Final Draft
of the original manuscript:

Economic potential of complex hydrides compared to conventional hydrogen storage systems

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Abstract

Novel developments of materials for solid hydrogen storage show promising prospects. Complex hydrides exhibit great technical potential to store hydrogen in an efficient and safe way. Nevertheless, so far an evaluation of economic competitiveness is still lacking. In this work, an assessment about the economic feasibility of implementing complex hydrides as hydrogen storage materials is presented. The cost structure of hydrogen storage systems based on $NaAlH_4$ and $LiBH_4$ / MgH_2 is discussed and compared with the conventional high pressure (700 bar) and liquid storage systems. The vessel construction for the complex hydride systems is much simpler than for the alternative conventional methods because of the milder pressure and temperature conditions during the storage process. According to the economical analysis, this represents the main cost advantage of the complex hydride systems.

Keywords

Economic analysis, hydrogen storage, metal hydride, $LiBH_4$ / MgH_2, $NaAlH_4$, reactive hydride composites

List of symbols

Symbol	Dimension	Description
a	(€ or \$) / kg H_2	Dilation factor
A_{cycl}	cycles	Minimum number of cycles
b	-	Exponential factor
k	kg sm / kg H_2	Hydrogen capacity factor as unit mass of total storage material (sm) per unit mass of stored hydrogen (H_2)
c_{ad}	€ / kg ad	Cost function of additive material (ad)
$C_{dep, TS}$	€ / kg H_2 / cycle	Depreciation of the tank system cost per cycle
c_E	€ / kWh	Energy cost
C_{HP}	\$ / kg H_2 / cycle	Total cost of hydrogen processing per cycle
C_{H_2}	\$ / kg H_2 / cycle	Total cost of the hydrogen gas per cycle
c_{H_2}	€ / kg H_2 / cycle	Cost of hydrogen gas per cycle
C_{SM}	\$ / kg H_2 / cycle	Total cost of the storage materials per cycle
c_{sm1}	€ / kg sm1	Cost function of storage material 1 (sm1)
c_{sm2}	€ / kg sm2	Cost function of storage material 2 (sm2)
C_T	\$ / kg H_2 / cycle	Total refuelling cost per cycle
C_{TS}	\$ / kg H_2 / cycle	Total cost of the tank system per cycle
c_{TS}	€ / kg H_2	Cost of tank system
E_{HP}	kWh / kg H_2 / cycle	Energy demand for hydrogen processing per cycle
l_{TS}	-	Factor for experience curve effect
p_{ad}	kg ad / kg sm	Ratio factor additive material (ad) as unit mass of additive material per unit mass of total storage material
p_{sm1}	kg sm1 / kg sm	Ratio factor storage material 1 (sm1) as unit mass of storage material 1 per unit mass of total storage material
p_{sm2}	kg sm2 / kg sm	Ratio factor storage material 2 (sm2) as unit mass of storage material 2 per unit mass of total storage material
x	kg H_2	Mass of stored hydrogen
x_0	kg H_2	Constant assumed to be 1 kg of H_2 for scaling purposes
x_{ad}	kg ad	Total share of additive material (ad)

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X_{min}	-	Minimum size of series
$X_{min,0}$	-	Initial size of series
x_{sm1}	kg sm1	Total share of storage material 1 (sm1)
x_{sm2}	kg sm2	Total share of storage material 2 (sm2)

List of indexes

Index	Description
liq	Liquid storage system
lm	Lithium boron hydride - magnesium hydride storage system
p	Pressure storage system
sa	Sodium alanate storage system

1. Introduction

It is widely accepted that the reserves of fossil fuels are continuously diminishing and, accordingly, the effort to convert energy resources into energy reserves will increase strongly [1]. This circumstance will have a major influence on our economy and daily life and might even lead to international conflicts. Therefore, a timely investigation of alternatives is necessary. Hydrogen technology could be one promising alternative [2, 3]. The use of renewable energy sources like wind, water or sun, the storage and distribution by hydrogen and the conversion to electric power by fuel cells, makes hydrogen economy conceivable. Meanwhile, improvements in all fields (supply, distribution and conversion) are necessary.

One of the major challenges is the storage of hydrogen. Because of the low energy density of hydrogen gas at room temperature and pressure, specific energy densities comparable to conventional fossil fuels are impossible. The most widely used alternatives are hydrogen stored at high pressures (≤ 700 bar) or liquefied at very low temperatures (-253 °C). These alternatives require a highly developed storage system at high costs, as it will be shown. Moreover, the safety aspect, with respect to the temperature and pressure, is another motivation to find alternative storage techniques.

Different metals and compounds have the ability to bond hydrogen and to form a hydride or a complex compound. These chemical reactions are in many instances reversible and need milder temperature and pressure conditions than the conventional storage systems mentioned above. In addition, the specific energy density of these hydride materials is very high [4].
Hydrogen storage systems based on different metal hydrides have been known for some years. In the past 20 years, several new promising materials have been discovered. Among them, the alanates investigated by Bogdanovic and Schwickardi [5] in 1997, as well as Reactive Hydride Composites (RHC's), discovered by Barkhordarian et al. [6] and Vajo et al. [7] some seven years later, are of particular interest. The results on these complex hydrides have shown a great technical potential so far, but it is yet unknown to what extent they are also economically competitive.

This paper provides a first concept and the associated essential key data and assumptions to compare the different technologies. A first comparison of the conventional high pressure and liquid storage systems with complex hydride systems is shown. As representatives of alanate and RHC families, sodium aluminium hydride ($NaAlH_4$) and lithium boron hydride / magnesium hydride ($LiBH_4$ / MgH_2) are investigated.

It should be mentioned here that previous studies have been carried out, however, they are based on different assumptions [8], the data meanwhile are out-dated [9-13] or the works have not been comprehensively published in peer-reviewed journals [14]. The data available come very often from presentations, which disclose only partial aspects of the models. A comparison with the official DoE target for storage system cost [15] is also not significant since the current value is under review. Therefore, any comparison with the present study is problematic.

2. The model

2.1. Cost structure

The model was developed with the aim of giving an answer to the following question: What would be the total costs of one refuelling process in a mobile application of a certain size using different

hydrogen storage techniques? In the proposed model, the total refuelling costs per cycle is composed of the following four cost fractions:

⇒ Costs of hydrogen gas C_{H_2}
⇒ Costs of hydrogen processing C_{HP}
⇒ Depreciation costs of the tank system C_{TS}
⇒ Depreciation costs of the storage materials C_{SM}

The sum of these four fractions forms the total refuelling cost per cycle C_T (eq. 2.1):

$$C_T = C_{H_2} + C_{HP} + C_{TS} + C_{SM} \qquad (2.1)$$

It is important to mention that the major part of the information about the costs was obtained in Euros since this work was carried out in Germany. Despite this fact and in order to provide a more straightforward comparison for the international field, the result of the economic analysis is expressed in US-\$[1].

