

EXPERIMENTAL AND NUMERICAL INVESTIGATIONS ON THERMO-CHEMICAL HEAT STORAGE

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Synopsis

The Institute for Thermodynamics and Thermal Engineering (ITW), University Stuttgart, is investigating chemical heat storage technologies for low-temperature applications. The focus of the work within the project “thermo-chemical heat storage” (CWS) is on the choice of the storage concept, on experimental investigation of suitable reaction systems as well as on modelling and simulation of the reaction behaviour and heat and mass transfer processes in order to develop an efficient reactor design and system concept. At ITW hydration / dehydration reactions of inorganic salts are investigated as storage material for thermo-chemical energy storages. The characteristic reaction behaviour of different materials has been experimentally analysed. In addition an energy assessment of a solar combisystem with a chemical energy store was performed for different system concepts taking the specific reaction behaviour of the thermo-chemical storage materials into account. In this paper, the results of the investigation of several thermo-chemical storage materials and of the energy assessment of a combisystem with a thermo-chemical heat store are presented and discussed.

1 Introduction

The increasing demand for thermal energy, the uncertainty in energy supply, soaring oil prices and the global claim for a significant reduction of greenhouse gas emissions are the main driving forces for an intensive research on a more effective usage of renewable energy sources for heating (and cooling) applications. Solar thermal systems for hot water preparation and space heating are already used since decades. Today with typical solar installations in Germany the fraction gained from solar energy for hot water preparation and space heating is about 20 % to 30 % of the total thermal energy demand. However, there is an increasing desire for higher solar fractions and a trend towards the “Solarhouse50+” with solar fractions of far more than 50 % and the “Solar Active House” which is totally heated by solar thermal energy. For these buildings hot water stores with a volume in the range of 5 to 50 m³ are at present state of the art [2]. To significantly reduce the storage volume and to increase the overall system efficiency new thermal energy storage concepts are crucial. Today, many institutes and researchers are engaged in improving existing and in developing new thermal energy storage technologies and concepts. One area of research is the investigation and development of new materials and concepts for thermal energy storage. Here, storage technologies based on thermo-chemical mechanism might provide a breakthrough for seasonal thermal energy storage due to their potential for high storage densities and minor heat losses. A great variety of hydration / dehydration reaction of inorganic salts have been investigated as storage material for thermo-chemical energy storage (e.g. [1], [3]). The specific material

characteristics (storage density, reaction behaviour) determine the boundary condition of the reaction such as reaction temperature and inlet air humidity. By means of an energy assessment the influence of these material parameters on the fractional energy savings and system sizes (store volume, collector area) have been analysed for different system concepts.

2 Experimental investigation of hydration reactions for chemical heat storage

The reaction behaviour of different inorganic salts with and without carrier matrix has been experimentally investigated at ITW in a laboratory test rig. A sketch of the test facility used to carry out the dehydration / hydration experiments is shown in figure 1. A detailed description of the test facility is given in [5].

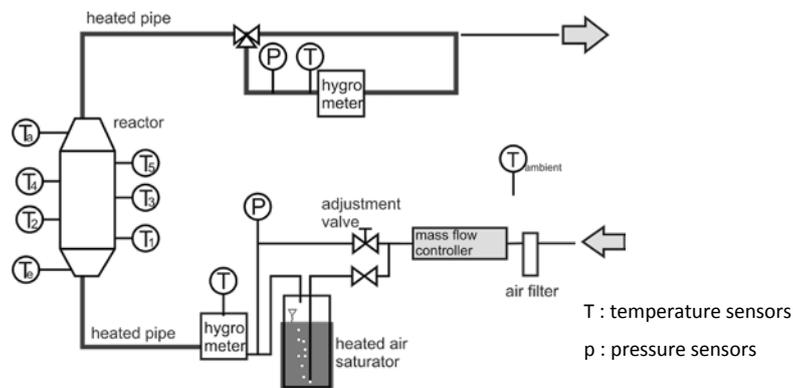


Figure 1: Sketch of the test rig for the hydration / dehydration experiments

In the following the reaction characteristics for pure salts as well as for salts on a carrier matrix will be described in more detail by the example of pure magnesium sulphate, zeolite impregnated with magnesium sulphate and bentonite impregnated with calcium chloride.

