Research and development for a metal hydride tank with double coil type heat exchanger below 1.0 MPa (G) operation

Akihiro Nakano ^{a,*}, Hiroshi Ito^a, Satya Shekhar Bhogilla^a, Theodore Motyka^b, Claudio Corgnale^b, Scott Greenway^c, Bjørn C. Hauback^d

^a National Institute of Advanced Industrial Science and Technology (AIST),
Energy Technology Research Institute,
1-2-1 Namiki, Tsukuba East, Tsukuba, Ibaraki 305-8564, Japan

^b Savannah River National Laboratory (SRNL), Energy Security Department,
999-2W, Room 124, Savannah River Site, Aiken SC 29808, U.S.A.

^c Greenway Energy LLC, 302 Gateway Drive, Aiken SC 29803, U.S.A

^d Institute for Energy Technology (IFE), Physics Department, P.O.Box40, NO-2027
Kjeller, Norway

*Corresponding author. Tel: +81-29-861-7250, Fax: +81-29-851-7523

E-mail address: a.nakano@aist.go.jp

Keywords: Hydrogen storage, Metal hydride, Reaction heat, Renewable energy,
Hydrogen system

Abstract

A metal hydride tank has been developed with the aim of recovering the reaction heat of a metal hydride for the Totalized Hydrogen Energy Utilization System application. The metal hydride tank, which had the same geometrical configuration as one previously evaluated at SRNL, was fabricated with a different composition of the metal hydride alloy for operation below 1.0 MPa (Gauge). The hydrogen mass flow data from hydrogen production by renewable energy (solar power) and the fuel cell operation, which were obtained at SRNL, were used for the testing at AIST. The relatively large heat leak from the tank support of the metal hydride tank at SRNL was confirmed, and thus the tank support was replaced in this work. Furthermore, a vacuum thermal insulator was developed and applied to the metal hydride tank. This resulted in overall tank size reduction without reducing the thermal insulation performance.

1. Introduction

The Totalized Hydrogen Energy Utilization System (THEUS) is a stationary hydrogen system for commercial building applications [1, 2]. THEUS is comprised of a Unitized Reversible (or Regenerative) Fuel Cell (URFC) and metal hydride tanks. It was developed for the purpose of load leveling. During the night, this system produces gaseous hydrogen by operating the URFC in a water-electrolysis mode and stores the hydrogen in metal hydride tanks. During the day, the URFC operates in fuel-cell mode by using the stored hydrogen to supply electricity to the building. THEUS can provide not only electricity but also thermal energy and hydrogen to the buildings. THEUS is an energy storage system, and therefore it can store renewable energy in the form of hydrogen. Moreover, we have a plan to apply this system for off-site hydrogen stations where the liquid hydrogen is dispensed [3]. All of the boil-off gas (BOG) generated from a liquid hydrogen tank can be stored in metal hydride tanks and utilized through THEUS. The versatility of hydrogen energy to the industrial, consumer, and transportation sectors can be enhanced through this stationary hydrogen system. The overall goal is to establish a robust hydrogen energy system.

Metal hydride based hydrogen storage is a suitable system for THEUS because it is one of the safest hydrogen storage methods and the reaction heat of the metal hydride alloy can be utilized. A horizontal type metal hydride tank, which had a double coil type heat exchanger, was developed at AIST and tested in the Applied Research Center (ARC) building at SRNL. The experimental results and the fundamental characteristics of the metal hydride tank have already been reported [4-6]. The operation pressure range was approximately from 0.5 to 3 MPa Gauge (G) [7]. However, THEUS has to operate at pressures lower than 1.0 MPa (G) in Japan because of the High Pressure Gas Safety Act. Otherwise, the cost of the system becomes about 5~10 times higher than the baseline case because qualified parts are required, and the system can only be operated in an approved facility. This makes commercialization very difficult. Thus metal hydride tanks should operate below 1.0 MPa (G) in Japan. Several metal hydride tanks that satisfy this requirement have been reported [2, 8, 9] but most reports result for operation above 1.0 MPa (G) [10-14].

