

METAL HYDRIDE STORAGE SYSTEMS: STATE OF THE ART ANALYSIS AND INTEGRATION IN FC-BASED ENERGY SYSTEMS

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Key words

Hydrogen storage; Metal hydride; Fuel Cell

Abstract

The technology of metal hydride hydrogen-storage systems has already reached a mature stage and several systems from different manufacturers around the world are now available for purchase. The main reasons for this development can be found in their intrinsic safety features, lower energy consumption and smaller size as compared to other more consolidated hydrogen storage technologies, such as hydrogen compression or liquefaction. However, metal hydride systems also present some drawbacks which limit their range of applications, above all their weight and poor kinetic performance.

Therefore, in this paper metal hydride based hydrogen-storage technology is reviewed in order to assess its performance, and several commercial systems are surveyed and compared to other well-established technologies.

Particular emphasis is given to the design constraints which must be taken into account when devising a proper layout for a FC-based energy system, with metal hydrides used to supply hydrogen to the FC. In order to achieve optimal efficiency, the thermal energy required by the storage system should be recovered from the system waste energy, and the most suitable accessories (batteries, supercapacitors) should be integrated in the system so as to extend its kinetic capabilities.

The authors have cooperated to the development of such an integrated system within two research programs (DEMAG, FEMAG) financed under the 6th EU Framework Programme. Details of the system layout implemented are given and discussed.

Nomenclature

c	Specific heat [$\text{J}\cdot\text{kmol}^{-1}\cdot\text{K}^{-1}$]	Greek symbols	
ΔH	Reaction enthalpy change [MJ/kmol]	β	Pressure ratio
m	Mass [kg]	ε	R/c_p ratio
\dot{m}	Mass flow rate [kg/s]		
MW	Molecular weight [kg/kmol]	Subscripts	
p	Pressure [bar]	a,d	Absorption, desorption
P	Power [kW]	eq	Equilibrium
\dot{Q}	Heat flux [W]	FC	Fuel cell
R	Universal gas constant [$\text{J}\cdot\text{kmol}^{-1}\cdot\text{K}^{-1}$]	MH	Metal hydride
ΔS	Reaction entropy change [$\text{MJ}\cdot\text{kmol}^{-1}\cdot\text{K}^{-1}$]	w	Wasted

1. Introduction

It is well known that one of the most powerful technology barriers to the widespread acceptance of hydrogen as energy vector is the storage problem [1,2]. Conventional ways to store hydrogen such as gas compression and liquefaction are bulky, expensive and raise important safety issues, and therefore several alternative ways to store hydrogen have been proposed.

Metal hydride hydrogen-storage systems represent a fairly good solution to some of the abovementioned problems, for they can store and release comparatively high amounts of hydrogen in a small volume at low pressure and at near room temperature: commercial tanks can in fact already be found on the market. The main drawback of these systems is their weight, which determines gravimetric densities of at most 1,5%, still very far from acceptable values especially for the transportation sector (for example, the U.S. DOE has set a target of 6 wt% for 2010, [2]). Anyway, it is unquestionable that metal hydride systems can be considered interesting as near- and medium-term solutions to the hydrogen storage problem, especially for their intrinsic safety features, at least for stationary and small portable applications, where they can successfully replace batteries as energy storage media.

The present work aims at surveying and discussing the strengths and limits of metal hydride hydrogen-storage units, the best options of integration between such units and fuel cells, and at describing possible applications of energy systems, including two research projects in which the authors are involved, where metal hydride tanks are used to store energy in a safe and compact way.

2. Metal hydride thermodynamic properties

Metal hydrides suitable for hydrogen storage are usually ternary systems, described by the chemical formula AB_xH_m , where element A belongs to the rare earth or alkali metal classes, and element B is a transition metal which, by itself, would give rise to unstable hydrides. The x ratio between metal B and metal A number of atoms present in the crystalline structure can be 0,5, 1, 2, or 5, depending on the metal elements, as in the following examples: Mg_2NiH_4 , $TiFeH_2$, $ZrV_2H_{5,5}$ and $LaNi_5H_6$ [3-5]. The hydriding process is described by the following equilibrium reaction:



The hydriding reaction is exothermic and therefore denoted by a negative enthalpy change ΔH , which, along with the hydriding entropy change ΔS , determine the reaction equilibrium pressure at a given temperature by means of the Van't Hoff law:

$$\ln \frac{p_{eq}}{p_0} = \frac{\Delta H}{RT} - \frac{\Delta S}{R} \quad (2)$$

The entropy change in the hydriding process is essentially due to the hydrogen molecule break-up into a pair of hydrogen atoms, and therefore it is usually very close to its standard entropy, i.e. $-130,7 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$, for all metal hydrides [3]. Thus, the stability of the hydride and its equilibrium pressure range at temperatures near to the ambient temperature are determined by the enthalpy change ΔH , which is much more variable among different metal hydrides.