2.1.1 Hydration reaction of magnesium sulphate

In the experiments performed in the test rig a fixed bed of magnesium sulphate monohydrate ($\text{MgSO}_4 \cdot \text{H}_2\text{O}$) powder was flown through by humid air with a flow rate of $1.5 \text{ m}^3/\text{h}$. In figure 2 the inlet temperature T_{in} and reactor temperatures at heights of 5 cm (T_1), 10 cm (T_2) and at the outlet (T_{out}) are plotted vs. time.

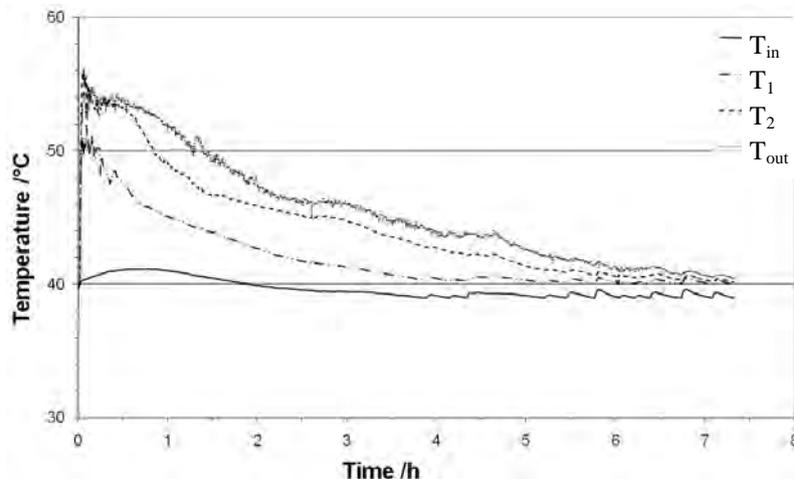


Figure 2: Temperature profiles of the reaction of $\text{MgSO}_4 \cdot \text{H}_2\text{O}$ with humid air versus time. The inlet temperature was $40 \text{ }^\circ\text{C}$, the humidity was kept at $x_{\text{H}_2\text{O},\text{in}} = 38 \text{ g}_{\text{H}_2\text{O}}/\text{kg}_{\text{air,dry}}$ (relative humidity of 80 %).

The temperature profile shows that the temperature increases almost uniformly over the whole reactor length to a peak of about 55 °C. With increasing water uptake the temperature lift decreases simultaneously. A slow but constant reaction rate has been observed even after 7 hours. However, the technical useful temperature lift is limited to the part of the experiment where the temperature lift is greater than 5 K.

In all experiments performed, the increase of weight was between 3 and 4 mol water per mol magnesium sulphate monohydrate which means that within the limited time of the experiment the fully hydrated state (6 mol H₂O per mol MgSO₄·H₂O) was not reached. Considering the reaction from mono to hexahydrate the maximum achieved energy gain of the experiments was about 50 % of the theoretical value. However, at low inlet humidities and inlet temperature of the air stream ($x_{\text{H}_2\text{O},\text{in}} < 20 \text{ g}_{\text{H}_2\text{O}}/\text{kg}_{\text{air,dry}}$, $T_{\text{in}} < 30 \text{ °C}$) the reaction takes place that slowly that the achieved temperature lift is too low for technical use e.g. for heating applications.

2.1.2 Hydration reaction of zeolite impregnated with magnesium sulphate

Magnesium sulphate impregnated on zeolite spheres significantly improves the reaction kinetic of the hydration reaction. Zeolite is a highly porous mineral and has a high ability of adsorbing water with a high adsorption enthalpy even at very low water vapour pressure. In a lab-scale prototype store with zeolite 4A monoliths as storage material an energy storage density of 130 kWh/m³ has been realised at ITW [5].

For the experimental investigation the zeolite spheres have been impregnated with a magnesium sulphate solution. A fixed bed using impregnated zeolite 4A particles (diameter of about 2 mm, 5 mass-% of magnesium sulphate) was flown through by an air stream with a humidity of $x_{\text{H}_2\text{O},\text{in}} = 13 \text{ g}_{\text{H}_2\text{O}}/\text{kg}_{\text{air,dry}}$ and an inlet temperature of 25 °C. The volume flow rate was set to 1.0 m³/h. In figure 3 the temperature distribution, measured at the five axial positions in the fixed bed, is shown.

