

An investigation of hydrogen storage methods for fuel cell operation with man-portable equipment

Darren Browning *, Peter Jones, Ken Packer

Defence Evaluation and Research Agency, Haslar, Gosport, Hants PO12 2AG, UK

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Abstract

Air breathing proton exchange membrane fuel cells (PEMFC) are being considered as a power source for man-portable equipment, such as army radios. In addition to the weight and volume of the fuel cell itself, the device producing hydrogen with which to fuel the cell is also of crucial importance. This paper describes a number of hydrogen storage methods and discusses their applicability to man-portable equipment.

Keywords: Fuel cells; Proton exchange membrane; Hydrogen storage; Applications/military

1. Introduction

Like civilian users, the UK Ministry of Defence wants to have longer-lasting, lighter-weight power sources for its equipment and this is the impetus for DERA research on fuel cells and their associated hydrogen storage systems. Fuel cells are being considered as a power source for man-portable equipment, e.g. army radio equipment to replace the current battery technology. The fuel cells will run on air and hydrogen. Hydrogen is an excellent fuel for use in a fuel cell due to its high energy density and simplicity. However, one of the major barriers that has prevented its widespread use is the difficulty involved in its storage and portability. Air is to be used instead of oxygen as this removes the need to carry oxygen, thereby reducing the weight of the overall system.

For man-portable applications there is a limit to how much weight can be easily carried. The development of lighter-weight power sources will enable more power-demanding equipment to be used. Although the volume of the system is not as critical as the weight, the system cannot be too bulky as this would hamper the user's mobility. Ideally the system would be easily carried in a backpack. Other factors of importance are the operating temperature, cost, reliability and safety of the storage system.

Hydrogen storage is an essential component of the fuel cell system. The main focus of this paper is primarily on the weight of this hydrogen storage system, with volume as a secondary consideration.

2. Review of storage methods

There have been many systems developed for the storage of hydrogen including those listed below:

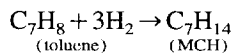
- liquid hydrogenated organic compounds
- hydrogen absorbed by glass spheres and zeolites
- hydrogen reversibly adsorbed (physically) on carbon at low temperature and moderate pressure
- liquid hydrogen
- gaseous hydrogen, compressed in cylinders, both of conventional and composite design
- hydrogen reversibly absorbed (chemi-sorbed) on a metal or alloy as a metal hydride
- non-reversible metal hydrides (hydrogen liberated by reaction with water)

Each of the various methods of hydrogen storage will be described in more detail to explain their principles and highlight any possible advantages and disadvantages. From the explanations it will become immediately obvious that some systems have little potential for man-portable applications but they have been included for completeness.

2.1. Liquid hydrogenated organic compounds

The idea behind this system is to hydrogenate an unsaturated hydrocarbon to store hydrogen and then reverse this process to release the hydrogen when needed, e.g. the hydrogenation of toluene to produce methyl cyclohexane (MCH), a process which is reversible [1,2].

* Corresponding author.



A temperature of 673 K using a noble metal catalyst has been suggested [2] for the dehydrogenation of the methylcyclohexane. The amount of reversible hydrogen carried by the fluid alone is around 6.1% on a weight basis, and for the MCH the hydrogen content is around 49 g dm⁻³. However, due to the large amount of equipment necessary for conversion and the high temperature involved this method is only applicable to the storage of very large amounts of hydrogen and is impractical for man-portable applications.

2.2. Zeolites/glass spheres

A further option for hydrogen storage is to use zeolites or microspheres. Hydrogen is forced into the space inside the microsphere, or in the case of zeolites into the molecular cage, under high pressure and at high temperatures. On cooling the hydrogen is trapped in the sphere or molecular cage and released again only on heating.

The amount of hydrogen trapped by the zeolite is small compared to alternative means of storage, and Weitkamp et al. [3] mention a best value of only 0.08% on a weight basis for hydrogen loaded into a zeolite (sodalite) at 573 K and 100 bar. Therefore at the present stage of development these systems are not an option.

It has also been suggested that hydrogen could be stored in microspheres. The process involves heating hollow glass microspheres, with diameters of 50–100 μm, at between 400 and 700 K so that they become permeable to hydrogen. They are then exposed to hydrogen at high pressure, up to 1500 bar, and cooled. When cooled the glass becomes impermeable

and so the hydrogen is trapped at high pressure in the microspheres until they are heated again. There is little published data on microspheres, with the technique still being in the laboratory stage [4]. This method has the potential of storing large amounts of hydrogen with theoretical figures of up to 40 wt.% H₂ [5] being discussed on the basis of the spheres alone. However, many technical difficulties need to be overcome before this figure is approached.

2.3. Cryo-adsorbed hydrogen

The use of high surface area materials to store hydrogen at modest pressures (up to 60 bar) and at low temperatures between 150 and 77 K, has aroused considerable interest [6]. With a material such as an active carbon and temperatures lowered to 77 K (liquid N₂ temperature) the amount of hydrogen stored at 60 bar is three times that expected by simple compression alone. Results obtained by Chahine and Bose [6] for cryo-adsorbents are shown in Fig. 1.

More recently, interest has been shown in using cryo-adsorbents at higher temperatures, such as could be reached by a relatively simple refrigeration plant. This development has stemmed from the discovery of a super-activated carbon (AX-31M) [7]. The use of the more active carbon gave hydrogen storage comparable with earlier results at liquid nitrogen temperature. The results of adsorption at the higher temperature of 165 K are shown in Fig. 2. Around 7.5% by weight of hydrogen was stored by the carbon at 40 bar pressure and 165 K.