The operating conditions of the metal hydride tanks used for THEUS are unique. Hydrogen is absorbed under high temperature and pressure, and desorbed under low temperature and pressure conditions. This is opposite to normal operating conditions for such systems. Furthermore the metal hydride tank of THEUS has to be operated below 1.0 MPa (G) in Japan, and thus relatively challenging operating conditions. During the absorbing process, the exothermic reaction heat needs to be

exhausted from the metal hydride tank to the environment via coolant. Assuming the exhaust heat is emitted by using conventional cooling towers, the coolant temperature from the outlet of the tank should be high enough to exchange heat even in a summer night. On the other hand, the endothermic reaction heat during the desorbing process can be utilized to cool the working fluid in a refrigeration system for a commercial building. Assumptions were made that the cooling tower worked well when the coolant temperature was above 32 °C, and the coolant was supplied to a refrigeration system when the coolant temperature was less than 12 °C. The operation conditions based on these assumptions were maintained in the THEUS operation mode tests. The individual and the continuous absorption–desorption tests were carried out and compared with the experimental results obtained at SRNL. Moreover, by using the mass flow controller, the data for the hydrogen production from renewable energy and the fuel cell at SRNL were reproduced at AIST. The reaction heat recovery rates of the metal hydride tank were investigated in this study, and the corresponding heat exchanging performance is reported. Furthermore, a vacuum thermal insulator for the metal hydride tank was developed and tested. The experimental results are discussed in this paper.

2. Experimental set-up

Fig. 1 shows the schematics of the horizontal type metal hydride tank. The fundamental design is the same as the tank tested at SRNL. The length is 1 m, with the outer diameter and the inner diameter as 165.2 mm and 155.2 mm, respectively. 50kg of MmNi₅ (AB₅) metal hydride alloy was placed in the tank. The grain size is 500 μm. The reaction heat for absorption is 28.934 kJ/mol_{H₂} and that for desorption is 27.867 kJ/mol_{H₂}, respectively. The composition of the alloy is different from the one tested at SRNL because of operation below 1.0 MPa (G). The total weight of the tank with the MmNi₅ hydride was approximately 85 kg. A double coil type heat exchanger, which was made of copper, was adopted for recovering the reaction heat of the metal hydride. Two sheath thermocouples, with one and three measurement points, were inserted in the tank to measure the temperature in each position. The thermocouple with one measurement point was set at 26 mm from the center of the side wall and 10 cm from the outer surface of the side wall (TC-C10). The thermocouple with three measurement points was fixed at 55 mm away from the center of the side wall and the measurement points were 25 cm, 15 cm, and 5 cm from the outer surface of the side wall (TC-O25, TC-O15, and TC-O5), respectively. The pressure in the metal hydride tank was measured by using a pressure transducer (Kyowa PGS-50KA). The safety

valve was set to 1.2 MPa. To reduce cost, a special designed hydrogen induction tube [15] was adopted and a commercial sintered stainless filter with a pore diameter of 0.5 μm was used outside of the tank. The tank was covered with thermal insulation foam (Areoflex) with a thickness of 6 cm to prevent heat loss from the outer surface.

The schematic of the experimental set-up is shown in Fig. 2. The hydrogen was provided from hydrogen cylinders. A Kofloc Model 8550 hydrogen mass flow controller (MFC) was used. The full scale (FS) value is 50 NL/min, and the accuracy of the controller is $\pm 1.5\%$ (FS). The hydrogen flow rate is controlled by a personal computer. When the experimental simulations were carried out, the mass flow data obtained from SRNL were read into the personal computer and the hydrogen flow rate was reproduced by the MFC. The hydrogen piping was plumbed on an aluminum plate. The solid arrows and dashed arrows in Fig. 2 show the hydrogen flow paths for absorption and desorption, respectively. The inlet and the outlet of the double coil type heat exchanger in the metal hydride tank were connected to a thermostat bath (SMC-HRS024). The temperature of the circulation water was maintained by the thermostat bath. The flow rate of the circulation water was measured by a flow meter (Keyence FD-SS2A) and controlled with a needle valve. The accuracy of the mass flow meter was $\pm 4\%$. The circulation water was a solution 85 vol% water and 15 vol% ethylene glycol. A small water pump (see Fig. 2) was installed on the water by-pass line. The circulation water can be flowed without entering the thermostat bath by using this pump. This pump and the by-pass line were used in the experiments for the absorption-desorption continuous test.

3. Experimental results and discussion

3-1. Absorption-desorption tests with constant hydrogen flow rate

THEUS was developed for the purpose of load leveling. The URFC produces gaseous hydrogen by using grid power during night, and the hydrogen is stored in metal hydride tanks. The URFC provides electricity in day time using the stored hydrogen. The hydrogen flow rate between the URFC and the metal hydride tank is constant in each process.