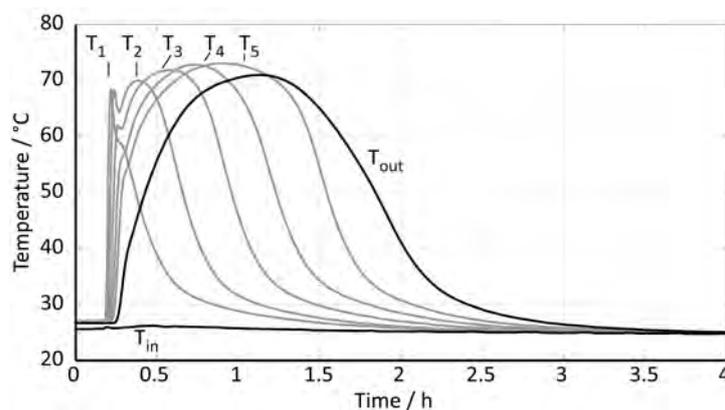


Figure 3: Temperature profiles of the reaction of MgSO₄ · H₂O impregnated on zeolite (5 mass % magnesium sulfate) with humid air versus time. The inlet temperature T_{in} was 25 °C, the humidity was kept constant at $x_{\text{H}_2\text{O},\text{in}} = 13 \text{ g}_{\text{H}_2\text{O}}/\text{kg}_{\text{air,dry}}$, (relative humidity of 65 %) the air flow was set to 1.0 m³/h.

Characteristic in these experiments are the sharp adsorption fronts moving through the reactor with a constant velocity. The inlet air humidity is completely adsorbed over a long time of the experiments followed by a step breakthrough.

The total water adsorbed of the impregnated zeolite is higher than of the pure zeolite. Furthermore, in these experiments the salt is reacting to a higher hydrate level even at low inlet humidities in a significant shorter time frame than in the experiments with pure salts.

In figure 4 the temperature in the fixed bed at the first temperature sensor ($h = 1.0$ cm) and at the outlet temperature is depicted for pure zeolite 4A (T_Z) and for the impregnated zeolite (T_{Z-MgSO_4}). The inlet conditions were the same in both experiments: an air stream of 1.0 m³/h with a humidity of 20 mbar and an inlet temperature of 25 °C. It can be seen that in the beginning of the experiments the slope of the curves are similar and also the temperature increase to around $T = 70$ °C is achieved in both experiments. However, due to the higher adsorption activity of the impregnated zeolite the reaction takes place over a longer period and the outlet temperature remains on a high temperature level over a longer time frame. This results in an increase of the energy storage density. In the experiments performed an increase of the energy storage density of 20 % (5 mass-% $MgSO_4$) up to 40 % (10 mass-% $MgSO_4$) compared to the pure zeolite was measured. It is expected that a further increase in the energy storage density with exceeding 200 kWh/m³ can be obtained by optimizing the impregnation of the zeolite.

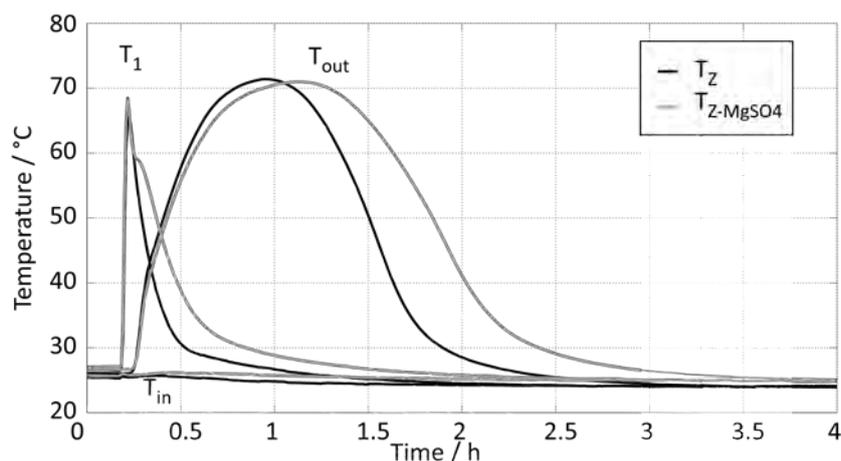


Figure 4: Temperature profiles of the reaction of pure zeolite (T_Z) and of zeolite impregnated with 5 mass % $MgSO_4$ (T_{Z-MgSO_4}) with humid air versus time. The inlet temperature T_{in} was 25 °C, the humidity was kept constant at $x_{H_2O,in} = 13$ g_{H₂O}/kg_{air,dry} (relative humidity of 65 %), the air flow was set to 1.0 m³/h.

