

## Gas-based hydride applications: recent progress and future needs

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Received 29 July 2002; accepted 15 November 2002

### Abstract

Hydride applications can be cataloged under five main categories: H<sub>2</sub> storage, thermal compression (including closed thermodynamic systems), gas separation, electrochemical and processing and miscellaneous. Recent commercial and R&D hydride applications activities are reviewed herein (excluding the NiMH battery). Growing interest in fuel cell power has stimulated interest in hydride storage of H<sub>2</sub>, with good near-term commercial potential in the subcategories of stationary, small portable and on-board vehicular storage. The competition for vehicular hydride storage is high pressure compressed H<sub>2</sub> held in lightweight composite cylinders. If hydrides are to compete, we must develop radical new concepts to substantially increase the gravimetric capacity at low desorption temperatures.

Published by Elsevier B.V.

*Keywords:* Hydrogen storage materials; Gas–solid reactions; Metal hydride applications

### 1. Professor Moshe Ron (1925–2001)

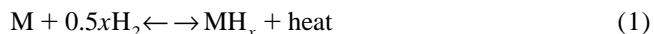
This paper is dedicated to the memory of Moshe Ron, an early pioneer in the properties and applications of reversible metal hydrides. One of his principal hydride applications interests was the hydride heat pump [1–3]. This spawned extensive work in the property areas of hydride heat transfer [4–6] and kinetics [7–9]. He led the development of ‘porous, metallic–matrix hydrides (PMH)’, whereby inert metallic matrices were used to simultaneously enhance the mechanical stability and heat conductivity of hydrides [4–6]. One of Professor Ron’s last publications gave us an extensive ‘normalized pressure dependence method’, a model for quantifying and generalizing hydride kinetics [9]. Extensive references to the work of him and his many students and coworkers can be found therein. The end to Moshe Ron’s direct contributions to the field of metal hydrides is to be regretted by the M–H community, but indirectly he will influence us for years to come.

### 2. Introduction

The ultimate utility of all our work on hydrogen–metal systems, be it fundamental or engineering, is to aid the

development of practical applications. The history of hydride applications R&D is long and rich. It is not our intent in this brief paper to review the entire history of hydride applications. Previous applications reviews have set that stage [10–13]. We shall instead focus on progress during the last few years. In one important area, vehicular H<sub>2</sub> storage, applications progress has somewhat stalled. We shall examine the competition and redefine the targets necessary to capture that market.

The two most important factors in the applications of metal hydrides are the easily reversed gas–solid chemical reaction:



and the well known van ‘t Hoff relation relating plateau pressure  $P$  to temperature  $T$ :

$$\ln P = \frac{\Delta H}{RT} - \frac{\Delta S}{R} \quad (2)$$

where  $\Delta H$  and  $\Delta S$  are the enthalpy and entropy changes of the hydriding reaction, respectively,  $T$  is absolute temperature and  $R$  is the gas constant. Of course, there are numerous other important hydride properties: plateau slope and hysteresis, H-capacity, activation, volume change, decrepitation, reaction kinetics, thermal conductivity, gaseous impurity resistance, cyclic stability, safety and cost [10,14].

Many applications for reversible metal hydrides have

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Table 1  
List of hydride applications

H <sub>2</sub> storage
Stationary
Portable
Vehicular
Thermal compression
H <sub>2</sub> compression
Closed thermodynamic systems
Heat storage
Heat pump
Cryocooler
Temperature sensor
Gas gap heat switch
Heat engine
Actuator
Gas separation
Isotope separation
H <sub>2</sub> gas concentration
Separation
Purification
Gettering
Electrochemical and processing
Catalyst
Batteries
NiMH
Segmented
Other
H <sub>2</sub> detector
Switchable mirror
Electrical devices
Miscellaneous

been proposed. A five-category list of the principal applications is shown in Table 1. Some of the applications listed have reached substantial commercial reality (e.g. NiMH batteries) and many have not. We shall briefly review recent activity on most of the hydride applications listed in Table 1. Because H<sub>2</sub> storage (especially for fuel cell vehicles) offers a large future potential (but with notable difficulties), we leave that area to last. We shall not cover the large electrochemical NiMH battery field, but will briefly mention the gas-phase ‘segmented’ hydride battery. In general, we shall not discuss the chemical hydrides that are hydrolyzed to generate H<sub>2</sub> gas and for which cyclic reversibility is a difficult task, e.g. the NaBH<sub>4</sub> solutions [15,16].

