Electric Energy Storage Using Aluminum and Water for Hydrogen Production On-Demand

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Abstract

The paper analyzes the potential electric energy storage resulting from a hydrogen-oxygen fuel cell fed by in-situ, on-demand production of hydrogen from aluminum-water reaction. The reaction is made practical by an original aluminum activation process using a small fraction (typically 1-2.5wt%) of lithium-based activator. The reaction provides 11% of hydrogen compared to the aluminum mass, with a practical yield of over 90%, which is equivalent to about 2200 Wh/kg Al of electric energy storage when combined with a fuel cell. It is shown that specific electric energy greater by up to 15 fold compared to common batteries may be obtained, depending on the specific application and operating time.

Keywords: Energy Storage, Hydrogen, Fuel Cell, Aluminum-Water Reaction, Activated Aluminum

1. Introduction

1.1 General

The increasing demand for energy around the world, together with the need to reduce the dependence on fossil fuels and find environmental friendly sources of energy, have led to an extensive search for alternative fuels and energy sources. The leading candidate to be the fuel of the future is hydrogen. It has an outstanding reaction energy, 142.9 MJ/kg (high heating value, HHV), about three times higher than hydrocarbon fuels (Figure 1), and its reaction product with air (only water in liquid or vapor state) is non-polluting. The use of hydrogen fuel involves difficulties of storage and transportation resulting from its very low density, 0.089 kg/m^3 (gas), about 14 times less than air, and 71 kg/m^3 (liquid), 14 times less than water, from its high flammability and explosion hazards, and from its higher cost than conventional fuels. In order to enable a wide use of hydrogen as a fuel a solution to those issues must be found.

The most efficient utilization of hydrogen energy for power generation is in hydrogen-oxygen fuel cells, e.g., proton exchange membrane (PEM) or alkaline fuel cells, which produce electric energy using (stored) hydrogen and oxygen (from the air). In fuel cells, the fuel (hydrogen) and oxidant (air) are not an integral part of the system, and are fed to the fuel cell stack whenever electric energy is required. Therefore, the fuel cell stack determines the power generated by the system, whereas the energy density of the entire system is dependent to a large extent on the hydrogen storage method used.

Hydrogen fuel cells have many advantages, as will be further discussed; compact hydrogen storage plays an important role in the implementation of hydrogen technology in various applications.

The objective of this work is to introduce a method for on-demand, in-situ hydrogen production and storage, based on the reaction between aluminum and water, and to compare its characteristics to other alternatives.

It is further sought to demonstrate its promising features as a method for electric energy storage. Initial discussion of the subject was previously presented by Elitzur, Rosenband & Gany (2013).

1.2. Common Methods of Hydrogen Storage

The challenge of hydrogen storage has been drawing much attention, and many methods and technologies for hydrogen storage have been developed, as described by Züttel (2004); Jena (2011); Zhou (2005). The main two requirements from a hydrogen storage technology are high hydrogen capacity and hydrogen release in moderate temperatures. Those requirements are especially important when applied in mobile applications (Felderhoff, Weidenthaler, Helmolt & Eberle, 2007; Schlapcach & Züttel, 2001). Today, three main methods are used for hydrogen storage: pressurized gas, liquid hydrogen, and metal hydrides. Pressurized hydrogen tanks to 200-700 bar contain only 2-6wt% of hydrogen. The second option of storing liquid hydrogen in cryogenic tanks requires cooling to about 20K, and the refrigeration process to this very low temperature implies investment of a substantial portion (about one third) of the hydrogen energy. Table 1 summarizes typical values of density and wt% of pure hydrogen in different storage alternatives.