2.2. Assumptions

2.2.1. General storage criteria

The model is based on the following assumptions to define the general criteria for the storage systems:

- construction as on-board system with an integrated fuel cell application
- manufacturing under the terms of a serial production
- minimum size of series X_{min} = 100 units
- initial size of series $X_{min,0}$ = 10 units
- hydrogen supply at ambient pressure for the loading
- hydrogen pressure after the unloading between 2 and 10 bar
- minimum number of cycles A_{cycl} = 1,000 cyles

The minimum and initial size of series opens the opportunity to include later cost effects for underdeveloped technologies. The minimum life expectancy, demonstrated by the minimum number of cycles [15], represents the basis for depreciations. To simplify the analysis, the depreciations are linear and neither inflation-adjusted nor charged with interest.

2.2.2. Evaluation criteria

In addition, a few fundamental assumptions for the evaluation criteria are necessary. They are mainly based on the theory that the relation between the relative price of an economic good and the ordered quantity is generally dominated by the quantity-discount-relationship. Hence, it is assumed that the amount of stored hydrogen has the most important influence on the total cost due to the correlation with the ordered quantity (e.g. storage material or tank system material). Therefore, the storage capacity x, in kilograms of hydrogen, is defined as the only variable factor. Moreover, different applications should be considered in the analysis. However, the applications are only distinguished on the basis of their amount of stored hydrogen. The field of observation is therefore defined between 0.1 and 10 kg of H_2. Only within this field of observation cost drivers are considered. This consideration is another simplification and implies that not each cost structure is taken into account in detail.

In many cases, only few data points regarding the costs are known and an interpolation is necessary. For all functions in this analysis, the power law function (eq. 2.2) has been selected for interpolations:

$$f(x) = a \cdot \left(\frac{x}{x_0} \right)^b \qquad (2.2)$$

The constants a and b are calculated by fitting to the available data. The constant factor x_0 is 1 kg H_2. If possible, all fittings are done including values outside of the field of observation. This ensures that the conclusions reached inside the field of observation are significant and can be used for realistic estimations. The power law function has been selected because the strong depression at the beginning corresponds to the characteristic fixed cost degression known in other economic scenarios and the slowly stagnation at the end, to the approximation to the variable costs as marginal cost.

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[1]based on currency rate from 27th of January 2011 (1 US-\$ ≡ 0,729 €)

One of the major challenges of this analysis is the economic evaluation of energy. In this work the energy is consistently evaluated by a German industrial consumer price for 1 kWh of electric energy: $c_E = 0.1$ € / kWh ($0.14 / kWh) [16].

2.3. Equations

In the following, the calculations for the four cost terms C_{H_2}, C_{HP}, C_{TS} and C_{SM} from equation 2.1 are described.

The cost of the hydrogen gas, C_{H_2}, is equal for all the considered storage systems and could therefore also be neglected for a pure comparison. However, to give a clear answer to the previous question concerning the refuelling cost in 2.1, this part is also included. The total cost of the hydrogen gas C_{H_2} corresponds to the hydrogen cost function c_{H_2} as shown in equation 2.3:

$$C_{H_2}(x) = c_{H_2}(x) \tag{2.3}$$

The hydrogen cost function is based on a current quote requests, done with two gas suppliers, and expected future cost of hydrogen produced by wind energy [17] as marginal cost. The maximum offered pressure of the quote requests was 300 bar (minimum purity 99.9 % at a purchase quantity up to 13 kg of H_2). The average marginal cost of the above mentioned requests represents the cost at low ordered quantities due to the fact that the hydrogen gas should be actually supplied at standard pressure in this analysis. The parameters a_{H_2} and b_{H_2} are shown in Table A.1 in appendix A.

The total cost of hydrogen processing C_{HP} is calculated by the multiplication of the energy cost c_E and the energy demand for hydrogen processing E_{HP} (eq. 2.4):

$$C_{HP} = c_E \cdot E_{HP} \tag{2.4}$$

The total cost of hydrogen processing is the only cost fraction in this analysis without dependency on the amount of hydrogen due to the intensive nature of the energy demand for hydrogen processing and therefore not written as a function of the storage capacity.

The total tank system cost function C_{TS} (eq. 2.5) equates the depreciation of the tank system cost $C_{dep,TS}$ (eq. 2.6) corrected by the experience curve effect [18]:

$$C_{TS}(x) = C_{dep,TS}(x) \cdot \left(\frac{X_{min}}{X_{min,0}} \right)^{\frac{\log(I_{TS})}{\log(2)}} \tag{2.5}$$

The linear depreciation of the tank system cost $C_{dep,TS}$ is calculated by equation 2.6, with the minimum life expectancy A_{cycl} as basis:

$$C_{dep,TS}(x) = \frac{c_{TS}(x)}{A_{cycl}} \tag{2.6}$$

The tank system cost function c_{TS} follows the power law function mentioned above (eq. 2.2). The constants a and b are fitted for each storage system. The experience curve effect implies that the value added costs of a product fall by 20 - 30 % for each doubling of the cumulative production volume. The correction term in equation 2.5 is a modified formula from Baum et al. [18] who provide also a simplified derivation. The correction with the experience curve effect is necessary because of different data for the tank systems. For instance, data from storage systems without a sophisticated production status in terms of a series production (e.g. solid storage systems) can be therefore corrected to compare these data with systems at a more developed production status (e.g. pressure storage systems).

The total storage materials cost C_{SM} is relevant only for the solid state storage systems, and is based on the amount of each storage material. The solid systems in the analysis rely on two storage material components reacting with hydrogen, and one additive material as reaction rate enhancer. The overall storage material results from the mass balance for each stoichiometric reaction. For instance, in the case of sodium alanate (sa), the reaction and its compounds are shown in equation 2.7 [19]:

$$2 \text{ NaH} + 2 \text{ Al} + 3 \text{ H}_2 \xrightleftharpoons{\text{TiCl}_4} 2 \text{ NaAlH}_4 \tag{2.7}$$

For the sodium alanate system sodium hydride (NaH) is the storage material 1, aluminium (Al) is the storage material 2 and titanium (IV) chloride ($TiCl_4$) is the precursor for the additive material [20]. The required quantity of each material is calculated by equations 2.8 - 2.10. The ratio p (following from the mass balance) is multiplied by the capacity factor k and the storage amount of hydrogen x:

$$x_{sm1}(x) = p_{sm1} \cdot k \cdot x \tag{2.8}$$

$$x_{sm2}(x) = p_{sm2} \cdot k \cdot x \tag{2.9}$$

$$x_{ad}(x) = p_{ad} \cdot k \cdot x \tag{2.10}$$

Through the share factors of equation 2.8 - 2.10, multiplied by the initial size of series $X_{min,0}$, the storage material costs c_{sm1}, c_{sm2} and c_{ad} are calculated. The multiplication of the initial size of series considers the purchasing cost under the assumed series initial conditions. These costs are multiplied by the share factors again and then added. The summation is multiplied by the inverse of the amount of hydrogen and the inverse of the number of cycles. Thereby the dimension of the total storage material cost function (eq. 2.11) is dollar per kilogram hydrogen and the cost is again expressed as depreciation for one cycle:

$$C_{SM}(x) = \left(\frac{1}{x}\right) \cdot \left(\frac{1}{A_{cycl}}\right) \cdot \left(\sum_{i=sm1}^{ad} c_i(x_i \cdot X_{min,0}) \cdot x_i\right) \tag{2.11}$$

The parameters for the cost functions and the share factors for the storage materials are again calculated for each material. The capacity factor is calculated for each storage system. Costs of processing the material, in particular for high energy milling, are neglegible due to the low energy consumption of the milling process, which is originated in the high efficiency of the device and the short processing time [21]. In addition, some recent concepts in the area of metal hydride synthesis, especially regarding plastic deformation, could lead to further remarkable energy savings in this field [22, 23].