Physical and thermodynamic properties of a wide range of metal hydrides are reported in the online database <http://hydpark.ca.sandia.gov/>, set up within the framework of the International Energy Agency Hydrogen Implementing Agreement (IEA HIA Task 12 – Metal Hydrides and Carbon for Hydrogen Storage – and Task 17 – Solid and Liquid State Hydrogen Storage Materials).

Figure 1 shows the Van't Hoff plots for several metal hydrides. It is possible to observe that, depending on their reaction enthalpy change, metal hydrides can be roughly divided in high-temperature alloys, when hydrogen release is possible at atmospheric pressure only with temperatures

higher than 100 °C, and low-temperature hydrides, which can instead release hydrogen at atmospheric pressure at temperatures lower than 100 °C. Mg-based alloys are an example of high-temperature hydrides, whereas LaNi₅ is a typical low-temperature hydride commonly used in storage applications.

In fact, high-temperature hydrides usually have larger storage capacity, but their application is hindered by the considerable high-temperature heat flux required, while low-temperature hydrides can even be used without any heat supply, the heat flux exchanged with the environment being enough, when the hydrogen flow rate required is sufficiently low. When the application needs higher flow rates, the storage system can be designed in such a way that the heat flux necessary for the hydrogen desorption can be recovered from fuel cell waste heat.

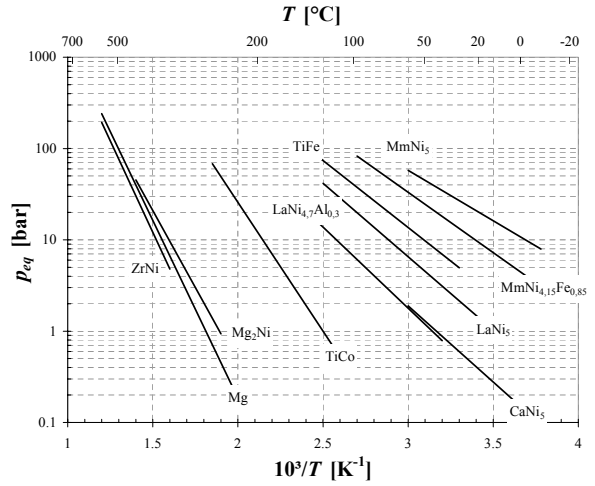


Figure 1 – Van't Hoff plots for several metal hydrides

3. Integration of metal hydride storage tanks in FC-based energy systems

In the preceding section the feasibility of the integration between metal hydride tanks and fuel cells has been briefly mentioned. This is based upon the fact that the storage tank operation involves a heat flux which, at least in the case of low-temperature hydrides, can be recovered from the waste heat dispersed by the fuel cell as a result of thermodynamic losses.

The heat flux required by the storage tanks in order to release hydrogen is, at steady state conditions:

$$\dot{Q}_{MH} = \dot{m} \Delta H \quad (3)$$

where \dot{Q}_{MH} is the heat flux, \dot{m} the hydrogen flow rate and ΔH the reaction enthalpy change of the hydriding process. On the other hand, the power lost at steady state conditions by the fuel cell as rejected heat is:

$$\dot{Q}_{wFC} = (1 - \eta) \dot{m} LHV \quad (4)$$

where η is the fuel cell efficiency based on hydrogen's lower heating value LHV (-241,8 kJ/mol).

The ratio between these two heat fluxes is therefore:

$$\frac{\dot{Q}_{MH}}{\dot{Q}_{wFC}} = \frac{1}{1 - \eta} \frac{\Delta H}{LHV} (\cong 25\% \text{ for LaNi}_5) \quad (5)$$

If we consider a 50% efficiency, and the common LaNi₅ as the storage medium ($\Delta H = -30,8$ kJ/mol), then the heat flux needed by the hydrogen release process is just approximately 25% of the total power lost by the fuel cell. Furthermore, LaNi₅ releases hydrogen at approximately 2 bar if its temperature is 25 °C, which is certainly lower than the fuel cell cooling fluid temperature, at least when there is a considerable power output required.

