] \cdot 13.6 \text{ eV}$$

$$2H_{fast}^+ + 2e^- \rightarrow 2H[a_H] + 54.4 \text{ eV}$$

wherein, the overall reaction is

$$10 \quad H[a_H] \rightarrow H\left[\frac{a_H}{(3)}\right] + [(3)^2 - 1^2] \cdot 13.6 \text{ eV}$$

81. A power source and hydride reactor of claim 80 wherein the catalyst comprises hydrinos in a catalytic disproportionation reaction wherein lower-energy hydrogen atoms, hydrinos, act as catalysts because each of the metastable  
15 excitation, resonance excitation, and ionization energy of a hydrino atom is  $m \cdot 27.2 \text{ eV}$ .

82. A method of producing power comprising:  
providing a reaction vessel constructed and arranged to contain a pressure in  
20 the range of lower, equal to, or greater than atmospheric pressure;

maintaining a pressure in the range of lower, equal to, or greater than atmospheric pressure;

providing hydrogen atoms in the reaction vessel from a first source of hydrogen atoms in communication with the reaction vessel;

5 providing a source of atomic hydrogen catalyst in communication with the reaction vessel comprising a reaction mixture of at least one reactant comprising the element or elements that form the catalyst and at least one other element, whereby the catalyst is formed from the source; and

heating the reaction mixture producing atomic catalyst from the source of atomic catalyst if the catalyst is not already present or the reaction to form the catalyst is not spontaneous at ambient temperature;

10

heating the reaction mixture to initiate the catalysis of atomic hydrogen in the reaction vessel if the reaction is not spontaneous at ambient temperature, whereby the catalysis of atomic hydrogen releases energy in an amount greater than about 300 kJ per mole of hydrogen.

15

83. The method according to claim 82, wherein the catalyst is atomic Li.

84. The method according to claim 83, further comprising reacting LiH, LiNH<sub>2</sub>, and Pd on Al<sub>2</sub>O<sub>3</sub> powder to form atomic Li catalyst and atomic hydrogen in the reaction vessel.

20

85. The method according to claim 84, further comprising the addition of H<sub>2</sub> to regenerate LiH and LiNH<sub>2</sub>.

86. The method according to claim 82, further comprising reacting Li,  $\text{Li}_3\text{N}$ , and hydrided Pd on  $\text{Al}_2\text{O}_3$  powder and optionally  $\text{H}_2$  gas to form atomic Li catalyst and atomic hydrogen in the reaction vessel.

- 5 87. The method according to claim 86, further comprising the removal of  $\text{H}_2$  to regenerate Li and  $\text{Li}_3\text{N}$  followed by hydriding the dissicator or reintroducing  $\text{H}_2$ .

88. A method of producing power comprising:

- providing a reaction vessel constructed and arranged to contain a pressure in  
10 the range of lower, equal to, or greater than atmospheric pressure;

maintaining a pressure in the range of lower, equal to, or greater than  
atmospheric pressure;

- providing a source of molecular hydrogen catalyst in communication with the  
reaction vessel comprising a reaction mixture of at least one reactant comprising the  
15 element or elements that form the catalyst and at least one other element, whereby  
the catalyst is formed from the source; and

heating the reaction mixture producing molecular catalyst from the source of  
molecular catalyst if the catalyst is not already present or the reaction to form the  
catalyst is not spontaneous at ambient temperature;

- 20 heating the reaction mixture to initiate the catalysis of atomic hydrogen in the  
reaction vessel if the reaction is not spontaneous at ambient temperature, whereby  
the catalysis of atomic hydrogen releases energy in an amount greater than about  
300 kJ per mole of hydrogen.

89. The method according to claim 88, further comprising providing hydrogen atoms in the reaction vessel from a first source of hydrogen atoms in communication with the reaction vessel.

5 90. The method according to claim 88, wherein the catalyst is molecular NaH.

91. The method according to claim 90, wherein the source of molecular NaH comprises Na metal and a source of hydrogen.

10 92. The method according to claim 90 wherein the reaction mixture comprises NaH and Pd on Al<sub>2</sub>O<sub>3</sub> powder.

93. The method according to claim 92, further comprising the addition of H<sub>2</sub> to regenerate NaH.

15

94. The method according to claim 90 wherein the reaction mixture comprises Na and R-Ni comprising about 0.5 wt% NaOH wherein Na serves as the reductant.

95. The method according to claim 90 wherein the reaction mixture comprises R-  
20 Ni comprising about 0.5 wt% NaOH wherein intermetallic Al serves as the reductant.

96. The method according to claims 94 and 95 wherein reaction mixture is regenerated by adding NaOH and NaH wherein NaH may serve as an H source and a reductant.

25

97. The method according to claim 90 wherein the reaction mixture comprises NaH, a lanthanide metal, and Pd on Al<sub>2</sub>O<sub>3</sub> powder.

98. The method according to claim 97, wherein the reaction mixture is  
5 regenerated by adding H<sub>2</sub>, separating NaH and lanthanide hydride by sieving, heating lanthanide hydride to form lanthanide metal, and mixing lanthanide metal and NaH.

99. The method according to claim 97, wherein the reaction mixture is  
10 regenerated by the steps of separating Na and lanthanide hydride by melting Na and removing the liquid, heating lanthanide hydride to form lanthanide metal, hydriding Na to NaH, and mixing lanthanide metal and NaH.

100. The method according to claim 90 wherein the reaction mixture comprises  
15 NaH, NaNH<sub>2</sub>, and Pd on Al<sub>2</sub>O<sub>3</sub> powder.

101. The method according to claim 100, further comprising the addition of H<sub>2</sub> to regenerate NaH and NaNH<sub>2</sub>.