These cryo-adsorbents have quite good weight and volume storage properties but for the very small scale storage of hydrogen being considered for man-portable applications the

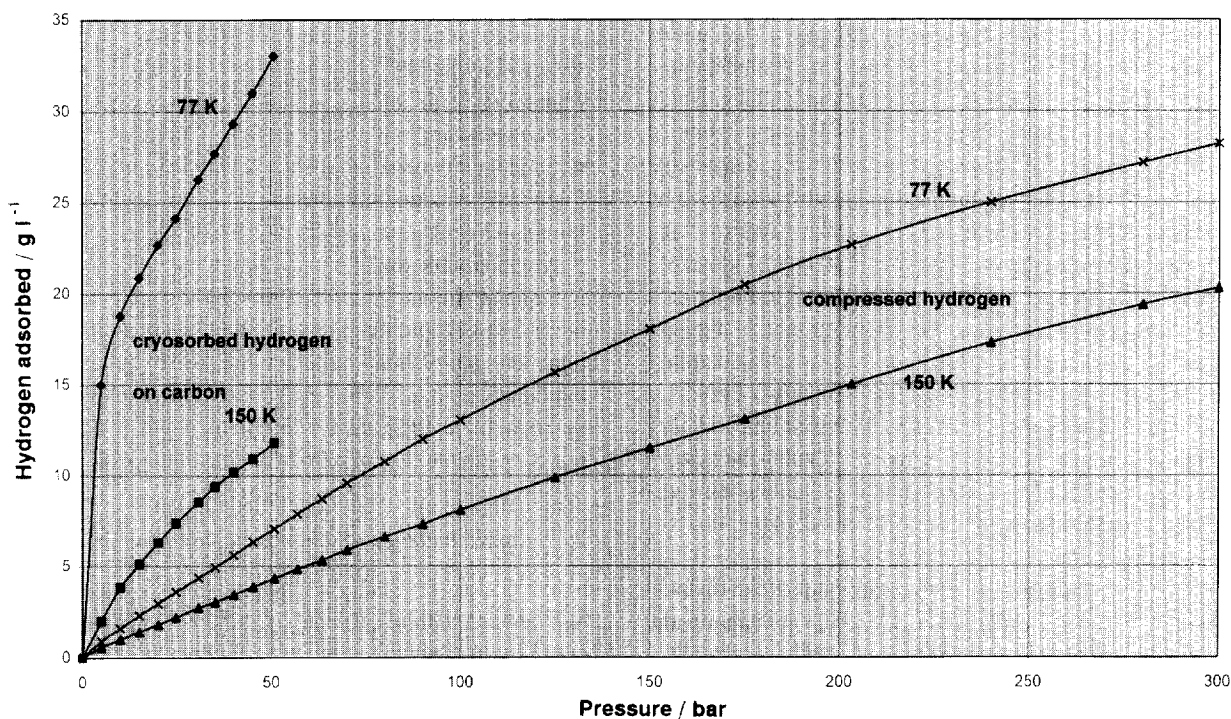


Fig. 1. Comparison of cryo-sorption on carbon with compressed gas storage.

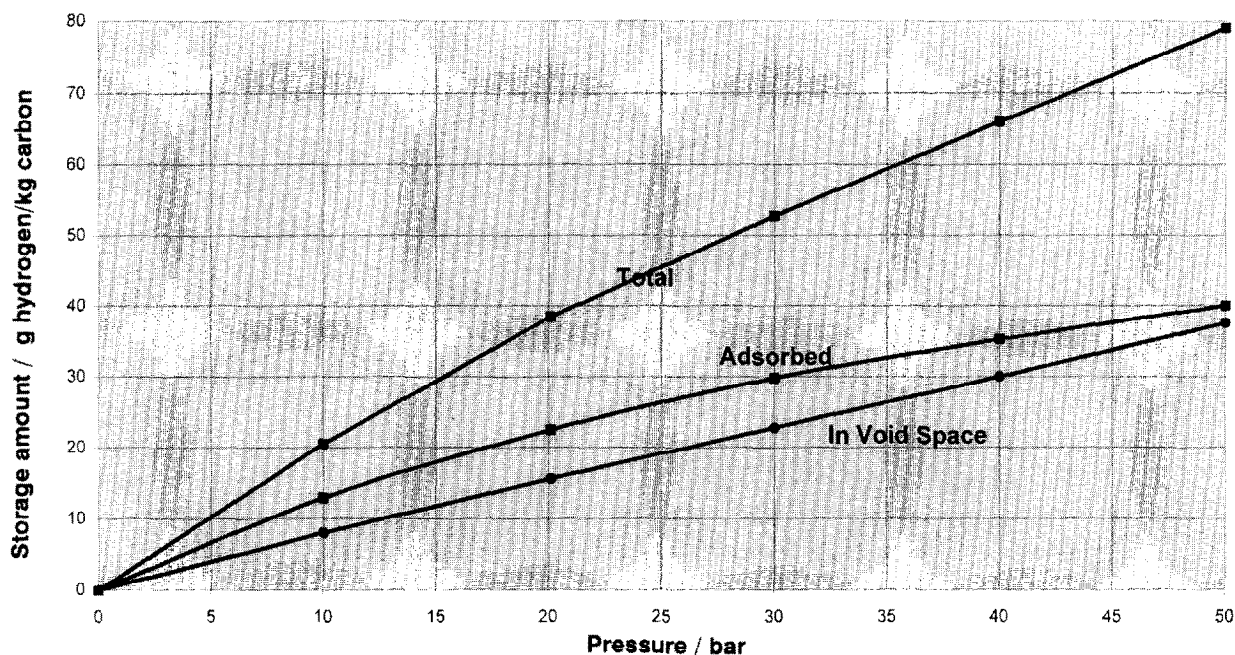


Fig. 2. Hydrogen storage on superactivated carbon at 165 K.

extra complexity of dealing with low temperatures makes these systems less viable.

