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Temperature distribution in a new composite material for hydrogen storage – Design study of different cooling concepts

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***Abstract*— A newly developed composite material has been explored based on metal hydrides in combination with polymers enriched with highly porous carbon. As metal hydride, a RHC (reactive hydride composite) was chosen (e.g., $MgH_2 + 2\ LiBH_4$). The hydride is infiltrated into the pores of the porous carbon suppressing the long-range phase separation of the two different hydrides by nano-confinement. The aim is to maintain fast kinetics and achieve cycle stability of the RHC (reactive hydride composite). The combination of RHC and porous carbon is then integrated into a polymer film to allow an easy and safe handling of the material. To produce a storage system out of such a film, the thin material is rolled in the same style like a rolled membrane module; i.e., it is rolled together with a thin spacer (e.g., steel mesh) allowing an easy hydrogen access to all parts of the membrane. The last step is the implementation of the rolled storage module into the tank shell. To analyze different design concepts and the behavior of this newly developed composite storage material, extensive FEM-simulations have been realized for different cooling structures. The latter is necessary to fulfil the thermodynamic requirements and to maximize the speed of hydrogen storage. Therefore, the temperature development within the storage during hydrogen feeding are investigated. Beside this, the hydrogen flow as well as the kinetics of the chemical reaction are analyzed. Based on such extensive simulations of different design concepts, the most promising overall storage systems are developed and systematically optimized. Finally, the total hydrogen content of the overall storage system is calculated and compared between different design concepts. Based on this, conclusions are drawn about robust criteria how to construct a cooling and heating device for this new storage material.**

Keywords — cooling concept, hydrogen storage, metal hydride storage, temperature development

I. INTRODUCTION

Hydrogen storage is an important option to integrate higher shares of renewable energy within our energy system, since hydrogen can easily be stored on the long-term and in large quantities (e.g., power-to-gas). This properties of hydrogen are also essential for the use of hydrogen in mobile applications – especially if fast refueling, long range and/or high power are required (e.g., heavy-duty traffic). Thus, for hydrogen storage several concepts have been and are under development. Currently, pressurized storage devices are most widely used at different pressure levels (e.g., 200, 350 or 700 bar). Also liquid hydrogen, metal hydrides and liquid organic hydrogen carrier (LOHC) are discussed for a wide range of applications including the transportation sector (Barthelemy et al., 2017; Baetcke and Kaltschmitt, 2018).

In particular, metal hydrides seem to be a quite promising option for the future. Metal hydrides can be used for several applications (e.g., hydrogen compressor, hydrogen purification, hydrogen storage) (d'Entremont et al., 2017; Eftekhari and Fang, 2017). But typically metal hydrides are mainly assessed as a hydrogen storage material due to the high volumetric energy density at low pressure.

Therefore, numerous different concepts for an efficient hydrogen storage based on metal hydrides have been developed in recent years. The storage concept assessed here is a further development based on some of these preliminary studies. It consists of a combination of different materials to optimize cycle stability and to improve reaction kinetics. Thus, the storage concept proposed here consists of four main components (Fig. 1): a composite storage foil, a spacer, a tank shell and a heating jacket. The composite storage foil consists of a polymer, nano-confined metal hydrides and highly porous carbons. These three elements are further processed into a

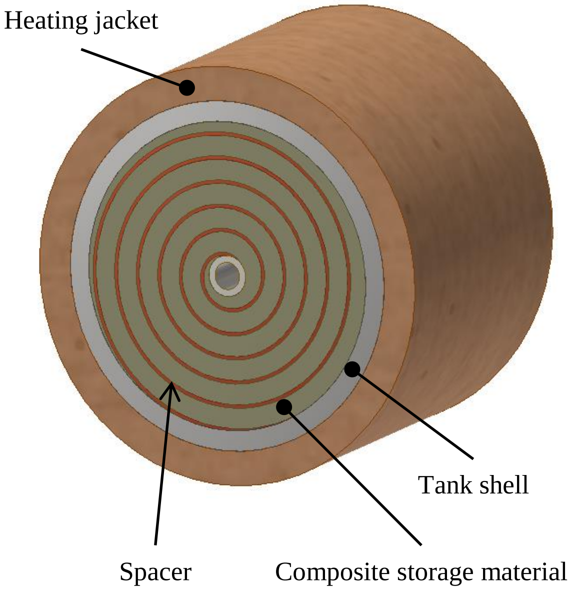


Fig. 1: Radial cut through the storage

polymer foil. This foil is called composite storage material representing the active part of the hydrogen storage. The spacer material consists of a simple metal mesh. The composite storage foil together with the spacer is wrapped to a spiral. Inside this spiral, the spacer ensures a fully hydrogen supply at all parts of the composite storage material. The overall spiral is located inside the tank shell. Around the tank shell, a heating jacket is wrapped to control the temperature inside the overall tank. This assembly of the storage and the special configuration of the composite storage material leads to a material combination with very specific properties (Ziemann, 2017; Hasse, 2016).

One important aspect while using metal hydrides as a storage material for hydrogen is the temperature development within the storage system. This is crucial since metal hydride need a constant temperature span for the desired hydrogenation reaction (Jepsen, 2014). Therefore, the simulations presented below analyze the effect of three different temperature control concepts within the metal hydride storage tank; namely these are fins, pipes and long spacer. These systems are compared to a spiral foil of the composite system using only natural convection at room temperature at the outer tank wall. This is the significant difference of the heat management systems proposed here compared to other studies; so far mainly internal heat exchanger with a forced heat transport medium have been analyzed (Hardy and Anton, 2009a, 2009b; Mohammadshahi et al., 2016a, 2016b; Singh et al., 2017; Zeaiter et al., 2015). Beside this the material which is considered here is unique and was firstly discussed in (Cao et al., 2018); thus this is the first analysis of the heat management of a polymer metal hydride combination.

Against this background, the overarching goal of this paper is to elaborate the best design of a heat transfer structure to control the heat inside the metal hydride storage material. To evaluate this different heat management systems, they will be analyzed during hydrogen adsorption (i.e., loading) including the heating of the storage material to the respective adsorption temperature level. The hydrogen desorption will not be considered separately since the process of desorption is endotherm; i.e., the heat management of the desorption is much simpler and the desorption reaction is more robust and much easier to control. Therefore, the working hypothesis is that the system which fits best to the heating as well as the subsequent hydrogen absorption also fits best for desorption, since from the thermal management point of view the same effects have to be considered for heating as well as for desorption. This is to transport the heat as simple and as fast as possible inside the storage material.

