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Aluminium alloy based hydrogen storage tank operated with sodium aluminium hexahydride Na_3AlH_6

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Abstract

Here we present the development of an aluminium alloy based hydrogen storage tank, charged with Ti-doped sodium aluminium hexahydride Na_3AlH_6 . This hydride has a theoretical hydrogen storage capacity of 3 mass-% and can be operated at lower pressure compared to sodium alanate $NaAlH_4$. The tank was made of aluminium alloy EN AW 6082 T6. The heat transfer was realised through an oil flow in a bayonet heat exchanger, manufactured by extrusion moulding from aluminium alloy EN AW 6060 T6. Na_3AlH_6 is prepared from 4 mol-% $TiCl_3$ doped sodium aluminium tetrahydride $NaAlH_4$ by addition of two moles of sodium hydride NaH in ball milling process. The hydrogen storage tank was filled with 213 g of doped Na_3AlH_6 in dehydrogenated state. Maximum of 3.6 g (1.7 mass-% of the hydride mass) of hydrogen was released from the hydride at approximately 450 K and the same hydrogen mass was consumed at 2.5 MPa hydrogenation pressure. 45 cycle tests (rehydrogenation and dehydrogenation) were carried out without any failure of the tank or its components. Operation of the tank under real conditions indicated the possibility for applications with stationary HT-PEM fuel cell systems.

Keywords: hydrogen storage, complex hydride, Na_3AlH_6 , aluminium alloy, extrusion moulding

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1. Introduction

Over the last years the use of different hydrogen based techniques for transportation, electricity production or energy storage has been widely discussed (Schlapbach and Zuttel, 2001; Weidenthaler and Felderhoff, 2011). Currently the hydrogen storage method for fuel cell powered vehicles seems to be 70 MPa compressed hydrogen (Eberle et al., 2012). Whereas the liquid hydrogen storage has been discussed to be used in aviation systems (Verstraete et al., 2010; Renouard-Vallet et al., 2012), the solid hydrogen storage in metal hydrides has already been implemented in submarines powered by fuel cells (Sattler, 2000; Psoma and Sattler, 2002). Besides of their high volumetric hydrogen storage density, metal hydrides offer high heat storage capacities (Bogdanović et al., 1989; Wierse et al., 1991). Therefore they could be used in applications like heat pumps, cooling systems (Willers and Groll, 1999; Muthukumar and Groll, 2010), heat transformers (Kang and Yabe, 1996), heat driven gas compressors (Li et al., 2010) or finally gas purifiers (Miura et al., 2012).

Sodium aluminium tetrahydride $NaAlH_4$ (sodium alanate) is a well known complex metal hydride, which kinetics was tremendously enhanced by Ti-dopants (Bogdanović and Schwickardi, 1997). Sodium alanate releases or consumes hydrogen in a two step reaction (cf. Equations 1 and 2). $NaAlH_4$, a low temperature hydride, has an equilibrium pressure of 0.1 MPa at approximately 300 K, whereas the second decomposition phase Na_3AlH_6 (sodium aluminium hexahydride), a medium temperature hydride, has an equilibrium pressure of 0.1 MPa at approximately 373 K. The reaction enthalpies are 37 kJ/mol_{H_2} for the first step and 47 kJ/mol_{H_2} for the second step respectively (Bogdanović and Schwickardi, 1997). Therefore $NaAlH_4$ or only Na_3AlH_6 , which characteristics have not yet been investigated, can be used as both hydrogen and heat storage material in CHP (Combined Heat and Power) systems for household applications, for instance combined with a HT-PEM fuel cell. Such additional heat storage possibility could prolong the operation time of the fuel cell based CHP system in stationary applications (Urbanczyk et al., 2012).