The metal hydride option offers hydrogen storage in compounds of metal and hydrogen. The hydrogen can be released from the compound by heating or pressure reduction. Hydrogen storage in solid metal hydrides is a safe and relatively compact (volume wise) method for hydrogen storage and it is currently used for different applications. However, practical metal hydrides which release hydrogen at moderate temperatures (e.g., LaNi5H6), store only a small amount of usable hydrogen, typically 1-2wt%. Table 2 summarizes some of the most studied hydrides (Chen & Zhu, 2008; Principi, Agresti, Maddalena & Russo, 2009) which release hydrogen via their decomposition. As can be seen in Table 2, among the listed hydrides, magnesium hydride has the largest gravimetric capacity, however, it exhibits major disadvantages such as the release of hydrogen at relatively high temperatures (~300°C), slow decomposition kinetics, high reactivity, high cost, and limited availability (Pain, Lal & Jain, 2010; Sakintuna, Lamari-Darkrim & Hirscher, 2007), making it impractical for common applications.

Some metal hydrides produce hydrogen when reacting with water. Lithium borohydride can theoretically produce the highest amount of hydrogen (37wt%), but it is expensive, very reactive, and sensitive to air and humidity. One of the more practical materials to use is sodium borohydride, because it is soluble in water. For reacting with water to produce hydrogen a catalyst is required. Theoretically 21.3 wt% of hydrogen (on the basis of the sodium borohydride mass) may be produced. However, the efficiency of hydrogen production from sodium borohydride and water is relatively low, and much attention is paid for the development of new catalysts to enhance its reaction with water (Kim J., Lee, H., Han, Kim, H., Song & Lee, J., 2004; Lu, Chen, M. & Chen, Y., 2012).

New hydrogen storage materials are constantly being investigated and developed, attempting to address the challenges of high gravimetric hydrogen density, decomposition at low temperatures, and faster decomposition kinetics (Greats, 2009; Klebanoff & Keller, 2012).

2. Hydrogen Production and Storage via Aluminum-Water Reaction

The aluminum-water reaction discussed in this article is:

$$\mathrm{Al} + 3\mathrm{H_2O} \rightarrow \mathrm{Al(OH)_3} + \frac{3}{2}\mathrm{H_2} \quad (1)$$

The stoichiometric reaction yields theoretically 11% hydrogen mass based on the aluminum mass, which is equivalent to 1.24 liter of hydrogen per 1 gram of aluminum in standard conditions. In addition, aluminum and water are readily available, simple and safe to work with, and can be stored for a long time, making the concept attractive and efficient for hydrogen storage.

However, the chemical reaction between aluminum and water does not practically occur in regular conditions due to a thin oxide layer that covers the aluminum surface, protecting the metal from further reactive interaction with the surrounding. A novel thermo-chemical process of aluminum activation, developed and patented at the Fine Rocket Propulsion Center of the Faculty of Aerospace Engineering of the Technion (Rosenband & Gany, 2014), enables a spontaneous and sustained reaction between activated aluminum particles and water at regular conditions (room temperature). A small fraction of a lithium-based activator (typically 1-2.5wt%) modifies the protective properties of the oxide or hydroxide film on the aluminum particle surface, allowing the water to react with the aluminum.

The influence of different parameters (e.g., water/aluminum mass ratio, fraction of activator, initial water temperature, size and shape of aluminum particles, and type of water) on the reaction rate and efficiency has been studied (Rosenband & Gany, 2010; Elitzur, Rosenband & Gany, 2013, 2014), demonstrating high reaction yields of more than 90% and high reaction rates, 200-600 ml/min/g Al (controllable).

It is generally found that higher water temperature causes higher reaction and hydrogen evolution rates. The reaction is exothermic and implies an increase of the water temperature during the reaction. Yavor, Goroshin, Bergthorson, Frost, Stowe & Ringuette (2013) and Vlaskin, Shkolnikov & Bersh (2011) demonstrated that at high temperatures (e.g., 200-300 ºC) micron-size aluminum may react with water without activation, giving relatively high yield of hydrogen production. However, operation at room temperature requires activation or catalysts. Elaboration on different studies of this subject is included in the works by Rosenband & Gany (2010) and Elitzur et al. (2014). With regards to the present technique, one of the main findings is that the reaction proceeds with any type of water (e.g., tap water, purified water, or sea water), making this method attractive for marine applications. Figure 2 demonstrates experimental results of hydrogen production from activated aluminum with different water types in a batch reactor at atmospheric pressure. The experiments were conducted using aluminum powder of 9 µm mean diameter, 2.5 wt% of activator, and starting with water at room temperature. The figure demonstrates the typical specific hydrogen production rate (a few hundreds ml/min hydrogen per gram of aluminum) and the typical reaction duration (a few minutes).