3. Model Calculations

3.1. Pressure storage system

Traditional pressure tanks are made of austenitic stainless steels [24]. However, the tensile strength of these materials is not high enough to withstand 700 bar of pressure. Therefore, novel tanks, like for instance the ones shown in the StorHy-Project [25], use a combination of materials. The basic construction is made from two materials: An inner-liner material that blocks the hydrogen from diffusing out and an outer-liner material that provides the tensile strength. In addition to the tank, the storage system consists of loading and unloading devices (e.g. filling coupling), safety devices (e.g. fusible safety plug) and control devices (e.g. pressure regulator).

It is assumed that the main cost drivers for the total cost of a pressure storage system over its life cycle are only the cost of hydrogen processing, the cost of the tank system and the hydrogen gas cost.

For the loading of a pressure storage system with a final pressure of 700 bar, a 25 % overpressure is necessary [26]. For the compression up to 875 bar, a standard piston compressor can be used [24]. However, for the calculation of the necessary energy a simplified adiabatic compression with an isentropic efficiency factor $\eta_{s,V}$ of 0.9 [27] is assumed.

The calculation is shown in appendix B. The total energy demand for hydrogen processing E_{HP}^{p} is 4.4 kWh / kg H_2. The calculation is based on thermodynamic data from the NIST database by using the software REFPROP[®] 9.0.

The cost of a pressure reducing regulator for the unloading process is neglected. For the tank system, it is assumed that the main cost driver is the pressure vessel. It is in particular relevant for storage systems with a capacity higher than 1 kg of H_2 (which corresponds to 90 % of the analysis).

The outer shell mostly consists of carbon fiber composite material. The inner-liner can be made of different materials like aluminium, steel or polyamide. The weight proportion of the inner-liner is only around 25 wt.% of the total tank. The main part and also the most expensive one is the outer load carrying shell and particularly the carbon fibre [25]. Based on quotes from producers, purchasers and other stakeholders, the fitting parameters for the cost function of a 700 bar tank system are ascertained. The parameters a_{TS}^p and b_{TS}^p, the respective standard errors and the coefficient of determination are shown in Table A.1 in appendix A.

Most of the data obtained from these quotes are based on series production aspects. Therefore, an experience curve effect is not expected: $l_{TS}^p = 1.0$.

Taking into account the described parameters, the total cost function for a pressure storage system (eq. 3.1) of 700 bar can be written as follows:

$$C_T^p(x) = \left[a_{H_2} \cdot x^{b_{H_2}}\right] + \left[c_E \cdot E_{HP}^p\right] + \left[\frac{a_{TS}^p \cdot x^{b_{TS}^p}}{A_{cycl}} \cdot \left(\frac{X_{min}}{X_{min,0}}\right)^{\frac{\log(l_{TS}^p)}{\log(2)}}\right] = \left[a_{H_2} \cdot x^{b_{H_2}}\right] + \left[c_E \cdot E_{HP}^p\right] + \left[\frac{a_{TS}^p \cdot x^{b_{TS}^p}}{A_{cycl}}\right] \qquad (3.1)$$

3.2. Liquid storage system

For a liquid storage system, is essential to keep an extremely low temperature by an excellent isolation. The construction of this isolation follows the principle of a Dewar vessel. The inner-jacket encloses directly the liquid gas, whereas the outer-jacket encloses the entire tank. The area between the inner- and outer-jacket is evacuated (vacuum: 10^{-5} - 10^{-6} mbar) to eliminate convective heat transfer. Furthermore the inner-jacket is wrapped with aluminium foil and glass fibre [28] to avoid heat transfer by radiation. Previous tanks used stainless steel for the inner- and outer-jacket. Novel tanks are made of aluminium, replacing the steel and thus achieving up to 50 % weight reduction. Further weight reductions are currently under investigation by using materials based on fibre-reinforced polymer composites with a copper coating [25]. The liquid storage system, in a similar way as the pressure storage systems, includes loading and unloading, safety and control devices.

It is assumed that the main cost drivers for the total cost of a liquid storage system, besides the hydrogen gas cost, are the costs of hydrogen processing and of the tank system. The cost of the storage materials is not applicable.

The energy demand for hydrogen processing, i.e. liquifaction in this case, $E_{HP}^{liq} = 15.2$ kWh / kg H_2 is taken from Züttel [24]. This is almost half of the energy content (lower heating value) of the hydrogen molecule. Because of the boil-off losses, the pressure is always slightly above atmospheric pressure, therefore a substantial pressure regulating valve for the unloading process is not necessary.

It is very difficult to determine the cost of such a storage system since there are no official informations accessible. Novel tanks are mostly produced as unique examples in manual work. The level of automation is estimated as 3 % [29]. The only available data sources are unofficial cost estimations that vary from 20,000 to 100,000 € ($27,440 to $137,200) for one vehicle tank system (~ 4 kg H_2) [30]. These cost estimates are extremely high because of the low level of automation during the manufacturing.

For this analysis, the average of this estimates at $20,580 (15,000 €) per kilogram for a 4 kg storage system is used. It is assumed that the cost can be reduced by $9,604 (7,000 €) per kilogram for a 50 kg storage system and by another $ 4,116 (3,000 €) per kilogram for a 100 kg storage system. Thereby, the parameters a_{TS}^{liq} and b_{TS}^{liq} for the tank system cost function (Table A.1) are calculated.

Owing to the potential to increase the level of automation by a higher quantity production and therewith a real potential of cost reduction, a first experience curve effect of $l_{TS}^{liq} = 0.8$ is selected.