20 102. The method according to claim 90, further comprising reacting NaOH with a reductant to form the molecular NaH in the reaction vessel.

103. The method according to claim 90, further comprising reacting at least one of;  
(1) a source of sodium;  
25 (2) a support material;

- (3) a source of hydrogen;
- (4) a displacing agent, and
- (5) a reductant or reducing agent to form molecular NaH.

5 104. The method according to claim 103, wherein the source of sodium comprises Na, NaH, NaNH<sub>2</sub>, NaOH, NaOH coated R-Ni, NaX (X is a halide), and NaX coated R-Ni.

10 105. The method according to claim 103, wherein the reductant or reducing agent comprises at least one of a metal such as an alkaline metal, alkaline earth metal, a lanthanide, a transition metal such as Ti, aluminum, B, a metal alloy such as AlHg, NaPb, NaAl, LiAl, and a source of a metal alone or in combination with reducing agent such as an alkaline earth halide, a transition metal halide, a lanthanide halide, and aluminum halide, metal hydrides such as LiBH<sub>4</sub>, NaBH<sub>4</sub>, LiAlH<sub>4</sub>, or NaAlH<sub>4</sub>, and  
15 an alkaline or alkaline earth metal and an oxidant such as AlX<sub>3</sub>, MgX<sub>2</sub>, LaX<sub>3</sub>, CeX<sub>3</sub>, and TiX<sub>n</sub> where X is a halide, preferably Br or I.

106. The method according to claim 103, wherein the source of hydrogen comprises H<sub>2</sub> gas and a dissociator and a hydride.

20

107. The method according to claim 103, wherein the displacing agent comprises alkali or alkaline earth metal.

108. The method according to claim 103, further comprising providing a source of NaH on a large surface area support that favors production of molecular NaH from the source, and reacting the source of NaH to form molecular NaH.
- 5 109. The method according to claims 103 and 108, wherein the support comprises at least one of R-Ni, Al, Sn, Al<sub>2</sub>O<sub>3</sub> such as gamma, beta, or alpha alumina, aluminates, sodium aluminate, alumina nanoparticles, porous Al<sub>2</sub>O<sub>3</sub>, Pt, Ru, or Pd/Al<sub>2</sub>O<sub>3</sub>, carbon, Pt or Pd/C, inorganic compounds such as Na<sub>2</sub>CO<sub>3</sub>, lanthanide oxides such as M<sub>2</sub>O<sub>3</sub> (preferably M= La, Sm, Dy, Pr, Tb, Gd, and Er), Si, silica,  
10 silicates, zeolites, Y zeolite powder, lanthanides, transition metals, metal alloys such as alkali and alkali earth alloys with Na, rare earth metals, SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> or SiO<sub>2</sub> supported Ni, and other supported metals such as at least one of alumina supported platinum, palladium, and ruthenium.
- 15 110. The method according to claim 89, wherein the source of hydrogen atoms comprises molecular hydrogen and the hydrogen atoms are formed from the molecular hydrogen using a dissociator.
111. The method according to claims 103 and 110, wherein the dissociator  
20 comprises at least one of Raney nickel (R-Ni), a precious or noble metal, and a precious or noble metal on a support where in the precious or noble metal may be Pt, Pd, Ru, Ir, and Rh, and the support may be at least one of Ti, Nb, Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and combinations thereof;
- Pt or Pd on carbon, a hydrogen spillover catalyst, nickel fiber mat, Pd sheet,  
25 Ti sponge, Pt or Pd electroplated on Ti or Ni sponge or mat, TiH, Pt black, and Pd

black, refractory metals such as molybdenum and tungsten, transition metals such as nickel and titanium, inner transition metals such as niobium and zirconium, and a refractory metal such as tungsten or molybdenum, and the dissociating material may be maintained at elevated temperature.

5

112. The method according to claim 90, further comprising forming  $\text{Na}^{2+}$  in the reaction vessel from the molecular  $\text{NaH}$ .

10

113. The method according to claims 82 and 88, further comprising removing reaction products from the vessel and regenerating the source of catalyst from at least a portion of the reaction products.

15

114. The method according to claims 82 and 88, further comprising converting the released energy to electrical energy.

20

115. The method of claims 82 and 88, whereby the source of hydrogen catalyst comprises at least one reactant having hydrogen and at least one other element, and  
the at least one reactant undergoes reaction such that the energy released is greater than the difference between the standard enthalpy of formation of compounds having the stoichiometry or elemental composition of the products and the energy of formation of the at least one reactant.

116. The method according to claims 82 and 88, whereby the source of hydrogen catalyst comprises at least one reactant having hydrogen and at least one other element, and

the at least one reactant undergoes reaction such that the energy released is  
5 greater than the theoretical standard enthalpy required to regenerate the at least one reactant from the products wherein the energy to replace any reacted hydrogen is the standard value.

117. A method for generating power comprising providing hydrogen and at least  
10 one other element that undergo reaction such that the energy released is greater than the difference between the standard enthalpy of formation of compounds having the stoichiometry or elemental composition of the products and the energy of formation of the reactants.

118. A method for generating power comprising providing hydrogen and at least  
15 one other element that undergo reaction such that the energy released is greater than the theoretical standard enthalpy required to regenerate the reactants from the products wherein the energy to replace any reacted hydrogen is the standard value for the combustion of the hydrogen.

20

119. The method of claims 82 and 88 further comprising the preparation or regeneration of the reaction mixture wherein preparation or regeneration is achieved by at least one of steps of mechanical mixing or separation, melting, filtration, hydriding, dehydriding, decomposition, vapor deposition, evaporation, vaporization,  
25 and sublimation, and ball milling.