II. BASICS

To simulate the loading of a metal hydride storage with $LiBH_4$+MgH_2 as storage material several effects have to be considered. The loading process consist of two steps: the first step is to heat up the storage material to reaction temperature and the second step is the hydrogen adsorption reaction. These steps influence each other directly because adsorption takes place at around 350 °C and starts when a temperature of 300 °C is reached inside the storage material. Due to the heating jacket located outside of the tank shell the temperature distribution inside the storage is not homogenous. Hence, the starting time of the hydrogen adsorption reaction within the metal hydride is different at various zones of the overall storage. Consequently, directly at the tank wall the adsorption starts first because most likely here are the highest temperatures. The adsorption process ends when the whole material is saturated. Due to the given constraints inside the storage material the overall hydrogen content follows an asymptotical development till the adsorption process is terminated. This happens after approx. 1200 min; i.e., at that time the simulations end. Thus to reach a theoretical fully loaded storage take too much time to be simulated, since the reaction gets very slow at the end.

The chemical reaction is described in equation (2.1). During this exothermic adsorption reaction, a reaction enthalpy of 46 kJ/mol_{H2} is released.

$$2\,LiH + MgB_2 + 4\,H_2 \leftrightharpoons 2\,LiBH_4 + MgH_2 \qquad (2.1)$$

Against this background, the most important physical phenomena defining the hydrogen absorption within a metal hydride based hydrogen storage are the reaction kinetics and the heat management; i.e., these two effects dominate the loading and unloading behavior of the storage. Which of this two phenomena has bigger impact on the storage behavior is analyzed in (Jepsen, 2014) showing that the size of the storage device defines, which of these two effects limiting the reaction velocity of the absorption in metal hydride storage devices. Thus in small quantities (e.g., samples for lab analysis) the kinetic of hydrogen adsorption within metal hydride is determined by the velocity of the adsorption reaction, since it is very easy to regulate the temperature within the whole sample. But in bigger applications (e.g., inside this simulation) the temperature distribution is the limiting factor for the reaction rate. This is caused by the effect that too high and too low temperatures of the metal hydride reduce the reaction rate (Emonts and Stolten, 2016). And, since metal hydrides do not show a high thermal conductivity, the heat cannot be transferred easily in and out of the storage device. Therefore, the metal hydride often overheats during the reaction, this reducing the reaction velocity. This explains why the heat management is the limiting factor for the hydrogen storage device discussed here (Jepsen, 2014).

Another physical phenomena effecting storage ability is the hydrogen flow inside the overall storage. But, the gas flow should not be a limiting factor. Since the material assessed here reacts at a pressure of 50 bar the hydrogen flow through the storage is typically faster than the heat flow or the reaction kinetics. Also, hydrogen diffusion through the polymer is faster than reaction kinetics (Weigelt et al., 2018; Cao et al., 2018). As a consequence, the hydrogen flow can be neglected as a limiting factor. Therefore, during the simulation performed here it is assumed that always enough hydrogen for a full reaction is present at the metal hydride surface.

Against this background, the basics for the reaction kinetics as well as the heat management are discussed in detail below.

A. *Reaction kinetics*

The reaction kinetics of metal hydrides mainly depends on their material properties (e.g., (Jepsen et al., 2018a), (Jepsen et al., 2018b) and (Cao et al., 2018)). The most important characteristic values of the reaction kinetics are the Arrhenius factor A and the activation energy E_a. And, the two most important parameter for the reaction kinetic are the pressure and temperature inside the storage material. They are controlled via the hydrogen valve and the heat management of the storage tank. Here only the adsorption kinetics are described and used for the simulations.

To model the temperature dependence of the reaction rate k_r the Arrhenius equation is used (equation (2.2)). The other determining factors are the universal gas constant *R* and the absolute temperature *T*.

$$k_r = A\ e^{\frac{-E_a}{R\ T}} \tag{2.2}$$

Beside material values and temperature, pressure influences the reaction rate of the material. Thus, the reaction rate is not only defined by equation (2.2). In addition, a driving force *f* is defined which is pressure dependent. The driving force *f* strongly depends on the reaction kinetics for the specific storage material; thus it has to be analyzed for each storage material. One common term for the driving force *f* of metal hydrides is shown in equation (2.3) (Jepsen, 2014).

$$f(p, p_{eq}) = \ln\left(\frac{p}{p_{eq}}\right) \tag{2.3}$$

Equation (2.3) describes the driving force depending on the total pressure *p* and the equilibrium pressure of the reaction p_{eq}. This equation fits well to the measured data of (Jepsen *et al.*, 2018a). Therefore, this term is used to create the overall equation for the reaction rate to be used within these simulations.

Equation (2.4) shows the combined equation for the reaction rate depending on temperature as well as on pressure. It is used to calculate the heat development and the concentration of the different materials inside the storage.

$$rate(T, p) = A\ e^{\frac{-E_a}{R\ T}}\ \ln\left(\frac{p}{p_{eq}}\right) \tag{2.4}$$

B. *Heat management*

The overall heat balance of this hydrogen storage is shown in equation (2.5). It contains the heat losses caused by the natural convection $\dot{Q}_{nat.\ convection}$ at the outer tank shell as well as the heat resulting from the chemical reaction $\dot{Q}_{reaction}$ and the heat provided by the heating jacket put around the hydrogen tank $\dot{Q}_{heating}$.

$$\dot{Q}_{heating} + \dot{Q}_{reaction} - \dot{Q}_{nat.\ convection} = 0 \tag{2.5}$$

Thus, for hydrogen adsorption (i.e., filling of the tank) one heat sink and two different heat sources are considered. This is on the one hand thermal energy coming from the heating jacket located around the tank and the heat resulting from the exothermic reaction of the metal hydride released during hydrogenation of the storage material. On the other hand, heat is lost by natural convection at the tank wall to the surrounding atmosphere.

At the hydrogen tank assessed here two different surfaces to the surroundings have to be considered. On the one hand side the heating jacket including a thermal isolation as well as on the other side the part of the storage device where the steel shell of the tank is directly surrounded by ambient air. The latter are the hydrogen inlet and the closed end of the tank. Thus $\dot{Q}_{nat.\ convection}$ includes the heat losses by heat transfer to the surrounding ambient air of the isolation and the steel shell.

