2003

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Abstract

Hydrogen energy offers great promise as an energy alternative. Hydrogen technologies can reduce and eliminate the release of carbon dioxide from fossil-fuel combustion, the main cause of global warming. One of the main challenges is hydrogen storage. Storing hydrogen in the solid-state hydride form holds a volumetric advantage over compressed and liquid hydrogen states. Solid hydrogen storage systems also have features of low-pressure operation, compactness, safety, tailorable delivery pressure, excellent absorption /desorption kinetics, modular design for easy scalability, and long cycle life.

In this paper, solid hydrogen storage systems (such as portable power canisters, lightweight fiber wrapped vessels, and aluminum tubular vessels, developed by Texaco Ovonic Hydrogen Systems LLC) will be discussed. A system of four canisters each storing approximately 80 grams of reversible hydrogen is shown to run a 1 kW PEM fuel cell for more than 247 minutes at full power. Canisters show no plastic deformation after more than 500 charge/discharge cycles. The measured strain on canister surfaces indicates that DOT stress limits are not exceeded. The canisters are in the early commercialization stage for uninterrupted power supply (UPS) and auxiliary power unit (APU) applications.

A lightweight fiber-wrapped vessel engineered with metal hydride and internal heat exchanger is being developed for onboard applications. At the system level, the vessel has a volumetric energy density of 50 grams of hydrogen per liter and a gravimetric density of 1.6 wt.%. The vessel is capable of storing 3 kg of hydrogen with a fast refueling capability. Ninety percent of the storable hydrogen can be refueled in 10 minutes at 1500 psig. The vessel can easily release the hydrogen at a rate of 350 slpm at 70^{o}C.

Aluminum tubular vessels are being designed and tested for bulk storage and infrastructure applications including stationary power, hydrogen shipment and hydrogen service stations. The tubular vessel dimensions may be designed for specific applications. For example, a tubular vessel 6 inches in diameter and 62 inches in length can store up to 1 kg of hydrogen.

Introduction

Fossil fuel-generated chemical energy is necessary for daily activities nearly everywhere on earth. Gasoline and heavy oil propel cars, trains, ships and airplanes. Natural gas and coal run industrial plants and provide heat for homes, offices, hospitals and schools. Over the course of the next fifty years, the use of fossil fuel will greatly diminish because of the environmental concerns, energy security awareness as well as the finite resources of the supply [1]. Fortunately hydrogen can provide the energy necessary for activities listed at the outset and do so without polluting. The use of hydrogen as a universal "energy carrier" will be an enormous benefit to society and to the environment of the whole earth [2-4].

The first hydride storage unit was introduced in 1976 by the Billings Energy Co. [5]. The unit was filled with TiFe or Ti(Fe,Mn) hydride alloy, had no internal heat exchanger and had a hydrogen storage capacity of 2500 STL. Subsequently, several other companies [6-10] began marketing their small storage containers with storage capacities between 28 to 2550 STL. Only a limited number of units have been made in the last 25 years because of relatively weak demand.

In 1974, Brookhaven National Lab built the first prototype large stationary hydride storage unit for a New Jersey Public Service Electric and Gas electric peak shaving experiment [11,12]. The system had a total weight of 564 kg of which 400 kg was Ti(Fe,Mn) type hydride alloy (1.6 wt.%). The system storage capacity was 6.4kg H_2. Since 1974, several additional large stationary storage units have been built [13], demonstrating the technical viability of large-scale H_2 storage systems.

Metal hydride onboard storage received much attention in the 70s and early 80s [14]. K.C. Hoffman presented the first SAE paper with the concept of using metal hydrides as a fuel source for vehicle propulsion in 1969 [15]. Many design challenges remained unresolved, and more development appeared to be needed to achieve practicality [16,17]. The quest of using hydrogen as a transportation fuel and the research required developing a better metal hydride onboard system declined and reached a low in the early 80s.

Upon entering into the 21st century, the urgency of energy security and the awareness of climate change resulted in the re-initiation of national and worldwide hydrogen programs [18]. In 2000, Texaco Ovonic Hydrogen Systems LLC, a 50-50 joint venture between Energy Conversion Devices, Inc. (ECD Ovonics) and a unit of ChevronTexaco Corporation, was formed to develop and advance solid-state metal hydride hydrogen storage systems for portable power, stationary and onboard applications. Some of the recent developments are discussed in this paper.