3-1-1. Results for the individual absorption-desorption test

The Pressure-Composition (P-C) isotherms, the temperatures in selected locations in the tank and the inlet temperature and the outlet temperature of the circulation water were assessed to determine the fundamental characteristics of the metal hydride tank. Fig. 3 shows the result of an individual absorption-desorption test

at AIST. The P-C isotherm is presented in Fig. 3a, while Fig. 3b and Fig. 3c show the time variations of the pressure, the temperature in selected positions in the tank and the inlet and outlet temperatures of the circulation water for absorption and desorption, respectively. The composition H/M of 0.18 was selected as the starting point, and hydrogen was absorbed for 9 hours. The hydrogen flow rate was 11.0 NL/min and the circulation water flow rate 1.12 L/min at 32 °C during the absorption process. For the 13 hours desorption step, the hydrogen flow rate was 7.6 NL/min and the circulation water flow rate 0.46 L/min at 12 °C. The solid lines with the circles and the triangles show the equilibrium pressures at 32 °C and 12 °C, respectively. The gray lines shown in Fig. 3b and 3c indicate the time variation of tank pressure for the absorption process and the desorption process. The hydrogen supply was stopped when the concentration, H/M, reached the value 0.94 corresponding to a tank pressure of 1.0 MPa (G). 5920 NL of hydrogen was absorbed during the experiment. The temperature data during the absorption process remained relatively steady, except the temperature data of the TC-O5. The measuring point of TC-O5 was very close to the side wall. It is noted from Fig. 3c that inhomogeneous temperature distribution occurred during the last stage of the desorption process. The temperature differences between the outlet and the inlet of the circulation water were kept at 3 °C and 5 °C during the absorption process and the desorption process, respectively. The recovered thermal energy, Q_{cw} , was calculated by using the following equation:

$$Q_{cw} = c_{pcw} \rho_{cw} \Delta T_{cw} \int \dot{V}_{cw} dt . \quad (1)$$

where c_p is the specific heat of the circulation water at constant pressure, ρ is the density, ΔT_{cw} is the temperature difference between the circulation water outlet and inlet, and $\dot{V}$ is the volumetric flow rate. The subscript cw indicates the circulation water. The recovered thermal energy was 6.84 MJ throughout the absorption process, and 6.62 MJ throughout the desorption process. The metal hydride reaction heat recovery rate, ε , can be obtained from the following equation:

$$\varepsilon = (Q_{cw}/Q_{MHreact}) \times 100, \quad (2)$$

where $Q_{MHreact}$ is the reaction heat of the metal hydride. The value of $Q_{MHreact}$ is given by $Q_{MHreact} = V_{H2} \times \Delta h$ where V_{H2} is the total volume of hydrogen and Δh is the metal hydride reaction heat per unit volume. The metal hydride reaction heat recovery rate was 89 % for absorption and it was 90 % for desorption, respectively. The

accuracy of the reaction heat recovery rate was about $\pm 5\%$, and it depends on the accuracy of the mass flow meter for the circulation water and for the hydrogen. The recovery rates are almost the same as those of the metal hydride tank tested at SRNL [4].

3-1-2. Results for the absorption-desorption continuous test

Fig. 4 shows the results for the absorption-desorption continuous test; the P-C isotherm in Fig 4a and the circulation water outlet temperature in Fig. 4b for the THEUS operation aimed at electric load leveling. The operating condition was a 24 hour cycle. The absorption and desorption process times were 9 and 13 hours, respectively, with an interval of one hour between each process.

The hydrogen flow rates for the absorption and desorption were the same as previously described. The circulation water flow rate and the circulation water inlet temperature control were different. During the first absorption, the temperature control bath supplied water at 32 °C and at a flow rate of 1.12 L/min. During the one hour interval between the absorption and subsequent desorption process, the inlet temperature of the circulation water was at 32 °C and the flow rate at 1.12 L/min to cool the metal hydride. After the one hour interval the hydrogen was desorbed, but the temperature of the metal hydride was initially close to 32 °C (i.e. higher than 12 °C). Until the output temperature of the circulation water reached 12 °C, the outflow of the circulation water was not fed through the temperature control bath, but directly to the inlet. A by-pass approach was adopted using a pump to cool the metal hydride using the reaction heat itself. This self-cooling continued until the outlet temperature decreased down to 12 °C. The flow rate during this self-cooling was arbitrarily set at 1.12 L/min. When the inlet temperature reached 12 °C, the circulation water was fed again into the thermostat bath, and the inlet temperature was maintained at 12 °C. The flow rate of the circulation water was reduced 0.46 L/min. During the one hour interval between the desorption process and subsequent absorption process, the temperature of the metal hydride was still below 12 °C, and cold energy was recovered with the circulation water at a flow rate of 1.12 L/min. The absorption after the preceding desorption and interval required self-heating of the cold metal hydride. Using the by-pass and the pump as in the self-heating, the exothermic reaction heat was used to heat up the metal hydride itself. After the inlet temperature reached 32 °C, the coolant was fed through the temperature control bath. The inlet temperature was maintained at 32 °C independent of the outlet temperature. The flow rate of the circulation water was 1.12 L/min. These processes were continuously repeated for three days. The first