One main advantage of this technology is safe operation. The hydrogen is produced in-situ and on demand. The aluminum powder and water are stored separately and produce hydrogen when and where it is needed. The hydrogen produced is channeled directly to a fuel cell, where it is used immediately, without accumulation, to generate electricity.

Examination of the technology for hydrogen storage according to the requirements previously mentioned proves its additional advantages: First, the reaction occurs at room temperature. Second, the reaction represents hydrogen storage at high gravimetric capacity. The overall hydrogen gravimetric capacity obtained with this technology depends on the application. Besides the aluminum powder weight, in certain applications water has to be carried along and its weight should be taken into account when calculating the overall hydrogen gravimetric fraction. On the other hand, there are applications where water is available from the surroundings and should not be considered as part of the system. Comparison between the hydrogen gravimetric capacity in the aluminum and water reaction and the hydrogen gravimetric capacity in different hydrides shows the promising potential of the present method. Figure 3 presents hydrogen storage gravimetric capacity of different hydrogen storage materials and methods versus temperature of application. The US Department of Energy (DOE) targets for hydrogen storage (Satyapal, 2007; Stetson, 2014) were introduced on the graph (dashed lines zone). One can see that the aluminum-water technology (marked as "Technion") meets well the DOE technical requirements (11wt%, room temperature) for applications where only aluminum weight is considered (and water is available); if water weight has to be considered as well (denoted as Technion Portable Application in Fig. 3), it still shows high percentage (about 4wt%) and is in convenient temperature range compared to most of the practical metal hydrides. One may notice, that except for the Al-water (Technion) method, there are currently no practical methods that comply with the DOE target.

3. Electric Energy Storage

The main problem with electric energy storage is its low specific energy (energy per unit mass) and energy density (energy per unit volume).

Most commonly, electric energy is stored in batteries. Batteries are divided into two categories: primary batteries (non-rechargeable) and secondary batteries (rechargeable). The energy density of primary batteries is typically higher than that of secondary (rechargeable batteries). However, the latter may be recharged and used tens and hundreds of times. Table 3 gives the specific energy, characteristics, and common applications of some primary (non-rechargeable) batteries (adapted from Aifantis, Hackney & Kumar, 2010; Linden & Reddy, 2002). Table 4 presents these properties for certain commonly used secondary (rechargeable) batteries.

One can note the low specific energy of batteries when comparing them to the overall specific chemical energy of common hydrocarbon fuels, about 11.7 kWh/kg (=42 MJ/kg), ow heating value LHV, and 12.4 kWh/kg (=44.5 MJ/kg), high heating value HHV (Lide, 2006).

The gross specific energy of hydrogen is yet about three times higher, about 33.6 kWh/kg (=120.9 MJ/kg), LHV, and 39.7 kWh/kg (=142.9 MJ/kg), HHV. Of course, the values above are the overall heating values. Conversion into electric energy may utilize only a fraction of these values. The most efficient way to convert reaction energy directly into electric energy is via fuel cells. When dealing with hydrogen fuel, the most practically used fuel cell alternative is the proton exchange membrane (PEM) fuel cell. Alkaline fuel cell is another option. Typically it is heavier than the PEM fuel cell and requires carbon-dioxide-free oxygen supply.

PEM fuel cells generate electric energy from hydrogen (supplied) and oxygen acquired from the ambient air in most applications (Fig. 4), according to the chemical reaction:

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \qquad (2)$$

The only byproduct of their operation is water (liquid or vapor), they are quiet and operate at low temperatures (typically 20-100°C). PEM fuel cells typically exhibit about 50% efficiency of electric energy generation compared to the HHV of the chemical reaction (this is considerably higher than the regular conversion of heat into mechanical energy and then to electric energy, e.g., in diesel generators).