Metal Hydride Alloy Selection

Hydrogen storage material is a critical component of solid hydrogen storage systems. After more than twenty years of research and development in metal hydride alloys, ECD Ovonics has gained an advantage in the design and manufacture of the low temperature multi-element intermetallic metal hydride (LTHM) alloys [19-21].

The intrinsic properties of LTMH alloys prepared in the lab are routinely characterized. The quantity of each batch is typically 10-20 grams of alloy. To successfully extrapolate the results from small batches to large scale production, it is necessary that the alloy from a large production run deliver the same properties as that of the small scale laboratory alloy. Ovonic Battery Company (OBC) has produced large scale alloy quantities for NiMH battery for many years. The LTMH alloys used in the various storage vessels discussed in this paper are supplied by OBC. All raw materials used in the OBC production runs are low grade scrapes available in the open market.

One of the favorable features of LTMH alloys is the tailorability of the equilibrium plateau pressure. Figure 1 shows an example of the pressure-temperature relationship of three Ovonic LTMH alloys. These alloys have a heat of formation in the range of 24-30 kJ/mole H_2. The criteria that are used to select an appropriate alloy for a particular application are 1) type of waste heat available, 2) required delivery pressure, and 3) allowed maximum charging pressure.

In one of the UPS systems presently under development, portable power canisters are integrated with an air-cooled fuel cell. The canisters designed for this application contain only a passive heat exchange system. Waste heat from the fuel cell stacks, in the form of air heated to between 25°C and 60°C (depending on fuel cell power level), is used to assist the discharge of H_2 from the canisters. Canisters are required to deliver at least 10 psig pressure at 5°C and the maximum pressure must be less than 250 psig at 30°C. OV694 is ruled out for the application because it cannot sustain the required deliverable flow rate at 5°C due to the lack of heat transferred by hot air from the fuel cell stack to the canister. OV610 has too high of a plateau pressure and this alloy cannot be charged with 250 psig at 25°C. OV679 alloy can meet all the requirements for the portable power canister application.

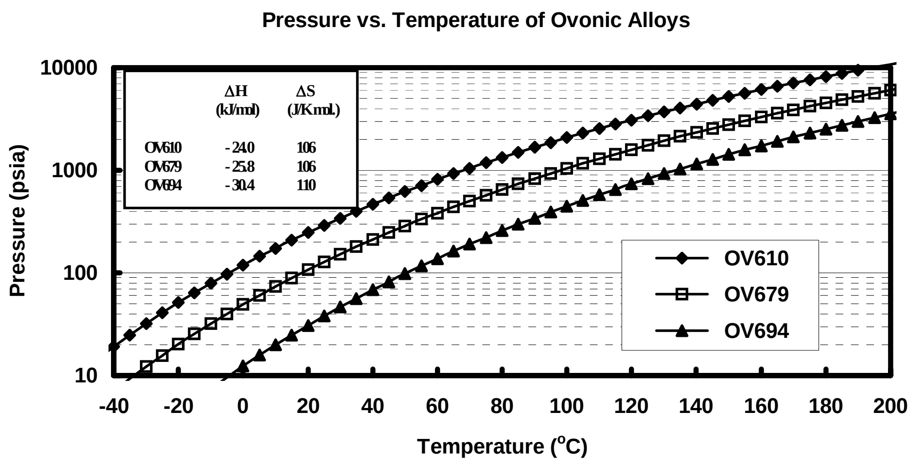

Figure 1. Pressure – temperature relationship of three Ovonic LTMH alloys.

The metal hydride storage vessel can integrate either with an internal combustion engine (ICE) or a PEM fuel cell for onboard vehicle application. There are two types of waste heat available from an ICE vehicle, the high temperature exhaust heat and the heat from engine cooling loop. For comparison, a PEM fuel cell vehicle provides heat only from the fuel cell cooling loop. The temperatures of the cooling loop from the ICE engine or the fuel cell stack are similar, in a range of 50 - 85°C. To be versatile, the onboard vessel is designed to utilize the heat only from the engine cooling loop coupling with the vessel's internal heat exchanger [22,23]. Due to the cold start demand, in the present case, a relatively high plateau pressure alloy, such as OV610, is more suitable for the onboard hydride storage vessel application.

Onsite hydrogen generation by an electrolyzer combined with the hydrogen bulk storage system is an attractive hydrogen stationary application. Typical deliverable pressure from an electrolyzer is in the range of 150-200 psig. OV694 is specifically designed to meet the low pressure charging for a bulk hydrogen storage system.

