ALTERNATIVE APPLICATIONS OF PELTIER THERMOPILES IN COOLING METAL HYDRIDE CONTAINERS
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Abstract: Hydrogen absorption into a metal hydride container is accompanied with generation of heat that must be removed during the process. In the case that the container is not cooled, the gas pressure rapidly increases and even with a small amount of stored hydrogen it exceeds the permissible value. Peltier thermopiles offer an interesting alternative to conventional cooling methods and they have been increasingly used. The present article describes the measurements aimed at identification of an optimal cooling method and the results of the comparison of surface cooling, circulation cooling, and the combined cooling methods in terms of thermodynamic and energy parameters during hydrogen absorption.

1 Introduction

The use of circulation heating and cooling of MH containers brings certain disadvantages with regard to the heat exchange between the heat-transfer fluid and the surrounding environment, as well as the heat gain from the rotating parts of the pump [1,2]. Thermal capacity produced by the PT is therefore reduced by a considerable portion; this may be eliminated by connecting the PT directly to the shell of the MH container. However, the surface heating (cooling) of containers is accompanied changing the internal heat transfer via the tubular spiral heat exchanger into the surface heating (cooling); this may, due to significantly low thermal conductivity of the powder MH material, prolong the time required for changing the container temperature and increase the pressure during hydrogen absorption as a result of insufficient heat removal [3-7].

2 Characteristics of the measurement stand with a metal hydride container

In order to facilitate the transport of thermal energy between the planar PT attached to the cylindrical surface of the HBond©500 container, it was necessary to construct a heat exchanger (Figure 1) which would also be used for accumulation in order to maintain low temperatures during heat generation in hydrogen absorption.

The surface heat exchanger consists of two mirrored aluminium parts that are screwed together and attached to the surface of the MH container (Figure 2) using the thermally conductive paste on the heat-transfer surfaces in order to reduce the thermal contact resistance.

For the purpose of determination of the difference between the temperatures of the heated side and the cooled side of the PT, temperature sensors T_9 to T_{12} were installed. T_{13} sensor was scanning the ambient temperature which is required for the correct identification of the heat losses and gains from the surrounding environment.

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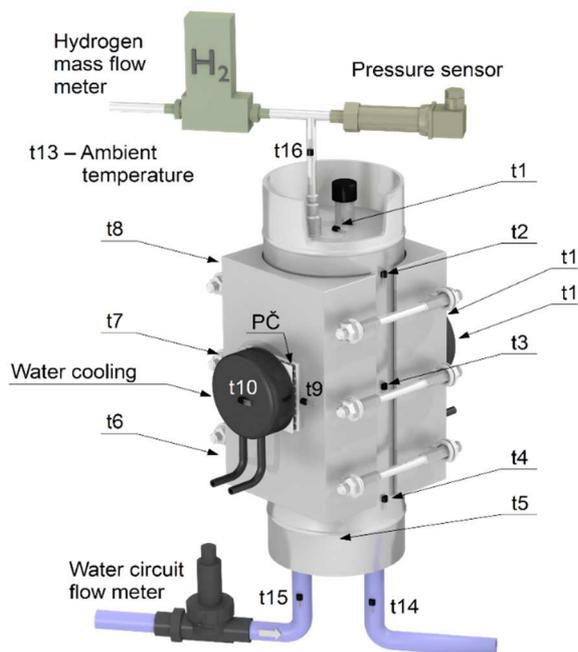
Figure 1 3D model of the MH container with aluminium heat exchanger, PT, water cooling and measuring probes
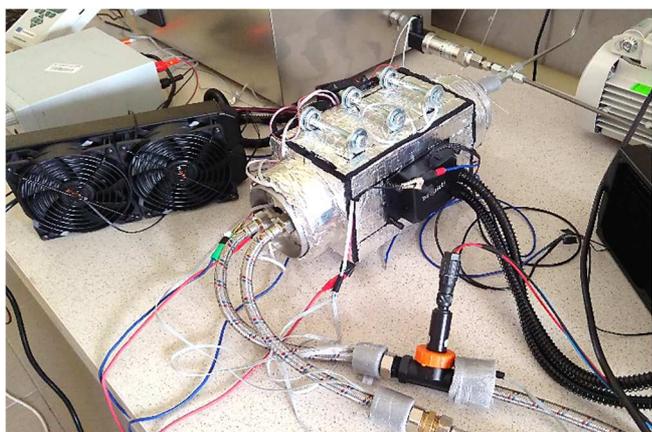
Figure 2 Image of the measurement stand with the MH container

After the sensors were installed, the container was thermally insulated with 10 mm thick foam rubber with the external reflective foil to eliminate the radiant heat flux.