2.1.3 Hydration reaction of calcium chloride impregnated on bentonite

Calcium chloride is a very hygroscopic material which can take up 6 mol of water per mol calcium chloride in an exothermic reaction. With a physical energy density of 380 kWh/m³ it is a very promising material for thermo-chemical energy storage. To prevent liquefaction (hydration reaction) or melting (dehydration reaction) of the calcium chloride the material is impregnated on bentonite, a highly porous mineral.

The experiments have been performed in the test rig described in figure 1. A fixed bed of impregnated bentonite (41.7 mass-% of calcium chloride) was flown through by an air stream with a humidity of $x_{H_2O,in} = 13$ g_{H₂O}/kg_{air,dry}, and an inlet temperature of 25 °C. The flow rate was set to 0.5 m³/h. In figure 5 the temperature distribution during the exothermic hydration reaction in the fixed bed is depicted. A reaction front is formed but not that distinctive as for the impregnated zeolite. Characteristic in these experiments is a high reaction kinetic at the beginning of the experiments associated with a high temperature increase and then a long period of an almost constant but slow reaction kinetic where the temperature increase is on a relatively low level ($\Delta T \approx 10$ K). The outlet humidity of the air stream during the reaction is relatively high ($x_{H_2O,out} \approx 2 \dots 7$ g_{H₂O}/kg_{air,dry}). In the experiments a maximum energy density of 185 kWh/m³ was achieved.

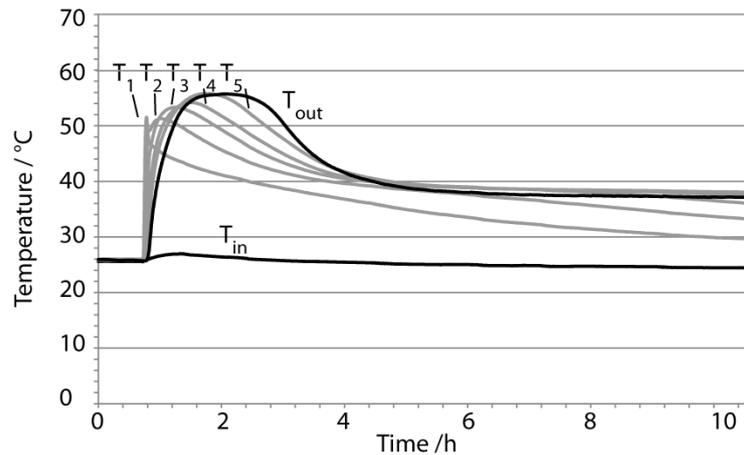


Figure 5: Temperature profiles of the reaction of calcium chloride (41.7 mass-%) impregnated on bentonite. The inlet temperature T_{in} was 25 °C, the humidity was kept constant at $x_{H_2O,in} = 13 \text{ g}_{H_2O}/\text{kg}_{air,dry}$, the air flow was set to 0.5 m³/h.

3 Energy performance of thermo-chemical heat stores in solar combisystems

Different concepts of integrating a chemical energy store into a solar combisystem have been investigated. In the following one of these concepts is described in more detail.

The main parts of the chemical energy store are a material reservoir and a reactor separated from the reservoir in which the hydration or dehydration reaction takes place. The decoupling of the reactor and material reservoir has several advantages such as a reduction of thermal masses during the hydration / dehydration process and lower pressure losses when air is flowing through the reactor. The reaction is performed in an open cycle which means that humid room exhaust air (ambient air) is flowing through the reactor and released into the ambient after leaving the reactor. The heat of the exothermic hydration reaction is transferred to a buffer store (combistore). The energy needed for the endothermic dehydration reaction is provided by high performance solar thermal collectors. In figure 6 the energy fluxes in the system are depicted for the heating mode (hydration of the material) and the regeneration mode (dehydration of the material).