The reversible hydrogen capacity is normally estimated from the PCT isotherm curves. A PCT measurement requires approximately 10 grams or less of alloy. A TOHS' designed alloy screening vessel with an active heat exchanger is also used to measure the reversible hydrogen capacity of the alloys. The amount of alloy loaded into the alloy screening vessel is 2.37 kg. The coolant temperature is set at 8°C during charging and desorption is performed by releasing the hydrogen through a flow meter with the rate of approximately 1 to 2 slpm while the heat exchange liquid stays at 85°C. Figure 2 summarizes the reversible capacity of the above three alloys as a function of charging pressure.

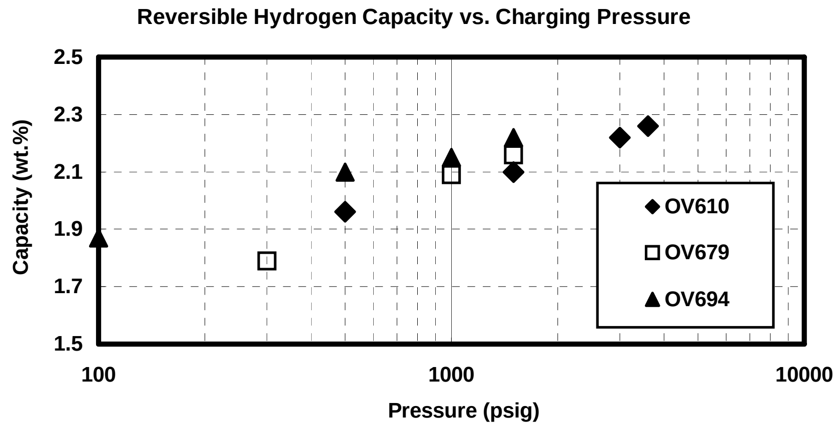

Figure 2. Reversible H_2 capacity vs. charging pressure of Ovonic LTMH alloys.

Portable Power Canister

The range of portable power applications varies from less than one watt (consumer electronic products) to several kilowatts (such as for UPS and backup power generators). Figure 3 is a diagram indicating portable power application categories for different power requirements. To satisfy the diverse power requirements of the portable market, TOHS strategy is to develop a family of portable canisters designed to fit identified market requirements. The initial product to be produced and marketed is a solid hydrogen storage canister for a UPS application. Figure 4 shows an example of a portable power canister (TOHS model number 85G250) that has been designed and developed for a UPS application.

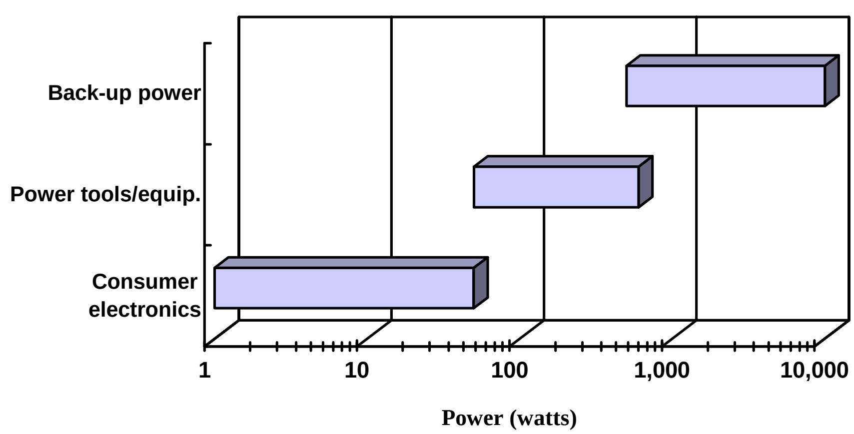

Figure 3. Diagram of portable power application category as a function of power requirement range.

The product has gone through extensive internal and external testing and shown to be in compliance with DOT issued exemption requirements, as well as other industry standards. For example, the canister (TOHS model number 85G250) has received more than 500 hydrogen absorption /desorption cycles without showing any visual sign of plastic deformation.

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- Size: 3.5” diameter x 16.6” length
- Reversible capacity: 74-80 g depending on air flow rate/temperature
- Weight: 6.5 kg (14.5 lbs)
- 250 psig max pressure at 25 ^oC
- Staubli RBE03 quick-connect interface

Figure 4. Example of a portable power canister for a UPS application.