absorption proceeded similarly to the absorption process of the individual absorption-desorption test. After the first absorption, the tank pressure and the circulation water outlet temperature showed the same profiles as indicated in Figs 4a and 4b. In this experiment, the reaction heat recovery rates for absorption processes were 87 % in Day1, 75 % in Day2, and 76 % in Day3. On the other hand, the reaction heat recovery rates for desorption processes were 73 % in Days 1 and 2, 72 % in Day3, respectively. After the first absorption process, the reaction heat recovery rates decreased as compared with the results in the individual absorption-desorption test because a part of the reaction heat was consumed in the heating or cooling of the metal hydride material and the stainless-steel tank. Even if the operating pressure range was different, it could be confirmed that the heat recovery ability was almost the same as that of the metal hydride tank at SRNL [16].

3-2. Absorption-desorption tests with intermittent hydrogen flow rate

The SRNL solar panel has a maximum power of 13.3 kW. The Water Electrolyzer (WE) at SRNL is a Proton Energy HOGEN RE S40 capable of producing hydrogen of 2.27 kg/day with the maximum DC stack power of 7 kW. The output pressure was about 1.5 MPa and the maximum hydrogen flow rate was 18 NL/min. The Fuel Cell (FC) is a Plug Power GenCore™ 5B48 capable of producing 5 kW net of DC power at a maximum hydrogen flow rate of 75 NL/min. A metal hydride tank with the same geometrical configuration was tested by using these facilities and the experimental results have already been reported [5]. The absorption-desorption tests were carried out for the metal hydride tank at AIST with hydrogen flow rate data obtained at SRNL as inputs for the tests.

3-2-1. Experimental simulation for absorption - Water electrolyzer (WE) was operated by solar power

Fig. 5 shows the experimental results of the hydrogen production using the WE at SRNL. The data obtained for clear day is presented in Fig 5-I, and the data obtained initially for a fair day, later becoming occasionally cloudy in Fig. 5-II. In the both figures, (a) shows the data of the irradiance measured by the pyranometer and the DC power at the cell stack in the WE, and (b) shows the time variation of hydrogen flow rate from the WE to the metal hydride tank at SRNL. When the power from the solar panels exceeded the maximum DC stack capacity of the WE of 7 kW, the surplus power went to the grid through a battery bank at SRNL. Therefore, the data of DC power did not exceed 7 kW. From both figures, it is noted that the hydrogen flow rates

are proportional to the DC power at the cell stack. Fig. 5-II-(a) and (b) also show strong reduction of the hydrogen flow rate when the clouds passed in front of the sun. In the case of THEUS operation, the absorption was carried out with a temperature of 32 °C. At this temperature, a pressure higher than 2.0 MPa is required for the absorption of the metal hydride in the tank at SRNL. However, the pressure of the hydrogen produced by the WE is lower than 1.5 MPa. Consequently the circulation water temperature was set at 12 °C in the experiments at SRNL. In both experiments, the composition of 0.15 was selected as the starting point and the circulation water flow rate was set to 1.72 L/min. The hydrogen flow rate data were read in a personal computer and the experimental simulation was carried out by using the mass flow controller and the metal hydride tank at AIST.

Fig. 6 shows the experimental results using the hydrogen flow rate data presented in Fig. 5-I(b). The circulation water temperature was set at 12 °C and the flow rate at 1.72 L/min, similar to the experimental conditions at SRNL. Fig. 6a shows the reproduced hydrogen flow rate with 6605 NL of hydrogen absorbed in the metal hydride tank. In the original experiment, Fig. 5-I(b), 6610 NL of hydrogen was absorbed. Fig. 6b shows the P-C isotherm. The final pressure was 0.518 MPa allowing the metal hydride tank to be fully charged with hydrogen. Fig. 6c shows the temperatures in each location in the tank, and the inlet temperature and the outlet temperature of the circulation water. All temperatures are decreased with the hydrogen flow rate during the last part of the experiment. In this experiment, the reaction heat recovery rate was 87 %. As mentioned before, the circulation water temperature could not be set at 32 °C in the SRNL experiment, but it could be set at the temperature in this experimental simulation. Fig. 7 shows the experimental results using the same hydrogen flow rate data under the condition that the temperature of circulation water inlet was equal to 32 °C during the experiment. The circulation water flow rate was also set at 1.72 L/min. The reproduced hydrogen flow rate is shown in Fig. 7a. The hydrogen flow rate was also almost completely reproduced, and 6606 NL of hydrogen was absorbed in the metal hydride tank. Fig. 7b presents the P-C isotherm. The tank pressure reached 1.047 MPa at the end of the experiment. In case of the absorption test with a constant hydrogen flow rate, it could not be fully charged. However, the full charge could be achieved in this experiment due to degradation of the pressure increment caused by the reduction of solar radiation. Fig. 7c shows the temperatures in the tank, and the inlet and outlet temperatures of the circulation water. The qualitative temperature profiles in each position are the same as the data shown in Fig. 6c, but all temperatures are higher than the temperatures shown in Fig. 6c. The metal