3 Hydrogen absorption into a metal hydride container without cooling

In the first part of the experiment, the measurements were performed with regard to the absorption of 200 litres of hydrogen into a metal hydride container while maintaining the average flow rate of approximately 9.4 l·min^{-1} without cooling. The container was only cooled by free convection from the insulation surface to the surrounding environment.

Hydrogen absorption into the intermetallic structure of the alloy was accompanied with generation of heat that increases the container temperature. Due to the fact that no additional cooling was used, the temperature significantly increased and this also proportionally increased the equilibrium pressure of hydrogen.

The development of the values of the relative pressure and the average container surface temperature, depending on the amount of the absorbed hydrogen, is shown in Figure 3.

Following the absorption of 200 litres of hydrogen into the La$_{0.85}$Ce$_{0.15}$Ni$_5$ alloy, weighing 3.125 kg, which lasted 1,270 seconds, the temperature increased in 32.9 °C and the pressure increased in 2.2 MPa. After the hydrogen supply was stopped, the temperature additionally increased due to residual hydrogen absorption.

During hydrogen absorption without cooling, the temperature significantly increased and the pressure increased up to the value close to the permissible maximum limit determined for the HBond©500 container, i.e., 2.5 MPa. It should be noted that the HBond©500 container facilitates storage of maximum 500 litres of hydrogen, so the critical pressure values are usually reached as soon as the container becomes filled in 40 % (at the average hydrogen flow rate of 9.4 l·min^{-1}).

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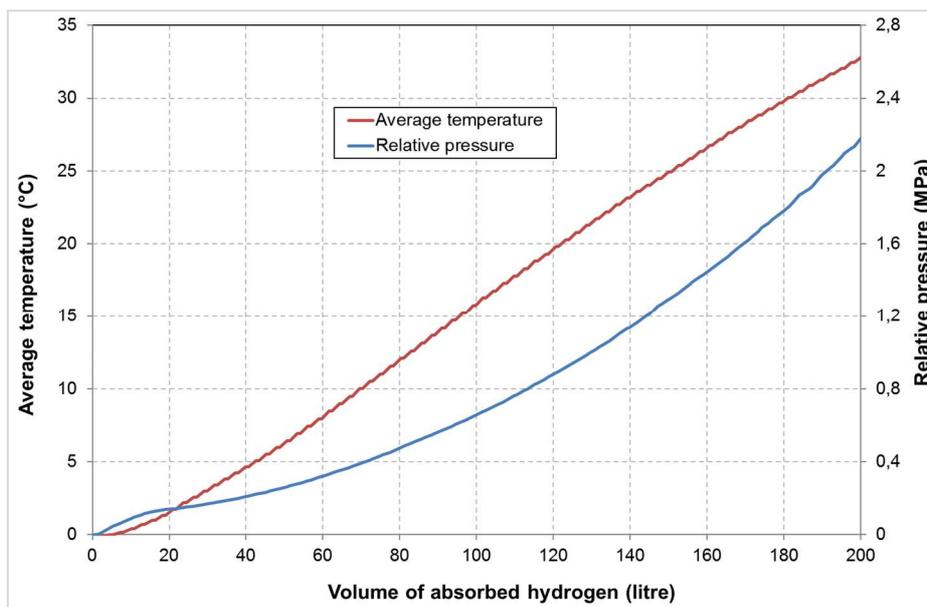
Figure 3 Curves of relative pressures and average container surface temperatures, depending on the amount of the absorbed hydrogen

4 Hydrogen absorption into a metal hydride container with surface cooling

In the second part of the experimental measurements we performed the absorption of 200 litres of hydrogen into an alloy while applying the surface cooling using two PTs arranged as shown in Fig. 1.