Figure 5 compares strain data for respective canisters at 5 and 503 cycles. Charging conditions included 250 psig H_2 pressure in a 25°C liquid bath for 3 hours while hoop strain was measured on the canister surface in the maximum absorbed condition. Following absorption, the canisters were desorbed at 50°C for a minimum of one hour. The data shows that the highest strain values reached at the end of absorption are 1300 με and 1800 με for the canisters at 5 and 503 cycles, respectively. The maximum value measured for canister cycled more than 500 cycles is below the maximum allowable (1854 με) according to DOT limits of for a pressure vessel with a minimum wall thickness of 0.145 inches constructed of 6061-T6 aluminum alloy. Furthermore, the test result from a DOT recognized independent agency confirmed that there was no change of burst pressure (>4500 psig) indicating no reduction of canister strength.

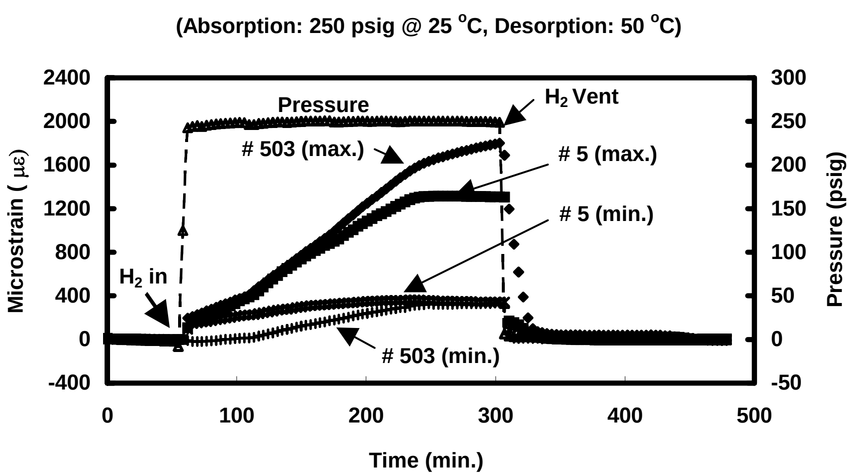

Figure 5. Microstrain data during absorptions for canisters on cycles #5 and #503.

Portable Power System

Portable canisters may be used individually or assembled together. The single or multiple canisters can be connected to other components (i.e. fuel cells) to create storage systems and power systems. Storage systems can be pre-assembled or manifolded for field installation and exchange of individual canisters.

Figure 6 shows a rack mount storage system with a set of four canisters manifolded together. Following a trend in fuel cell integration, the rack mount system was designed to demonstrate that hydrogen could be safely and efficiently stored in the same equipment rack used to mount the fuel cell and the electronic load being powered.

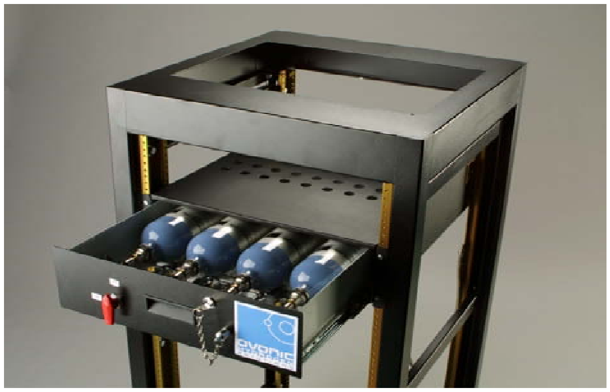

Figure 6. Rack mount storage system using four TOHS 85G250 canisters.

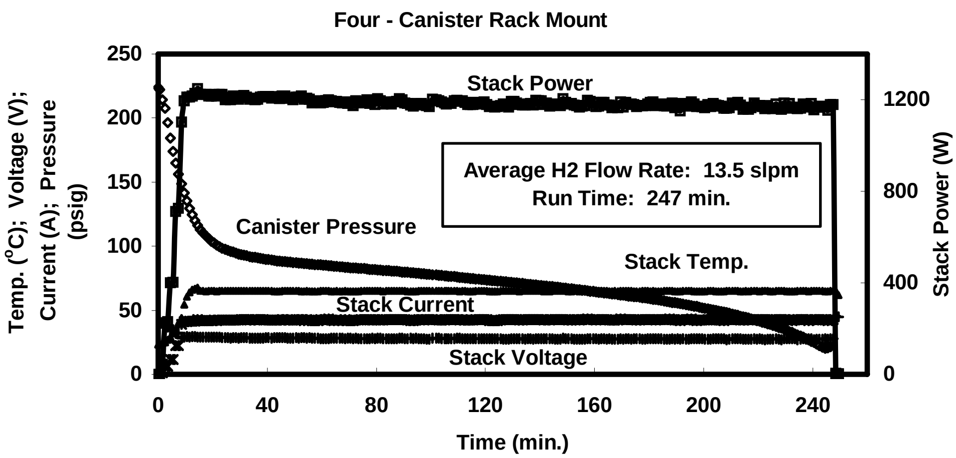

Figure 7. Typical experimental data from a four-canister rack mount solid hydrogen storage system running a Ballard Nexa 1kW fuel cell at close to full power.