Similar remarks apply for alloys other than LaNi₅, so that it is possible to conclude that for low-temperature metal hydrides the hydrogen release process usually takes place at pressure and temperature conditions that are compatible with the heat sources available from the fuel cell, although it may be possible that at very low power output the heat discharged by the fuel cell is lower than that

required by the metal hydride tank [6]; in this case, the tank cools down until the temperature difference between the tank itself and the hot stream is such that sufficient heat flux takes place.

Figure 2 shows how a fuel cell can be coupled with a metal hydride storage system when the system performance is critical: the fuel cell cooling fluid is collected at the FC outlet and sent to a heat exchanger, where it releases heat to the metal hydride heat transfer fluid, which is then sent to the storage tank where it releases the heat necessary to sustain the dehydrating reaction. The storage tank itself is in this case designed as a heat exchanger between the fluid and the metal hydride bed.

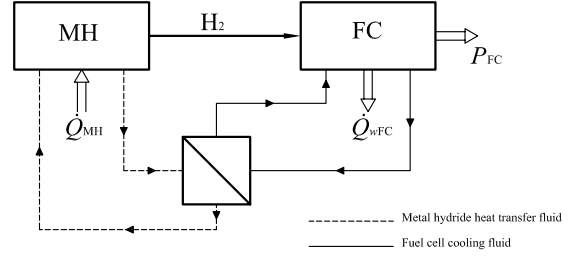


Figure 2 – Schematic of a fuel cell system coupled with a metal hydride hydrogen storage tank

This scheme allows to obtain good kinetic performance by designing the heat exchanger and the metal hydride tank in order to achieve the maximum heat exchange efficiencies. If the kinetic performance is not a significant issue for a specific application, then the integration between fuel cell and metal hydride tank may be simpler: for small applications it is even possible to place the storage tank just directly into the FC-cooling air stream without any need of heat exchangers. Details on the main factors influencing the hydrogen absorption and desorption may be found in [7-9].

4. Comparison among current commercial hydrogen storage technologies

In order to assess metal hydride storage systems potentiality and limits, in this section a comparison with more mature technologies, such as hydrogen liquefaction and compression, is carried out, taking into consideration only those systems that have already reached a commercial development stage. Therefore, technologies involving hydrogen storage in carbon nanotubes or glass microspheres are not included in this assessment.

The comparison is based on the following features:

- energy density (chemical energy stored per unit volume, kWh/l);
- gravimetric density (ratio of mass of stored hydrogen to the total tank mass $\text{kg}_{\text{H}_2}/\text{kg}$);
- energy required for the hydrogen storage, as a percentage of the hydrogen LHV;
- cost of the storage system.

The data concerning metal hydride storage tanks were collected from the leading manufacturers and suppliers, such as Treibacher Industrie AG [10], Japan Steel Works ltd. [11], Ergenics – HERA [12], Ovonic Hydrogen Systems LLC [13], Labtech Int. Ltd. [14], Tianjin Highland Energy Technology Development Co., Ltd. [15] and udomi [16].

Sources for liquefied and compressed hydrogen technologies were both a report from IEA [17] on hydrogen production and storage, and a survey of several manufacturers and suppliers.

The energy required for compressing hydrogen is evaluated with reference to a two-step interrefrigerated adiabatic compression, with polytropic efficiency $\eta_y = 0,85$, carried out at optimal conditions ($\beta_1 = \beta_2 = \sqrt{\beta}$; ambient temperature as starting point for both compression phases). Under these assumptions the mechanical energy required is:

$$W = 2 c_p T_1 \left(\beta^{\frac{\varepsilon}{2\eta_y}} - 1 \right) \quad (6)$$

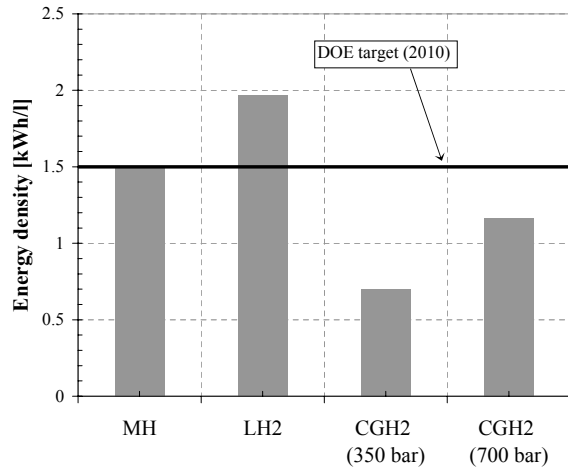
where $c_p = 14,5 \text{ kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$, $T_1 = 25 \text{ }^\circ\text{C}$, and $\varepsilon = R/(MW c_p) = 0,2844$.