Both batteries and fuel cells convert chemical energy into electric energy. The difference between those options for energy storage is the location of the active material (fuel and oxidant). In batteries the fuel and oxidant are part of the device, whereas in fuel cells they have to be supplied externally. This difference leads to several advantages of fuel cells over batteries: fast recharging, longer continuous run-time (approximately two- to ten- times longer), constant voltage and lack of power decrease during operation, greater durability in outdoor environment under wide temperature range, and less maintenance (DOE Report, 2011).

Although electric energy production via fuel cells includes certain additional mass besides the fuel (i.e., the fuel cell with its regulator and controls and the fuel container or source), one may estimate the specific electric energy on the basis of the fuel, particularly when dealing with an extended operating time (when the overall electricity production mass is mainly the fuel mass).

Depending on the storage density of hydrogen, PEM fuel cell systems may exhibit a much better energy density than batteries. The appropriate comparison should be made according to the application or mission, i.e., to primary batteries if one considers a single use, and to secondary (rechargeable) batteries when aiming at multiple applications. Further comparison between the energy storage characteristics of batteries and fuel cells is conducted by Winter & Brodd, 2004.

When applying our hydrogen production method for electric energy storage, one may consider a number of scenarios with relation to specific energy or energy density according to the aluminum-water reaction (for hydrogen production) and the hydrogen-oxygen reaction in the fuel cell.

Typically, for land devices (e.g., emergency electric generators) aluminum has to be provided, water is usually available, and oxygen is obtained from the ambient air.

In portable devices, as well as in automotive applications, aluminum and water have to be carried along, whereas oxygen is available from the surroundings. Nevertheless, in many cases water may be readily added, and does not have to be provided in full for an extended operation.

For aeronautical vehicles the entire mass of both the aluminum and water has to be accounted for. Oxygen is obtained from the surroundings. Exceptions may be auxiliary power units (APUs) and cabin power generators in commercial airplanes, where water is already available on-board.

Marine surface vessels have to carry only the aluminum. Water and oxygen (air) are available.

Marine underwater vehicles should carry both the aluminum and oxygen (e.g., in pressurized tanks or in liquid oxygen containers). Water is available from the ambience. If the operation is not deep under the sea, then in some cases oxygen may be acquired from the ambient air as well.

The electric energy that can be produced by a PEM fuel cell is equal to 19.8 kWh per kg hydrogen, considering that hydrogen reaction energy (high heating value HHV) is 142.9 MJ/kg and that PEM fuel cell efficiency is 50%.

In a complete reaction between aluminum and water, one ninth (0.11) kg of hydrogen is produced per 1 kg of aluminum. This means that the specific electric energy (per unit mass of aluminum) generated by the combination of the activated aluminum-water reaction and a PEM fuel cell is 2.2 kWh per kg aluminum.

Table 5 presents the specific electric energy that can be obtained by applying the activated aluminum reaction with water to produce hydrogen, combined with a PEM fuel cell, at different operating modes. It should be noted that our calculations take into account 50% efficiency of the fuel cell. Fuel cell mass is approximately scaled with its power. In this work a conservative estimate of fuel cell system mass (including reactor and auxiliary systems) of 10 kg/kW has been used, though modern fuel cells, particularly for automotive application may be as light as 2 kg/kWh (DOE Research, 2011). The water produced by the fuel cell reaction (Eq. 2) may be recycled back to the aluminum-water reactor, reducing by half the overall need for external water supply. One may assume that the design of fuel cell electricity generation systems should take it into account. Therefore, the specific energy obtained by using stoichiometric amount of water accounting for the recycled water from the fuel cell is presented in Table 5, whereas the values corresponding to the aluminum-water reaction without recycling the water produced by the fuel cell are given in parenthesis. The latter may be applicable to small, batch type, short operating time systems. Note that the energy density (in kWh/liter) has somewhat higher value compared to the specific energy (in kWh/kg), taking into account that water density is 1 kg/liter and volumetric aluminum powder density is approximately 1.4 kg/liter.