2.4. Liquid hydrogen

One of the most efficient methods of storing hydrogen on a weight basis is as a liquid, hydrogen liquid having a very low density, 0.071 g cm^{-3} .

The principal difficulty is the extremely low temperatures required, the boiling point (20 K at 1 bar) demanding storage in very well insulated containers, with special attention also for hydrogen lost in natural boil off. Good containers should lose no more than 0.3% per day in boil off for large containers or 1–1.5% for the more modest containers appropriate to man-portable applications.

Although liquid hydrogen is efficient on a weight and volume basis for large and medium scale applications the extra complexity of dealing with such low temperatures makes it unsuitable for small scale man-portable usage.

2.5. Gaseous hydrogen

Storing hydrogen in its natural form as a gas is a widely used and simple method to exploit.

Compression increases the amount (weight) of gas stored per unit volume in the cylinder, but not in direct proportion to the pressure applied. At high pressures hydrogen deviates significantly from the ideal gas laws.

The deviation of hydrogen from the 'perfect' gas law has been calculated from the van der Waals equation at a series of temperatures, and is shown in Fig. 3. With the commonly used storage pressure of 175 bar and an ambient temperature of 298 K the amount of hydrogen contained in the cylinder is about 82% of what would be expected if the hydrogen

obeyed the rules of an ideal gas under compression. Raising the storage pressure to much higher values does not bring the expected benefits as the actual amount of hydrogen trapped in the cylinder deviates yet further from the ideal and the storage container wall thickness has to be increased.

The amounts of hydrogen carried by steel cylinders that are normally available from suppliers of compressed gases are close to a value of 1% by weight. This figure refers to cylinders of around 50 dm^3 and would decrease significantly for the small cylinders appropriate for man-portable use.

The weight percentage of hydrogen carried can be more than tripled if composite-wound pressure vessels are used. This technology involves using a seamless inner liner, usually aluminum or a thermoplastic, which is then overwound with carbon fibre to provide the cylinder's strength. The precise geometric arrangement of the overwinding is calculated to maximise the strength of the cylinder.

2.6. Reversible metal hydrides

A considerable amount of literature has been published on metal hydrides, with many developments over the past 30 years. As a means of storing hydrogen it was a favoured option for many vehicle applications and was studied by BMW in the 1970s for their hydrogen-powered car programme [8]. Much development has also occurred in connection with nickel–metal hydride batteries. A large number of metal alloys have been researched to find one with the most suitable properties. In general, the hydrogen is absorbed reversibly with an isotherm of the type shown in Fig. 4, the available hydrogen being that shown by the flat part of the curve. In practice the hydrogen is taken up at a lower temperature, and released when required by heating the metal hydride to a higher temperature.

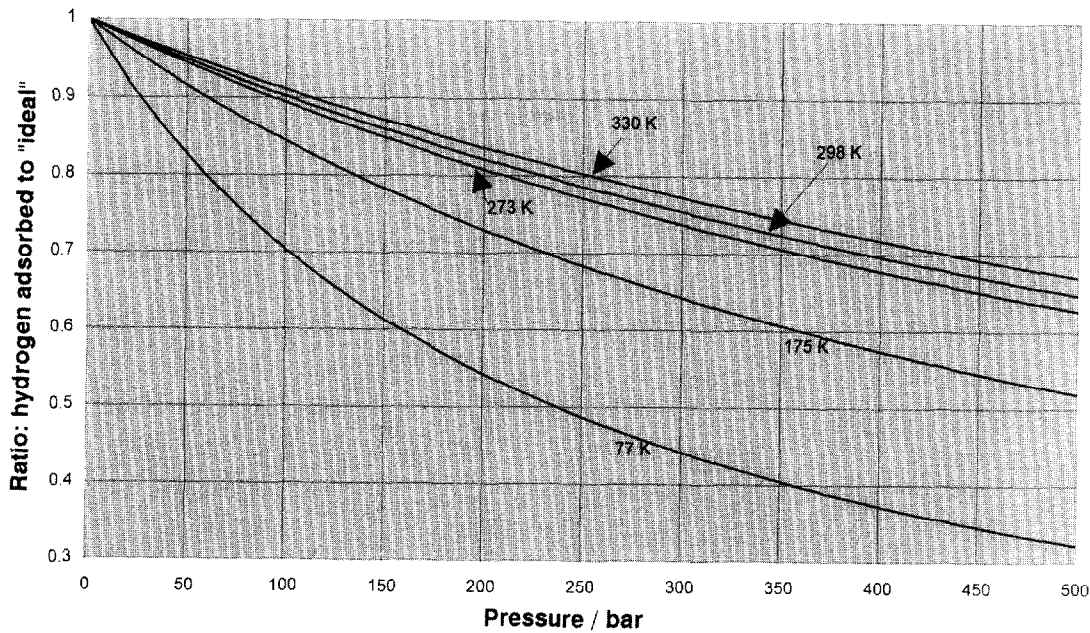


Fig. 3. Deviation of hydrogen from ideal gas law during compression.