In the case of liquefied hydrogen, most sources indicate an energy requirement of 30% based on hydrogen LHV [17], but some authors suggest that by adopting optimized liquefaction processes, even with current technologies this figure could be lowered down to 15% [18].

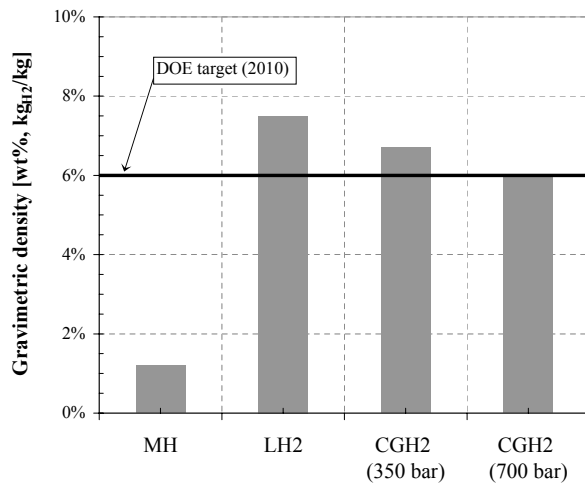
Figure 3 gives a graphical illustration of the comparison among the abovementioned storage technologies. It is immediately apparent that the main assets of metal hydride systems are their high energy density (Figure 3a), which translates into small footprint, and low energy requirement (Figure 3c): in fact, a charging pressure of 25 bar is generally suitable for low-temperature hydrides and guarantees adequate charging times. The mechanical work needed, calculated by means of equation (6), is 5,1% of hydrogen's LHV. In Figure 3c the heat flux that must be drained from the tank has been neglected, because it is usually possible to cool the tank by means of ambient-temperature water, and therefore the heat flux involved does not represent a significant economical or technical challenge and it is indeed negligible from an exergetic point of view.

On the other hand, the low gravimetric density (Figure 3b) attainable by metal hydride systems represent a major technological barrier that severely limits their applications in transport applications. In fact, a big research effort is being devoted to the development of advanced materials capable of absorbing hydrogen with much higher gravimetric densities. Task 17 (Solid and Liquid state Hydrogen Storage Materials) of the IEA-HIA (International Energy Agency Hydrogen Implementing Agreement) has set a goal of 5 wt%, with operating temperatures lower than 80 °C [19]. The targets set by the U.S. DoE (represented in Figure 3) for the year 2010 are even more demanding: 6 wt% for the gravimetric density and 1,5 kWh/l for the energy density [2]. The physical absorption of hydrogen into solid materials is therefore still an interesting research area, also because of its intrinsic safety features, which are discussed in the next section.

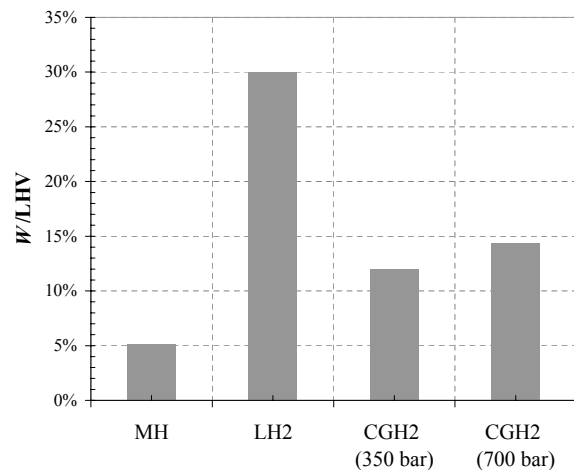
On Figure 4 the specific cost of commercially available hydrogen storage systems is reported, as a function of the energy stored in the device.



a – energy density



b – gravimetric density



c – energy required for storage purposes

Figure 3 – Comparison among commercial hydrogen storage technologies

A scale effect is clearly visible: small systems designed for demonstrative-academic purposes cost an order of magnitude more than other bigger metal hydride tanks.