During the heating up of the tank to the reaction temperature the heating jacket is the only heat source. So, the value of $\dot{Q}_{reaction}$ equals zero for the heating part inside the simulations. This constellation is very similar to the desorption behavior, since there the value $\dot{Q}_{reaction}$ is negative and thus it acts as another heat consumer. All three heat flows are shown in Fig. 2.

To calculate the heat transfer and the heat distribution inside the overall storage device the respective characteristic material values for the different parts of the hydrogen storage are used to calculate the heat and temperature development of the different materials as well as the heat transfer between the different materials inside the storage. Here it is taken into consideration that each material has a specific value for thermal conductivity and heat capacity (i.e., the composite material, the spacer, the hydrogen). Additionally, the porosity of the different materials, especially of the spacer, influence the heat flow. This is also taken into consideration.

The excess heat resulting from the storage loading process are calculated according to equation (2.6). This heat released from the adsorption reaction depends mainly on the reaction enthalpy H_r measured in (Jepsen et al., 2018a) and the reaction rate described in equation (2.4).

$$\dot{Q}_{reaction} = rate(T, p)\ H_r \tag{2.6}$$

The heat losses due to natural convection at the outer rim of the overall storage device $\dot{Q}_{nat.\ convection}$ is calculated based on equation (2.7). It depends on the heat transfer coefficient α, the surface area *A* and the temperature difference ΔT between the surface and the ambient temperature.

$$\dot{Q}_{nat.\ convection} = \alpha\ A\ \Delta T \tag{2.7}$$

The heat provided by the heating jacket $\dot{Q}_{heating}$ is described by equation (2.8). Within the storage assessed here the heating jacket is characterized by an installed power Pheating jacket of 1200 W. Additionally, the factor *X* can vary between zero and one. This factor describes the control of the heating system. *X* is determined by the temperature of the storage material directly at the tank wall to ensure, that the composite storage material, especially the polymer, do not get too hot. The heating jacket shuts off at an upper boundary of 355 °C.

$$\dot{Q}_{heating} = X\ P_{heating\ jacked} \tag{2.8}$$

The external heating of the storage tank with the help of a heating jacket is necessary, because the reaction starts at about 300 °C and the ideal reaction temperature are reached at 350 °C. Therefore, the adsorption reaction of the hydrogen cannot start without externally provided thermal energy. The heat required to allow hydrogenation reaction to start is provided by the heating jacket. After the adsorption reaction has initially started within the storage additional heat is no longer required. Due to the exothermic behavior of the adsorption reaction enough heat is provided internally within the storage device. But this is only true for a short time, where the reaction velocity is very high. After that period the external heating is again necessary to allow a full reaction of the metal hydride with the remaining hydrogen. Because the heat released from the exothermic adsorption reaction is emitted to the environment (i.e., the heat is lost), without the heating

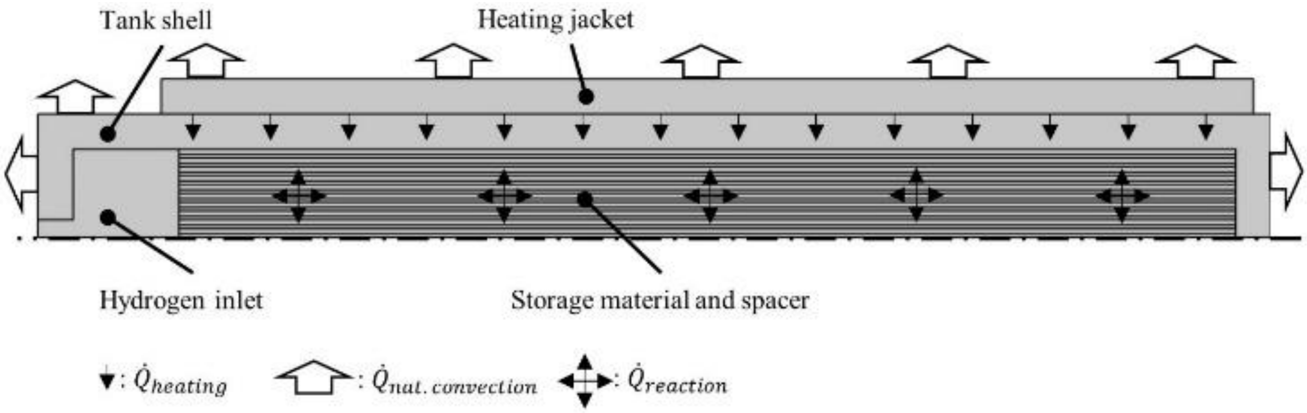


Fig. 2: Axial cut through the storage including the heat flows

jacket no full reaction (i.e., not full saturation of the overall metal hydride with hydrogen) would be possible. After the loading reaction is completely finished, the tank can cool down to ambient temperature; throughout the overall storage time no additional heat is required. Only for the desorption process again thermal energy is needed; i.e., the tank as to be re-heated to allow the hydrogen to be released from the metal hydride.

For the simulation of the temperature distribution inside the storage device equation (2.9) is used. This formula describes the heat flow inside the porous storage media including the spacer and the composite material. Therefore, the heat flow inside the spiral representing the actual hydrogen storage is described. For this calculation, the characteristic values of the different materials of the analyzed areas are taken into account. The effective values for the heat capacity $(\rho \cdot c_p)_{eff}$ and the thermal conductivity k_{eff} of the composite material as well as the spacer layer are estimated via the relation of the weight percent of the different materials inside the porous media.

$$Q = \left(\rho \ \ c_p\right)_{eff} \frac{\partial T}{\partial t} - k_{eff} \ \ \nabla T \qquad (2.9)$$

All the different calculations are combined in one overall model interacting between the chemical reaction and the heat transfer. The control variable for the simulation is the temperature T in relation of the time.

III. CASE STUDY DEFINITIONS

The case study presented below analyze different design concepts of such hydrogen storage related to the heat management of the storage material by varying the geometry and the amount of the different internal heat exchanger. Therefore, first the boundary conditions and the simulation parameters are described in chapter III.A. After that, the reference storage device and the used heat transfer geometries as well as the parameters which are varied are described in chapter III.B.