The lowest temperature, as to the container surface, was observed on the external cylindrical surface, i.e., at the place where the container is thermally attached to aluminium exchangers. This resulted in the increased concentration of the stored hydrogen, especially in the close vicinity of the external cylindrical surface of the container. However, a disadvantage of the shell cooling is the minimum heat removal from the area along the container axis, as was also indicated by a significant increase in pressure, comparable to the pressure observed when the cooling was not applied. The development of the average surface temperature and the relative pressure of the container, depending on the volume of the absorbed hydrogen, is shown in Figure 4.
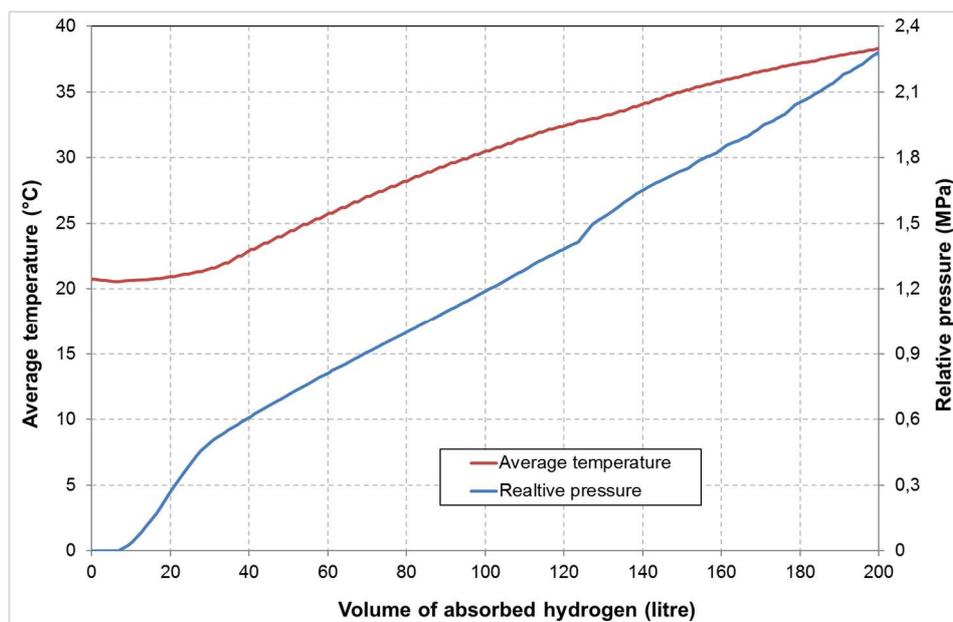
Figure 4 Curves of average surface temperatures and relative pressures of the container, depending on the volume of the absorbed hydrogen

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The total increase in the surface temperature, following the absorption of 200 litres of hydrogen, was 17.6 °C; this value was much lower than the one observed during the absorption process without cooling and was primarily caused by the surface cooling without having a more significant impact on the internal thermal field, as indicated by the above mentioned relative pressure in the container.