Both theoretical (Jensen et al., 2005; Pfeifer et al., 2009; Reddy and Jayanti, 2012) and practical investigations (Urbanczyk et al., 2011; Weiss-Ungethüm et al., 2014; Urbanczyk,

2014) were carried out to demonstrate the thermal coupling of $NaAlH_4$ based hydrogen storage tanks in combination with a HT-PEM fuel cell. While in one case air was proposed as a heat transfer medium between the tank and fuel cell (Utz et al., 2011), in other cases a liquid was used for the thermal coupling. Another complex hydride ($2LiNH_2 - 1.1MgH_2 - 0.1LiBH_4 - 3 \text{ mass-}\% \text{ ZrCoH}_3$) and a combination of it with a metal hydride ($LaNi_{4.3}Al_{0.4}Mn_{0.3}$) was also proposed as a solid hydrogen storage system thermally coupled with a HT-PEM fuel cell (Bürger et al., 2014b,c,a).

For the evaluation of a storage system the overall performance of the complete system (tank system plus storage material) is quite important. Therefore a lightweight material for the tank systems in combination with a hydrogen storage material with even relatively low hydrogen capacity promises several advantages (for example regarding the gravimetric hydrogen storage capacity of the system). To the best of our knowledge, until now, aluminium alloy based tank systems in combination with complex metal hydrides were never explored as a hydrogen storage system. Ti-doped Na_3AlH_6 was selected as hydrogen storage material, because its properties (453.15 K operating temperature, $p_{max} = 2.5 \text{ MPa}$) fit very well to the mechanical load limits of the selected aluminium alloy.

2. Materials and calculations

2.1. Synthesis of doped Na_3AlH_6

According to equations 1 and 2 sodium alanate can be decomposed in a two step reaction. In the first reaction theoretical 3.7 mass-% of hydrogen can be released. The hexahydride phase decomposes with theoretical 1.8 mass-% hydrogen content (Bogdanović et al., 2007).

Without additional aluminium, sodium aluminium hexahydride releases theoretical 3 mass-% hydrogen, according to reaction 3.

Doped and undoped Na_3AlH_6 was already investigated by Bogdanović and Schwickardi in 1997 (Bogdanović and Schwickardi, 1997). This complex hydride was doped with both $TiCl_3$ (2.0 mol-%) and $Ti(OBu)_4$ (2.0 mol-%). The doped sodium aluminium hexahydride showed superior hydrogen storage capacity and cyclic stability than the undoped material. The cyclic tests for the dehydrogenation of $TiCl_3$ doped Na_3AlH_6 were conducted at 543.15 K and the hydrogenation tests at 473.15 K and 6 MPa hydrogen pressure. The cyclic tests for the dehydrogenation of $Ti(OBu)_4$ doped Na_3AlH_6 were conducted at 503.15 K between 3 and 4.2 MPa and the hydrogenation tests at 443.15 K between 4.2 and 3 MPa hydrogen pressure. The first mentioned sodium aluminium hexahydride (doped with $TiCl_3$) was cycled for 25 times and achieved storage capacities between 2.1 and 2.3 mass-%. The second one (doped with $Ti(OBu)_4$) was cycled for 100 times.

The starting materials, for Na_3AlH_6 synthesis in the work presented here, NaH (60% dispersion in mineral oil, Sigma-Aldrich Chemical) and $NaAlH_4$ (95% technical grade, Sigma-Aldrich Chemical) were further purified in our laboratory. $TiCl_3$ (Sigma-Aldrich Chemical), Al (99%, Aluminium Rheinfelden), and doped $NaAlH_4$ (doped with 4.0 mol-% $TiCl_3$, prepared in house by mechanochemical synthesis), were used as received. The sodium aluminium hexahydride (250 g) was prepared by mechano-chemical synthesis (reaction 4) under an inert atmosphere using hardened steel grinding bowls and grinding balls in a Fritsch Planetary Mill PULVERISETTE 5.