For the latter the specific cost in terms of quantity of hydrogen stored is, at present, approximately 11 €/g_{H2}, or 330 €/kWh in terms of chemical energy stored.

It is clearly evident that these costs would not be sustainable for a more widespread utilization of metal hydride tanks as hydrogen storage devices; on the other hand, these systems must still be regarded as niche products, even though they are already commercially available, and therefore their cost could be significantly be lowered in case of mass production (in fact, these materials are not unlike those used for the production of some modern rechargeable batteries).

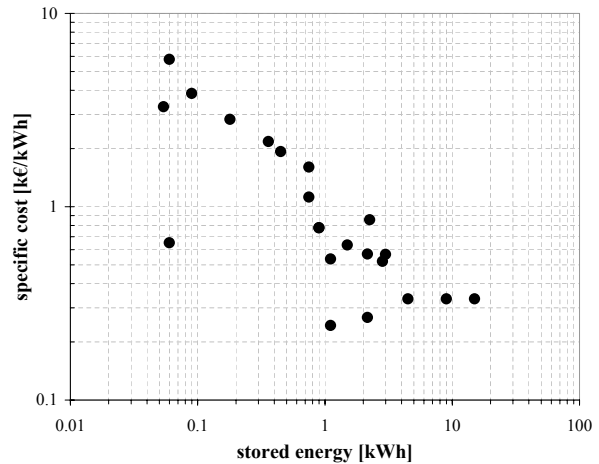


Figure 4 – Specific cost of commercial metal hydride storage systems

4.1. Safety considerations about hydrogen storage technologies

Metal hydride systems exhibit positive characteristics from the safety point of view, especially with reference to the other commercial storage technologies, essentially because they allow to store in solid phase great amounts of hydrogen at relatively low pressures. Two main benefits can be ascribed to metal hydride storage systems with respect to the compressed hydrogen technology.

The first benefit concerns the system response if the tank walls are cracked in the case of a serious incident: the consequent hydrogen release would be relatively small, because the reaction would not be favoured from a kinetic standpoint, given the absence of active heating of the metal hydride bed and of a forced hydrogen removal (which both would be beneficial for the reaction, on the basis of the reaction kinetics [9] and of Le Chatelier – Braun principle [20]). On the contrary, if a crack were to appear on the surface of a compressed hydrogen cylinder, the quantity of hydrogen released to the environment per unit time would be much higher, and also the hydrogen jet, being discharged from reservoirs at pressures as high as 350 bar or even 700 bar, poses a non-negligible safety risk by itself.

The second benefit is related to the storage system behaviour in response to a temperature increase, that is a situation of practical interest, considering a storage tank affected by some heat source (for example direct sunlight). In the case of metal hydride systems, the relationship between pressure and temperature is represented by Van't Hoff equation (2), which determines smaller pressure rises with respect to compressed hydrogen cylinders.

Figure 5 graphically describes the abovementioned behaviour, with reference to an initial temperature $T_0 = 300$ K. The data for the

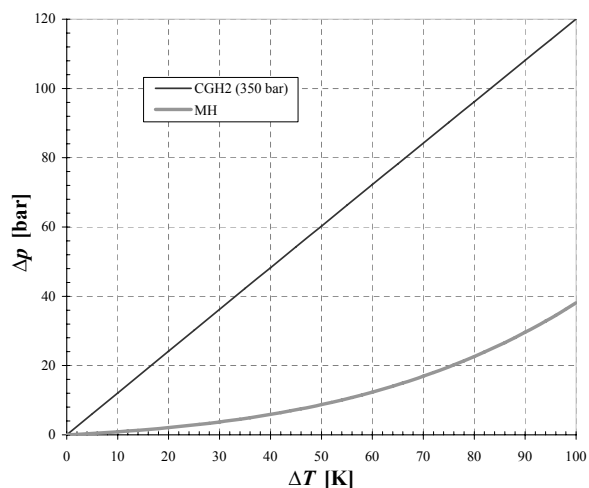


Figure 5 – Tank pressure increase as a result of temperature rise for metal hydride and compressed hydrogen storage systems

compressed hydrogen at $p_0 = 350$ bar have been obtained from the NIST database [21], therefore taking into account its real gas behaviour, but it is evident from Figure 5 that for an isochoric process the linear relationship between pressure and temperature, as predicted by the ideal gas equation of state, is perfectly adequate to describe also the real process.