When comparing to electric energy storage by batteries, the present method reveals prominent superiority, particularly for long operating time of stationary power generators as well as marine and underwater vehicles. The specific electric energy is presented in Figure 5 as a function of the operation duration and application. It can be seen that this technology has clear advantages in long duration applications, such as electricity supply for remote communication posts and long duration UUV's (unmanned underwater vehicles) and UAV's (unmanned aerial vehicles).

4. Conclusions

Hydrogen and electric energy production and storage by on-demand, in-situ hydrogen generation via the reaction between powdered activated aluminum and water, further supplied to a hydrogen-oxygen (PEM) fuel cell for non-polluting electricity generation, have been analyzed and tested. Original patented activation process employing a small (1-2.5%) fraction of lithium-based activator, has been proven to be an effective means for reacting aluminum spontaneously with water of any type (including sea water) at room temperature, producing hydrogen at a high rate and a high yield exceeding 90%. The 11wt% of hydrogen produced compared to the aluminum weight implies generation of specific electric energy of about 2200 Wh/kg Al via a hydrogen-oxygen fuel cell. Considering different operating scenarios it is shown that depending on the specific application and operating duration, the overall specific electric energy of the aluminum-water system may be noticeably superior to that of primary and secondary batteries. Stationary and marine applications where water is available from the surrounding and does not have to be carried along, present the highest overall specific energy. All missions will show better overall specific energy for extended operating times, since the weight fraction of the reactant (Al, water) will play a more dominant role in the overall system weight (which includes the fix weight of the fuel cell, and auxiliary systems).

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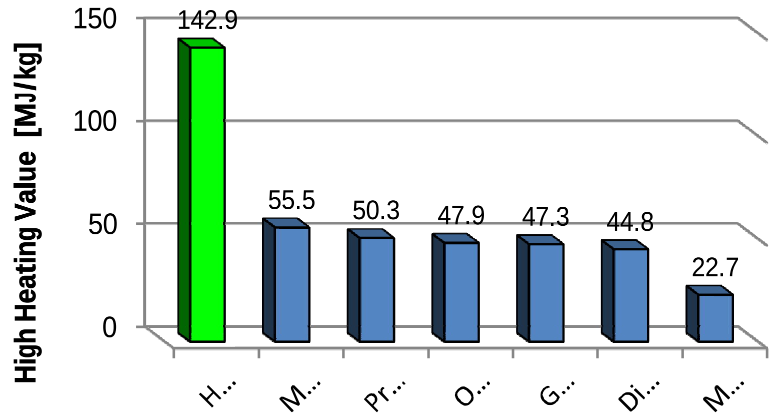

Figure 1: Reaction Energy (high Heating Value, HHV) of Hydrogen Compared to Different Fuels