At TOHS laboratories in Rochester Hills, MI, experiments have been performed using the type of rack mount system shown in fig. 6. The initial storage of hydrogen in the canisters involved charging at 250 psig at 25°C for three hours in a liquid bath. The canisters provided hydrogen to a Ballard Nexa 1 kW fuel cell unit for a series of experiments. Figure 7 shows representative data of a rack mount system running a 1kW fuel cell at its full power. The run time was 247 minutes with the fuel cell stack power at 1.20 kW. The fuel cell stack voltage and current ranges are 28.1V ± 0.9V and 42.1A ± 0.8A respectively. The canister pressure is shown to quickly drop from the initial 225 psig to less than 100 psig in 20 min. The average H_2 desorption rate was 13.5 slpm from a set of four canisters. During the continuous 247 minutes running, temperatures inside the fuel cell stack and on the canister surface were 65 ^oC and 41 ^oC, respectively.

Lightweight Onboard Vessel

TOHS has developed a lightweight carbon fiber-wrapped vessel for onboard application. The vessel is incorporated with a proprietary heat exchanger and liquid tube manifold [22]. Figure 8 shows the Ovonic prototype onboard vessel. At opposite ends are hydrogen solenoid valve and liquid heat exchanger port. Vessel dimensions include 33 inch (84 cm) length and 13 inch (32.8 cm) outer diameter. The total vessel weight including alloy is 190 kg and the internal volume is 50 liters. The fiber wrapped outer shell was provided by Dynetek (Calgary). A prototype was hydrostatically cycled between 290 and 4500 psig pressure for 15,000 cycles without leakage and then hydrostatically burst. The burst pressure of 13,801 psig is above the min. required burst pressure of 10, 800 psi or 3 times the 3600 psig service pressure.

Figure 8. Ovonic prototype lightweight onboard vessel.

Table 1 is a comparison of the storage capacity and weight of the metal hydride and compressed hydrogen using the same fiber vessel (an internal volume 50 liters).

Table 1

	Metal Hydride @ 1,500 psig	Compressed H_2 @ 3,600 psig
H2 Capacity (kg)	3.0	0.88
Total weight (kg)	190	25.2
Weight %	1.58	3.49
Volume (g / liter)	60	17.6

Several vessels have been instrumented with ten strain gages and eight thermocouples. The strain gages are mounted on the aluminum liner underneath the carbon composite outer wrapping. Thermocouples were placed at coolant inlet and coolant outlet locations, at locations embedded in the metal hydride alloy and on the aluminum liner surface. The instrumented vessel was subjected to absorption/desorption performance testing at the ChevronTexaco Richmond Technology Center (CRTC) in Richmond, CA.

Figure 9 represents a hydrogen absorption experiment preformed on the vessel. The figure shows hydrogen delivery pressure (triangles), absorption capacity (squares), metal hydride bed temperature (TC1) and aluminum liner surface temperature (TC6) each as functions of absoption time. The flow rate of cooling liquid though the internal heat exchanger is about 20 gallons per minute with the coolant temperature maintained at 8°C. The hydrogen delivery pressure started at 1000 psig and then gradually ramped up to 1700 psig for a total of 20 min. charging time. Total storage capacity was 3 kg of which 2.7 kg (90% of the capacity) was charged in 10 min.