### 3. Non-storage hydride applications

#### 3.1. Thermal compression

This broad category ranges from the open-ended compression of H<sub>2</sub> gas to closed-cycle thermodynamic systems that utilize a hydride compressor as the driver. In all cases, H<sub>2</sub> is effectively compressed via the van ‘t Hoff equation

(Eq. (2)) utilizing a heat input that is direct or through  $I^2R$  electrical means.

##### 3.1.1. H<sub>2</sub> compression

Hydride-based hydrogen compressors have been an application of commerce more than 20 years. They are manufactured and sold mainly by Ergenics, Inc. in single- or multistage-designs [17]. In recent years, a number of new designs have been introduced, including a single stage, electrically-driven unit that is capable of output pressures greater than 400 bar (40 MPa), levels that are particularly useful for filling the high pressure gaseous H<sub>2</sub> tanks commonly being used in prototype fuel cell vehicles [18].

##### 3.1.2. Closed thermodynamic systems

Although numerous prototype systems have been build over the years, most of the closed hydride systems are not yet commercial reality. There are notable exceptions to that, however. As far as we know, the hydride-based aircraft fire detector (a temperature sensor) [19] is still being manufactured and widely used after a quarter century.

Large-scale heat storage remains of limited interest because of the relatively high cost of hydriding alloys, but a design based on Ni-doped MgH<sub>2</sub> and AB<sub>2</sub> alloys offers hope for high-grade solar heat storage and steam generation [20]. There are also special small-scale heat storage possibilities that may tolerate moderately high hydride cost. One is the hydride cold start heater which is designed to store the >350 °C waste heat of an automotive exhaust for later use to rapidly (5 s) heat the catalytic converter, thus quickly reducing the undesirable startup emissions [18,21].

Heat pumps (both temperature upgrading and refrigerator types) remain in positions of relatively strong research and prototype development, although it is not clear that they are approaching commercial viability. Good prototype demonstrations have been recently made in Germany [22,23], Korea [24,25], Finland (with two-stage compressor) [26], Russia [27] and the USA [18]. A conceptual design of an oscillating bed heat pump has been patented [28]. Ergenics, Inc. demonstrated a benchtop automobile air conditioner design capable of 9,000–18,000 BTU/h (9,500–19,000 kJ/h) cooling capability at 7 °C [18]. The unit weighed about 30 kg, including 14 kg of AB<sub>5</sub> hydride. In addition to these experimental demonstrations, there have been some theoretical heat pump models published recently [25,29,30].

Work on hydride heat engines and actuators, the direct conversion of heat to mechanical energy has been relatively inactive in recent years. A solar powered water pump was recently demonstrated in Russia [31]. Automatically resetting fire sprinklers have been developed and offer an interesting commercial possibility [18,32].

The concept of using a La(Ni,Sn)<sub>5</sub>-based thermal H<sub>2</sub>

compressor and Joule–Thomson expansion for aerospace cryocooling is being actively worked on by one of us [33,34]. Included in the cryocooler system are gas gap heat switches, spaces which can be reversibly switched from vacuum insulation to thermally conducting  $H_2$  by using an electrically heated hydride-based  $H_2$  dispenser (e.g.  $ZrNiH_{1.5}$ , a relatively stable hydride) [35]. The concept of hydride-based gas gap heat switches has also been developed for controlled heat insulation/conducting glass windows for buildings [36].