The storage device analyzed here is built as a rolled membrane module; i.e., two thin material layer are rolled up to a spiral. One of these layers is the composite storage material containing the metal hydrides as well as the polymers and the other layer is the metal mesh acting as a spacer to allow a good hydrogen flow towards and from the active storage material. These two layers are rolled to a spiral with an outer diameter of 50 mm and a length of 300 mm. This spiral in then located within a shell. The containing shell is made of steel and have an inside length of 350 mm. It is important that the tank shell is longer than the spiral, since free space is needed to secure a sufficient hydrogen flow inside each spacer layer. Such a design is intended to ensure a rapid hydrogen distribution inside the overall tank, since it is necessary to deliver hydrogen at each spot inside the tank on the shortest possible way without any significant delays.

The overall assembly of the storage is presented in Fig. 1 and Fig. 2. The composite material has a thickness of 1.4 mm and the spacer a thickness of 1 mm. The polymer material thickness is defined by the material properties of the polymer, therefore especially the hydrogen diffusion and the brittleness are important. The spacer thickness is defined by the thickness of the polymer and by possible thicknesses of the supplier

Fig. 2 shows the axial cut through the hydrogen storage device. The hydrogen inlet of the overall storage tank is at the left side. This side is also the inlet for the rolled storage foil with the integrated spacer. After the rolled storage material is in position, a cap is welded on the shell to close the overall storage tank gastight. This procedure has the benefit, that the otherwise required flange can be omitted. Important for the simulation outlined below is that it is no longer necessary to heat up a big flange to the reaction temperature, since this needs additional energy as well as time. Another benefit for some possible applications is a significant reduction of steel, which leads to a weight reduction.

A. *Simulation parameters and boundary conditions*

The metal hydride assumed here are 2 $LiBH_4$ + MgH_2. This material is infiltrated inside carbon shells made out of a highly porous activated carbon. These metal hydride carbon composites are then embedded inside the polymer matrix made of Polymethylpentene (TPX). The detailed storage assembly are shown in Fig. 3.

The different material values of the storage material must be customized, due to the composition of the overall storage material. The chemical reaction is defined by the active material (here: 2 $LiBH_4$ + MgH_2). The heat capacity as well as the thermal conductivity are determined by a mixture of the different materials as explained in chapter II.

As spacer material, a steel mesh is chosen here. Steel is selected, since it shows a higher thermal conductivity than the polymer foil, it is stable at high temperatures and no chemical

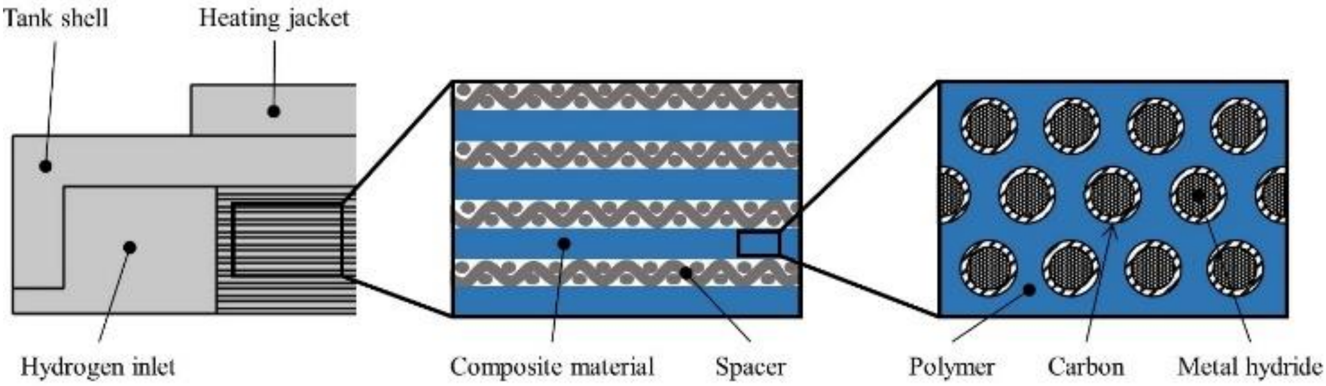


Fig. 3: Schematic storage assembly and storage material structure

reactions are expected between the steel spacer and the polymer foil.

The temperature and the pressure inside the storage are determined by the hydrogenation reaction taking place within the metal hydride; i.e. with an ongoing hydrogenation reaction these values might change. The storage material operates best at a temperature of 350 °C and a pressure of 50 bar. Therefore, the goal during the storage process is it to ensure a more or less constant temperature within the overall storage during the entire adsorption process.

Therefore, the simulation starts at 200 °C by modelling the heating process. This starting temperature is chosen to simulate the loading of the storage after a break of usage. During this time the overall storage device is cooled down from its desorption temperature level. For reasons outlined above it is very important that the internal heat transfer geometries lead to a fast heating up to the reaction temperature of 350 °C. Compared to that the pressure control is not a significant problem since this can easily be done by controlling the hydrogen feed stream during the ongoing adsorption process within the storage device. The ambient temperature level is assumed with to be constant at 20 °C; this determines the heat convection into the surrounding atmosphere.

