

# Background and Strategies for Identification and Design of Materials for Thermochemical Energy Storage and Conversion

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Within the past decade, ecological issues accompanying energy generation and utilization have gained increasing interest, thereby creating a need for new scientific and technical solutions on the path to a sustainable energy future. Besides switching the basis of the electricity sector from fossil fuels to renewables, also the heat sector is to be transformed. A major obstacle accompanying this energy transition is the temporal intermittency of power generation from renewables. However, these hurdles can be overcome by design of systems for energy storage and conversion. Within the growing field of solutions for thermal storage, thermochemical systems move into the focus as they provide comparably highest storage densities but at the same time also options for heat conversion. This concise review summarizes the background and the scope of possible applications discussed in recent literature. A focus is set on the identification and modification of new reaction systems, criteria for material selection are presented and different classes of reaction systems are discussed with regard to their operating temperature ranges. It is concluded, that an evaluation of possible use cases with precise definition of their respective thermal boundary conditions would be of high value for a purposeful continuation of future screening approaches.

### Introduction

With regard to the probable human impact on the earth's climate by emission of greenhouse-gasses as  $CO_2$  as product of combustion of fossil fuels, a global transition to energy generation from renewables has begun.

However, the energy demand is increasing steadily, as is the demand for affordable energy for all population groups. In order to enable an ecologically and economically feasible switch to renewables as the main source of primary energy, a solution for a major drawback of renewable energy generation has to be found: This drawback is the temporal intermittency of energy generation from the sun and wind, which is not only fluctuating on a daily basis, but in most regions of the earth also on a seasonal scale. For example, concentrated solar thermal power plants (CSP) are yet commercially not competitive, since their base load capability cannot be ensured. In order to balance demand and supply, generated excess energy would have to be stored in the form of heat. Therefore, thermal storage technologies are required, which allow for short- and longterm storage periods without significant losses.

Beside this contribution to primary energy generation, thermal storages can also contribute to a more efficient energy utilization in other applications and sectors, for example when used for transformation of waste heat into process heat or for heating in housing. Another major opportunity is a contribution of thermal storage systems to sector coupling, where excess energy can be transferred and stored in form of heat for later reintegration.

In general, energy storage is classified by the physical form in which energy is contained in a respective medium into chemical, electrical mechanical and thermal storage (**Fig. 1**, top) [1].

A category of chemical energy storages are batteries or accumulators, which store energy in form of the chemical potential of the electrodes or charge carriers. In contrast, electrical storages store electric current for example in capacitors or in form of magnetic fields in coils. Mechanical energy is either stored in form of kinetic or potential energy as for example in pump storage plants or by flywheels. All of the mentioned classes contain several subgroups.

This is also true for thermal storages, which are again classified according to the respective operational principle of the utilized storage medium into sensible, latent and thermochemical storages [2-5].

Sensible storages are probably the most familiar. Their working principle is based on simply transferring heat from a primary source to the storage medium, with the amount of stored heat proportional to the specific heat capacity of the medium and its available mass (**Fig. 1**, bottom left). Respective storage technologies are already implemented

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in domestic applications, where mostly water is used as the storage medium. High temperature storage could also be facilitated, using thermally stable media as for example ceramics.

Another option is found in latent heat storage, where heat is stored isothermally during a reversible phase transition as melting/solidification (**Fig. 1**, bottom middle). The amount of stored heat is proportional to the enthalpy of phase transition and hence comparably higher than in case of sensible material, also when neglecting the fraction of sensible stored heat.

However, for an economically feasible implementation in technical scale, both storage types demand for a thorough and oftentimes very costly insulation during the storage period, especially when a long-term storage period is required [6,7].



**Fig. 1.** Classification of energy storage by the physical form, in which energy is stored (top) and the three principles of thermal energy storage (bottom): Sensible (left), latent (middle) and thermochemical (right) with their respective functional principle, a schematic illustration of the energy incorporated during the storage period, the calculation of the amount of stored heat along with some example materials.

Thermochemical energy storage (TES) is based on reversible chemical reactions, with the charging procedure initiated by input of heat equivalent to the energy needed for thermal decomposition of the reactants.

Usually those are a solid and a gaseous species (sometimes referred to as the work fluid), since these two can easily be separated to initialize the storage period. Upon discharging, both reactants recombine under release of heat proportional to the reaction enthalpy to form the initial solid educt (Fig. 1, bottom right). As long as no side reactions occur during the storage period, a long-term and loss free storage of heat can be facilitated.

Since in case of thermochemical storage media the enthalpy of a chemical reaction is a main factor for the amount of stored heat, the storage density is up to four times higher than in case of latent storage media [2].

# The principle beyond thermochemical heat storage and conversion

In order to achieve technical maturity, experimental testing and evaluation of thermochemical storage materials has to be transferred from laboratory scale to reactor scale, where the materials are repeatedly charged and discharged in bulk scale. Beside the fundamental thermophysical and mechanistic investigation in lab scale, this allows to probe the materials performance under inclusion of possible bulk effects accompanying thermochemical cycling. For this purpose, mostly fixed reaction beds without internal movement of the material are filled with several grams or kilograms of the material [**8-11**].

A simple setup is given by a reactor in so-called direct operational mode, where the gaseous reaction partner dissolved in a carrier gas is flowing through the reaction chamber, thereby additionally facilitating the heat transfer during charging and discharging. The storage capacity of the overall fixed bed setup is limited by the mass, filled into the reactor unit (**Fig. 2**).