### 3.2. Gas separation

#### 3.2.1. Isotope separation

As summarized in an earlier (1995) review [11], the major industrial use of hydrides for hydrogen isotope separation is in the Savannah River (USA) tritium plant where a Pd-based system is used. There have been rather limited isotope separation activities (using other hydrides) in more recent years, notably in Japan [37,38] and China [39]. A predictive, non-isothermal model for the separation of hydrogen isotopes using a Pd column was published [40].

#### 3.2.2. $H_2$ gas concentration

With the exception of removing  $H_2$  from ammonia purge gas in a small plant in China [41], separation does not seem to have developed into a commercial process. But interest remains. Modeling of the separation process has been performed in Russia [42,43] and the USA [44]. A new process for the separation of  $H_2$  from mixed gas using a hydride sol–gel composite has been developed at Savannah River [45].

The purification of  $H_2$  by metal hydrides is becoming more important in the growing new era of low-temperature PEM (proton exchange membrane) fuel cells. PEM fuel cells do not tolerate impurities like CO very well and hydrides supply exceptionally pure  $H_2$  ideal for PEM fuel cell use [46,47]. For this purpose, Ergenics is designing a combined purifier–compressor capable of handling quite impure  $H_2$  [48]. This is made possible by special bed designs that locally filter the impurities [49].

$H_2$  gettering is a long-established commercial process that is used widely in vacuum technology and inert gas purification [50]. It is beyond the scope of this brief review.

### 3.3. Electrochemical and processing

#### 3.3.1. Catalysts

There has been some renewed interest in using hydrides for catalytic purposes, for example, hydrogenation/dehydrogenation of organics [51,52], methanation [53] or the destruction of hazardous organic wastes [54].

#### 3.3.2. Batteries

The ubiquitous NiMH battery (MH anode) represents the most widespread commercial application of reversible hydrides. They are used for high power applications and hybrid gasoline–electric vehicles, but they are being more and more displaced by lithium-ion batteries for low power applications such as cell phones or laptop computers.

A new kind of bipolar battery has been developed by Ergenics that does not use the hydride as an electrode but rather as a gas charged and discharged component [18,55]. In essence, it is a derivative of the nickel–hydrogen battery that is used for aerospace and military applications, but incorporating a ‘segmented’ MH to maintain low  $H_2$  pressure in the charged state. The problem of protecting the MH from  $H_2O$ -saturated  $H_2$  has been solved [49]. Because of its long cycle life and ability to be charged and discharged at extremely rapid rates, the bipolar ‘segmented’ battery has good potential for use in hybrid electric vehicles.

### 3.4. Miscellaneous other applications

Other interesting hydride applications have surfaced in the last few years. One is the switchable mirror whereby one makes use of the metal $\leftrightarrow$ insulator transition  $YH_2\leftrightarrow YH_3$  to change optical properties of a film from mirror to transparent, respectively [56]. This work, originated in The Netherlands, has spawned many interesting fundamental studies and potential practical optical applications. Similarly, the complex semiconducting hydride  $Mg_2NiH_4$  exhibits profound changes in optical properties with temperature and mechanical pressure, suggesting a number of possible electronic applications [57]. We note from the recent patent literature the application of hydrides to certain electrical devices, e.g. a spark plug for internal combustion engines [58] or an end-of-life arc suppression feature for fluorescent light bulbs [59].

## 4. Hydrogen/hydride storage

We have left hydrogen storage to last because it bears more focus because a growing perceived potential for new commercial applications, stationary, portable and vehicular. There have been a number of companies that have offered commercial hydride storage containers for many years: Ergenics, Inc. [17], Hydrogen Components, Inc. (FuelCellStore) [60], Gesellschaft für Electrometallurgie [61], Japan Metals and Chemicals [62], among others [11]. In the last year, two new major joint companies have formed in North America, specifically to present a bold position to cover the future hydride storage market. On the USA side, ChevronTexaco joined with ECD Ovonic to form Texaco Ovonic Hydrogen Systems, L.L.C. [63], establishing an alloy production, container manufacture and R&D facility in Rochester Hills, MI, USA. The