Due to these parameters the total cost function for a liquid storage system (eq. 3.2) can be expressed as follows:

$$C_T^{liq}(x) = \left[a_{H_2} \cdot x^{b_{H_2}}\right] + \left[c_E \cdot E_{HP}^{liq}\right] + \left[\frac{a_{TS}^{liq} \cdot x^{b_{TS}^{liq}}}{A_{cycl}} \cdot \left(\frac{X_{min}}{X_{min,0}}\right)^{\frac{\log\left(l_{TS}^{liq}\right)}{\log(2)}}\right] \tag{3.2}$$

3.3. NaAlH₄ storage system

The basic construction of a metal hydride storage system resembles that of a heat exchanger. The tubes filled with the storage material are typically surrounded by a heat transfer medium [31]. Other configurations use cooling tubes inside a hydride filled pressure resistant hull [32, 33]. The heat transfer medium controls the loading or unloading process: the heat of reaction that is produced by the absorption of hydrogen during the loading process is discharged and the heat of reaction that is necessary for the desorption of the hydrogen during the unloading is provided. Thus, the use of a metal hydride storage system makes sense if a heat source, like an engine or a fuel cell, is available to supply heat at the appropriate temperature level. According to the heat of reaction, the systems can be classified in low, medium and high temperature systems. The temperature and pressure conditions depend on the storage material, thus also the design and the type of heat transfer medium. First results for magnesium hydride [34, 35] have shown a high correlation between the heat transfer medium and the reaction time (corresponding to the loading and unloading time). For this analysis only research systems are considered, because commercial products for middle and high temperature hydrides have not been developed so far.

With an operational temperature of 125 °C - 160 °C, the sodium aluminium hydride (sodium alanate NaAlH₄) storage material belongs to the group of medium temperature hydrides. It is assumed that the main cost drivers are the costs of hydrogen gas and processing as well as the depreciation cost of the tank system and the storage materials.

For the NaAlH₄ storage material, a temperature of 125 °C and a H_2 pressure of 100 bar is used during the absorption process [19]. The calculation of the energy necessary for the compression follows what was applied above for the pressure storage system. According to equation B.3 in appendix B the total energy demand for hydrogen processing is $E_{HP}^{sa} = 3.04 \text{ kWh / kg } H_2$. The desorption pressure can be at 3.75 bar by creating a suitable backpressure [19]. Costs for a compressor during discharging are therefore not applicable.

For the tank system costs, it is assumed that the main cost drivers are the heat exchanger and the storage material vessels. In comparison to the pressure tank vessel, these vessels can be produced at much lower cost because of the lower operating pressure and the resulting possibility of using lower cost tank vessel materials. Around 80 % of the system costs of the so far produced research storage systems resulted from the heating system, and in particular from the thermostat. These costs are neglected in the present calculations due to system integration aspects with the fuel cell.

For the tank system cost function, two different research tanks are considered that were developed in our laboratories. In both cases, the heat exchanger enclosed the storage material vessel in a tube-and-shell arrangement [31]. The first design was a prototype single tube storage tank for approximately 100 g of H_2 and costs of ~ 900 € ($1,234). The second system was designed in cooperation with the Hamburg University of Technology and comprised 8 kg NaAlH₄ [31], with a H_2 capacity slightly less than 0.5 kg. In additions, the costs for a capacity of 50 kg of H_2 are fitted.

The resulting parameters a_{TS}^{sa} and b_{TS}^{sa} for the tank system cost function are shown in Table A.1. Because of the single-unit production, similar to the liquid storage tank, a potential of cost reduction due to series production of $l_{TS}^{sa} = 0.8$ is expected.

The basic reaction of the storage material was already described in equation 2.6. The starting materials are sodium hydride (storage material 1) with a ratio factor of $p_{sm1}^{sa} = 0.437$, aluminium (storage material 2) with $p_{sm2}^{sa} = 0.492$, and 5 mol-% of titanium (IV) chloride (additive) with $p_{ad}^{sa} = 0.071$. The ratio factors are calculated by the mass balance of equation 2.6 plus the 5 mol-% of the additive material. For each storage material or additive, a request for a cost estimation was sent to different suppliers. Based on these quotes, the parameters a_{sm1}^{sa} and b_{sm1}^{sa} for the storage material 1, a_{sm2}^{sa} and b_{sm2}^{sa} for the storage material 2 and a_{ad}^{sa} and b_{ad}^{sa} for the additive are defined (Table. A.1). Quotes were requested for quantities at gram, kilogram and ton scale, however these were not received in all cases.

Experimentally, a storage capacity of ~ 3.5 wt.% was reached [19]. Hence, the factor of storage capacity is calculated to be $k^{sa} = 28.57$. There were no additional measurable losses during the cycling of the material [19].

Taking into account these parameters, the total cost function for a NaAlH$_4$ storage system (eq. 3.3) can be calculated by:

$$C_T^{sa}(x) = \left[a_{H_2} \cdot x^{b_{H_2}}\right] + \left[c_E \cdot E_{HP}^{sa}\right] + \left[\frac{a_{TS}^{sa} \cdot x^{b_{TS}^{sa}}}{A_{cycl}} \cdot \left(\frac{X_{min}}{X_{min,0}}\right)^{\frac{\log(l_{TS}^{sa})}{\log(2)}}\right] + \left[\left(\frac{1}{x}\right) \cdot \left(\frac{1}{A_{cycl}}\right) \cdot \left(\sum_{i=sm1}^{ad}\left(a_i^{sa} \cdot \left(x_i^{sa} \cdot X_i\right)^{b_i^{sa}}\right) \cdot x_i^{sa}\right)\right] \qquad (3.3)$$

3.4. LiBH$_4$ / MgH$_2$ storage system

With an operational temperature of 350 °C - 400 °C, the lithium boron hydride / magnesium hydride (LiBH$_4$ / MgH$_2$) storage material belongs to the group of high temperature hydrides. In LiBH$_4$ / MgH$_2$, Mg and B react exothermally upon desorption of hydrogen, thus compensating for the required endothermal energy for desorption. Accordingly, hydride systems like this have been named "Reactive Hydride Composites (RHC)" [37, 38].

Analogue to the NaAlH$_4$ storage system, it is assumed that the cost drivers for the total cost of a LiBH$_4$ / MgH$_2$ storage system are all four cost fractions.

For LiBH$_4$ / MgH$_2$, a temperature of 350 °C and a H$_2$ pressure of 50 bar is used [39] during the absorption process. Using the formulae from the pressure storage system, the total energy demand for hydrogen processing is calculated as $E_{HP}^{lm} = 2.74$ kWh / kg H$_2$.

For the desorption pressure, it is known that a minimum backpressure of a few bar of hydrogen is necessary to avoid the formation of elementary boron and concomitant loss in capacity [38, 40, 41]. Like for the other storage systems, additional cost of hydrogen processing (e.g. recompression due to low delivery pressure) during the discharging process can therefore also be neglected.

In the same way as for the NaAlH$_4$ system, it is assumed that the main cost drivers for the tank system costs are the heating and the storage material vessel.

For the tank system cost function, no commercial data are available. There is only one experimental test tank constructed by us that can be used for the calculations. The construction is comparable to the NaAlH$_4$ storage system mentioned above. The costs for this prototype tank system are ~ 3,400 € ($4,663). The theoretical storage capacity is well below 100 g. For storage capacities up to 50 kg of H$_2$ it is assumed again that the production cost can be reduced to at least $2,744 (2,000 €) per kilogram. Hence, the parameters a_{TS}^{lm} and b_{TS}^{lm} for the tank system cost function (Table A.1) can be calculated.