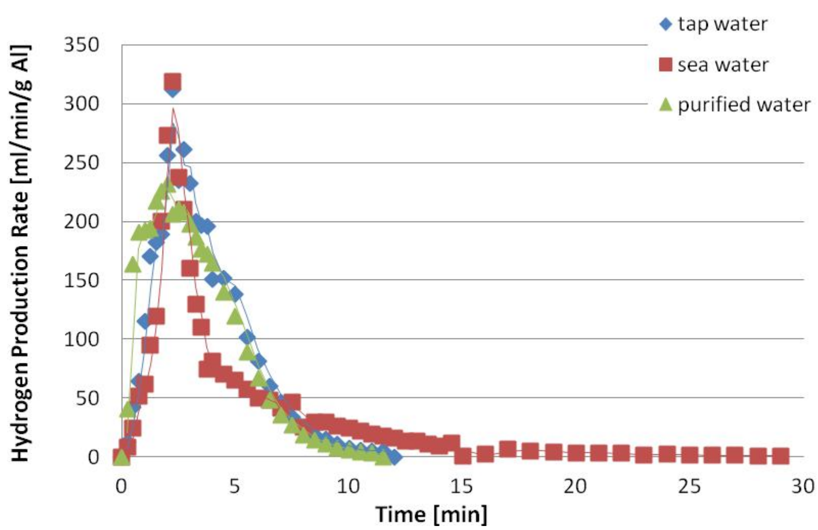

Figure 2: Hydrogen Production Rate vs. time for Different Types of Water

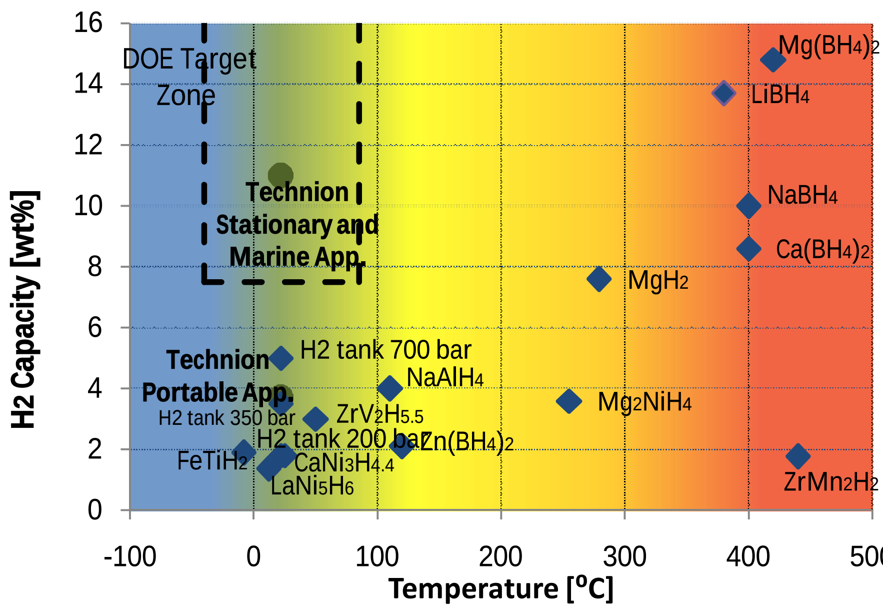

Figure 3: A Map Describing Hydrogen Storage Gravimetric Capacity and Temperature of Hydrogen Release for Different Hydrogen Storage Materials and Methods