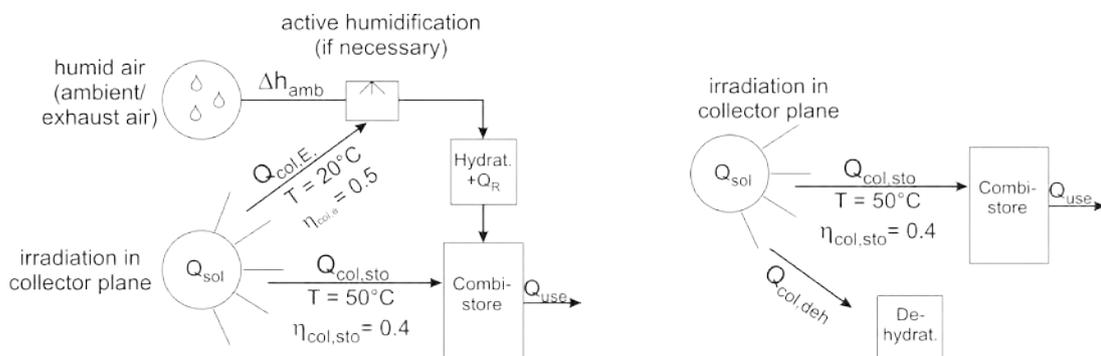


Figure 6: Heat and enthalpy-fluxes during the heating mode (left) and regeneration mode (right)

Heating mode:

If the solar irradiation is too low so that the energy from the solar collector field ($Q_{col,sto}$) cannot completely cover the heat demand the chemical heat store is used as backup. The heat of the exothermic hydration reaction is released by flowing humid room exhaust air through the reactor. If

necessary the air is actively humidified using energy from the solar collector field on a low temperature level ($T \approx 20 \text{ }^\circ\text{C}$, $Q_{\text{col,E}}$).

Regeneration mode:

The regeneration of the storage material takes place during times of high solar irradiation. When the energy provided by the collector field exceeds the heat demand for hot water preparation and space heating the available solar energy is used for the endothermic dehydration reaction of the storage material.

3.1 Energy analysis

For the energy assessment of the system concept an energy balance of the heat sources (solar irradiation, ambient humidity) and heat demands (space heating, hot water preparation) is performed on a daily basis as a first step. The daily value of the heat supply is obtained on the basis of a weather data file for a specific location, the daily value of the space heating demand is obtained from a thermal simulation of a building. The domestic hot water demand was set to 200 l/d at a hot water temperature of 45 °C. The dynamic behaviour and hence a possible time shift between demand and supply within one day is not considered. This is synonymous to the behaviour of a system with an ideal one day buffer store. Depending on the daily heat demand and heat supply the system is operating in the heating mode ($Q_{\text{demand}} > Q_{\text{col,sto}}$) or in the regeneration mode ($Q_{\text{demand}} < Q_{\text{col,sto}}$). The energy provided by collectors field for direct heating ($Q_{\text{col,sto}}$) and for providing heat for water evaporation ($Q_{\text{col,E}}$) is estimated using an effective collector efficiency η_{col} at the respective operating temperature for the collector field (cf equation 1):

$$Q_{\text{col}} = \eta_{\text{col}} \cdot Q_{\text{sol,South45}} \quad (1)$$

The heat released in the hydration reaction is calculated with equation (2) using the specific material parameters described in table 1.

$$\begin{aligned} Q_{\text{hyd}} &= m_{\text{air}} \cdot \Delta x_{\text{air}} \cdot \Delta h_r \\ &= m_{\text{air}} \cdot x_{\text{air}} \cdot \mu \cdot \Delta h_r \end{aligned} \quad (2)$$

with

$$\Delta x_{\text{air}} = x_{\text{air,in}} - x_{\text{air,out}} \quad (3)$$

$$\mu = \Delta x_{\text{air}} / x_{\text{air,in}} \quad (4)$$

In the equation \dot{m}_{air} [kg/h] is the mass flow rate of the air, Δx_{air} [g_{H₂O}/kg_{air,dry}] the difference of the inlet ($x_{\text{air,in}}$) and outlet humidity ($x_{\text{air,out}}$) of the air flow and Δh_r [kJ/kg_{H₂O}] the mean reaction enthalpy per unit mass absorbed water.

Based on the results from the experimental investigations of the storage material further limitations concerning the reaction behaviour of the materials are necessary. The following assumptions and specifications are made (cf. table 1):

- For the hydration reaction a minimum water content of the inlet air flow of $x_{\text{H}_2\text{O,min}}$ is necessary. Optional an active humidification of the air flow can be performed if the humidity of the inlet air is below that value. The collector field is then acting as energy source for the humidification. If the collector field cannot provide sufficient energy for the humidification the heat demand cannot be completely covered.
- The storage material is adsorbing only a part of the water content of the inlet air flow. The mean ratio of the absorbed water to the inlet humidity of the air flow is specified with μ (cf. equation 3 and 4).