Figure 5 depicts the development of temperatures on the cold and hot sides of the PT measured by T_9 a T_{10} temperature sensors; the difference between the temperatures was a decisive parameter for the determination of the cooling power, as shown in the graph in Figure 1.
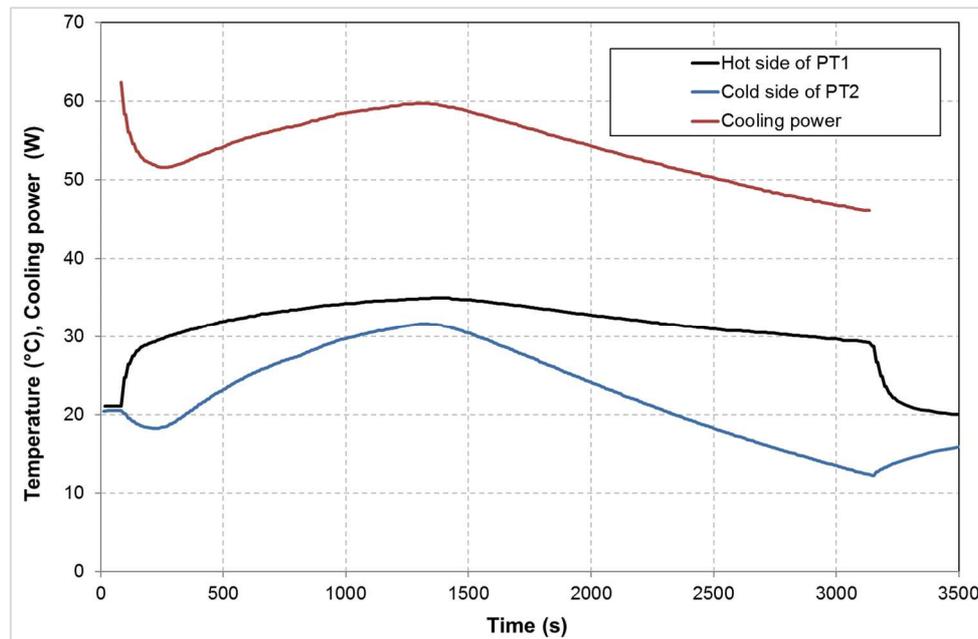
Figure 5 Curves of temperatures on the cold and hot sides of the PT measured by T_9 and T_{10} temperature sensors

The average cooling power of the PT was 108 W (2·54 W); this means that if the average input power is respected, the coefficient of performance of Peltier thermopiles COP cooling is 0.68.

5 Hydrogen absorption into a metal hydride container with the circulation cooling based on the principle of Peltier thermopiles

In the third part of the experimental measurements we performed the absorption of 200 litres of hydrogen into an alloy while applying the circulation cooling using a PT-based cooler.

When compared to the previously applied surface cooling, this cooling method differs in the fact that the water cooled in the external cooler comprising four PTs TEC1-12710 enters the metal hydride container through the inlet of the internal spiral heat exchanger; it is therefore the circulation internal cooling. During the whole period of

performing the measurements, the flow rate of the cooling water was 1.18 l·min^{-1} at the maximum performance of the circulation pump. The flow rate value was limited by a small internal diameter of the piping inside the spiral exchanger. The average initial surface temperature of the container was 19.5 °C. Hydrogen absorption began in second 100 and 200 litres of hydrogen were supplied into the container in second 1,400.

The cooling power of the external cooler was identified applying an indirect method, i.e., measurements of the water flow rate, input and output water temperature, with subsequent application of the calorimetric equation.

The maximum cooling power removed from the alloy during hydrogen absorption was 118 W and the internal source of heat generated during the absorption reached the average value of 145 W; this resulted in a gradual increase in the average temperature of the container (Figure 6).

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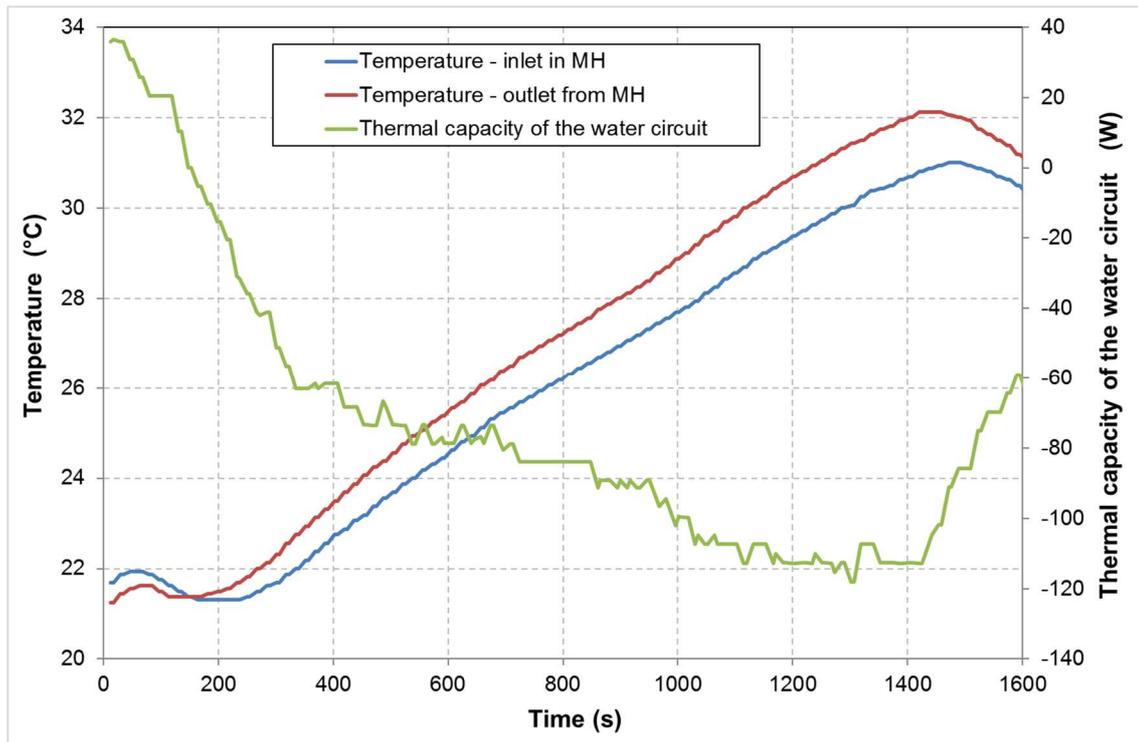
Figure 6 Curves of temperatures and thermal capacities removed by the cooling water