For the LiBH$_4$ / MgH$_2$ storage system, a cost reduction potential of $l_{TS}^{lm} = 0.8$ is also expected. The basic reaction of the storage material can be described by equation 3.4 [39]:

$$2 \text{ LiH} + \text{MgB}_2 + 4 \text{ H}_2 \xleftrightarrow{\text{Nb}_2\text{O}_5} 2 \text{ LiBH}_4 + \text{MgH}_2 \qquad (3.4)$$

The starting materials are lithium hydride (storage material 1) with a ratio factor of $p_{sm1}^{lm} = 0.21$, magnesium (II) boride (storage material 2) with $p_{sm2}^{lm} = 0.606$ and 5 mol-% of niobium (V) oxide (additive) with $p_{ad}^{lm} = 0.184$. These ratio factors are calculated again by the mass balance of equation 3.4. The same requests as for the NaAlH$_4$ system were also set in place to define the parameters a_{sm1}^{lm} and b_{sm1}^{lm} for the storage material 1 (lithium hydride), a_{sm2}^{lm} and b_{sm2}^{lm} for the storage material 2 (magnesium (II) boride) and a_{ad}^{lm} and b_{ad}^{lm} for the additive (niobium (V) oxide) in Table A.1.

The material showed an experimental reversible capacity of 7 wt.% [39]. Other additives have led to capacities above 9 wt.% [42] but they were not as cost effective. Therefore the factor of storage capacity of $k^{lm} = 14.29$ is calculated. Until now, measurable losses during the cycling of the material were not observed for this system.

The total cost function for a LiBH$_4$ / MgH$_2$ storage system (eq. 3.5) can thus be calculated by:

$$C_T^{lm}(x) = \left[a_{H_2} \cdot x^{b_{H_2}}\right] + \left[c_E \cdot E_{HP}^{lm}\right] + \left[\frac{a_{TS}^{lm} \cdot x^{b_{TS}^{lm}}}{A_{cycl}} \cdot \left(\frac{X_{min}}{X_{min,0}}\right)^{\frac{\log\left(l_{TS}^{lm}\right)}{\log(2)}}\right] + \left[\left(\frac{1}{x}\right) \cdot \left(\frac{1}{A_{cycl}}\right) \cdot \left(\sum_{i=sm1}^{ad}\left(a_i^{lm} \cdot \left(x_i^{lm} \cdot X_i\right)^{b_i^{lm}}\right) \cdot x_i^{lm}\right)\right] \qquad (3.5)$$

4. Results

Figure 1 shows the comparison of the results from equation 3.1, 3.2, 3.3 and 3.5 with the deduced parameters from this work. However, for a better understanding, the respective total refuelling costs are multiplied by the storage capacity. Hence, the result is shown as absolute cost and the unit is $ per cycle (instead of $ per kilogram hydrogen per cycle).

Fig. 1: Total refuelling cost per cycle against the storage capacity

The NaAlH$_4$ storage system has the lowest cost per cycle, whereas the liquid system has the highest, and, interestingly, this is independent from the storage capacity. The total refuelling cost functions for the pressure storage system and LiBH$_4$ / MgH$_2$ system are almost equal. The pressure system is slightly more favourable than the LiBH$_4$ / MgH$_2$ system up to a theoretic break-even-point at 16.38 kg H$_2$ (beyond the field of observation). Apart from the liquid system, the average cost of one refuelling process is between $4 and $75 for applications with capacities between 100 g and 10 kg of H$_2$. For instance, for an automotive application (capacity ~ 4 kg H$_2$, which is then about comparable to the range of current vehicles based on fossil fuels) a refuelling process can cost between $36.87 and $73.79 (including the liquid storage system). Thereby, it is shown in Figure 2 that the depreciation of the tank system has the greatest share of the total cost at 4 kg of H$_2$, besides the hydrogen cost. Hydrogen processing and storage material costs are rather less important.

Fig. 2: Cost distribution of the total refuelling cost per cycle for each storage system at a storage capacity of 4 kg of H$_2$

This relation persists over the considered range from 100 g to 10 kg of H$_2$. The absolute value for a refuelling process for an automotive application based on a NaAlH$_4$ storage system of approximately $37 equals $0.28 per kWh.

5. Discussion

5.1. Field of observation

The field of observation and thus the scope of this analysis was defined from 100 g to 10 kg of H$_2$. The cost trends inside this field, in accordance with the quantity-discount-relationship and following the power law function, are considered as realistic. However, the field of observation at lower capacities should be evaluated in a different way. The assumptions of the main cost drivers in this area are too unprecise. For example: Besides the vessel material, the almost constant cost of supply units like pipe connections, safety installations and control devices become more important with decreasing capacity. In this area, the analysis is therefore critical. However, the taken assumptions were chosen in order to cover a wide range of the analysis.

5.2. Operation and additional technically required cost

Operation costs have been defined here as the cost of external energy to the overall storage system. Almost no operation costs have been therefore expected for all storage systems: the pressure storage system requires only a pressure reduction valve that operates without any external energy supply. The complex hydride storage system requires external energy to release the hydrogen, but this is assumed to be supplied by the fuel cell integrated with the storage system. The feasibility of this operation mode without external energy has been shown first through simulation by Pfeifer et al. [43] and then experimentally by Urbanczyk et al. [32]. The liquid storage system requires energy during the operation as well. To use the hydrogen in gaseous form, it is necessary to evaporate the liquid hydrogen. The thermal energy of evaporation can be supplied by the fuel cell, as in the case of the hydride storage system. It can also be supplied by a heat exchanger that works with the ambient air. In both cases there are no costs associated with the supply of this energy, since it is freely available. On the other hand the permanent evaporation inside the liquid hydrogen tank leads to what we call additional technically required cost. These costs, due to the so called "boil-off losses" of the system, strongly depend on the duration of standstill or idle periods, when the vehicle is not used; and the

quality of the heat insulation of the system. Naturally, the duration of the standstill periods is particularly difficult to predict for every user. The influence of these costs to the total costs depends also on the economical evaluation of the losses. This can be done in a straightforward way by using the cost of hydrogen and considering the lost hydrogen as hydrogen that has to be bought in addition to the regular required amount. However, this approach cannot be used here, firstly, because it would mean that the "boil-off" costs are proportional to the costs of hydrogen, which was left out of this analysis, secondly, because we have no way at present to predict the idle time of the storage systems. Thus, in the present study, the boil-off losses of the liquid hydrogen storage system have simply been neglected. Due to this boil-off in real life, liquid hydrogen storage systems will be even more disadvantageous economically than shown in Fig. 1 and 2.

5.3. Potentials and challenges

For calculation of some tank system data, certain assumptions and estimations have been made. However, in all cases the assumptions are made carefully and in a transparent way for the discussion. The cost distribution (Fig. 2) illustrates the potentials and challenges for each storage system. The main cost fraction for all considered systems is the tank system cost, which is dominated by the vessel cost. The pressure storage system benefits independently of the storage capacity from the fact that it does not incur storage material costs. On the other hand, this limits the possible fields for improvement to hydrogen processing and tank system cost. Since the hydrogen processing costs are always present and are difficult to reduce, because of a physical minimum of work that needs to be carried out in the compression, only the tank system costs remain for further cost reductions. Thus, the cost of carbon fibre in the future will have a great influence on the total cost of this storage method. However, the historical evolution of carbon fibre cost does not show much promise for reduced cost for the pressure storage system due to the ongoing increase in demand [44].