hydride reaction heat recovery rate was 83 %, which is about 4 % smaller than the previous case. This experiment was carried out over the ambient temperature. The room temperature was set at 22 °C in all experiments. The thermal energy flowed from the metal hydride tank to the surroundings. The experimental condition at 12 °C in Fig. 6 is lower than the ambient temperature. The thermal energy flowed from the surroundings to the metal hydride tank increasing the reaction heat recovery rate. The difference between the reaction heat recovery rates was observed for that reason.

Fig. 8 presents the experimental results using the hydrogen flow rate data shown in Fig. 5-II-(b). The circulation water temperature was set at 32 °C with the circulation water flow rate equal to 1.72 L/min. Fig. 8a shows the reproduced hydrogen flow rate. The hydrogen flow rate was also successfully reproduced and 6451 NL of hydrogen was absorbed in this case. For comparison, 6403 NL of hydrogen was absorbed in the original experiment shown in Fig. 5-II. Fig. 8b shows the P-C isotherm. The maximum pressure was 0.974 MPa. The hydrogen was also fully charged in the metal hydride tank. Fig. 8c shows the temperature in the tank, and the inlet and outlet temperature of the circulation water. Temperature drops are presented in Fig. 8c, when the hydrogen flow rate was diminished. It is also noted from Figs. 8b and 8c that the temperature is more sensitive to the hydrogen flow rate as compared with the pressure. The metal hydride reaction heat recovery rate was 83 % which is approximately the same as the value described above. However, compared to the reaction heat recovery rate of the absorption process in the individual absorption-desorption test, the reaction heat recovery rate was smaller than the result in the individual absorption-desorption test. Previously, it has been considered that the temperature variation improved the reaction heat recovery rate. The temperatures in the tank and the circulation water outlet gradually came back to the initial level at the end of the experiment. If the thermal energy stored in the outer shell of the tank and the metal hydride material could be recovered that might improve the recovery rate. However, the experimental results did not support this hypothesis. Further investigation is necessary for an improved understanding. In addition, the experimental simulation under the condition of the circulation water inlet temperature of 12 °C was also carried out and the metal hydride reaction heat recovery rate was 88 %.

The experimental reaction heat recovery rates for the metal hydride tank at SRNL at 12 °C, which corresponds to the experiments shown in the Fig. 5, were 102 % [5]. The wooden tank support was adopted in the experiments at AIST, but the metal hydride tank was placed on an aluminum support with the compressed thermal insulating foam in the experiments at SRNL. The thermal insulating foam was pressed

between the metal hydride tank and the aluminum support. At the beginning of the experiment, (i.e. when the absorption-desorption test with constant hydrogen flow rate were carried out) the foam maintained the thermal insulation function. However, the insulating foam had been pressed flat by the tank weight as time passed. The thermal insulation was lost during the absorption-desorption tests with intermittent hydrogen flow rate. This resulted in large difference of the heat introduction from the surroundings through the support and the large difference observed in the reaction heat recovery rate data. For the reaction heat recovery rate tests, care must be taken to avoid the heat introduction and heat leakage from the tank support.