During the absorption of 200 litres of hydrogen into the intermetallic structure with concurrent circulation cooling, the temperature increased in 14.5 °C. With an increasing temperature, the relative pressure of hydrogen increased up to the value of 1.41 MPa (Figure 7).
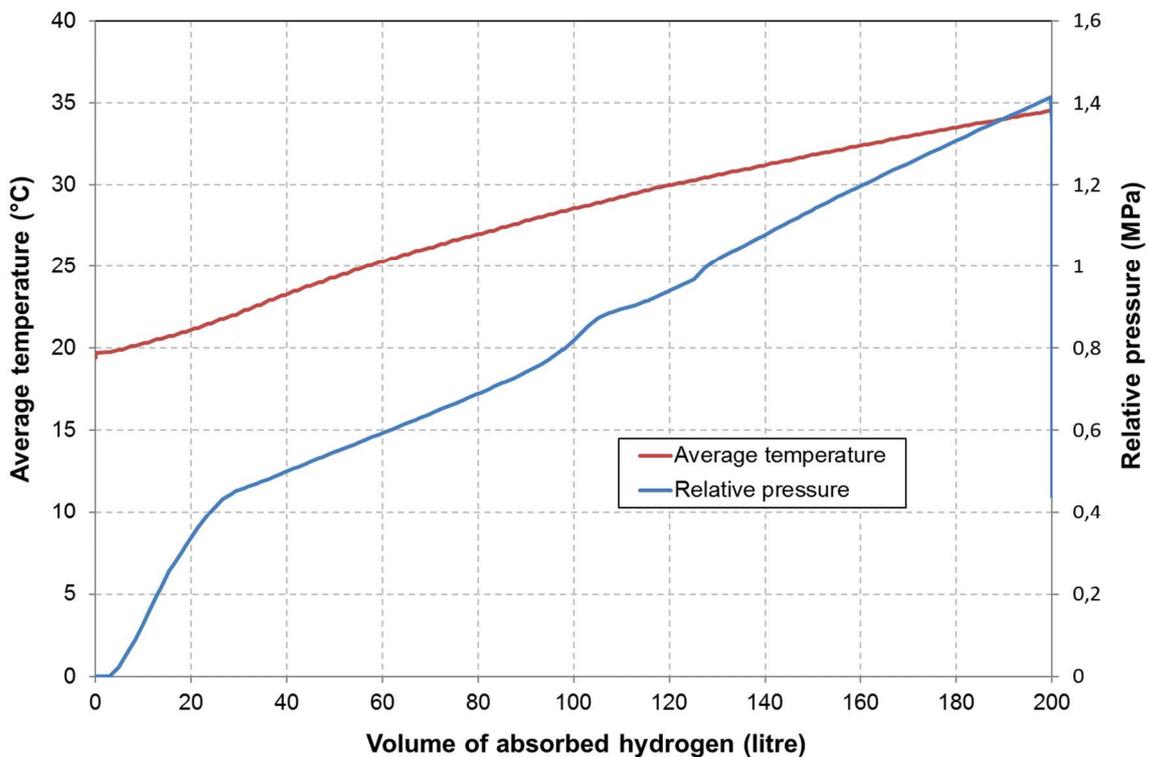
Figure 7 Curves of average temperatures and relative pressures of hydrogen, depending on the volume of the absorbed hydrogen