The main challenges of the liquid storage system are the processing and tank system costs. According to the previous evaluation, the hydrogen processing costs cannot be avoided. In view of this background, liquid storage will always have difficulties to compete in the future. This holds despite the high potential for cost reduction for the tank system, because of a less mature development status in comparison with the compressed gas systems.

The assumptions for the complex hydride systems arise from first experience with systems at laboratory scale and it can therefore be assumed that the actual cost will likely be significantly lower for a serial production. The complex hydride storage systems have the advantage of their lowest tank system cost. As seen above, the tank system costs of the $NaAlH_4$ system are below the costs of the $LiBH_4 / MgH_2$ system. This fact can be ascribed to the higher operation temperature of the $LiBH_4 / MgH_2$ storage system. Nevertheless, for both hydride tank systems, the design is simpler and the construction materials are less costly than for the pressure and liquid hydrogen alternatives. It is conceivable that this cost advantage is also transferable to other metal hydride systems. In any case, the $NaAlH_4$ system has the lowest cost per cycle of all the considered storage systems.

6. Conclusion

Focusing on one refuelling process, a $NaAlH_4$ storage system has an economical advantage over a conventional high pressure (700 bar) or liquid storage system. This is due to a simpler structure of the tank vessel, which can be achieved because of the lower pressure and more moderate temperature conditions. A $LiBH_4 / MgH_2$ storage system also benefits from this advantage, but due to higher depreciation of the storage material and the tank system, the cost function is almost equal to the pressure storage function. Among the different alternatives, the hydride-based systems have the highest potential for further cost reduction.

7. Outlook

From an economic perspective, the research efforts on hydrogen storage in metal hydrides need to focus particularly on the tank development. According to the trends identified in the present study, improvements in the cost effectiveness of hydrogen storage tanks should endorse metal hydrides and in particular the complex hydride systems $LiBH_4 / MgH_2$ and $NaAlH_4$ as economically highly promising storage methods.

For further economic analysis of the complex hydride system at larger scale, it is necessary to gather more experience. In addition, more accurate data about the tank system costs, especially of liquid hydrogen storage systems, need to be collected for more detailed comparisons. More precise data would also enable a comparison with other energy storage systems like accumulators or batteries. However, an important precondition would be a reliable cost estimate of hydrogen gas based on the

real manufacturing cost plus a realistic profit margin. Then, hydrogen economy could finally also be compared to the main competitive energy carrier: the fossil fuels.

Acknowledgments

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Appendix A

Table A.1: Fitting parameter

Appendix B

A simplified thermodynamic calculation is shown to give a reasonable estimate on the energy input which is necessary for the compression of the hydrogen. From a thermodynamic point of view, the change in enthalpy and entropy of a fluid necessary to reach a given end state is independent of the way this state is reached. If mechanical power and heat fluxes are calculated, these process dependent values are influenced by the specific process steps involved. For this calculation a two step compression with intermediate cooling is chosen, as shown in Fig. B.1. This is to avoid high temperatures where high cost materials might be necessary for the equipment.

Fig. B.1: Process chain of a two-stage compression with 3 cooling processes

Each compression stage is followed by a cooling process. A possible heat recovery of these two cooling processes is neglected, the heat flux is discarded to the environment. The final cooling process is necessary for the precooling of the gas before it is loaded to the tank. Otherwise, there will be local overheating because of the poor heat conductivity of the liner materials [25].

Therefore, the calculation of the total energy demand for hydrogen processing E_{HP}^P includes the mechanical compression energy $E_{HP,12}^P$ of step 1 and $E_{HP,34}^P$ of step 2, and additional mechanical or electrical energy to cool down the gas $E_{HP,56}^P$ to a temperature of -80 °C.

$$E_{HP}^P = E_{HP,12}^P + E_{HP,34}^P + E_{HP,56}^P \tag{B.1}$$

Initially, the hydrogen is compressed to 50 bar in step 1 and finally to 875 bar in step 2. The energy which is necessary for these compression steps is calculated using the first law of thermodynamics. The resulting energy balance (kinetic and potential energy is neglected) [27] is given in equation B.2:

$$\frac{dE}{dt} = 0 = \sum Q + \sum P + \sum_{in} \dot{m}_{in} \cdot h_{in} - \sum_{out} \dot{m}_{out} \cdot h_{out} \tag{B.2}$$

Assuming a stationary state and an adiabatic compression process, the derivative of the energy with respect to time and the heat flow are zero. The engine of the piston compressor is the only considered power source ($P = E_{HP,nm}^P$), and we assume a constant mass flow $\dot{m}$ of 1 kg / h over the entire process ($\dot{m}_{in} = \dot{m}_{out}$). For a single compression stage, equation B.2 can be therefore simplified to:

$$E_{HP,nm}^P = \dot{m}(h_m - h_n) = \frac{\dot{m}}{\eta_{s,V}}\left(h_m^{is} - h_n\right) \tag{B.3}$$

The isentropic efficiency $\eta_{s,V}$ is defined as $\eta_{s,V} = \left(h_m^{is} - h_n\right)/\left(h_m - h_n\right)$ and gives the ratio of the ideal isentropic enthalpy change necessary for a given compression step as compared to the true enthalpy change in reality. In this context h_m^{is} is the isentropic enthalpy $h_m^{is} = h(T_m, s_m = s_n)$. The isentropic compression efficiency is arbitrarily assumed to be $\eta_{s,V} \overset{!}{=} 0.9$ [27].

The necessary energy for the compression of the first stage $E_{HP,12}^P$ is 2.74 kWh / kg H_2 and 1.88 kWh / kg H_2 for the second stage $E_{HP,34}^P$.

The final cooling of the hydrogen gas is thought to be performed by a reversible refrigeration cycle. The coefficient of performance (COP) for such an ideal refrigeration cycle [27] is given as:

$$COP_{rev} = \frac{Q_{HP,56}^P}{P_{rev,56}} = \frac{T_0}{T_{amb} - T_0} \tag{B.4}$$

Where $T_0 = 193\,K$ is the lowest thermodynamic temperature of the cold side, T_{amb} is the ambient temperature ($T_{amb} = 298\,K$). Using the COP and again the first law of thermodynamics (eq. B2) for the calculation of the heat flow $Q_{HP,56}$ the cooling power ($E_{HP,56} = P_{rev,56}$) can be calculated by:

$$E^p_{HP,56} = \frac{\dot{m}(h_6 - h_5)}{COP_{rev}}$$

(B.5)

Thus the mechanical or electrical power needed for cooling down the hydrogen in the last process step is (at least) 0.24 kWh / kg H_2.