3-2-2. Experimental simulation for desorption – Fuel Cell (FC) operation

The FC at SRNL was capable of producing 5 kW net of DC power. Since the hydrogen supply capability of the metal hydride tank was small, 0.5 kW and 1 kW loads were selected in the experiments at SRNL. In spite of these small load selections, the hydrogen consumptions were still large, which resulted in large temperature drops in the metal hydride tank. Due to freezing issues, the temperature of the circulation water was set at 22 °C in the SRNL experiments, while the operating temperature of THEUS was 12 °C. Fig. 9 shows the results of the experimental simulation using the hydrogen flow rate data with the load of 0.5 kW at SRNL. The circulation water flow rate was 1.14 L/min, being the same circulation water flow rate during the SRNL experiment [5]. The ambient temperature was also set about 22 °C. Fig. 9a shows the time variation of the hydrogen flow rate. The dashed gray line shows the data obtained at SRNL, and the mass flow controller was attempted to reproduce the flow rate. The black solid line shows the flow rate from this experimental simulation. The hydrogen flow rate varied ranging from 15 NL/min to 25 NL/min with the average flow rate about 20 NL/min. It is noted that the hydrogen flow rate cannot reproduce the SRNL data in the last 30 min. Fig. 9b presents the P-C isotherm and the circles with the solid line indicate the equilibrium pressure of desorption process at 22 °C. The starting composition, H/M, was 0.998 and 6256 NL of hydrogen was desorbed. The onset pressure in the tank was 0.618 MPa and the minimum pressure was 0.197 MPa. In the SRNL experiment the onset and final pressure in the tank was 1.68 MPa and 0.44 MPa, respectively. The hydrogen supplying capability of the metal hydride tank is lower than the metal hydride tank at SRNL due to the lower working pressure range design. Fig. 9c shows the temperature in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water. The reaction heat recovery rate was 86 %. On the other hand, the reaction heat recovery rate was 96 % in case of the original

experiment at SRNL. The temperature difference of 5 °C between the inlet temperature and the outlet temperature of the circulation water was maintained during the original experiment at SRNL. However, the temperature difference of 5 °C could not be maintained in the last stage in the experimental simulation as shown in Fig. 9c. The wooden tank support was adopted, but the operation temperature was still approaching the ambient temperature. The large difference between these reaction heat recovery rates was mainly from the decline of the desorption performance.

Fig. 10 shows the results of the experimental simulation using the hydrogen flow rate data when the load of 1 kW was adopted. The ambient temperature was also set at 22 °C. The circulation water flow rate was adjusted to 1.78 L/min which was the experimental condition at SRNL and the average flow rate was 1.77 L/min. Fig. 10a shows the time variation of the hydrogen flow rate. The hydrogen flow rate varied ranging from 24 NL/min to 35 NL/min and the average flow rate was about 28 NL/min. 6137 NL of hydrogen was desorbed in 3 hour 38 min (about 2 hours shorter than the previous case). The hydrogen flow rate cannot follow the SRNL data in the last 40 min. The P-C isotherm is presented in Fig. 10b. The starting composition, H/M, was 0.978 and the final composition was 0.19. The onset pressure in the tank was 0.619 MPa and the minimum pressure was 0.157 MPa. Fig. 10c shows the temperature in each location in the tank, and the inlet and the outlet temperature of the circulation water. The temperature in the metal hydride tank is lower than that for the 0.5 kW load. The minimum temperature in the tank was 3.5 °C due to the large hydrogen flow rate. The reaction heat recovery rate was 87 % in this experimental simulation. It was almost the same as the former case. The desorbing time was shorten, and the decline of the desorption performance started in the early stages. However, the short running time has an advantage for the reaction heat recovery rate when the same amount of hydrogen is desorbed or absorbed.

The experimental results of the reaction heat recovery rate for the metal hydride tank at SRNL, those corresponding to the experiments shown in the Figs. 9a and 10a, were 96 % and 101 %, respectively. The temperature difference between the inlet and the outlet of the circulation water was maintained at 5 °C for long time as compared with the experimental simulations. Compared with the results of the experimental simulation, the reaction heat recovery rates become large and these values seem reasonable. However, thermal introduction from the surroundings reduces the reaction heat recovery rate in the desorption tests. The good reaction heat recovery rates at SRNL cannot be fully explained but as previously mentioned, the accuracy of the reaction heat recovery rate is about ± 5 %.

3-3 Development of the vacuum thermal insulator for the metal hydride tank

For the efficient utilization of metal hydride reaction heat, the thermal insulation is an important factor. The thermal insulation foam (Aeroflex) has been used to insulate the metal hydride tank. However, the thickness of the foam needed was large to obtain the required thermal insulation. For this metal hydride tank, the diameter including the foam was 28.5 cm and the total length was about 110 cm even though the tank diameter and the length were about 16.5 cm and 100 cm. **It is preferable to reduce the total size of the metal hydride tank for the construction of THEUS.** To reduce the total diameter, a flexible vacuum thermal insulator was developed and tested. The vacuum thermal insulation was manufactured by using a banding band, a plastic bag, and a vacuum deaerating sealer. The flexible interlining was made using the banding band. The handmade interlining was set in the plastic bag and vacuum-packed. The thickness of the vacuum thermal insulator was about 6 mm. The vacuum thermal insulator was put around the metal hydride tank. After that, the Aeroflex with the thickness of 1 cm was wrapped around the vacuum thermal insulator to improve the appearance of the metal hydride tank. The total diameter of the tank was about 20 cm. To test the thermal insulator, the individual absorption-desorption test was carried out and the experimental results were compared with the data shown in the section 3-1-1.