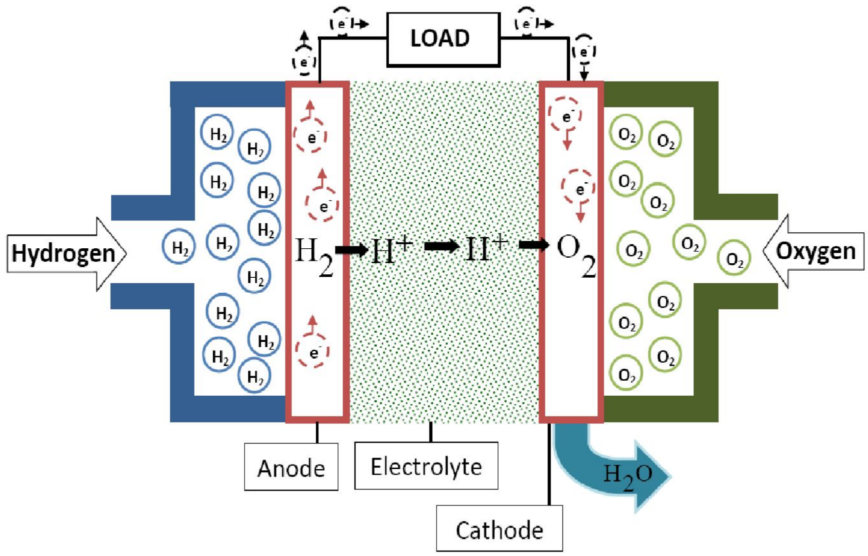

Figure 4: PEM Fuel Cell Employing Hydrogen and Oxygen

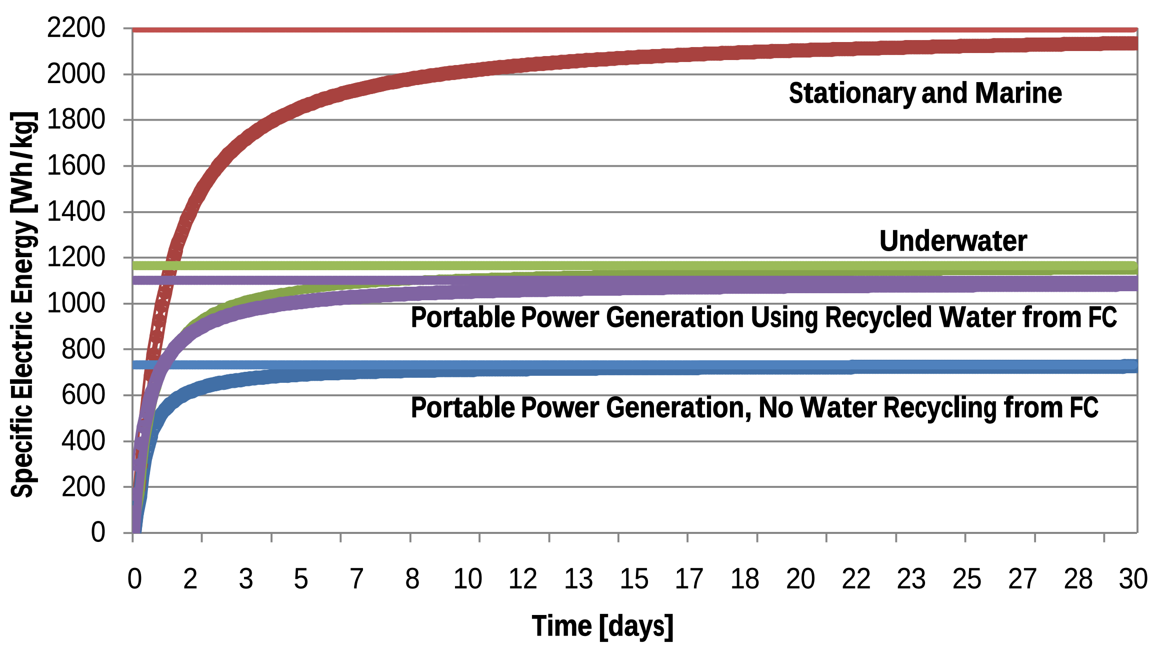

Figure 5: The specific electric energy of the current aluminum-water hydrogen producing reaction combined with a PEM fuel cell vs. time for different applications. The dashed lines present the asymptotic values for very long operation times

Table 1: Pure Hydrogen Approximate Density and Typical Wt% for Different Liquid or Gas Storage Alternatives

Storage Type	**Hydrogen Density [kg/m^3]**	**%Weight**
Liquid hydrogen	71	5-9
Metal tank 200 bar	15	1.6-2
Metal tank 350 bar	25	3-5
Metal tank 700 bar	40	4-6

Table 2: Characteristics and Hydrogen Capacity of Some of the Most Studied Hydrides (Chen Et Al., 2008; Principi Et Al., 2009)

Metal/Alloy	Hydride	Hydrogen Capacity (wt%)	T for 1 bar (°C)
$LaNi_5$	$LaNi_5H_6$	1.37	12
$CaNi_3$	$CaNi_3H_{4.4}$	1.8	25
FeTi	$FeTiH_2$	1.89	-8
ZrV_2	$ZrV_2H_{5.5}$	3	50
Mg_2Ni	Mg_2NiH_4	3.59	255
$ZrMn_2$	$ZrMn_2H_2$	1.77	440
Mg	MgH_2	7.60	279

Table 3: Specific Energy and Characteristics of Selected Primary (Non-Rechargeable) Batteries (Adapted from Aifantis Et Al., 2010; Linden & Reddy, 2002)