B. *Reference storage and heat transfer concepts*

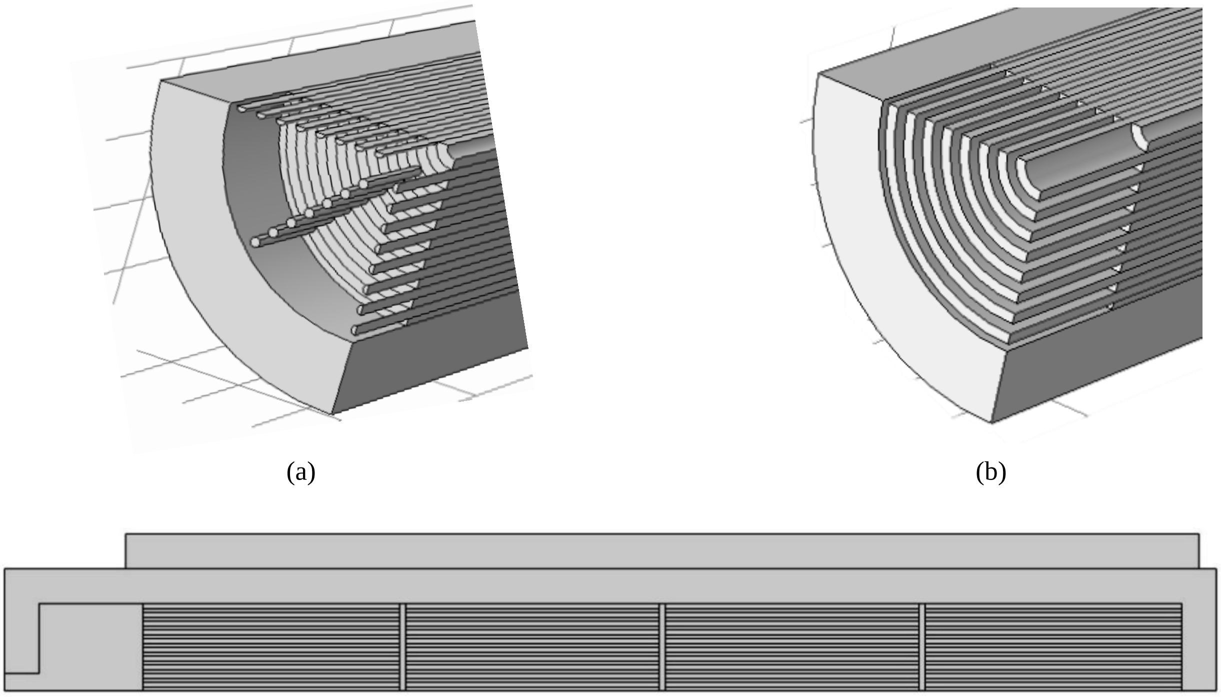
As a reference, a hydrogen storage module designed as spiral foil consisting of the composite material and a steel mesh inside the storage is used. This reference storage system does not have any additional heat transfer structures inside the storage. This very basic design can be seen in Fig. 1 and Fig. 2. This reference design of the hydrogen storage respectively the results of the simulation define the base line for the comparison of the different internal heat transfer structure to be assessed. They have to show a better performance compared to the storage without any special heat transfer structure. This is valid for all characteristic values (e.g. temperature development, heating time and reaction rate).

To improve the heat transfer within the hydrogen storage system relative to the reference design concept, different technological solutions are developed and compared with the reference storage defined above. This is realized based on two steps. First, three different geometrical concepts are compared to each other. Second, the most promising concept out of these three options will be simulated based on different geometrical parameter of this concept.

The three different geometries analyzed here are axial pins, a long spacer and radial fins. These three heat transfer concepts are shown in Fig. 4.

- The first geometry are pins (Fig. 4(a)) which are integrated axial inside the spacer material to transport the heat to the front and back surface. The idea behind this design approach is the optimization of the axial heat distribution and especially the usage of the cap at the hydrogen inlet as heat sink.
- The second geometry, called long spacer (Fig. 4(b)), are quite similar to the pins. But, here the spacer is realized longer, so that it hits the cap at the inlet. Therefore, it becomes possible to transport the heat to the front surface without the integration of an additional heat transfer structure. The benefit of this axial concept from a construction point of view is that it does not reduce the hydrogen storage capacity of the storage. Additionally, for this concept are no extra parts necessary making this approach cheap and easy to implement.
- The third concept is called fins (Fig. 4(c)). These fins are radial integrated inside the storage material and interrupt the storage material (i.e., they divide the spiral-formed storage material into several compartments). The fins are made of the spacer material. This approach has the benefit, that the hydrogen can flow easily through the material and the heat can be distributed easily over the radius of the enrolled storage material.

The important difference between the last and the first two design concepts are the direction of the modified heat transfer. The fins are the only concept allowing for a radial heat flow within the storage device. But this approach is characterized by the disadvantage that the possible amount of storage material inside the tank need to be reduced accordingly. In comparison, the axial systems only modifying the spacer layer. Hence, no reduction of the storage capacity occur. Therefore, it is important to analyze, if a radial optimization of the heat transfer is necessary for faster reaction kinetics and therefore to allow for an improved loading and unloading of the hydrogen storage.


(a) (b)

(c)

Fig. 4: Different heat transfer geometries ((a) pins, (b) long spacer, (c) fins)

Furthermore, the most promising concept will be analyzed more closely to improve the geometry until it shows the highest impact for a better heat distribution and a faster reaction related to the overall storage. Therefore, the amount of items (e.g., pins, fins) and the thickness of the geometry implemented will be varied. The details of the different variation will be described in chapter IV.B.b.

IV. CASE STUDY RESULTS

Below the results for the case study described in chapter 3 are discussed in depth. First, the results of the reference case are analyzed. Then the different geometries are presented. After that, the optimized geometry is investigated. This is realized regarding two effects taking place during the use of the storage. These are the heating to reaction temperature (i.e., heating up till the exothermic adsorption reaction can start) and the impact of the heat transfer geometries on the average reaction rate during the adsorption process.

A. *Reference storage*

Below, the reference storage will be examined in more detail. This should clarify, why a heat transfer structure is required to get a better storage behavior.

Fig. 5 shows the temperature distribution inside the reference storage after 50 min of heating and shortly before the control unit reduce the heating power to avoid overheating of the composite material directly at the tank shell. The graphic makes it obvious that the area inside the storage with the lowest temperature lays lightly left to the center of the storage. Moving along the centerline of the storage from the center towards the hydrogen inlet the temperature stays lower and in the direction to the closed end the temperature rises towards the upper end of the temperature range. Therefore, it would be beneficial if the heat distribution to the middle of the storage and towards the hydrogen inlet can be improved.

This temperature distribution shows that it is important to improve the heat transfer through the composite material. But, until the cold parts are in the middle and in the direction of the hydrogen inlet, it cannot be clearly defined if it is better to implement a radial or an axial heat transfer geometry. Thus, both options will be analyzed to find the best solution for this storage concept.

B. *Heat transfer concepts*

According to the definitions in chapter III in total three different heat transfer concepts with different geometries are analyzed. The most promising geometry is further analyzed with the variation of different parameter.

a) Variation of the heat transfer geometrie: The results of the three different heat transfer geometries as well as the reference storage described in chapter III.B will be discussed here. The comparison of the different geometries in Fig. 5 shows that the implementation of the different heat transfer structure helps to reach a more equal temperature development. It also shows that the fins (Fig. 5(d)) have the most homogeneous temperature distribution inside the storage material.