Parameters for the synthesis are given in Table 1. Prior to filling material into the hydro-

Table 1: Ball milling parameters for synthesis of Na_3AlH_6

volume ¹ [ml]	No. of balls [–]	Na_3AlH_6 [g]	rotat. freq. [rpm]	milling time [h]	BPR ² [–]
500	25	50	250	2	30:1

¹ grinding bowl without balls

² Ball to Powder Ratio

gen storage tank it was tested to assess the cycle-life kinetics. The test was carried out

using PCTPro (Setaram), a fully automated Sieverts instrument for measuring gas sorption properties of materials. 2 g of material was placed into a specially designed tube like sample holder. In total 275 two-hour-hydrogenation/dehydrogenation cycles were achieved without degradation. Every hydrogenation cycle was performed at 2.5 MPa hydrogen pressure and 443.15 K, while every dehydrogenation cycle was performed at ambient hydrogen pressure and 443.15 K. 1.7 mass-% of hydrogen was evolved and absorbed during hydrogenation or dehydrogenation (Figure 1). 80% of the total hydrogen amount is evolved or absorbed during first 30 min of every cycle.

Figure 1: Long term cycling of $Na_3AlH_6 + 4.0 \text{ mol-}\% \text{ } TiCl_3$

2.2. Comparison between the tetra and hexa aluminium hydrides

A typical hydrogenation pressure for $NaAlH_4$ is 10 MPa and higher (Lozano et al., 2009; Johnson et al., 2011). The typical hydrogenation pressure for the Na_3AlH_6 formation is approximately four times lower, compared to the hydrogenation pressure of $NaAlH_4$ at the same temperature. The hydrogenation of $3NaH + Al$ system can be carried out at a pressure

around 2.5 MPa. The energy demand (specific work w_t) for hydrogen compression from the low pressure p_1 up to the high pressure p_2 can be calculated by using the equation for the isentropic enthalpy difference Δh_s . The specific compression work w_t from the starting temperature T_1 can be written as shown in Equation 5 (Baehr and Kabelac, 2009):

$$w_t = \Delta h_s = \frac{\kappa}{\kappa - 1} \cdot R \cdot T_1 \cdot \left[\left(\frac{p_2}{p_1} \right)^{\frac{\kappa-1}{\kappa}} - 1 \right] \quad (5)$$

R is the specific gas constant and κ the isentropic exponent for hydrogen.

The isentropic work for hydrogen compression can be put in relation to the low heating value of hydrogen LHV_{H_2} , which is shown in Figure 2. It can be seen in the figure that the

Figure 2: Energy demand for isentropic compression of hydrogen. Thermophysical data according to (Baehr and Kabelac, 2009)

energy demand for the compression of hydrogen up to 10 MPa is approximately 1.8 times higher compared to the compression up to 2.5 MPa. It should be noted that compressors are not working isentropic. They work irreversible with entropy production and some of them can be cooled down using several cooling loops (Jepsen et al., 2012). This additional cooling shifts the compression closer to the ideal isothermal compression process, in which

the entropy production is reduced and less energy for compression is necessary. When isothermal compression is considered (cf. Equation 6 (Baehr and Kabelac, 2009)),

$$w_{t,is} = R \cdot T_1 \cdot \ln \left(\frac{p_2}{p_1} \right) \quad (6)$$

the LHV (Low Heating Value) based energy demand is only 1.4 times higher for 10 MPa in comparison to 2.5 MPa end pressure. Increasing the end pressure p_2 , the cooling procedures becomes more important.

Beside of some energy merits also advantages of hydrogen storage density can be accomplished. The same or even higher hydrogen storage system density can be achieved by using low weight materials for the pressure tank. A steel pressure tank manufactured from an austenitic steel (W.-Nr. 1.4571 (EN 10027-2, 1992)) and charged with $NaAlH_4$ can be compared to an aluminium alloy (EN AW 6082 T6 (EN 573-2, 1994; EN 515, 1993)) based pressure tank, which is charged with Na_3AlH_6 . The assumptions for the comparison are given in Tables 2 and 3. The gravimetric hydrogen storage density is calculated from the

Table 2: Assumptions I for considered complex hydrides

	density [kg/m ³]	max. pressure [MPa]	ξ_{H_2} [mass – %]	ψ_{dopant} [mol – %]
$NaAlH_4$ ¹	720	10	3.5	3 ²
Na_3AlH_6	variable	2.5	1.95	4 ³

¹ (Mosher et al., 2007)