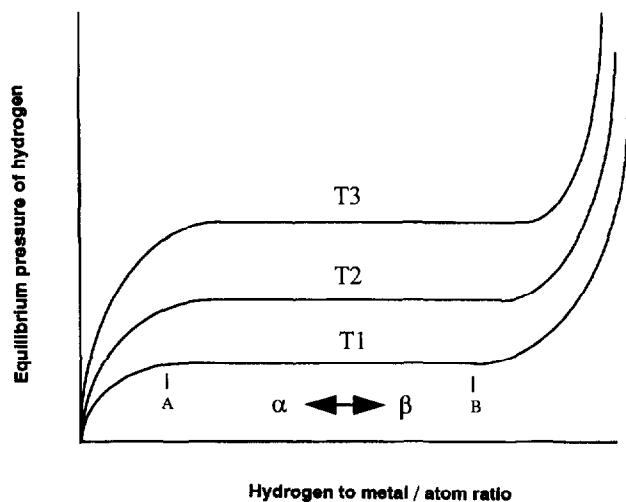


Fig. 4. Idealised P-T-C isotherm for a reversible metal hydride.

The problem with reversible hydrides is their high cost, and in general, high density. The latter property means that on a weight basis they do not achieve better than 1 or 2 wt.% of hydrogen stored for ambient temperature systems.

Reversible metal hydrides are a heavy option, but on a volume basis are very competitive and in some cases better than liquid hydrogen itself. However, the packing density of the hydride cannot be too high as channels need to be left to allow hydrogen gas to enter or exit the bulk storage. In addition, the hydriding process is exothermic, so the mass has to be cooled, and conversely the same heat exchange system has to input heat to desorb the gas when output is required. Overall this can add a volume at least as much as the actual bulk of the metal alloy, as well as adding further weight.

There are a wide range of alloys employable for hydrogen storage and the selection of the best alloy depends on the precise conditions required. The magnesium based alloys can be discounted for this application as they require high tem-

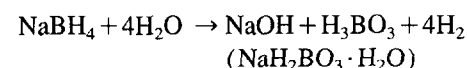
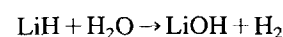
peratures for charging and discharging. The two other major types of alloys are the FeTi based alloys and the LaNi₅ based alloys although small additions of other transition metals can be used to alter their properties slightly. They both have similar capacities on a volume and weight basis. FeTi is by far the cheaper of the two alloys but is less resistant to poisoning and more difficult to activate than LaNi₅. At 298 K the desorption plateau pressure of FeTi is 5.2 bar and of LaNi₅ is 1.6 bar, although these can be altered by addition of other metals to give a range of desorption pressures from 0.4 to 25 bar at 298 K. The effect of some of these additions is shown in Table 1 for the commercially available HYSTOR range of alloys [9].

The plateau pressures of the alloys vary as a function of temperature, with the plateau pressure increasing as the temperature increases. The effect of temperature on the desorption pressures of several common alloys is shown in Fig. 5.

2.7. Non-reversible metal hydrides

For applications where the amounts of hydrogen required are relatively small consideration should be given to using 'one-shot' hydrides, such as LiH, NaBH₄ or LiAlH₄. The amount of hydrogen carried per unit weight is high.

Hydrogen is evolved by the exothermic reaction with water as shown by the equations below:



The non-reversible hydrides contain lighter metals than the reversible hydrides which generally contain heavier transition metals. A further advantage of these 'one-shot' hydrides over

Table 1
Properties of commercially available hydrides

Hystor alloy	Nominal atomic composition	Practical storage capacity (wt.% H ₂)	Hydrogen pressure (bar) at 298 K		Heat of reaction (kJ mol ⁻¹ H ₂)
			Absorption	Desorption	
101	FeTi	1.75	9.9	5.2	-28.0
102	Fe _{0.9} Mn _{0.4} Ti	1.79	8.7	4.6	-29.3
201	CaNi ₅	1.39	5.4	0.46	-31.8
202	Ca _{0.7} M# _{0.3} Ni ₅	1.60	4.1	3.8	-26.8
203	Ca _{0.2} M# _{0.8} Ni ₅	1.08	36.5	24.7	-24.3
204	M#Ni ₅	1.41	118.4	22.7	-20.9
205	LaNi ₅	1.43	2.0	1.6	-31.0
207	LaNi _{4.7} Al _{0.1}	1.36	0.43	0.41	-33.9
208	M#Ni _{4.5} Al _{0.5}	1.20	4.2	3.8	-28.0
209	M#Ni _{4.15} Fe _{0.85}	1.15	12.8	10.9	-25.1

M# denotes Mischmetal, a mixture of rare earth metals.