Fig. 11 shows the results for the individual absorption-desorption test with the vacuum thermal insulator. The P-C isotherm is presented in Fig. 11a. Fig. 11b and Fig. 11c show the time variations of pressure and the temperature in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water in cases of absorption and desorption, respectively. The P-C isotherm is almost the same as in Fig. 3. The pressure history shown in Figs. 11b and 11c are the same as those shown in Figs. 3b and 3c. From Fig. 11b, the small temperature fluctuation is observed in the tank, but the temperatures at all position vary as in Fig. 3b. The temperature profiles in Fig. 11c are similar to the data in Fig. 3c. The metal hydride reaction heat recovery rates were **89 %** for absorption and desorption, respectively. It was, therefore, shown that the tank size could be successfully reduced using the vacuum thermal insulator without reducing the thermal insulation performance [17].

4. Conclusion

A metal hydride tank with a double coil type heat exchanger for operation below 1.0 MPa (G) was prepared and tested at AIST. The experimental data were

compared with the data obtained from the metal hydride tank with the same geometrical configuration as previously tested at SRNL. From the absorption-desorption tests with constant hydrogen flow rate, the ability of the metal hydride tank was comparable with the metal hydride tank at SRNL. From the absorption tests with intermittent hydrogen flow rate, hydrogen could be full charged below 1.0 MPa (G) even for the case of the circulation water inlet temperature of 32 °C. This is due to the fact that the rate of pressure increase became low as the hydrogen flow rate became low, caused by the reduction of solar radiation. From the desorption tests with intermittent hydrogen flow rate, it was confirmed that the hydrogen supplying capability of the metal hydride tank was lower than the metal hydride tank tested at SRNL due to the low working pressure range design. In these experiments with intermittent hydrogen flow rate, the reaction heat recovery rates were about 10~14 % less than that of the results obtained at SRNL depending on the experimental conditions. For the reaction heat recovery rate tests, the heat introduction and heat leakage from the metal hydride tank support needs to be considered. Finally, the vacuum thermal insulator was developed and tested successfully, with a reduction of the tank size without reducing the thermal insulation performance.

Acknowledgements

This study was supported from U.S.-Japan joint research under the program of U.S.-Japan clean energy action plan based on a grant from the Ministry of Economy, Trade, and Industry (METI). A part of this study was also supported from CONCERT-Japan joint research program of the Japan Science and Technology Agency (JST).

REFERENCES

- [1] Masuda M, Kato A, Takahashi A, Maeda T. Energy efficiency evaluation of totalized hydrogen energy utilization system for commercial buildings. Proc. of 17th world hydrogen energy conference 2008; Paper No.260, CD-ROM.
- [2] Tange M, Maeda T, Nakano A, Ito H, Kawakami Y, Masuda M, Takahashi T. Experimental study of hydrogen storage with reaction heat recovery using metal hydride in a totalized hydrogen energy utilization system. Int. J. Hydrogen Energy 2011; 36: 11767-76.
- [3] Nakano A, Maeda T, Ito H, Masuda M, Kawakami Y, Tange M, Takahashi T, Nishida K. Study on absorption/desorption characteristics of a metal hydride tank for boil-off gas from liquid hydrogen. Int. J. Hydrogen Energy 2012; 37: 5056-62.