² $TiCl_3 \cdot \frac{1}{3} AlCl_3$

³ $TiCl_3$

Table 3: Assumptions II for considered tank materials

	density [kg/m ³]	pressure [MPa]	yield strength [MPa]	temperature [K]
austenitic steel ¹	8000	10	$Rp_{1,0} = 190$	473.15
aluminium alloy ²	2700	5	$Rp_{0,2} = 162.5$	473.15

¹ 1.4571 (DIN EN 10027-2:1992)

² EN AW 6082 T6 (DIN EN 573-2:1994 and DIN EN 515:1993; (Ostermann, 2007))

110

mass of stored hydrogen and the mass of the tank, while the volumetric hydrogen density

depends on the mass of stored hydrogen and the volume of the tank. Higher hydrogen mass is achievable if either high hydrogen content of the hydride is available or the hydride powder is densified. For energy density comparison between an Aluminium Alloy Tank charged with Na_3AlH_6 (AAT) and a Steel Alloy Tank charged with $NaAlH_4$ (SAT) a variable x is defined, which describes the ratio of the respective hydride densities of Na_3AlH_6 and $NaAlH_4$, (Equation 7).

$$x = \frac{\rho_{Na_3AlH_6}}{\rho_{NaAlH_4}} \quad (7)$$

The gravimetric energy density e_g of the system means the ratio of stored hydrogen energy ($m_{H_2} \cdot LHV$) per kg of the system mass m_{Sys} (cf. Equation 8).

$$e_g = \frac{m_{H_2} \cdot LHV}{m_{Sys}} \quad (8)$$

The volumetric energy density of the system e_v means the ratio of stored hydrogen energy ($m_{H_2} \cdot LHV$) per m^3 system volume V_{Sys} (cf. Equation 9).

$$e_v = \frac{m_{H_2} \cdot LHV}{V_{Sys}} \quad (9)$$

In Figure 3 the ratio Z of the gravimetric energy densities of both systems AAT and SAT

$$Z = \frac{e_{g,AAT}}{e_{g,SAT}} \quad (10)$$

is shown on the left ordinate, whereas on the right ordinate the ratio Y of volumetric energy densities

$$Y = \frac{e_{v,AAT}}{e_{v,SAT}} \quad (11)$$

is shown.

The data of a hydrogen content of 3.5 mass-% and a hydride density of 720 kg/m^3 (sodium alanate), that was used in the calculations, was taken from Mosher et al. (Mosher et al., 2007). Other research groups have achieved different results of hydrogen content and hydride density of the used hydrides. A hydrogen content of 4.5 mass-% and a hydride density of 1600 kg/m^3 were achieved by Lozano et al. (Lozano et al., 2011), while a hydrogen content of 3.4 mass-% and a hydride density of 1470 kg/m^3 were accomplished by van Hassel et al. (van Hassel et al., 2012).

Figure 3: Ratios of gravimetric $Z = \frac{e_{g, AAT}}{e_{g, SAT}}$ and volumetric $Y = \frac{e_{v, AAT}}{e_{v, SAT}}$ hydrogen densities vs. the hydride density ratio $x = \frac{\rho_{\text{Na}_3\text{AlH}_6}}{\rho_{\text{NaAlH}_4}}$.

Wall thickness of the pressure tank, dimensions of flanges and lids were calculated according to technical data and safety regulations (AD Merkblätter, 2002). Based on these dimensions the volumes and from known densities of the tank alloys (steel and aluminium in Table 3) the masses of the tanks were calculated. The calculated wall thickness depends on the diameter of the tank (cylindrical shape) and influences the mass of the tank (AD Merkblätter, 2002). Because of the different material densities and hydrogen contents of Na_3AlH_6 and $NaAlH_4$, the respective hydride volumes are not the same for storage of equal mass of hydrogen. The mass and volume of the SAT were calculated according to the values in Tables 2 and 3 and were kept constant. For comparison, only the length of the respective aluminium tank (AAT) was changed according to Na_3AlH_6 density, while the inner exemplary diameter (117.4 mm) of the both tank systems AAT and SAT was left the same .