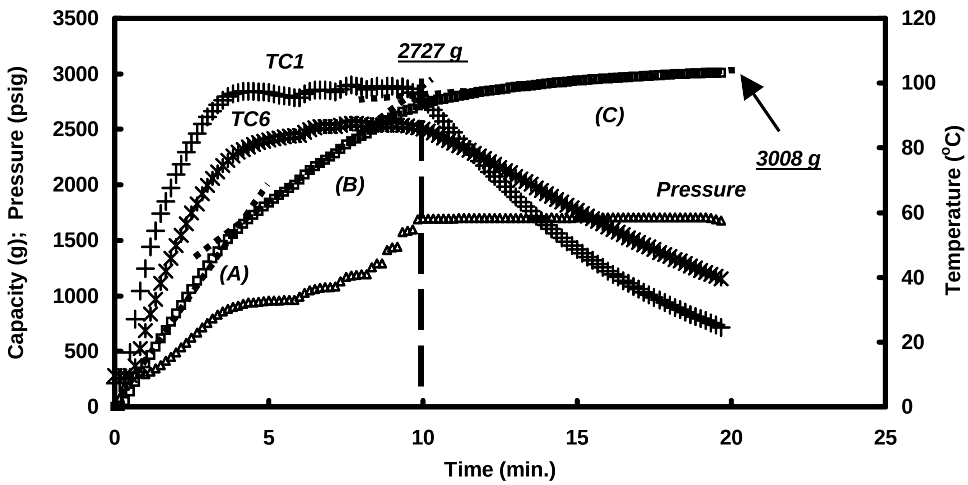

Figure 9. H_2 absorption experiment recorded from an Ovonic lightweight onboard vessel.

The capacity curve indicates that the refilling process involves three stages, marked as (A), (B) and (C). The first stage (A) is characterized by the rapid H_2 uptake, in which 1.5kg of H_2 is absorbed in the first 3 min. The fast charge capability demonstrated in the first stage of charging is mainly due to the high heat capacity of the MH alloy. The flow rate is governed by the difference between the H_2 delivery pressure and the pressure in the metal hydride bed. The rapid reaction rate in this stage is accompanied by a rapid rise in both temperature and pressure.

During the second stage, an additional 1.2 kg of H_2 is stored in the next seven minutes. The H_2 delivery pressure is gradually ramped up while the temperatures of the MH bed (TC1) and on the aluminum liner (TC6) are kept fairly constant so that the heat generation and heat removal rate remain in balance. In this stage, the hydride reaction is determined by the effectiveness of the internal heat exchanger. 2.7 kg of hydrogen is absorbed at the end of second stage in 10 min.

The third stage (C) is the end of the process where the vessel pressure approaches the delivery pressure. As a result, the hydrogen flow rate and the hydride reaction rate are dramatically decreased. The vessel temperatures begin to fall due to the slow reaction. Additional 300 g of hydrogen is charged to the MH in 10 min.

Figure 10 illustrates the relationship between the strain and the capacity. The data shows that in the first 10 minutes of absorption, at 90% of the total capacity, 2.7 kg, the strain is 1200 $\mu\varepsilon$ that is 33% of the allowable strain. The figure shows that the strain rapidly increases as hydrogen is further absorbed into the vessel. The strain value is more than doubles for the last 0.3 kg hydrogen. Nevertheless, the maximum strain of 3000 $\mu\varepsilon$ measured at the completion of charging remains below the maximum allowable working limit of 3600 $\mu\varepsilon$.

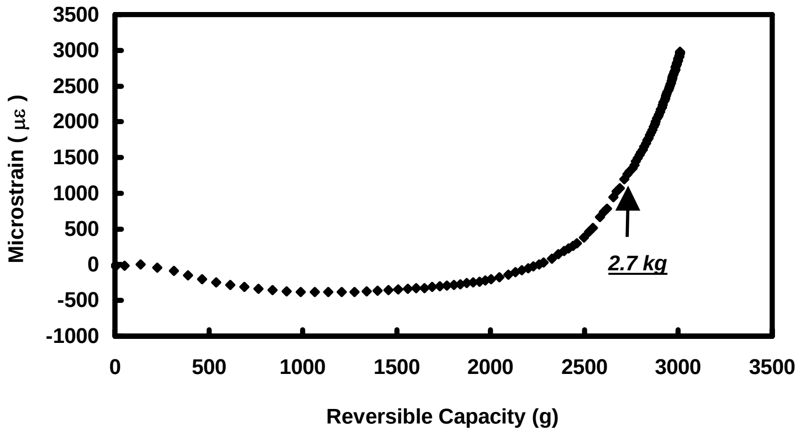

Figure 10. Microstrain as a function of reversible capacity during absorption.

Figure 11 is a typical desorption experiment, the data including hydrogen flow rate, desorption capacity, internal vessel pressure, liquid inlet temperature and metal hydride bed temperature as functions of discharge time. The liquid temperature was maintained at 75°C during the entire desorption experiment. The desorption flow rate is set at 350 slpm (31.25 g/min.) for approximately 90 minutes equivalent to 2.8 kg hydrogen. For hydrogen ICE vehicle, a hydrogen range of 17-300 slpm at 30 psi pressure is required. In this case, the vehicle can continue run until the vessel is almost empty.