- The maximum adsorption/hydration capacity of the storage material depends on regeneration temperature (T_{dehyd}). With decreasing regeneration temperature less water is desorbed / dehydrated during the regeneration process and the water remaining on the material (remaining water charge) increases. The maximum amount of water vapour adsorbed per unit mass dehydrated storage material is specified with $\Delta x_{\text{H}_2\text{O,max}}$.
- A constant heat of reaction is assumed. The reaction enthalpy per unit mass adsorbed water is specified with Δh_r . The heat released is completely available for heating purpose.
- The mass flow rate \dot{m}_{air} through the reactor is controlled in such a way that the heat demand can be completely covered or a maximum mass flow of rate $\dot{m}_{\text{air,max}} = 150 \text{ kg/h}$ (if not specified otherwise) is reached
- The humidity of the room exhaust air (reactor inlet air) is set to $x_{\text{air,in}} = x_{\text{amb}} + 3 \text{ g/kg}_{\text{air,dry}}$ with the daily mean water content x_{amb} of the ambient air for the considered location
- The collector efficiency and the heat losses in the pipes depend on the operating temperature. Based on the efficiency curve of a vacuum tube collector, the following effective efficiencies are assumed for the collector loop:
 - for active humidification at $T \approx 20 \text{ }^\circ\text{C}$: $\eta_{\text{col,E}} = 0.5$
 - for direct heating/hot water preparation at $T \approx 50 \text{ }^\circ\text{C}$: $\eta_{\text{col,sto}} = 0.4$
 - for material regeneration: $T \approx 150 \text{ }^\circ\text{C} \Rightarrow \eta_{\text{col}} = 0.3$; $T \approx 180 \text{ }^\circ\text{C} \Rightarrow \eta_{\text{col}} = 0.2$

The energy assessment enables to combine the available heat sources (irradiation and air humidity) in an optimal way to cover the daily heat demand as both values daily demand and supply is known beforehand from the boundary conditions. With the energy assessment a statement can be made about the storage material volume needed and the fractional energy savings obtained with the system for defined boundary conditions (collector area, climatic data, heating and hot water demand).

3.2 Results

An energy assessment of the system concept described above has been performed with the following boundary conditions:

- total heat demand: 7519 kWh/a (hot water demand: 200 l/d at 45 °C; space heating demand: 35 kWh/m²/a)
- location: Würzburg, Germany
total irradiation in collector plane: 1230 kWh/m²/a
- solar thermal system: vacuum tube collectors orientated south, 45 °C tilted

For the verification of the energy assessment a long term prediction of a solar combisystem (store volume 900 l, CPC vacuum tube collectors) for the same boundary condition was performed with the dynamic simulation software TRNSYS. The fractional energy savings calculated with TRNSYS are compared to the fractional energy saving obtained by the energy assessment for a system without a sorption store ($V_{\text{TCM}} = 0$). This system represents a conventional solar combisystems with an ideal one day buffer store. The results of the fractional energy savings obtained with the two methods are in good agreement (cf. figure 7).

Within the energy assessment, the reaction behaviour is characterized by the parameters described in table 1. The values are derived from the experimental investigation of the thermo-chemical storage materials described in section 2 (cf. assumptions made in section 3.1).

In table 1 q_v is the energy density of the storage material calculated with:

$$q_v = \Delta x_{H_2O, \max} \cdot \rho_{\text{eff}} \cdot \Delta h_r \quad (5)$$

In equation 5 ρ_{eff} is an effective storage material density which takes a gas void fraction of $\epsilon = 0.4$ into account.

Table 1: Parameter describing the reaction behaviour of different storage materials

Material	T_{dehyd} [°C]	$\Delta x_{H_2O, \max}$ [g _{H2O} /kg _{anhyd}]	ρ_{eff} [kg/m ³]	Δh_r [kJ/kg _{H2O}]	μ [-]	$x_{H_2O, \min}$ [g _{H2O} /kg _{air, dry}]	q_v [kWh/m ³]
Zeolite 4A	180	180	700	3600	0.99	4.0	125
Zeolite 4A	150	140	700	3600	0.99	4.0	98
Zeolite 4A + 10 mass-% MgSO ₄	180	230	797	3500	0.99	4.0	178
Bentonite + 40 mass-% CaCl ₂	150	200	695	3500	0.65	6.0	135
<i>Case 1</i>							
Bentonite + 40 mass-% CaCl ₂	150	260	695	3250	0.5	8.0	163
<i>Case 2</i>							