Therefore for compressed hydrogen the derivative of pressure with respect to temperature can be estimated as follows:

$$\frac{dp}{dT} = \frac{p_0}{T_0} = 1,167 \text{ bar/K} \quad (7)$$

In the case of a metal hydride storage system, on the other hand, the same derivative can be calculated as:

$$\frac{dp_{eq}}{dT} = -\frac{\Delta H}{RT_0} \frac{p_{eq0}}{T_0} = 0,078 \text{ bar/K} \quad (8)$$

where the numerical data refer to LaNi_5 , which exhibits an equilibrium pressure of $p_{eq0} = 1,9$ bar at ambient temperature ($T_0 = 300$ K).

Therefore metal hydride tanks can be considered safer than compressed hydrogen cylinders in the case of accidental temperature increase or heat exposure, because the resulting pressure increment would be significantly lower (e.g. by an order of magnitude for a 50 °C temperature increase).

On the other hand, this behaviour is reversed if gaseous hydrogen at low pressures is considered. For the abovementioned pressure of $1,9$ bar, which represents the equilibrium pressure at 300 K for a LaNi_5 system, the pressure increase for gaseous hydrogen for a unit temperature rise is much lower:

$$\left. \frac{dp}{dT} \right|_{\substack{p=1,9\text{bar} \\ T=300\text{K}}} = \frac{p}{T} = 0,0063 \text{ bar/K} \quad (9)$$

This means that by exposing a metal hydride canister or a gaseous hydrogen container (at low pressure) to a heat flux, the former can release hydrogen at higher pressures than the latter, i.e. it is possible to devise a metal hydride based hydrogen compressor whose only energy input is a heat flux at relatively modest temperatures [22-25].

5. Metal hydride hydrogen-storage applications

5.1. Mobile application: FEMAG project

The authors have been involved in the FEMAG (Flexible Ecological Multi-purpose Advanced Generator) project, a CRAFT project financed within the 6th EU Framework Programme. This project intends to explore optimised integration of components and power aggregates, delivering an energy generator, closed, of small power, based on the integration of a fuel cell with a battery pack and ultracapacitors, for the flexible supply at variable power of small portable non-automotive devices, in the power range of 125 to 1000 W.

FEMAG methodology is based on the integration of commercial and pre-commercial devices and components. The aggregated FEMAG generator design is based on the criterion of minimising fuel cell rated power, entrusting to backup batteries and ultracapacitors the supply of power transients, and put the cell in the condition to work near fixed power output conditions, thus extending its life and increasing overall efficiency. The project involves both experimental and computational optimisation of aggregated systems, and exploits experimental design to set up rigorous testing activities.

The project's goal is to develop two prototype power systems, namely an electric wheelchair for disabled people and an industrial AGV for the transportation of finished goods inside an industrial area. The hybrid structure of the FEMAG power unit is especially helpful in these applications, due to