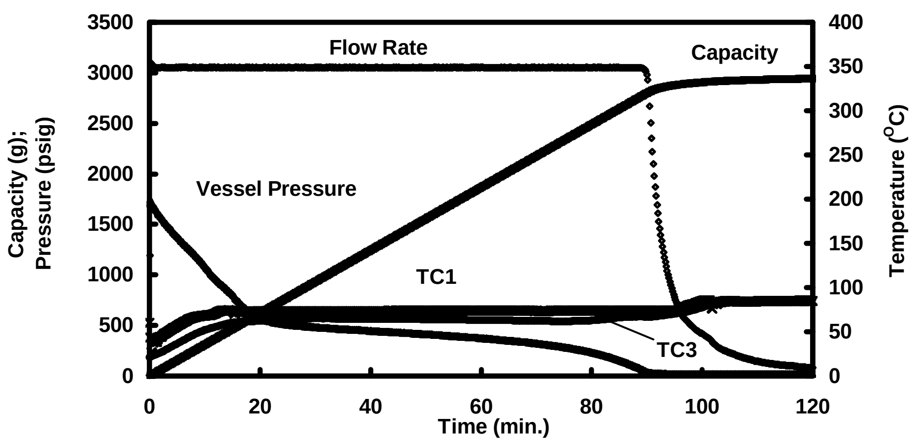

Figure 11. Desorption data recorded from an Ovonic lightweight onboard vessel.

One of the advantages of hydrogen storage using metal hydrides is the low pressure operation. As shown in fig. 11, even though the filling pressure is about 1500-1700 psig, the vessel pressure quickly decreases from the initial charging pressure to below 500 psig within 20 minutes and continuously operates at the pressure below 500 psig for most of the time. In contrast, the pressure a compressed hydrogen vessel either 3600 or 5000 psig, would remain above 500 psig until close to empty.

Bulk Storage System for Stationary Power Application

TOHS has developed a bulk hydrogen storage system with a modular approach. The basis unit is an aluminum tubular vessel that can be fabricated within a reasonable range of diameter and length dimensions, to enable custom geometries and performance requirements. TOHS pressure vessels are designed according to ASME requirements and incorporate a proprietary internal heat exchanger and liquid tube manifold. An example of an aluminum tubular vessel is shown in fig. 12, showing a vessel with 6 inches outer diameter and 62 inches in length. The figure shows hydrogen solenoid valve and liquid heat exchanger ports at opposite ends of the vessel.

Figure 12. Ovonic aluminum tubular vessel.

An example of aborption experiment was illustrated in fig. 13. The charging pressure was kept at 130 psig to simulate the delivery pressure of an electrolyzer. The figure shows H_2 delivery pressure, H_2 flow rate, absorption capacity, and temperatures of aluminum vessel and coolant inlet. The coolant inlet temperature was set at 10 ^oC. Total hydrogen absorption capacity was 1.02 kg after 90 min charging.

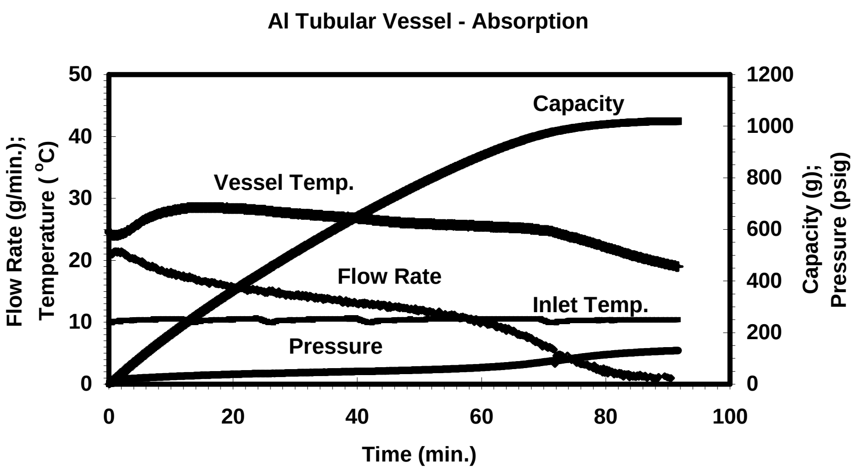

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Figure 13. H_2 absorption recorded from an Ovonic aluminum tubular vessel.