Figure 7 shows the fractional energy savings calculated by the daily energy balancing under the assumptions and boundary conditions defined above for different adsorption storage volumes (V_{TCM}) and collector areas (A_{col}). The calculations are performed for the storage material zeolite 4A with a desorption temperature of $T_{\text{dehyd}} = 180^\circ\text{C}$. In addition the fractional energy savings are depicted for two typical combisystems with a hot water store of $V_{\text{H}_2\text{O}} = 0,9 \text{ m}^3$ and $V_{\text{H}_2\text{O}} = 15 \text{ m}^3$ determined by a detailed system simulations with the dynamic simulation software TRNSYS.

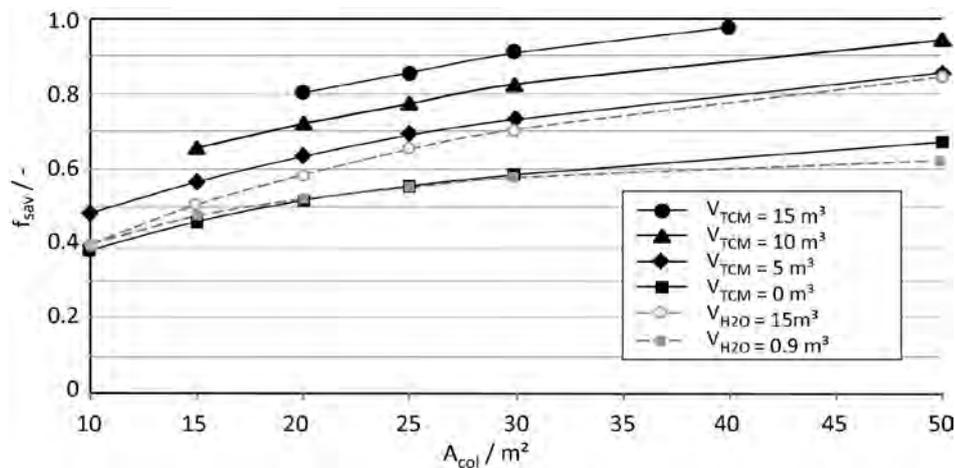


Figure 7: Solid line: Fractional energy savings calculated by daily energy balancing for different adsorption storage volumes (Zeolite 4A) in dependency of the collector area

dashed line: Fractional energy savings of a solar combisystems with a hot water store of the volume $V_{\text{H}_2\text{O}}$ based on a long term prediction performed with the dynamic simulation software TRNSYS

The results of the energy assessment show that by integrating a sorption store into the solar combisystem the fractional energy savings can be significantly increased compared to a conventional solar combisystem with a hot water store of the same volume. With collector areas

$A_{col} > 40 \text{ m}^2$ and sorption store volumes $V_{TCM} = 10 \dots 15 \text{ m}^3$ the heat demand can already be covered by more than 90 % by solar thermal energy.

In a next step the energy analysis has been performed for the same boundary conditions but different storage materials. The reaction behaviour and the desorption or dehydration temperature of the different materials are listed in table 1. Figure 8 shows the effect on the store volume needed for reaching a fractional energy saving of 80 %. The collector area was set constant to $A_{col} = 20 \text{ m}^2$.