Figure 3 shows, that the gravimetric energy density (gravimetric hydrogen density) is higher in case of the AAT for all hydride density ratios. The volumetric energy density (volumetric hydrogen density) is lower in case of the AAT if the hydride density ratio is less than 1.5. In this comparison the mass and the volume of connectors, hydrogen distribution and heat transfer system were not taken into account. It can be concluded, that, despite of the lower hydrogen content of Na_3AlH_6 in comparison to $NaAlH_4$, an exemplary aluminium alloy tank filled with Na_3AlH_6 has a considerable potential with regard to the operational procedure (Figure 2) and gravimetric energy density (Figure 3) of the whole system.

2.3. Aluminium alloy based hydrogen storage tank

The hydrogen storage tank and the heat exchanger were manufactured from aluminium based alloys (EN AW 6082 T6 and EN AW 6060 T6) characterized according to the European standards ((EN 12392, 2000; EN 13445, 2009)). The pressure vessel was developed for maximum operational temperature of 473.15 K, although such a high temperature was not expected. The yield stress of EN AW 6082 T6 of 162.5 MPa at 473.15 K was taken from (Ostermann, 2007). While decreasing of the yield stress at higher temperature over time ($> 2h$) has been unknown, the tank has been designed for higher hydrogenation pressure

161 (5 MPa) than applied under realistic conditions (2.5 MPa cf. Table 2). At 2.5 MPa hydrogen
 162 pressure the tank could be theoretically operated up to 563.15 K. In addition a safety factor
 163 of 1.5 was used for the calculation of dimensions (AD Merkblätter, 2002). The tube of the
 164 heat exchanger has orthogonal fins to increase material strength against the pressure of the
 165 reacting metal hydride at the outer side of the heat exchanger. Thermal oil flows inside
 166 of the tube at a pressure, which is high enough to overcome the pressure drops in the oil
 167 circuit. In one of the strength experiments, which were carried out in a similar steel tank
 168 without Na_3AlH_6 , the heat exchanger withstood an outer pressure of 20 MPa (at ambient
 169 temperature), whereas inside of the heat exchanger tube only ambient pressure prevailed.
 170 It was constructed as a bayonet heat exchanger with reversal oil flow inside of the tube,
 171 achieved by a small steel capillary, which was inserted into the finned heat exchanger tube.
 172 One end of the tube was closed by laser welding while the other end was laser welded to the
 173 lid. The aluminium tank with integrated aluminium heat exchanger is shown in Figure 4.

Figure 4: Aluminium alloy tank. The heat exchanger can be seen from the open side of the tank.

174 The dimensions of the bayonet type heat exchanger, made by extrusion moulding of

175 EN AW 6060 T6 aluminium alloy, are given in Table 4.

Table 4: Data of the bayonet type EN AW 6060 T6 heat exchanger

number of orthogonal fins ¹	28	[–]
number of radial fins ²	4	[–]
fin thickness ³	0.6	[mm]
fin clearance ⁴	2.8	[mm]
fin height (minimal/maximal)	4.5/10	[mm]
fin length	205	[mm]
heat exchange area	0.103	[m ²]
mass of finned length	118.1	[g]
heat exchange tube (inner diameter/wall thickness)	10.1/1.1	[mm]
bayonet tube (outer diameter/wall thickness)	8/0.5	[mm]

¹ fin distribution shown in Figures 7 and 9

² basis for orthogonal fins

³ at fin tip, orthogonal fins

⁴ at fin tip, orthogonal fins

176 All mass of the most important components of the tank can be found in Table 5. The over-
 177 all mass of the tank system consisting of aluminium alloys based components was 1380.4 g
 with a free inner volume of 414 ml available for the metal hydride. The total mass of the

Table 5: Mass of the particular components of the tank in [g]

tank	lid I	lid II	sealings	H_2 -distribution	heat exchanger	bayonet tube	screws
698.2	303 ¹	263.5 ²	9.4	75,3	115.7	17.6	913.4