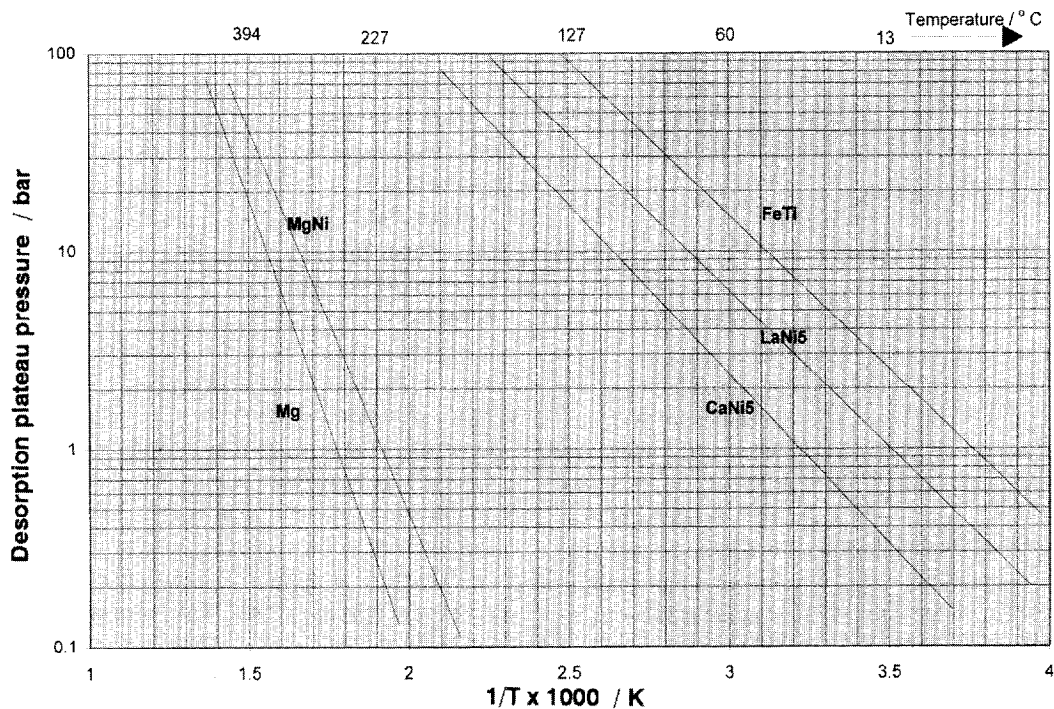


Fig. 5. Variation of desorption pressure with temperature for the reversible hydrides.

the reversible hydrides is that they release twice the amount of hydrogen stored in the alloy itself as half of the hydrogen is derived from the water with which the hydrides react.

Given that the fuel cell itself produces water, there is the possibility of engineering the system such that the waste water produced by the fuel cell is used to react with the metal hydride to liberate hydrogen. It is envisaged that this could drastically reduce the amount of water to be carried with only a small amount for start up of the fuel cell being required.

3. Discussion

The current solution to providing power to man-portable equipment is by the use of batteries and any replacement fuel cell based system would have to be lighter than these, when

the combined weight of the fuel cell, the hydrogen and its storage system are considered.

We will assume a typical weight for a man-portable power source of 3 kg, as being easily carried, and then compare the various methods of storing hydrogen on the basis of how much energy is available for a 3 kg system.

A separate programme aims to develop a man-portable fuel cell capable of delivering 5 Ah at 24 V and weighing 1.4 kg or less with an estimated volume of 0.14 dm³. Therefore for energy systems which require a fuel cell the available weight for the storage system is reduced to 1.6 kg. A fuel cell efficiency of 60% has been assumed throughout this paper.

A number of the possible systems of hydrogen storage discussed earlier are impractical for man-portable use because of their complexity, requiring either too high or too low a temperature for operation, or because the technology is not

Table 2
Hydrogen stored in a 1.6 kg reversible hydride store of FeTi and LaNi₅

	FeTi	LaNi ₅
Percentage H ₂ stored in alloy alone (wt.%)	1.75	1.40
Percentage H ₂ stored in entire container (allowance for cylinder and heat exchange) (wt.%)	0.875	0.70
Weight of H ₂ carried in a 1.6 kg hydride storage container (kg)	0.014	0.0112

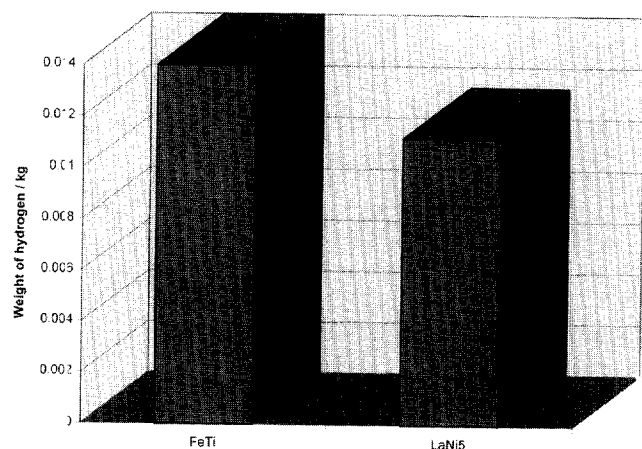


Fig. 6. Weight of hydrogen contained in a 1.6 kg reversible hydride storage system.

sufficiently understood at present. This leaves a choice between reversible hydrides, non-reversible hydrides and gaseous hydrogen stored in filament-wound cylinders.

Hydrides, both reversible and non-reversible, have an advantage over cylinder storage from the standpoint of safety. For the reversible hydrides, in the event of a leak the hydrogen can only be desorbed at the rate the heat of desorption enters the hydride bed, hence in the absence of a substantial heat source the reaction is self-limiting. For the non-reversible hydrides, the hydrogen will only be desorbed when water is available for reaction and therefore once again a puncture in the storage container will not liberate hydrogen immediately but only if a water source is available.