Fig. 6 shows the average temperature development inside the composite material. It points out that the radial fins are most helpful to reduce the necessary time to heat the system up to a temperature of around 350 °C. They are approx. 5 min faster compared to the pin concept. The long spacer reaches the reaction temperature in 55 min and the reference system do not reach the reaction temperature within a timeframe of 60 min at all. This result suggests that fins could be the best option.

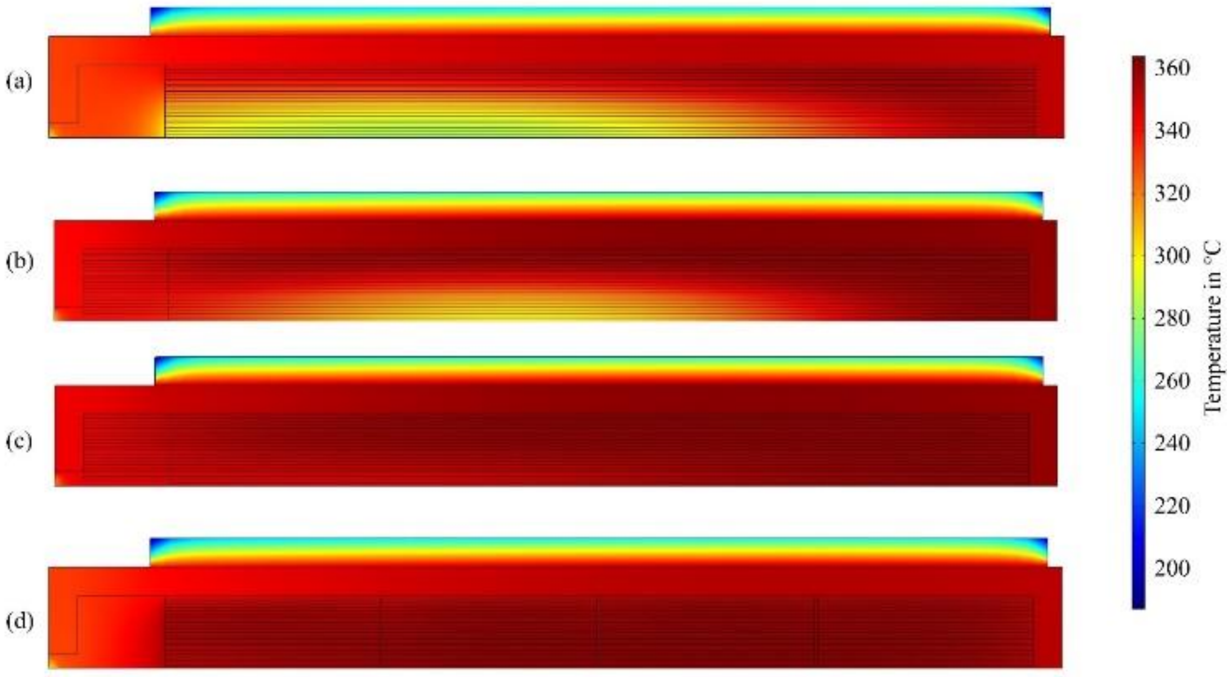


Fig. 5: Temperature distribution after 50 min heating time; (a) reference model, (b) long spacer, (c) pins and (d) fins

Fig. 7 presents the development of the amount of hydrogen stored inside the metal hydride in weight percent of hydrogen. Here it becomes obvious that fins help most to increase the reaction rate, since the storage system with the fins shows the fastest increase of the stored amount of hydrogen. In comparison to the reference case the pin concept is characterized by approximately the same reaction rate and the long spacer concept results in an even lower reaction rate. This can be explained by the fact that axial heat distribution leads to a better average temperature, but the local temperature values in parts of the storage material stays longer beneath the reaction temperature, since heat is needed to increase the temperature of the cap and the hydrogen inside the inlet area resulting in a slower average reaction rate. The fact that both parameter, heating time and reaction rate, are best with fins as internal heat transfer geometry, justifies a more detailed assessment of different variations of fins (see below).

b) Variation of the parameter of the most promising concept: Chapter IV.B.a shows that fins are the most efficient heat transfer structure for an improvement of the heating as well as the reaction time. Based on this finding, the parameters thickness of fins and number of fins inside the storage are varied here. Additionally, it will be analyzed which parameter combination leads to the best heat distribution as well as to the fastest absorption time. Beside this, also the loss of hydrogen storage capacity through the fins is taken into account.

The amount of fins will be analyzed from one up to six and the thickness is varied between 1.0 mm and 1.8 mm in 0.2 mm steps. All combinations of these parameters are simulated. The most interesting variation are shown below.

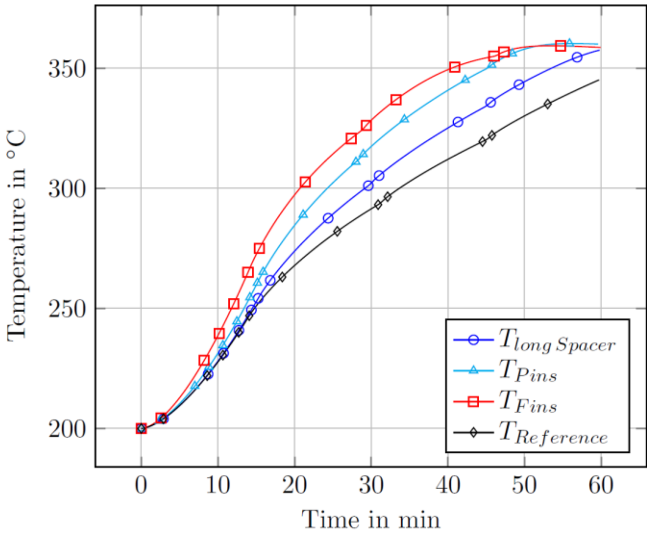


Fig. 6: Temperature development in the composite material with the different heat transfer structures