Following this calculation based eq. B.1, the total mechanical (or electrical) energy demand for hydrogen processing E^p_{HP} is 4.4 kWh / kg H_2. The values for the enthalpy and entropy used in this calculation are from the software REFPROP® 9.0 (NIST).

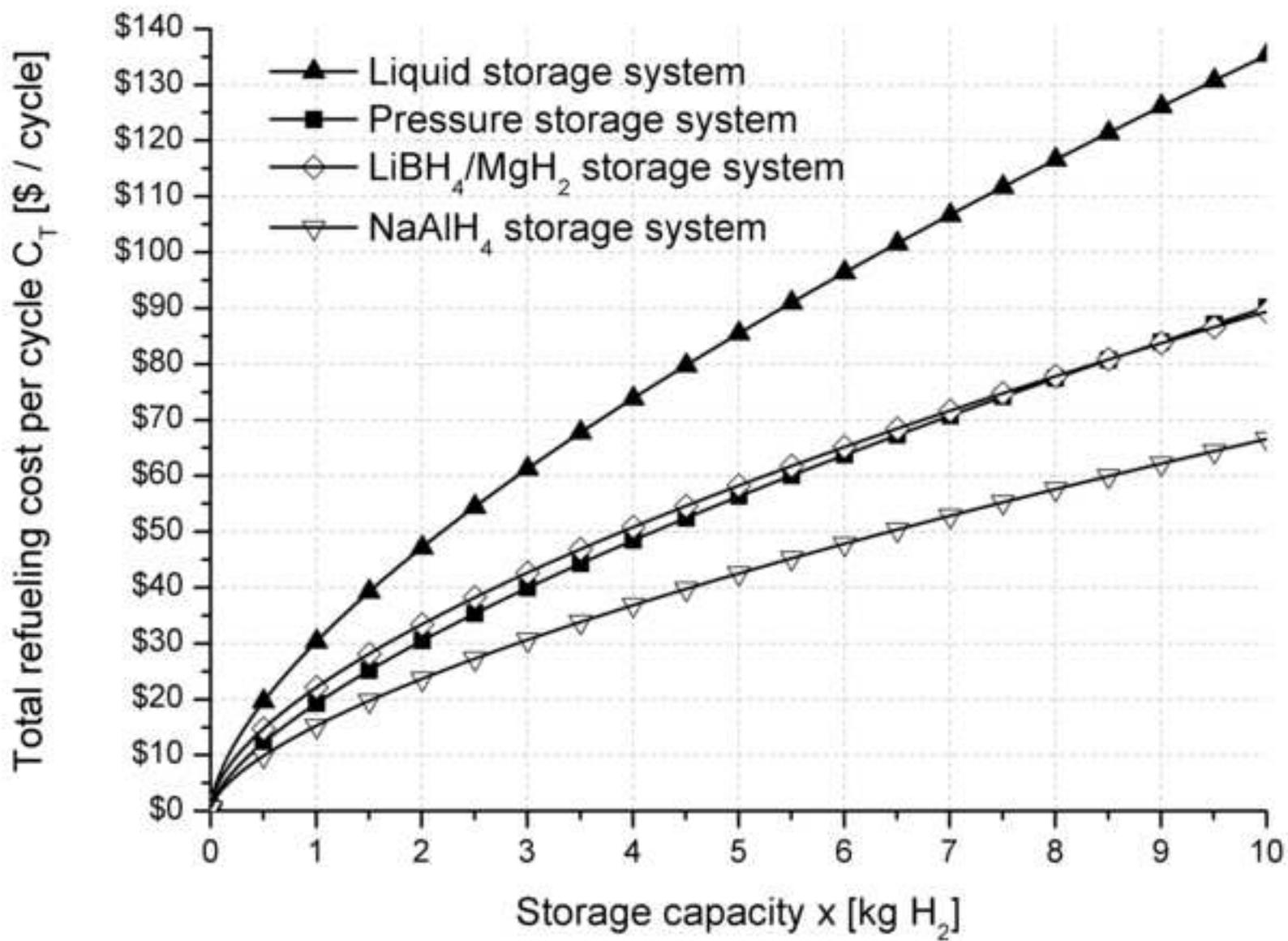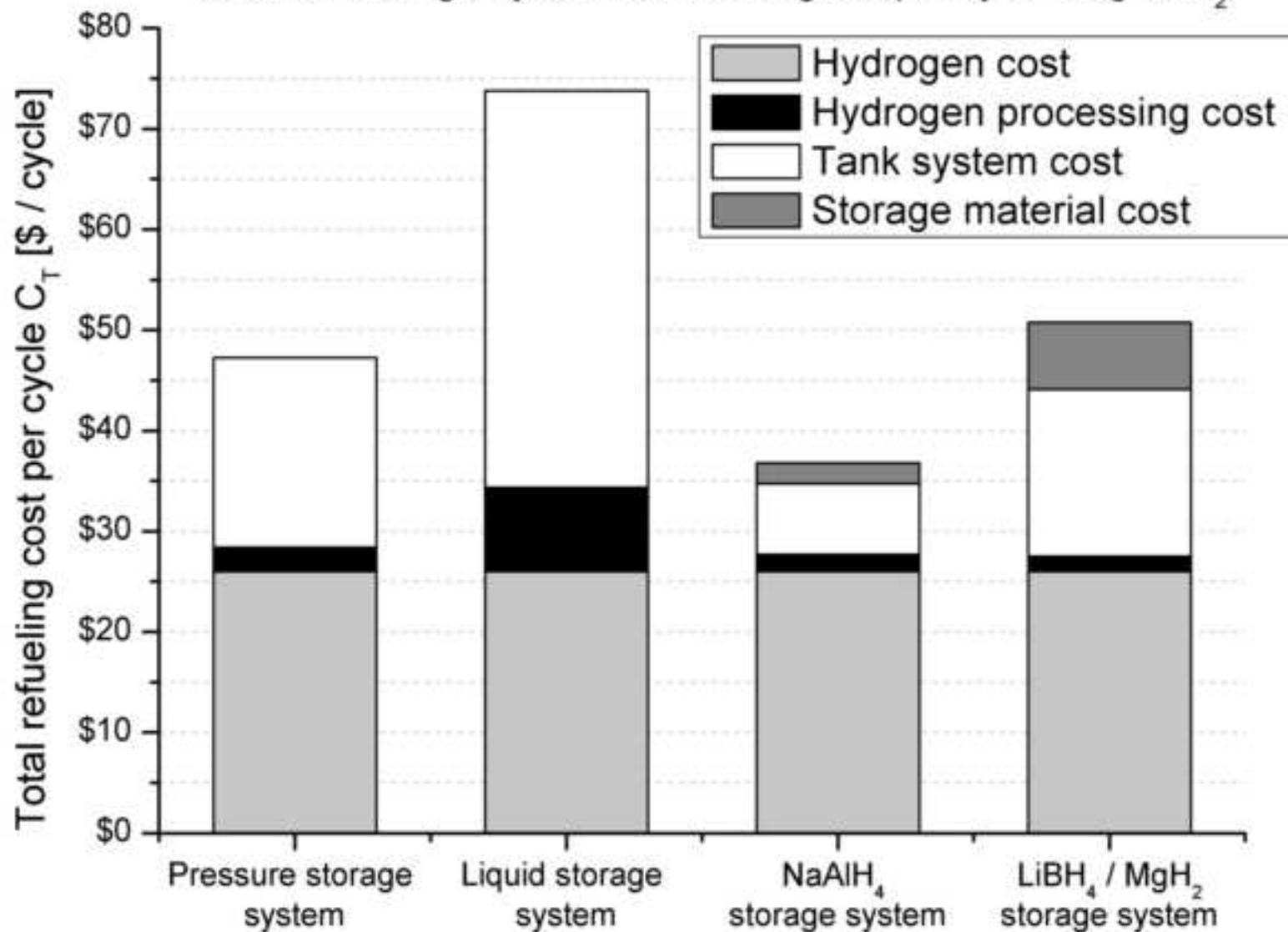Fig. 1: Total refuelling cost per cycle against the storage capacity