The reversible hydrides are traditionally thought of as a heavy but safe method of hydrogen storage. Currently available ambient temperature hydrides can store approximately 1.8 wt.% hydrogen based on the weight of the alloy alone. However if the storage container and heat exchangers are also considered this figure is likely to halve.

Table 2 and Fig. 6 shows the amount of hydrogen which can be stored in a 1.6 kg metal hydride container.

Table 3
Hydrogen stored in a 1.6 kg non-reversible hydride storage system

	LiH	LiAlH ₄	NaBH ₄
Percentage H ₂ evolved from hydride alone (wt.%)	25.2	21.1	21.3
Percentage H ₂ evolved from reactants (i.e. hydride + water) (wt.%)	7.71	7.32	7.33
Percentage H ₂ evolved from total system (i.e. hydride + water + container) (wt.%)	6.43	6.10	6.11
Weight of H ₂ contained in 1.6 kg store (kg)	0.1030	0.0976	0.0978

Note: a stoichiometric amount of water has been assumed although some excess might be required for complete reaction.

The heavy reversible hydrides have the advantage of being a low-volume storage method. The two most common hydrogen storage alloys — FeTi and LaNi₅ — have densities of 5.47 and 6.59 g cm⁻³ [10] when hydrided which corresponds to 0.091 and 0.093 g of hydrogen per cm³, respectively. For the 1.6 kg storage system this equates to a volume of hydride of 0.146 dm³ for FeTi and 0.121 dm³ for LaNi₅. When the bulk of the storage container and heat exchanger is considered the volume is likely to approximately double to give volumes for the entire storage systems of 0.292 and 0.242 dm³ for FeTi and LaNi₅.

Non-reversible metal hydrides are much lighter than their reversible counterparts with sodium borohydride being able to liberate 21.3 wt.% hydrogen based on the weight of the hydride alone. The actual percentage stored will be less than this when the weight of the storage container, the water required to react with the hydride and the mixing device is considered. As the pressure of the hydrogen liberated can be kept low, only a thin walled container will be required and we will assume a further 20% weight for the hydrogen and water containers, mixing device and control valves. If we assume all the water required for the reaction is carried, then the hydrogen available from a 1.6 kg storage system of some common non-reversible hydrides is listed in Table 3, and is summarised in Fig. 7.

Values of up to 6 wt.% H₂ have been quoted for non-reversible hydride stores. This agrees with the estimates in Table 3.

It can be seen from Table 3 that the non-reversible hydrides provide similar amounts of hydrogen to each other and are much more favourable than the reversible hydrides on a weight basis even when the weight of the water reactant is considered. The 'one-shot' hydrides are not as advantageous as the reversible hydrides on a volume basis, especially when the volume of reactant water is included. The waste hydroxides produced by the reaction take up more space than the original hydrides and so space must be left in the hydride

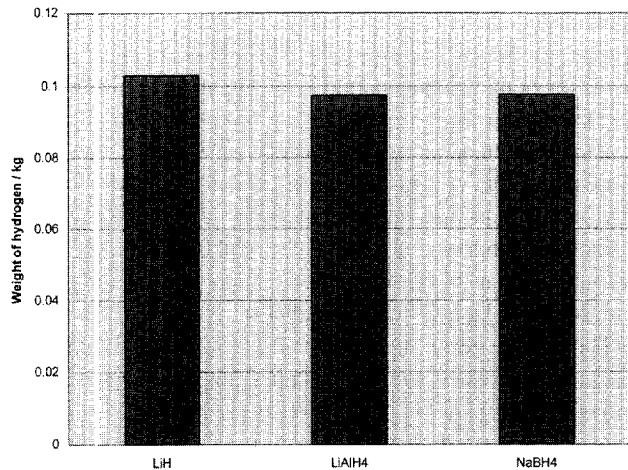


Fig. 7. Hydrogen contained in a 1.6 kg non-reversible metal hydride storage system.

container for swelling upon reaction. A commercial system [11] is available for 100 Wh (3 g H₂) in which the volume of the system is approximately double that for the hydride and water reactants when the ancillary equipment and provision for hydride swelling is included.

Fig. 8 shows the volume of a 1.6 kg non-reversible hydride storage system based on LiH, LiAlH₄ and NaBH₄.

Cylinder storage, as the most established and simplest method of storing hydrogen, must also be considered. As stated earlier, traditional steel cylinders can store approximately 1 wt.% of hydrogen (for cylinders of around 50 dm³). With the advent of composite cylinders, values of more than 3 wt.% H₂ have been quoted by manufacturers. However, for man-portable equipment the amount of hydrogen required is quite small. At these small volumes it seems unlikely that the cylinders will be as efficient as the 3% quoted.

The smallest composite cylinder we have found which is commercially available [12] has a volume of 4.7 dm³, a weight of 2.8 kg and a working pressure of 200 bar. Therefore this cylinder could store 0.065 kg of H₂ (corrected for non-ideality) which corresponds to 2.3 wt.%. However the weight of the cylinder does not decrease linearly as the volume decreases. In fact at low pressures the weight decreases almost exponentially with the volume. If this is the case then it has dire consequences for the weight percent of hydrogen stored at smaller volumes. For a cylinder as small as 1.6 kg the weight percentage is likely to drop to around 0.6%.

The weight of the regulator for the cylinder also has to be considered, as on a small scale this may contribute a significant proportion of the weight. Lightweight regulators are available, weighing approximately 0.2 kg. This will decrease the weight percentage of hydrogen stored in a small composite cylinder still further to approximately 0.5%.