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We may state that when the circulation cooling of the container was applied, the operating parameters of the absorption improved due to the decreased temperature and relative pressure. This was a result of low thermal conductivity of powder metal hydride with which the even distribution of heat-transfer surfaces in the entire volume is more advantageous for heat removal, when compared to the surface cooling. Obviously, increased cooling power was caused by higher performance of four PTs in the circulation cooler, when compared to two PTs applied in the surface cooling.

6 Hydrogen absorption into a metal hydride container with combined circulation and surface cooling methods

In order to point out potential intensification of the cooling effect and subsequent improvement of operating parameters during hydrogen absorption into the alloy, we performed the measurements while combining the surface and circulation methods of cooling the container during hydrogen absorption. During the measurements, the cooling water flow rate was the same as in the previous case, i.e. 1.18 l·min⁻¹.

Temperatures of water at the inlet to and outlet from the MH container and the values of cooling power are shown in Figure 8. The maximum power with the combined cooling was only 85 W.

The equality of the internal heat source and the amount of the heat removed per time unit may also be depicted as the curve of average container surface temperatures, depending on the absorbed quantity of hydrogen (Fig. 9) which was almost constant. With a constant container temperature, we may only expect a minimum increase in pressure with an increasing mass concentration of the absorbed hydrogen. However, the real pressure increase was rather high and at the end of absorption of 200 litres of hydrogen it reached the value of 1.18 MPa.
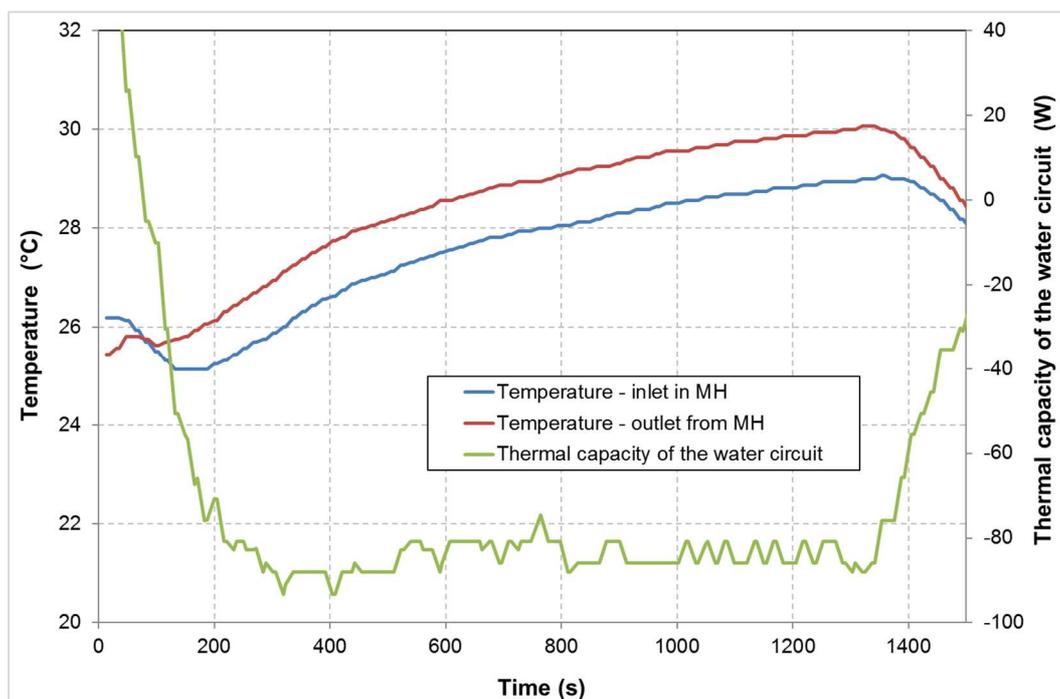
Figure 8 Curves of temperatures and thermal capacities removed by cooling water in the combined cooling of the MH container