hydride technology for Texaco Ovonic comes from ECD Ovonic. On the Canadian–Dutch–German side, Hydro-Québec, Shell Hydrogen and Gesellschaft für Electrometallurgie formed HERA Hydrogen Storage Systems based in a new production and R&D facility in Longueuil, Canada [64]. This combines the low-temperature, inter-metallic hydride expertise of GfE with the nanocrystalline expertise of Hydro-Québec. In addition, a joint development agreement was recently executed between the US company Air Products and Chemicals, Inc. and Japan Metals and Chemicals [65].

#### 4.1. Stationary and portable storage containers

We would estimate that a few thousand small hydride storage units have been made over the last 25 years and used for a variety of stationary and portable purposes. Designs (at least the internal construction) are usually proprietary, but a wide variety of individual products are currently available [17,60–64]. What seems to be driving the near-future market is stationary and portable fuel cell power supplies for instantly available emergency backup power. For example, the Canadian company Hydrogenics now offers a 500 W fuel cell power generator that is supplied with H<sub>2</sub> from a chemical hydride [66]. In the USA, the Coleman Powermate Company reports that it is about to introduce a fuel cell power supply that is supplied by a rechargeable, low temperature hydride [67]. Most of the major fuel cell companies have interest in hydride supply of H<sub>2</sub> for portable and stationary power applications, e.g. United Technologies, Ballard, H-Power, Plug Power and others. There are numerous suitable low

Table 3

Volume and weight comparisons for state-of-the-art on-board vehicular storage of 3 kg H<sub>2</sub> [76]

Technology	System volume (l)	System weight (kg)	Weight % H <sub>2</sub>
5,000 p.s.i. (34.5 MPa) compressed H <sub>2</sub>	145	45	6.7
10,000 p.s.i. (69 MPa) compressed H <sub>2</sub>	100	50	6.0
Low-temperature metal hydride	55	215	1.4
Liquid H <sub>2</sub>	90	40	7.5

temperature hydrides available [13], so we feel no major new R&D is needed for this particular application.

#### 4.2. Vehicular storage

In the last few years, immense commercial interest has developed in fuel cell vehicles as an answer to poor internal combustion efficiency, pollution and fuel dependence. The need for lightweight, compact onboard H<sub>2</sub> fuel supply is acute. Storage is a major barrier in the minds of the automobile manufacturers. This has led to suggesting the concepts of onboard reforming of hydrocarbon fuels and chemical hydrides, both of which have debatable viability and economics. As shown in Table 2, some recent demonstration vehicles have used reversible metal hydrides [68–75]. But most have used high-pressure compressed gas. The reason for this can be seen from Table 3, which compares the properties of compressed, liquid and hydride storage of H<sub>2</sub> [76]. Compressed hydrogen (C-H<sub>2</sub>) offers

Table 2

Recent vehicles using hydride tanks [68–75]

Maker	Designation	Power <sup>a</sup>	Size (kW)	Hydride	Year
GM Opel	Precept FCEV	FC	75	?	2000
Honda	FCX-V1	FC	60	JMC <sup>e</sup>	1999
Mazda	Cappella	ICE	?	JMC	1994
Mazda	Demio	FC	50	?	1997
Toyota	RAV4 FCEV	FC	20	?	1996
Toyota	FCHV-3	FC	90	JMC	2001
John Deere <sup>b</sup>	Gator 1	FC hybrid	8.5	Mm(Ni,Al) <sub>5</sub>	1998
John Deere <sup>b</sup>	Gator 2	FC hybrid	8.5	Ti(Fe,Mn)	1998
SRTC Bus <sup>b</sup>	Augusta	ICE hybrid	75 <sup>i</sup>	Lm(Ni,Al) <sub>5</sub> <sup>f</sup>	1996
FCPI/SNL <sup>c</sup>	Mine Locomotive	FC	12	(Ti,Zr)(Mn,V,Cr,Fe) <sub>2</sub> <sup>d</sup>	2001
ECD <sup>g</sup>	Motor Scooter	ICE	?	ECD	2002
Germany	U212 Submarine	FC hybrid	300	GfE <sup>h</sup>	2004

<sup>a</sup> FC = Fuel cell; ICE = internal combustion engine.