Hence on a basis of weight considerations alone these systems are much less favourable than 'one-shot' hydrides and surprisingly even less favourable than the reversible hydrides which are traditionally thought of as the heavy option for hydrogen storage. However cylinder storage still has the benefit that it is the most widely understood and simplest method of storing and transporting hydrogen and this might outweigh its poorer weight and volume performances. Further developments of composite cylinder technology should decrease the weight of the cylinders in addition to increasing their strength and allowing higher pressures to be used. This will increase the weight percent of hydrogen stored in a composite cylinder.

The hydrogen stored in a 1.6 kg composite cylinder with hydrogen at 200 bar is given in Table 4.

The volume of cylinder systems is generally taken as the volume of the gas plus a further 10% for the volume of the

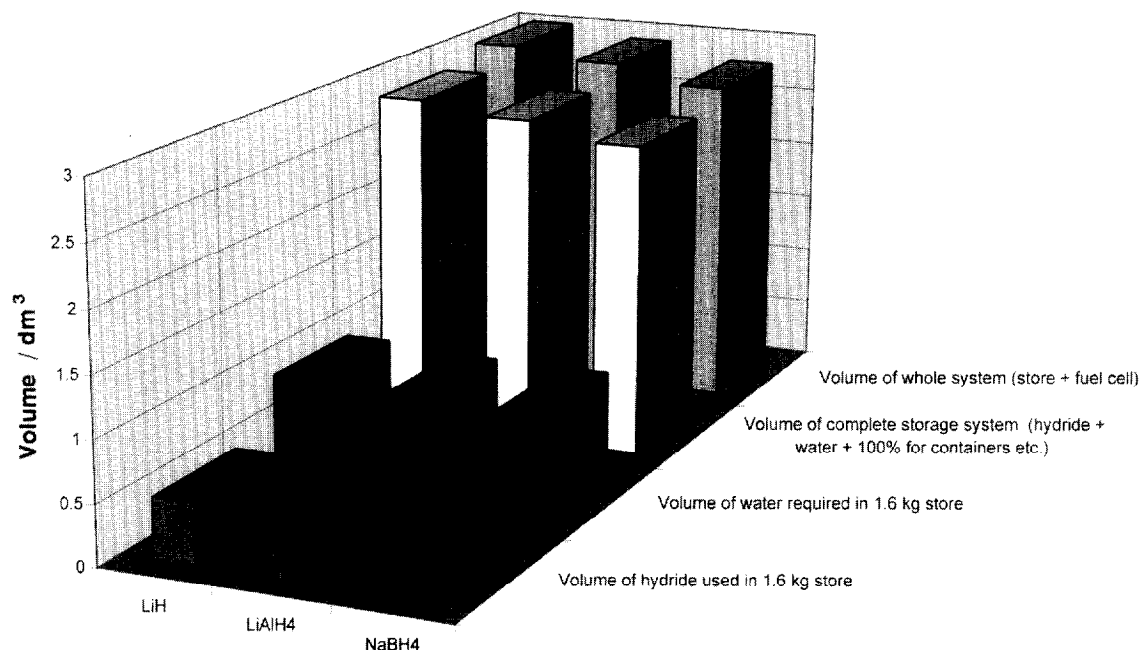


Fig. 8. Volume of a 1.6 kg non-reversible metal hydride storage system.

Table 4
Hydrogen stored in a 1.6 kg composite cylinder at 200 bar

Weight of cylinder (kg)	Percentage H ₂ stored (wt. %)	Weight of hydrogen stored (kg)
1.6	0.5	0.008

cylinder. As was the case for the weight, the proportion of the volume taken up by the cylinder itself will be larger for a small amount of hydrogen. We have taken a value of 20% as the proportion of the total volume occupied by the cylinder and regulator of a small 1.6 kg cylinder. This corresponds to a cylinder size of 0.59 dm³ for the small 1.6 kg cylinder.

4. Summary

Table 5 compares the amount of hydrogen and energy stored in a 1.6 kg storage system based on reversible metal hydrides, non-reversible metal hydrides and composite cylinder storage.

From Table 5 it can be seen that by far the most weight efficient method of storing hydrogen for man-portable applications is as a non-reversible, 'one-shot', metal hydride. The reversible metal hydrides are the most efficient on a volume basis but volume is a secondary consideration. Although the non-reversible metal hydrides are classed as 'one-shot', and cannot be recharged with hydrogen, the power source itself is recharged by replacing a cartridge of spent hydride with a cartridge of unreacted hydride.

If fuel cell and hydrogen storage systems are to be considered as a replacement for battery systems in man-portable

applications then they must offer better energy densities than competing battery technologies. Table 5 showed the energy densities of the hydrogen storage systems. However, to convert the energy stored as hydrogen into electricity a fuel cell is also required. It is the total weight of the hydrogen storage system and fuel cell system which must be considered when any comparison with batteries is made. As mentioned earlier a separate programme is developing a fuel cell which can deliver 5 Ah at 24 V, with an efficiency of 60%, and has a weight of 1.4 kg and a volume of 0.14 dm³.

The battery currently in use is a rechargeable nickel/cadmium with an energy density of approximately 30 Wh kg⁻¹ and 65 Wh dm⁻³. The next generation of rechargeable batteries (lithium-ion) has an energy density of 70 Wh kg⁻¹ and 200 Wh dm⁻³. Even higher energy densities of up to 200 Wh kg⁻¹ have been suggested as being achievable with this technology, in the long term.