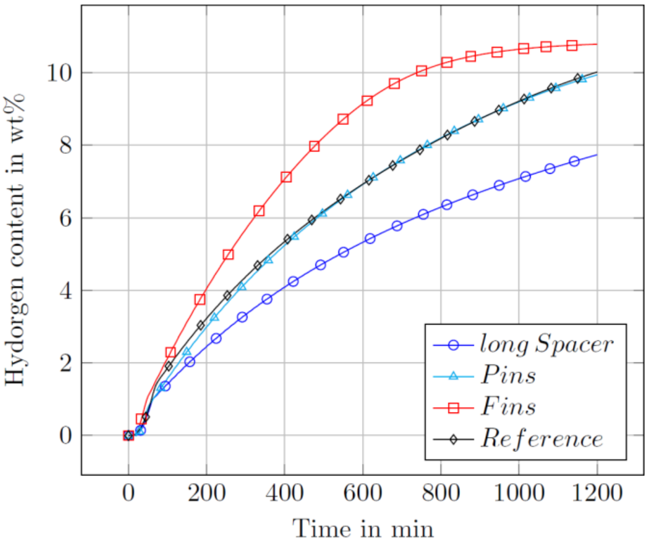


Fig. 7: Hydrogen content inside the metal hydride combined with different heat transfer structures

Fig. 8 shows the temperature development of hydrogen storages with a different amount of fins inside the storage material. All fins are characterized by the same thickness of 1 mm. The amount of fins varies between one and six. They are distributed equally over the length of the storage material. Also, the reference storage without fins are shown. It can be seen that the amount of fins has a significant impact on the heating time. The heating time of the storage with six fins is reduced by around 20 min compared to the storage with one fin.

Fig. 9 shows the development of the hydrogen loading inside the hydrogen tank. Again, the number of fins is varied between one and six and all fins have the same thickness of 1 mm. The development of the hydrogen loading shows that the storage with six fins allow the fastest reaction and reach the maximum hydrogen loading approx. 400 min before the storage with only one fin comes close to the storage maximum. Therefore, a rising number of fins has a positive influence on the overall reaction velocity of the storage. This result is based on the optimized heat transfer inside the storage improving the conditions for the chemical reaction. Overall, more fins result in a more equal and faster temperature distribution and also to a faster reaction of the storage.

Since the storage with six fins shows the best values, fins with a different thickness are assessed with a storage system that contains six fins. The thickness is varied between 1.0 mm and 1.8 mm in 0.2 mm steps. The results of this variation are shown in Fig. 10 (temperature development) and Fig. 11 (reaction rate). These graphics show small benefits through the thicker fins, but in comparison to the effect of the amount of fins this effect is negligible. This leads to the conclusion that many thin fins are the best way to optimize the reaction velocity as well as the heating time of such storage tanks, while probably the capacity loss is minimized.

Finally, the hydrogen capacity losses due to the volume loss caused by the fins are considered. Therefore, many thin fins and some thick fins are investigated to compare the thermal and kinetic improvement with the resulting hydrogen capacity loss. Fig. 12 shows the capacity losses at each considered amount of fins and thickness. The capacity losses increase linear with the amount and the thickness of the fins.

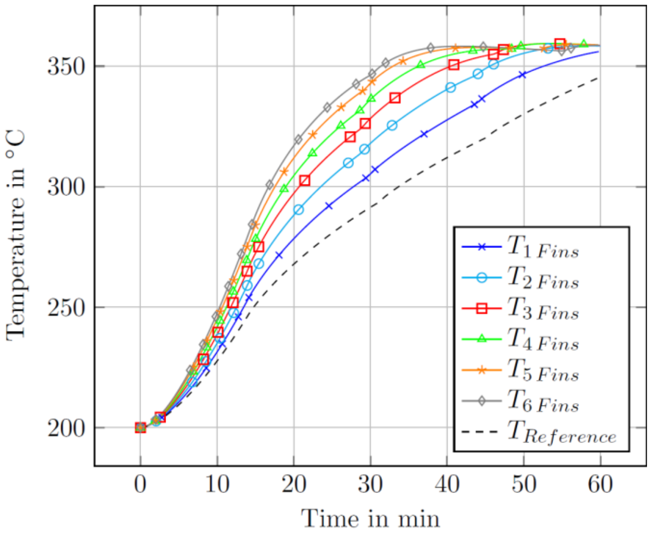


Fig. 8: Temperature development of the composite material with different amount of fins with 1 mm thickness

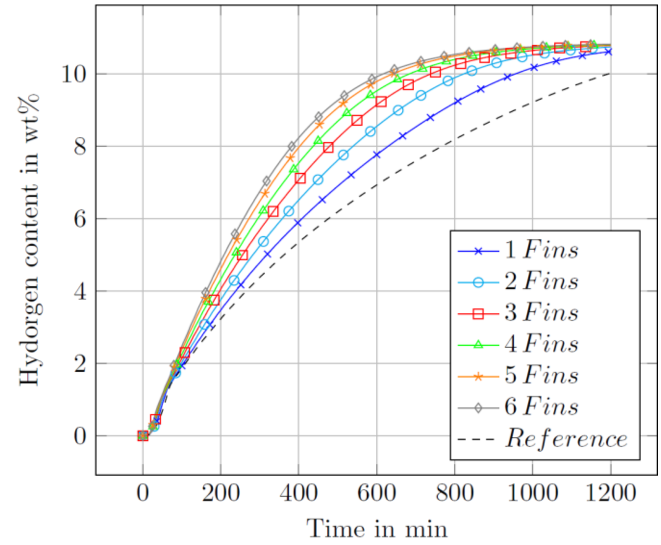


Fig. 9: Hydrogen content inside the metal hydride with different amount of fins with 1 mm thickness

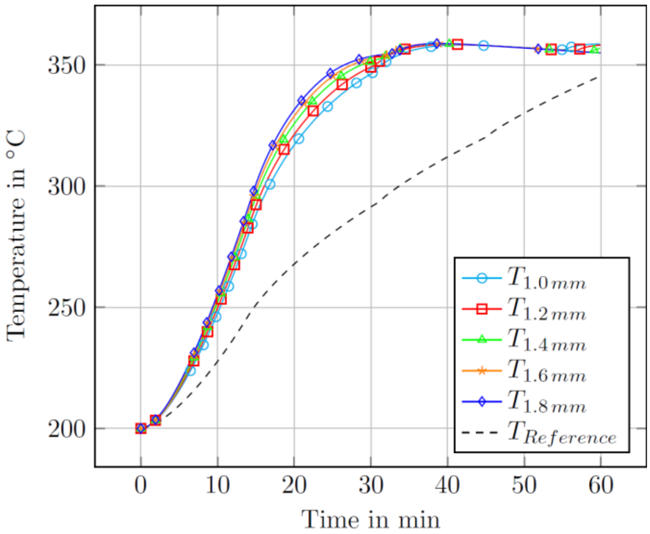


Fig. 10: Temperature development of the composite material with six fins with different fins thickness