- [4] Nakano A, Maeda T, Ito H, Motyka T, Perez-Berrios MJ, Greenway S. Experimental Study on a Metal hydride Tank for the Totalized Hydrogen Energy Utilization System, *Energy Procedia* 2012; 29: 463-8.
- [5] Nakano A, Ito H, Maeda T, Munakata T, Motyka T, Corgnale C, Greenway S, Perez-Berrios MJ. Study on a metal hydride tank to support energy storage for renewable energy, *J. Alloys & Compounds*. 2013; 580-S1 15: S418-22.
- [6] Corgnale C, Motyka T, Greenway S, Perez-Berrios MJ, Nakano A, Ito H, Maeda T. Metal hydride bed system model for renewable source driven Regenerative Fuel Cell. *Journal of Alloys and Compounds*, 2013; 580-S1: S406-09
- [7] Motyka T. Savannah River National Laboratory Regenerative Fuel Cell Project, SRNL-STI-2008-00388, OSTI ID: 941112, Nov. 11, 2008.
- [8] Miura S, Fujisawa A, Ishida M. A hydrogen purification and storage system using metal hydride, *Int. J. Hydrogen Energy* 2012; 37: 2794-99.
- [9] Blinov VD, Borzenko IV, Dunikov OD, Romanov AI. Experimental investigations and a simple balance model of a metal hydride reactor, *Int. J Hydrogen Energy* 2014; 39: (In Press).
- [10] Muthukumar P, Prakash Maiya M, Srinivasa Murthy S. Experiment on a metal hydride-based hydrogen storage device. *Int J Hydrogen Energy* 2005; 30: 1569-81.
- [11] Kikkinides ES, Georgiadis MC, Stubos AK. On the optimization of hydrogen storage in metal hydride beds. *Int. J Hydrogen Energy* 2006; 31: 737-51.
- [12] MacDonald BD, Rowe AM. Impacts of external heat transfer enhancements on metal hydride storage tanks. *Int J Hydrogen Energy* 2006; 31: 1721-31.
- [13] Forde T, Eriksen J, Pettersen AG, Vie PJS, Ulleberg O. Thermal intergration of a metal hydride storage unit and a PEM fuel cell stack. *Int J Hydrogen Energy* 2009; 34: 6730-9.
- [14] Souahlia A, Dhaou H, Askri F, Sofiene M, Jemni A, Nasrallah SB. Experimental and comparative study of metal hydride hydrogen tanks. *Int J Hydrogen Energy* 2011; 36: 12918-22.
- [15] Nakano A, Ito H, Maeda T. Hydrogen induction tube for a metal hydride tank, Japanese Patent Toku-gan 2012-125583 (PATPEND)
- [16] Maeda T, Nakano A, Ito H, Motyka T, Perez-Berrios MJ, Greenway S. Effect of the metal hydride tank structure on the reaction heat recovery for the Totalized Hydrogen Energy Utilization System, *J. Int. Council on Elect. Eng.* 2013; 3-2: 103-109.
- [17] Nakano A, Ito H. Vacuum thermal insulator and method for manufacturing the same, Japanese Patent Toku-gan 2014-172127 (PATPEND)

Figure captions

Figure 1 Schematic of the metal hydride tank.

Figure 2 Schematic of the experimental set-up.

Figure 3 Results for the individual absorption-desorption test. (a) P-C isotherm. (b) Temperature in the tank and circulation water in case of absorption. (c) Temperature in the tank and circulation water in case of desorption.

Figure 4 Results for the absorption-desorption continuous test. (a) P-C isotherm. (b) The circulation water outlet temperature.

Figure 5 Experimental results of the hydrogen production using the water electrolyzer (WE) in SRNL. (I) Experimental data obtained when there was not a cloud in the sky during a day. (II) Experimental data obtained when a day of fair, later, occasionally cloudy. (a) Irradiance measured by the pyranometer and DC power at the cell stack in the WE. (b) Time variation of hydrogen flow rate from the WE to the metal hydride tank set in the SRNL.

Figure 6 Experimental simulation results using the hydrogen flow rate data shown in Fig. 5-I-(b). (a) Reproduced hydrogen flow rate. (b) P-C isotherm. (c) The temperatures in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water.

Figure 7 Experimental simulation results using the hydrogen flow rate data shown in Fig. 5-I-(b) under the condition that the temperature of circulation water inlet was 32 °C. (a) Reproduced hydrogen flow rate. (b) P-C isotherm. (c) The temperatures in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water.

Figure 8 Experimental simulation results using the hydrogen flow rate data shown in Fig. 5-II-(b) under the condition that the temperature of circulation water inlet was 32 °C. (a) Reproduced hydrogen flow rate. (b) P-C isotherm. (c) The temperatures in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water.

Figure 9 Results of the experimental simulation using the hydrogen flow rate data when a 5 kW fuel cell in SRNL was operated with the load of 0.5 kW. (a) Time variation of the hydrogen flow rate. (b) P-C isotherm. (c) The temperatures in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water.

Figure 10 Results of the experimental simulation using the hydrogen flow rate data when a 5 kW fuel cell in SRNL was operated with the load of 1 kW. (a) Time variation of the hydrogen flow rate. (b) P-C isotherm. (c) The temperatures in each position in the tank, and the inlet temperature and the outlet temperature of the circulation water.

Figure 11 Results for the individual absorption-desorption test with the vacuum insulator. (a) P-C isotherm. (b) Temperature in the tank and circulation water in case of absorption. (c) Temperature in the tank and circulation water in case of desorption.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11