¹ only for opening/closing purpose

² this lid is welded to the heat exchanger

178
 179 tank and the listed components including 216.6 g of hydrogenated Na_3AlH_6 (3.6 g H_2) was
 180 2612.7 g. As a result the gravimetric hydrogen density of 0.14 mass-% and the volumetric
 181 hydrogen density of 3.7 kg/m³ (overall tank volume 990.3 ml) was achieved. The mass of the
 182 assembled tank including thermocouples with appropriate fittings and oil distribution sys-
 183 tem for the bayonet heat exchanger was 3805.4 g. Although these values are far away from
 184 the results achieved for example by Mosher et al. (2.0 mass-% and 21.0 kg/m³ with carbon

185 fibre composite vessel filled with doped $NaAlH_4$ (Mosher et al., 2007)) high improvements
 186 regarding mass reduction of the proposed system are possible. The aluminium mass con-
 187 tributes to 36% of the total tank mass. Several steel parts as screws and fittings offer a high
 188 potential for further reduction of the mass of the tank.

189 3. Experimental results

190 3.1. Test rig

191 The hydrogenation and dehydrogenation experiments were carried out in the test rig
 192 shown in Figure 5. The heat transfer was conducted by circulation of a thermal oil. The
 193 heated oil entered the annulus of the bayonet heat exchanger. After the oil transferred the
 194 heat to/or from the heat exchanger, it flew into the bayonet tube in the central position
 195 (cf. drawings in Figures 7 and 9). In case of the dehydrogenation tests the necessary
 196 temperature was controlled by a thermostat and additional electrical heating. The energy
 197 flow was estimated by measuring the temperature and the mass flow of the oil. The hydrogen
 198 flow was controlled with a thermal mass flow controller and the temperatures with T type
 199 thermocouples. The thermal oil circuit, the pathways of hydrogenation and dehydrogenation
 procedures are depicted in Figure 5.

Figure 5: Aluminium alloy tank integrated in test rig

In case of hydrogenation experiments the hydride was heated by both thermostat via thermal oil and electrical heating until the temperature in the material reached a value between 423.15 and 443.15 K. Afterwards hydrogen at a pressure of 2.5 MPa was introduced into the tank. The hydrogen (5.0 purity grade) was provided from 50 l, 20 MPa cylinders. Because of heat evolution during the hydrogenation, electrical heating was switched off and in some experiments the thermostat cooled down the tank.

It was important to hold the temperature during the dehydrogenation step constantly above 453.15 K, because of non-optimized kinetics of the hydride. Dehydrogenation was started after the controlled temperatures inside the tank reached more than 453.15 K.

3.2. Tests of the sodium aluminium hexahydride

The tank (inner volume 414 ml) was filled with 213 g of dehydrogenated Na_3AlH_6 , doped with 4 mol-% of $TiCl_3$. The hydrogenation pressure was mostly 2.5 MPa and the operating temperature of the hydrogen storage material was varied between 423.15 and 443.15 K. For the monitoring of the temperature evolution eight thermocouples were installed at different positions inside the tank. Three of them were located 183 mm from the inner surface of the lid connected to the heat exchanger. The other five were installed 40 mm from the same lid. The temperatures of the hydrogenation and dehydrogenation process as well as the position of the thermocouples are shown in Figure 7 and Figure 9. The hydrogen pressure in the tank and the hydrogen flow to the tank during the hydrogenation as well as the hydrogen flow from the tank during the dehydrogenation are shown in Figure 6 and Figure 8 respectively. The hydrogen flow during a hydrogenation test has been automatically changed according to the status of the hydrogen saturation of the hydride. The highest hydrogen flow achieved in the beginning of the test was about 53 l/h. It decreased to zero when the cooling of the tank system started. The additional electrical heating was switched off before hydrogen was introduced into the reactor. The small changes of the hydrogen pressure dramatically influence the hydrogen flow (flow peaks at 110 min and at 121 min), as can be seen in Figure 6. At the beginning of the hydrogenation process the temperature was not equally distributed inside the powder, thus a temperature difference of 15 K was measured between

Figure 6: Hydrogen flow and pressure during hydrogenation

TC3 and TC5. At the end of the hydrogenation process, after 373 min, the temperature was evenly distributed and the highest temperature difference measured in the hydride bed was only 4.3 K between TC3 and TC5. During this time the temperature difference (between TC1 and TC3) at the heat exchanger wall was 6.2 K.