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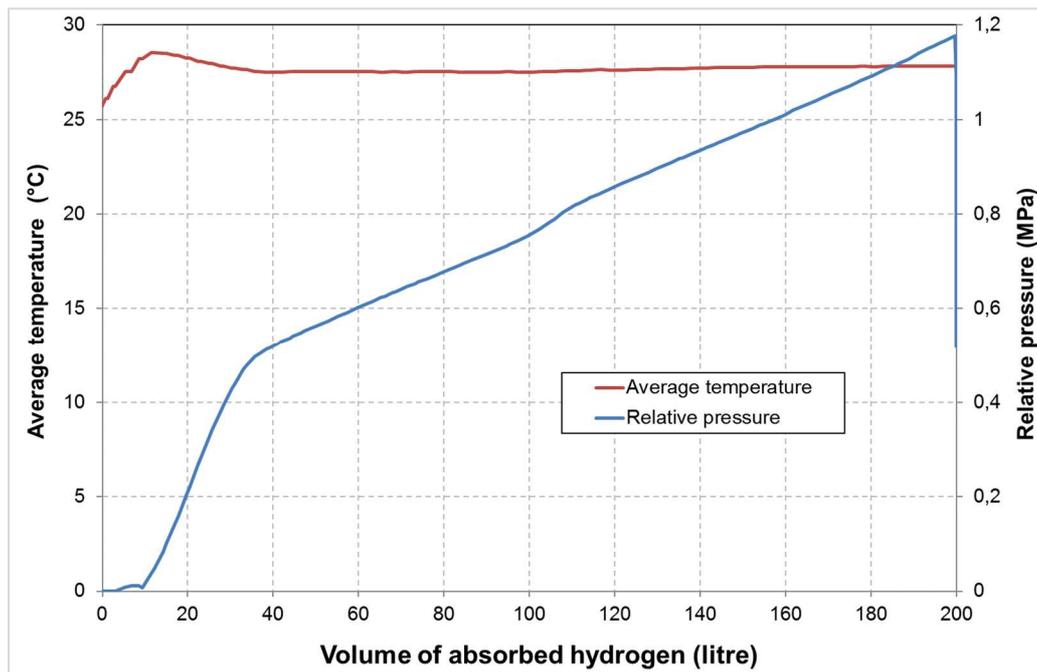
Figure 9 Curves of average temperatures and relative pressures of hydrogen, depending on the volume of the absorbed hydrogen

This was caused by the fact that cooling was concentrated primarily on the surface of the container where the temperature sensors were located; however, along the container axis and at its terminal parts the temperature is expected to rise and, accordingly, the relative pressure would rise too. Nevertheless, the pressure increase was the lowest out of all performed experimental measurements. Moreover, it seems that certain problems may also arise as a result of low thermal capacity of cooling of the circulation heating which is caused by mutual effects of the two independent cooling systems.

7   Conclusion

On the basis of the above described experimental measurements performed in the process of cooling the MH container we may state that due to low thermal conductivity of powder alloys the worst alternative is the surface cooling in which the increases in pressure and temperature observed during hydrogen absorption were the highest.

The circulation cooling resulted in significant pressure decrease. However, there is a potential of optimising the shape of the spiral heat exchanger aimed at increasing the quantity of the heat removed from the terminal parts of the container. The lowest increase in pressure was observed when we combined the surface and the circulation cooling methods; this, however, resulted in a significant increase in the electric energy consumption by the cooling systems.

Chemically bound energy of 200 litres of the stored hydrogen, derived from its combustion heat, was 0.71 kWh. Article shows the energy consumption for individual methods of cooling the MH container, including the percentages of the consumed electric energy required for cooling the container of the total energy contained in hydrogen. The switch from the surface cooling to the circulation cooling resulted in a significant increase in the consumption of energy required for cooling, not to mention the combined method in which the reached values of pressure and temperature were very good, but as much as over 27 % of the stored energy were used for cooling the container.

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