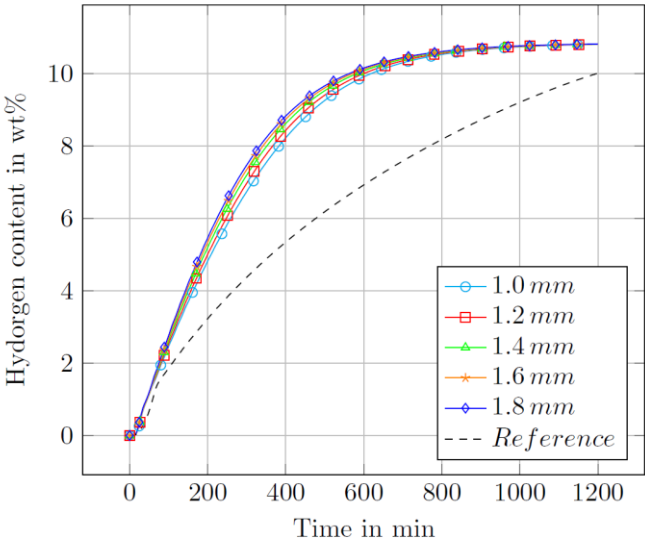


Fig. 11: Hydrogen content inside the metal hydride with six fins and different thicknesses

It can be seen that the hydrogen losses for the different combinations of amount and thickness of the fins, results in a hydrogen loss of 0.2 % up to 2.2 % of the total hydrogen capacity of 10 g. Fig. 13 presents the temperature development in the composite material of a storage with six 1 mm fins and five 1.8 mm, 1.6 mm and 1.4 mm fins. According to this, the storages with five fins with 1.8 mm and 1.6 mm have a small benefit, but the six thin fins (1 mm) still provide a better heat distribution then the five fins with 1.4 mm. As a benefit for the storage with six 1 mm fins, this design concept has the lowest hydrogen capacity loss (Fig. 12). Therefore, it can be concluded that many thin fins are in total the best option for improving the heat distribution and reaction rate combined with the lowest possible hydrogen storage capacity losses.

V. CONCLUSION

The overarching goal of this paper is the simulation and analysis of different design concepts for a new developed composite material for hydrogen storage in metal hydrides. Therefore, an expensive simulation model allowing to analyze and to investigate the temperature distribution during heating as well as the hydrogen adsorption is developed. This model analyzes the adsorption reaction influenced by the temperature inside the material as well as the heating of the material. The desorption process is not analyzed individually, since the behavior of the thermodynamics of desorption are very similar to the heating process and have therefore no further impact on the analyzed heat transfer structure.

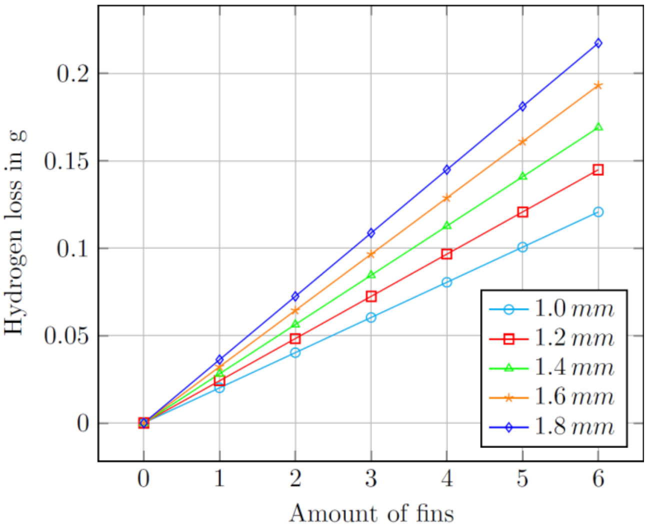


Fig. 12: Total hydrogen losses

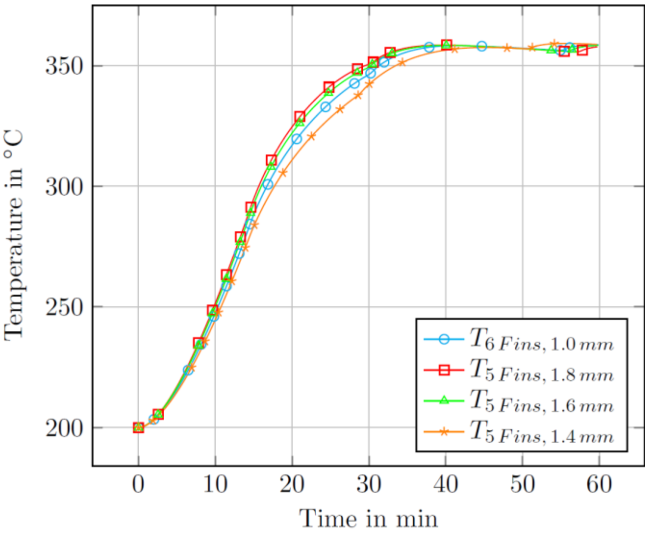


Fig. 13: Temperature development of different fin combinations

Based on this simulation model different heat transfer geometries are considered. The geometries are long spacer, pins and fins. A comparison of these three geometries points out that fins show the best results for the reaction rate as well as the temperature development inside the composite material. Furthermore, the amount of fins as well as the thickness of the fins are varied. Finally, the hydrogen loss of the different fins is compared to check if less thick fins or many thin fins provoke a smaller reduction of the hydrogen capacity at the same benefit for the heat transfer. To conclude, the main results of the simulations can be summarized as follows.

- A radial improvement (i.e., implementation of fins) of the heat transfer inside the storage give the best effects for the improvement of the heating as well as the reaction.
- The thickness of the fins has only a small effect on the improvement.
- The number of fins has a big effect on the improvement of the heating as well as the adsorption kinetic.
- Many thin fins produce less hydrogen capacity losses then a few thick fins.

All this points leads to the conclusion that many thin fins are the best solution to improve the heat transfer as well as the reaction rate inside this kind of hydrogen storage.

The next step of this research is to build a test rig for this new hydrogen storage concept. There the simulated and optimized hydrogen storage can be analyzed. The expected experimental results will indicate where further improvements are possible. Also further analyses of the combination with different hydrogen sources and consumers are planned for the near future.

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