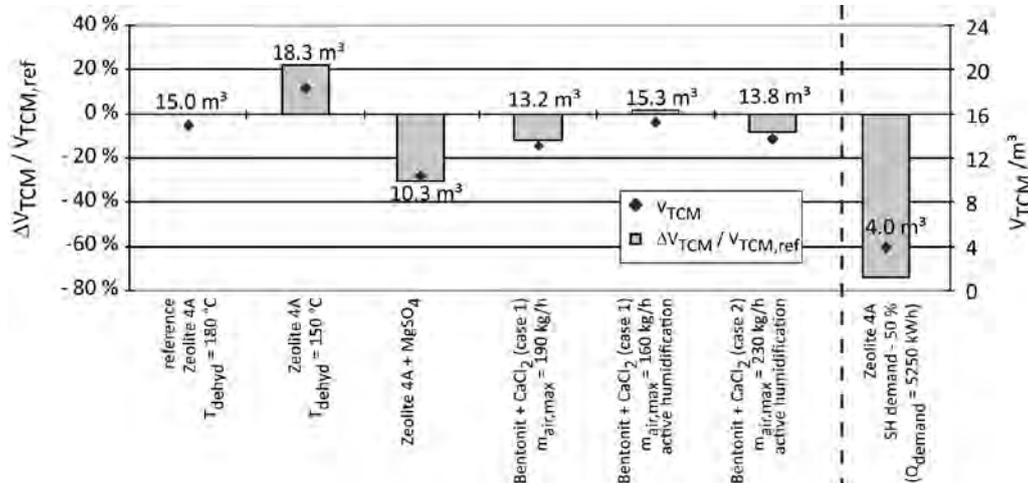


Figure 8: Material volume needed for reaching a fractional energy saving of 80 %; values are obtained from the energy assessment.

For the reference calculations with zeolite 4A using room exhaust air about 15 m^3 of storage material is needed to reach a fractional energy saving of 80 %. While technical feasible, the effort for regenerating the material at a desorption temperature of $T_{dehyd} = 180 \text{ }^\circ\text{C}$ is high. When the desorption temperature level is reduced to $T_{dehyd} = 150 \text{ }^\circ\text{C}$ the remaining water charge on the zeolite increases. This results in a decrease of the maximum water uptake $\Delta x_{H_2O,max}$ during the exothermic reaction and hence the amount of storage material rises by about 20%.

The contrary effect can be observed in the case of the composite material: with the impregnated zeolite (about 10 mass-% of magnesium sulphate) the water charge could be significantly increased and a fractional energy saving of 80% is predicted for about 10 m^3 store volume of the composite material zeolite 4A+ MgSO_4 .

For the salhydrate CaCl_2 on the passive carrier bentonite higher air humidities are required for the exothermic reaction and the content of water vapor in the outlet air stream is significantly higher than for zeolites. Two cases have been evaluated: for the material labelled with “Bentonit+ CaCl_2 , Case 2” the hydration process is continued to a higher hydration level than in the case of “Bentonit+ CaCl_2 , Case 1” (cf. table 1). The higher hydration level can only be achieved with higher inlet air humidities which results in a higher mean outlet air humidity. A fractional energy saving of 80% can only be achieved for the “Bentonit+ CaCl_2 -Case 2” when the system design allows high air massflows of $m_{air,max} = 230 \text{ kg/h}$ combined with active humidification. Even though parts of the energy provided by the collector field has to be used for humidification, the material is promising a lower store volume compared to zeolite 4A at lower regeneration temperature.

If the total heat demand is reduced to $Q_{demand} = 5250 \text{ kWh/a}$ (space heating demand of $18 \text{ kWh/m}^2/\text{a}$) a significant reduction in the store volume can be obtained. In this case a sorption store with a volume of only 4 m^3 of zeolite 4A is needed to obtain a solar fraction of $f_{sav} = 80 \%$. This is a reduction in store volume of more than 70 %.

4 Summary and outlook

From the experimental investigation of the different storage materials characteristic parameters describing the reaction behaviour have been obtained. These parameters are needed for an energy assessment of different system concepts.

The energy assessment presented in chapter 3 allows a detailed energy analysis of solar thermal heating systems with chemical energy stores. Statements can be made about the material volume needed and the fractional energy savings obtained with the system for defined boundary conditions (collector area, weather data, space heating and hot water demand). The energy assessment also shows the importance of taking the reaction behaviour of the thermo-chemical storage material into account.

With the thermo-chemical storage material investigated high solar fractions can be obtained already with moderate sizes of collector area and store volume even for heat demands of typical energy renovated house. By increasing the store volume and collector area sizes a 100 % solar coverage of the heat demand can be realized. On the other hand, to achieve the thermal performance of solar combisystems with high solar fractions the energy assessment predicts a significant reduction in the store volume (up to 1/3) or collector areas (up to 1/2) when using a sorption store (cf. figure 7, example 15 m³ water store volume, 50 m² collector area). A further reduction of the system size can be achieved by using the enhanced storage materials as shown in figure 8.

Current research at ITW is concentrating on the improvement of the reaction behaviour and increasing the energy storage density of the thermo-chemical storage materials by using different materials, impregnation technologies and various carrier matrices.

In parallel a dynamic simulation tool is in development which allows a detailed analysis of the system performance under dynamic boundary condition and will allow a more detailed system analysis than possible with the energy assessment presented in this paper. The dynamic interaction between the different components of the system (e.g. reactor, collector, buffer store) as well as thermal capacities of the system can be taken into account.

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