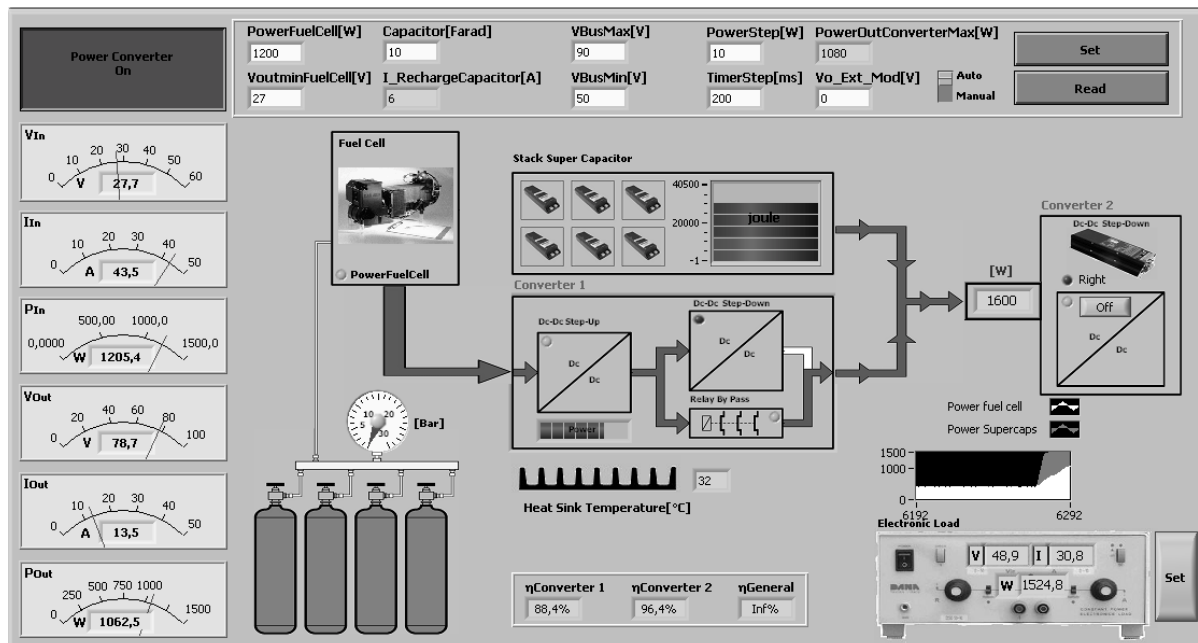


Figure 6 –FEMAG system control panel

their wide load variability and above all the possibility of very high loads for short time intervals (eg. to go over a sloping path with the wheelchair, or to lift a heavy load with the AGV).

A similar concept has been recently deployed also by Hydrogenics Corporation in a lift truck powered by a hybrid system composed of one FC, ultracapacitors and batteries [26]. The main difference from the FEMAG project is that compressed hydrogen storage tanks are used to store hydrogen on-board. Hydrogenics' lift truck has been successfully tested in Canadian GM and FedEx plants.

Figure 6 illustrates the FEMAG prototype control panel during its operation. In the situation represented in the picture, the system was subject to a sudden increase in load (from 400 W to 1600 W) which required the ultracapacitors to come into operation, because a rule is set in the system management policy software so that the fuel cell is able to increase its power output only at a pre-determined rate. The chart on the bottom right shows the power output of both the fuel cell (white area) and the ultracapacitors (grey area) over time.

Figure 6 gives also an insight on the integration among the different modules of the FEMAG prototype: the metal hydride tanks supply hydrogen to the fuel cell, whose electrical output is intercepted by a first DC/DC converter and then combined with the ultracapacitors' output. A second DC/DC converter is finally required in order to supply the power to the electric load represented on the bottom right.

Overall, metal hydride hydrogen storage, even though at present inapplicable for transport applications due to excessive weight, can still represent an interesting option for those mobile applications that are currently powered by batteries, because of lower weight and footprint.

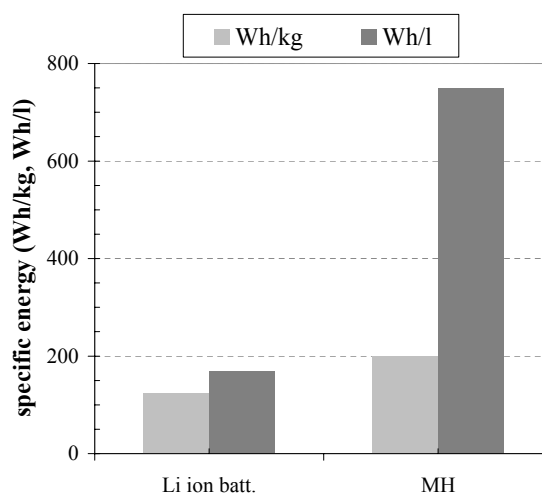


Figure 7 – Comparison between Li-ion batteries and MH-FC systems

For modern Li-ion batteries the specific energy can be estimated as 125 Wh/kg and 170 Wh/l. In the case of metal hydride tanks coupled to fuel cells, both these figures are considerably higher. In fact, the specific chemical energy stored in metal hydrides is, according to Figure 3, of the order of 400 Wh/kg and 1500 Wh/l. Taking into consideration the FC efficiency (roughly 50%) the useful energy stored in a metal hydride – fuel cell system is 200 Wh/kg and 750 Wh/l. This comparison is graphically described in Figure 7.

Several mobile applications have been developed substituting conventional batteries with a fuel cell powered by metal hydride cartridges; among them a professional video-camera application developed by Jadoo Power Systems, Inc. is particularly remarkable [27,28].