<sup>b</sup> Savannah River Technology Center.

<sup>c</sup> Fuel Cell Propulsion Institute/Sandia National Laboratories.

<sup>d</sup> GfE C15, 1.8 wt.% H<sub>2</sub>.

<sup>e</sup> Japan Metals and Chemicals.

<sup>f</sup> Lm<sub>1.06</sub>Ni<sub>4.96</sub>Al<sub>0.04</sub>, where Lm = La-rich mischmetal.

<sup>g</sup> Energy Conversion Devices.

<sup>h</sup> GfE Gesellschaft für Electrometallurgie.

<sup>i</sup> ICE only (approximate).

excellent gravimetric H-capacity, more than 11 wt.% in advanced designs, far better than we in the hydride community can provide. Composite high pressure gas cylinders are simple and require no heat exchange for release of the contained H<sub>2</sub> for the vehicle use or recharging. But C-H<sub>2</sub> offers poor volumetric H<sub>2</sub> capacity compared to hydrides, not to mention the compression energy and safety disadvantages of C-H<sub>2</sub> at 350–700 bar (35–70 MPa) pressure. From the gas storage proponents' points of view, however, the concept of C-H<sub>2</sub> is safe and composite tanks have been officially certified for high pressure H<sub>2</sub> use [76].

In January 2002, the US Department of Energy announced the FreedomCAR initiative setting cost and performance targets for future fuel cell vehicles [77]. We are asked to provide a complete onboard storage system that provides 6 wt.% H<sub>2</sub>. When we consider the hydride container and heat exchange components, we probably need a hydride medium with 7.5+ wt.% H<sub>2</sub> capability. Ideally speaking, we need to have relatively low temperature thermodynamics such that we can use the <100 °C waste heat of the vehicle fuel cell to provide the endothermic heat of H<sub>2</sub> desorption without burning some of the desorbed H<sub>2</sub> for that purpose. This is far beyond the present state-of-the-art for all known reversible metal hydrides and provides a very strong challenge for us.

## 5. Conclusion

Hydride applications R&D is alive and well. The most important future application will be elemental H<sub>2</sub> storage for fuel cell vehicles. But we need to develop a new generation of hydrides with far greater gravimetric H-capacity than the present families of metallic (intermetallic compound and solid solution) hydrides, yet with near-ambient thermodynamic and kinetic properties. Recent work on the catalyzed Na-alanates may offer hope, especially if the concept can be extrapolated to the transition metal complexes and the borohydrides [78]. We need new 'breakthrough' concepts. Our task is very difficult, but the technical, environmental and economic rewards will be immense if we succeed.

## Acknowledgements

G.S. wishes to thank the US Department of Energy Hydrogen Program, Sandia National Laboratories and the International Energy Agency Hydrogen Implementing Agreement for support and encouragement. R.C.B. wishes to acknowledge that the Jet Propulsion Laboratory is operated by California Institute of Technology under a contract with the National Aeronautical and Space Administration. We wish to also express our gratitude to the following individuals and organizations for providing

information for this paper and especially for our oral presentation at MH2002: David DaCosta (Ergenics, Inc.), Marc Hubert (HERA Hydrogen storage Systems), Ted Motyka (Savannah River Technology Center), Bill Replogle (Sandia National Laboratories), Kenichi Sakamachi (JMC [USA], Inc.), Krishna Sapru (Energy Conversion Devices, Inc.) and Ned Stetson (Texaco Ovonic Hydrogen Systems, L.L.C.). If we were to look back over our long professional lives in hydride applications, there would be many others to thank, including Moshe Ron.

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