Fig. 2: Cost distribution of the total refuelling cost per cycle for each storage system at a storage capacity of 4 kg of H_2
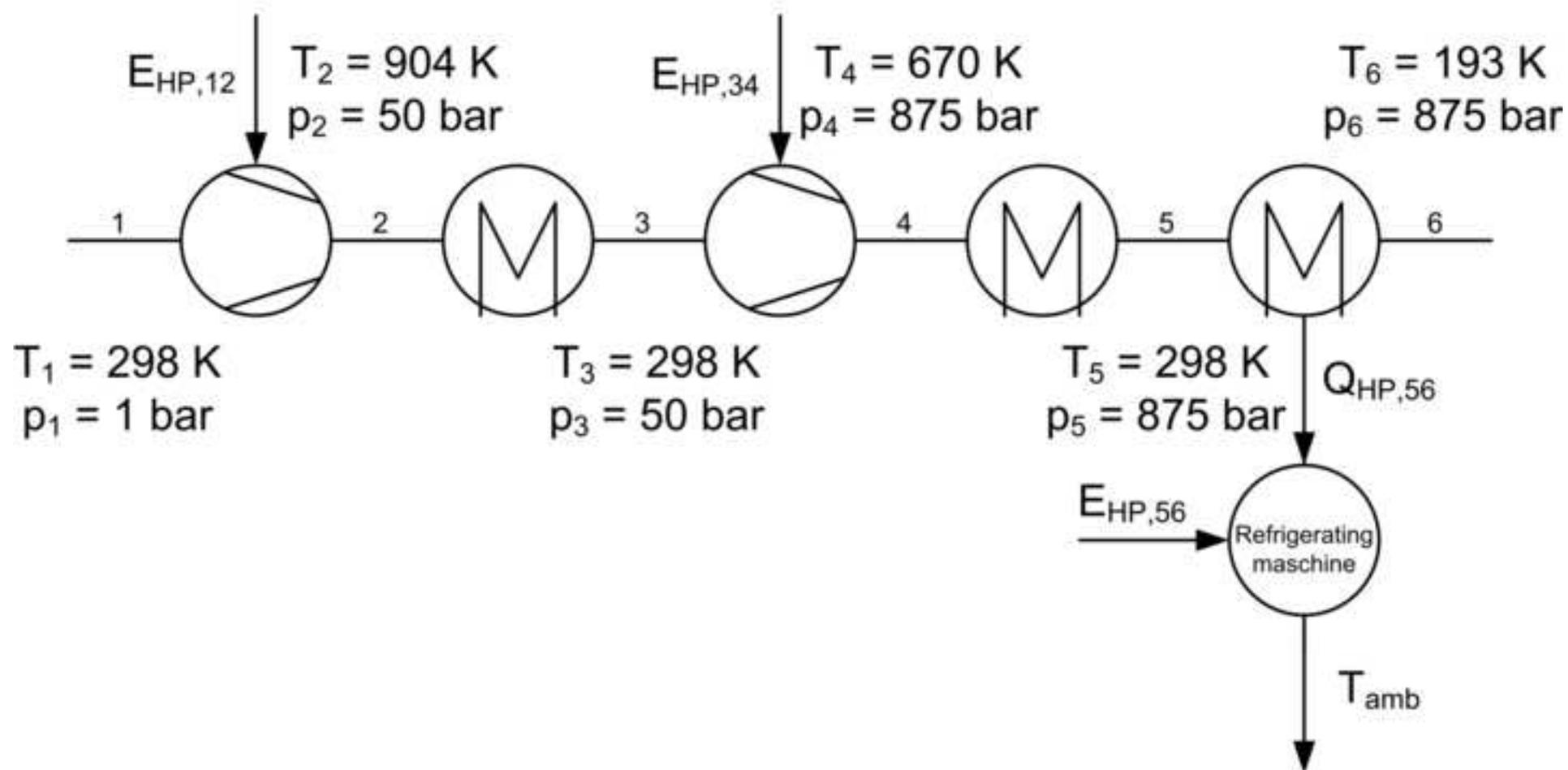
Fig. B.1: Process chain of a two-stage compression with 3 cooling processes

Table A.1: Fitting parameter

Parameter	Value	Standard error	Coefficient of determination
a_{H_2}	8.14 € / (kg H_2)^2	0.23	0.99914
b_{H_2}	-0.389	0.013	
a_{TS}^{p}	5192.11 € / (kg H_2)^2	1411,43	0.37898
b_{TS}^{p}	-0.299	0.175	
a_{TS}^{liq}	25,964.23 € / (kg H_2)^2	2993.53	0.97002
b_{TS}^{liq}	-0.392	0.053	
a_{TS}^{sa}	4134.39 € / (kg H_2)^2	914.73	0.89787
b_{TS}^{sa}	-0.307	0.108	
a_{sm1}^{sa}	340 € / (kg H_2)^2	-[1]	0[1]
b_{sm1}^{sa}	-0.464	-[1]	
a_{sm2}^{sa}	64.31 € / (kg H_2)^2	1.57	0.99649
b_{sm2}^{sa}	-0.3	0.022	
a_{ad}^{sa}	33.32 € / (kg H_2)^2	0.065	1
b_{ad}^{sa}	-0.587	8.869E-4	
a_{TS}^{lm}	11,956 € / (kg H_2)^2	-[1]	0[1]
b_{TS}^{lm}	-0.457	-[1]	
a_{sm1}^{lm}	447.43 € / (kg H_2)^2	22.75	0.9149
b_{sm1}^{lm}	-0.203	0.06	
a_{sm2}^{lm}	2,121 € / (kg H_2)^2	-[1]	0[1]
b_{sm2}^{lm}	-0.707	-[1]	
a_{ad}^{lm}	317.83 € / (kg H_2)^2	26.22	0.8124
b_{ad}^{lm}	-0.151	0.049	

[1]No standard error / coefficient of determination because only 2 data points for the interpolation were available.

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