Battery Type Cathode/Anode	Specific Energy [Wh/kg]	Characteristics	Applications
Mercad (Cd/HgO)	45	Expensive. Good low and high temperature performance. Low energy density.	Applications that require operations under extreme temperature conditions and long life.
Zink/Carbon	65	Low cost. Common. Low energy density. Available in various sizes and shapes. Poor low temperature performance. Poor shelf life (1.5 years).	Flashlights, portable radios, toys.
Alkaline ($Zn/Alk/MnO_2$)	95-145	Moderate cost. Good low temperature and high rate performance.	Most popular for general purposes: used in various portable applications.
Magnesium (Mg/MnO_2)	105	High capacity. Long shelf life.	Military receiver-transmitters, aircraft emergency transmitters.
Mercury (Zn/HgO)	105	Expensive. High energy density. Flat discharge. Good shelf life.	Hearing aids, medical devices (pacemakers), photography, detectors, military equipment (limited use due to hazard of mercury).
Silver/Zinc Zn/Ag_2O	130	Expensive. High energy density. Flat discharge. Good shelf life.	Hearing aids, photography, electric watches, missiles, underwater and space applications (large size).
Li/MnO_2	200-230	Low cost. Small. Low drain application. High energy density. High specific energy. Wide operating temperature range.	Memory applications, watches, calculators, cameras, toys.
Li/SO_2	260-280	High cost. Pressurized system. Excellent low temperature performance. High energy density. Long shelf life.	Military, industrial, and space applications.
Zn/Air (Zn/O_2)	290	Low cost. Limited power output. High energy density. Long shelf life-sealed. Short activated life.	Hearing aids, pagers, medical devices, portable electronics.
$Li/SOCl_2$	300	Low rate. High energy density.	Long operation applications. Memory backup, military applications, emergency back-up power source.

Table 4: Specific Energy and Characteristics of Selected Secondary (Rechargeable) Batteries

Type	Specific Energy [Wh/kg]	Characteristics	Applications
Lead-acid	35-40	Popular. Low cost. Low energy density. Moderate specific energy. Available in various sizes and shapes.	Automotive, golf carts, lawn mowers, tractors, aircraft, marine, industrial trucks, portable electronics, emergency power
NiMH	75-80	Maintenance free.	Portable electronics. Electric and hybrid vehicles.
NiCd	35-55	High cost. Good low temperature and high rate performance. Excellent life cycle. Maintenance free. Environmental not friendly - strongly regulated.	Biomedical, aircraft.
Li-ion, Li-Polymer	120-200	Moderate cost. High specific energy. High energy density. Low life cycle. Long shelf life.	Portable electronic equipment, electric vehicles, space applications.

Table 5: The Specific Electric Energy of the Current Aluminum-Water Hydrogen Producing Reaction Combined with a PEM Fuel Cell

Application, Mission	Specific Energy [Wh/kg]				Remarks
	Without Fuel Cell Mass(1)	With Fuel Cell Mass(2) Recycling Water from the Fuel Cell(3) (Stoichiometric Al-Water Reaction) Operating Time			
		1 hr	10 hr	100 hr	
Stationary power generation	2200 (2200)	96 (96)	688 (688)	1803 (1803)	Al mass only. Water and air available
Portable power generation	1100 (733)	92 (88)	524 (423)	991 (683)	Al and water mass. Ambient air
Automotive	1100 (733)	92 (88)	524 (423)	991 (683)	Al and water mass. Ambient air
Aeronautical	1100 (733)	92 (88)	524 (423)	991 (683)	Al and water mass. Ambient air
Marine surface vessels	2200 (2200)	96 (96)	688 (688)	1803 (1803)	Al mass only. Water and air available
Marine underwater vehicles	1165 (1165)	92 (92)	538 (538)	1043 (1043)	Al and oxygen mass. Water available

(1) Al mass 0.455 kg/kWh.
(2) Estimated system mass of fuel cell, reactor, and auxiliary equipment 10 kg/kW.
(3) Water recycled from the fuel cell reduces the required amount of water supply by half vs. the stoichiometric Al-water reaction.