5.2. Stationary application: DEMAG project

The authors have also been involved in the DEMAG (Domestic EMergency Advanced Generator) project, a CRAFT project financed within the 6th EU Framework Programme. DEMAG’s goal was to investigate the indoor domestic application of advanced hydrogen technologies to life-saving emergency energy generators, and deliver an emergency power supply, rated 10 kWh, based on the integration of a PEM Fuel Cell with ultracapacitors and metal hydride tanks for the hydrogen storage: the FC is expected to provide a basic power output, whereas ultracapacitors can efficiently supply temporary peak loads. This hybrid configuration allows to design the fuel cell and hydrogen storage system for small loads (it is currently rated 1,2 kW), thus reducing cost, weight and size of the system, which is anyway able to meet significantly higher peak loads (even 100% increase at least for brief intervals) thanks to the ultracapacitors’ intervention.

Hydrogen storage by means of metal hydride canisters is a key feature in this project because safety concerns are obviously very important for an indoor domestic application; furthermore, in this stationary case the system’s weight does not play a significant role.

The prototype is currently equipped with 7 canister of the MHS 1000 IHE type produced by Treibacher Industrie AG, with a nominal capacity of 1 Nm³ each; the system can therefore store approximately 21 kWh of chemical energy (based on hydrogen LHV), thus meeting the target of 10 kWh of energy to the end-user, considering a system efficiency of 50%.

According to the scheme illustrated in Figure 8, the Demag Emergency Power Supply operates as follows: the Master Switch Interface disconnects the domestic grid from the main one and communicates to the Central Unit that it has to start. Then the Central Unit starts, while the Automatic Disconnection Modules disconnect from the local grid high-consumption and not emergency related devices and appliances (e.g. a washing machine). Ultracapacitors can supply power exceeding 1 kW for a limited time; if this happens, DEMAG emits a sound signal informing the user if the system has been overloaded and if it is necessary to disconnect further loads: the sound can be intermittent, increasing its speed if the overload keeps going on and if there is the risk of disconnection. If the load exceeds also the temporary capacity of ultracapacitors, the system disconnects, and waits some seconds before reconnecting; simple modules based on the same communication

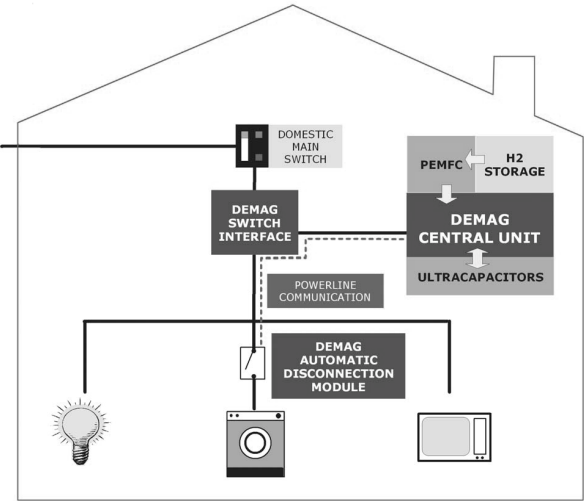


Figure 8 – DEMAG architecture

standards can be placed in the house to repeat the sound signal where required if the Central Unit needs to be placed far from living areas. When the main power is back, the system reconnects automatically, with no disconnection.

Metal hydride hydrogen storage has also been successfully tested and demonstrated in backup systems installed in remote locations, such as telecommunication stations [29], which can be subject to disconnections from the grid, or in renewable-energy systems as a safe solution for storing wind or solar energy in the form of hydrogen [30,31] in order to make remote systems self-sufficient and independent from fossil fuels supplies.

6. Conclusions

In this paper a survey of the integration between metal hydride-based hydrogen storage units and fuel cells has been proposed, along with an analysis of metal hydride systems' state of the art, both from a technological and a commercial point of view, with particular emphasis on the comparison between this relatively new storage technology and more mature ones, such as compressed and liquefied hydrogen or electrochemical batteries.

Metal hydride systems perform well in terms of size, safety and energy required for the storage, but their weight is considerably higher with respect to other options of hydrogen storage. On the other hand, the comparison with electrochemical batteries is favourable for the metal hydride units also in terms of weight, and therefore FC systems with hydrogen storage in metal hydride tanks (or cartridges) can certainly be competitive in those applications that currently rely on batteries.

Two research projects, one for emergency backup stationary applications, and one for small mobile applications, have also been presented and some details on their hybrid architecture have been given.

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