In this typical hydrogenation test an amount of 3.6 g (1.7 mass-%) of hydrogen was stored in the hydride. During the hydrogenation process the hydrogen flow was measured by the mass flow meter (cf. MFM, Figure 5). When the hydrogenation had finished and the hydride has cooled down, the left over amount of hydrogen was exhausted by the mass flow controller (cf. MFC, Figure 5).

During the dehydrogenation test the hydrogen flow was stepwise reduced, to keep the hydrogen pressure in the tank at 0.65 MPa (cf. from 140 to 215 min in Figure 8). In the beginning of the dehydrogenation process the hydrogen flow was about 62 l/h. After the hydrogen flow was decreased to 22 l/h, the hydrogen pressure increased slightly. At decreasing hydrogen pressure the hydrogen flow was kept constant at 5.6 l/h until the pressure reached

Figure 7: Temperatures during hydrogenation process and positions of the thermocouples

Figure 8: Hydrogen flow and pressure evolution during dehydrogenation

0.1 MPa.

The dehydrogenation test was started when the temperatures of the hydride reached 453.15 K (cf. vertical line in Figure 9). The lowest temperature of 450.15 K was measured at TC3, which is located near the wall of the tank. At the beginning of the dehydrogenation all the temperatures inside the hydride and at the walls of the heat exchanger (TC1, TC4 and TC8) dropped. The temperature gradient in all locations (in fin ground and in 3 mm and

Figure 9: Temperatures during dehydrogenation process and positions of the thermocouples

7 mm distance from the fin tip) was almost the same indicating an equal heat distribution in the tank. To avoid underpressure during the cooling process an inert gas (argon) was introduced into the tank (cf. Figure 8). The influence of the parasitic heat losses along the oil flow through the annulus of the heat exchanger have no important effect on the temperature development in the hydride (TC1 and TC4 are almost the same; cf. Figure 9). The temperature difference between the levels A (TC2) and B (TC5) during the dehydrogenation process was lower than 5 K. During the hydrogenation the measured temperature difference was even less significant. Previously carried out hydrogenation or dehydrogenation exper-

iments showed, that the temperature distribution around the investigated heat exchanger was quite equal at the two measured levels and thus independent from the not centrally located hydrogen distribution tube.

The inner reactor diameter was 53 mm and the edge length of the heat exchanger was 30 mm, the maximal length from the end of the fin tip to the internal wall of the tank was 11.5 mm. This design of the tank with this heat exchanger was less than ideal, because approximately 70% of the free volume was not occupied by the heat exchanger. As a result the temperature difference in the hydride at 7 mm distance from the fin tip towards the wall was quite high (≈ 15 K in Figure 9). In a total 45 cycling test (hydrogenation and dehydrogenation tests) were carried out with this aluminium alloy tank charged with doped Na_3AlH_6 .

4. Conclusions

A light weight aluminium alloy based tank system operated with a Ti-doped complex metal aluminium hydride Na_3AlH_6 was constructed and tested under hydrogenation and dehydrogenation conditions. Both the aluminium alloy parts of the tank as well as welded connections between these parts have withstood the operational pressure of 2.5 MPa during the hydrogenation and a temperature up to 463.15 K during the dehydrogenation. After a 45 cycling test neither failure of the aluminium alloy parts nor leakages were detected. During the hydrogenation/dehydrogenation processes up to 3.6 g (1.7 mass-%) of hydrogen were stored and released. At this stage of development we were not interested in the optimization of the kinetics and capacity of the complex hydride material. The successful operation of the tank system was the main target. A forthcoming publication will address optimization of the complex metal hydride properties and construction of an aluminium alloy based tank system for 3 kg of the hydrogen storage material.

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