Table 6 compares the best example of each of the three potential hydrogen storage systems (reversible hydride, non-reversible hydride and cylinder storage) with the current nickel/cadmium and the lithium-ion battery on the basis of weight and volume.

The gravimetric and volumetric densities of each of the methods for a man-portable 3 kg total unit are shown in Fig. 9.

The potential for man-portable fuel cell systems therefore seems quite promising. For a total weight of 3 kg, all three combined hydrogen storage and fuel cell systems offer better energy levels than the current nickel/cadmium rechargeable batteries. Composite cylinder and reversible hydride systems offer similar energies to the lithium-ion battery, which will be the next generation of rechargeable batteries for military use. However, the best system is the non-reversible metal

Table 5
Comparison of the amount of hydrogen and energy stored in a 1.6 kg storage system between the reversible metal hydrides, non-reversible metal hydrides and composite cylinder technologies

	Reversible hydride (FeTi)	Non-reversible hydride (LiH)	Composite cylinder
Weight of H ₂ carried (kg)	0.014	0.098	0.008
Energy stored (Wh)	457	3365	262
Volume of storage system (dm ³)	0.29	2.85	0.59
Gravimetric energy density for 1.6 kg storage unit (Wh kg ⁻¹)	286	2103	164
Volumetric energy density for 1.6 kg storage unit (Wh dm ⁻³)	1576	1180	444

Table 6
Comparison of energy stored in a man-portable fuel cell and hydrogen storage system (fuel cell = 1.4 kg, hydrogen store = 1.6 kg) with nickel-cadmium and lithium-ion batteries

	Reversible hydride (FeTi)	Non-reversible hydride (LiH)	Composite cylinder	Nickel-cadmium battery	Li-ion battery
Energy available from 3 kg total system (60% efficient fuel cell + store) (Wh)	274	2019	157	90	210
Total volume (store + fuel cell) (dm ³)	0.43	2.99	0.73	1.38	1.05
Gravimetric energy density for 3 kg unit (Wh kg ⁻¹)	91.3	673.0	52.3	30	70
Volumetric energy density for 3 kg unit (Wh dm ⁻³)	637.2	675.3	215.1	65	200

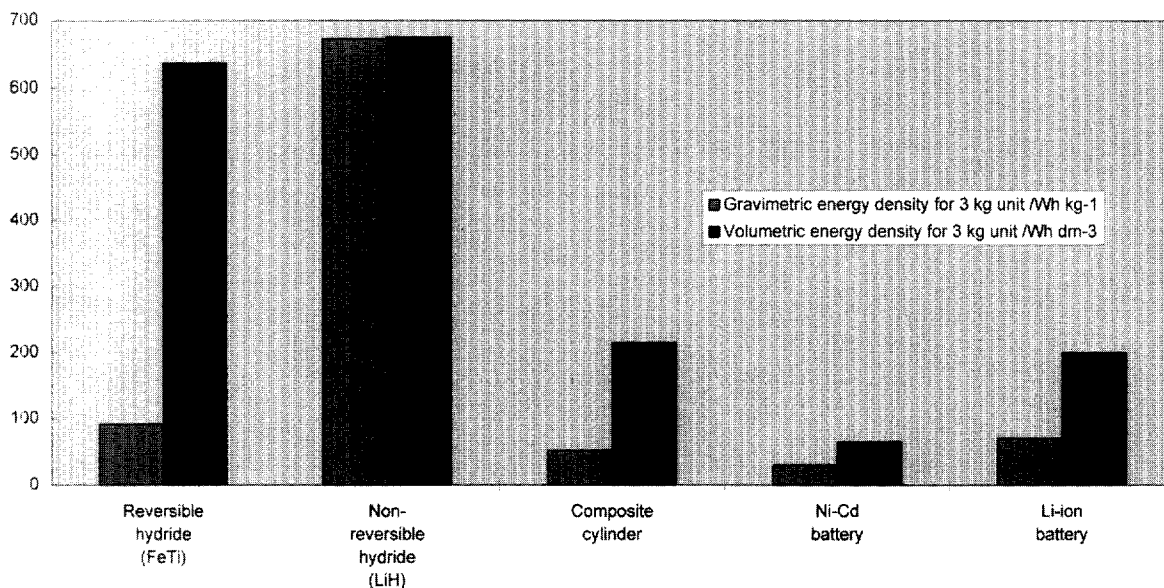


Fig. 9. Gravimetric and volumetric energy densities for a 3 kg unit of each of the candidate power sources.

hydride and fuel cell system which offers the potential of providing over 20 times the energy of the present nickel/cadmium battery for the same weight. The energy density of these non-reversible metal hydride and fuel cell systems (673 Wh kg^{-1}) is potentially over three times better than the predicted future energy density of the lithium-ion batteries (200 Wh kg^{-1}).

One operational benefit of increasing the amount of energy available for a set weight is that the operating time of the equipment is increased resulting in extended mission duration.

Fuel cell and hydrogen storage systems will become even more competitive as the weight allowed for man-portable power supplies increases as although the weight and volume of the store need to be increased the fuel cell can remain the same.

Where possible, the values discussed in this paper are present-day values and although the energy density of lithium-ion technology is predicted to increase to around 200 Wh kg^{-1} , it is likely that the performance of the various hydrogen storage methods and fuel cells will also improve with further research.

It should also be remembered that the values quoted in this paper are specific for small scale man-portable applications

and the relative advantages of each of the systems reviewed will differ for larger scale applications.

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