TOHS has designed and constructed a prototype stand-alone bulk hydrogen storage system, shown in fig. 14. The system includes ten tubular vessels (each 6” OD x 62” L). Each vessel can store 1 kg H_2 and has its own thermal management components, UPS and PLC. The system absorbs and stores hydrogen at low pressure directly from an electrolyzer or from a reformer/PSA and provides fuel to a fuel cell or ICE unit for back-up power unit. The thermal management system is designed to utilize waste heat from a fuel cell/ICE or a fuel processor for desorption and remove heat from a radiator during absorption. The valves, flow meters, fan and liquid pump are automatically controlled and monitored through the PLC system. The system is also equipped with a UPS system for start-up in the power outage condition. The system is designed for outdoor usage. However, the initial operation and testing was performed in the testing lab at Rochester Hills facility. Hydrogen source comes from an onsite electrolyzer (manufactured by Vandenborre Technologies Co.), which has a hydrogen generation rate of 1 kg/hr at 130 psig with a dew_point of -80 ^oC or a water level of < 1ppm.

Figure 14. An Ovonic prototype of 10 kg bulk storage system.

Figure 15 shows the absorption data recorded from the prototype 10 kg bulk storage system taking the hydrogen directly from the onsite electrolyzer. The straight line of H_2 capacity versus time in the figure indicates the system has an ability of absorbing hydrogen at a constant hydrogen flow rate of 160 slpm generated by the electrolyzer. It took about 800 minutes to fill the 10 vessels to 10 kg H_2. During the charging, the vessel pressure increased monotonously and finally reached to the set point of the electrolyzer. The kink in the pressure curve is due to the fact that an additional coolant was circulated in the cooling loop to remove the heat from the vessel more efficiently. Since the system was originally designed for outdoor usage as such a radiator provides the cooling. However, the system is now operated in a closed testing lab, the heat released through the radiator was not able to dissipate effectively, which caused the room as well as the coolant temperature steadily increasing. When the coolant temperature and room temperature reached equilibrium, the heat removal rate was approaching zero. At this point, an additional coolant was added into the coolant loop from the heating loop, which was about 20 ^{o}C and circulated to outside the testing lab. The dew_point was - 66 ^{o}C or < 5ppm water level at the beginning of absorption and leveled off at -80 ^{o}C or <1ppm until the completion of absorption.

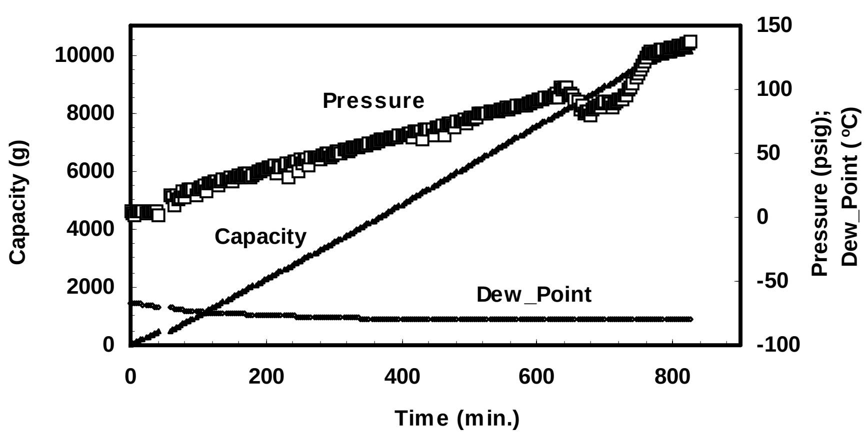

Figure 15 Refill the hydrogen to a prototype 10 kg bulk storage system from an electroyler

Conclusion

TOHS has advanced hydrogen storage technology from small scale units in the lab to prototype size systems in the real world. The portable power canister is ready for initial commercialization for UPS and APU applications. The basic design concept has demonstrated the feasibility of prototype vessels for onboard and stationary power applications. Long term performance and reliability of the vessels are currently under the life cycle testing. In a mean time, TOHS continues to advance today's technology for hydrogen storage systems. Many challenges remain, including increasing the gravimetric H_2 capacity in the hydride families. In time, the major technical challenges will be overcome.

Acknowledgments

We acknowledge Larry Veverka of ChevonTexaco countless time to set up the testing lab at the CRTC facility, Ming Wang and John Slot for onboard and tubular vessels testing, Jeff Schmidt for the10 kg storage system performance testing, V. Wang, K, Tessmar, J. Lewis and R. Colenso for portable power canister life cycle testing, Alexander Gerasmimov, Nester Kropelnyckyj, and Isaak Fidel for vessel design and assembly.

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