**Fig. 2.** A schematic drawing of the principle of thermochemical energy storage based on a gas-solid reaction in a thermochemical reactor with a fixed reaction bed and direct operational mode.

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However, for commercial implementation a scale-up from reactor to pilot scale has to be facilitated. Therefore, oftentimes moved or fluidized reaction beds are considered [12-14]. These scenarios of thermochemical cycling are comparably more realistic, since respective setups move the storage material from a silo to the reactor for charging or discharging and hence allow to detach the reactors power from its capacity.

The overall power is then, beside the materials storage density, determined by the mass of the storage medium inside the silos (compare also Fig. 1).

However, recent investigations on the reaction system  $Ca(OH)_2/CaO$ , for example, have shown that severe bulk effect of the powdery storage material inhibit its movement or fluidization, as the averaged particles diameter becomes too small for further movement or fluidization [15] by making the particles highly cohesive [16].

Therefore, in case of this very prominent reaction system, but also in case of other materials with a similar behaviour, a lot of research is spent on suitable options for persistent stabilization of the particle size over a multitude of reaction cycles [17-21].

Concerning the pure storage material, the storage density plays a major role for mutual comparison and hence some important values have to be calculated upon first consideration. At first, of course, the reaction enthalpy of the respective charging / discharging reaction has to be determined. In most cases, this can be facilitated using tabulated enthalpies [**22,23**] of formation of the involved solids of the charged (c) and discharged (d) as well as x mole of the gaseous phases (Eq. 1). In the case of some very rare compounds, especially hydrates, some tabulated data may be missing. However, the reaction enthalpy can then be determined experimentally be differential scanning calorimetry or similar caloric methods [**24**].

$$\Delta H_R = \Delta H_{Fc} + x \cdot \Delta H_{Fgas} - \Delta H_{Fd} \qquad \text{Eq. 1}$$

With the determined reaction enthalpy, the gravimetric storage density can be calculated by consideration of the molar mass of the charged species (Eq. 2).

$$\Delta H_{grav} = \frac{\Delta H_R}{M}$$
 Eq. 2

The most important value is probably the volumetric storage density which is defined as the product of the gravimetric storage density and the true density of the charged storage material. Although this is only one piece of the puzzle, the obtained value is more realistic with regard to a later application than a consideration of the discharged species would be.

Additionally, by utilization of the true density of the material, the porosity present in bulk scale is neglected. In case of a fixed reaction bed, it would be suitable to determine the volumetric storage density by putting the raw density of the material into the equation. However, this is not feasible in case of moved or fluidized reaction beds.

$$\Delta H_{vol} = \Delta H_{grav} \cdot \rho_c \qquad \qquad \text{Eq. 3}$$

Beside a simple storage of energy in form of heat, thermochemical systems can also be used for heat conversion (TEC), as the temperature required for charging or discharging of a respective gas-solid reaction is dependent on the partial pressure of the gaseous reaction partner. This principle could be used to significantly enhance the efficiency of heat utilization, since it allows for the conversion of waste heat to a reutilizable temperature level.

Waste heat, which is released at a middle temperature  $T_m$ , could be used for induction of thermal decomposition (e.g. charging) of a thermochemical system at a low partial pressure of its respective gaseous reaction partner. Thereby, the gaseous reaction partner, as for example water, is released and condensed at a lower temperature  $T_1$ . Upon demand, the reaction partner is evaporated and recombined to the solid reactant at a high partial pressure  $p_h$ .

As a consequence, discharging will occur at a higher temperature level  $T_h$  and could be utilized within other processes (**Fig. 3**). This scenario could be of high advantage for a large number of applications in industry, especially in chemical, steel or cement industry where large amounts of waste heat are generated [**25-27**].

By running this process in the opposite direction, it is also possible to downgrade waste heat to a lower temperature level, what could be suitable for a variety of cooling applications.



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# Requirements and restrictions on materials for technical implementation

The selection methodology of materials for thermochemical energy storage and conversion contains numerous criteria, which can vary in literature since respective works mostly focus on only one possible application. However, a generalized list can be made with regard to the basic principles of operation [**28-30**].

#### Fundamental requirements and restrictions

The most fundamental criterion for potential candidate reaction systems is of course the unlimited reversibility of the chemical charging and discharging reaction under the applied conditions of temperature and pressure. Therefore, possible side reactions in any phase of the process must not occur, since otherwise continuously thermochemically reactive material would be converted into species, which do not contribute to the charging or discharging reaction. This criterion also includes, that the thermochemical material must not have corrosive properties towards the reactor, which is in most cases made of steel [**31**].

Since large scale industrial applications are considered for thermochemical systems and hence economic considerations play a dominant role, the material itself should be of good availability at low cost.

Another criterion is the environmental sustainability and non-toxicity of the materials. Possible options for later recycling, also in other applications, would further increase the respective materials benefits.

In case of materials considered for thermochemical storage, of course the gravimetric and volumetric storage density should be as high as possible. In contrast, this is not a major aspect during the selection of materials for thermochemical energy conversion [24,32]. In this case, the reaction hysteresis between charging and discharging plays a major role, since it determines the extent of the upgrade of waste heat, as illustrated in the van't Hoff plot depicted in Fig. 3.

### Application specified requirements

The criterion of highest relevance when selecting a thermochemical material for a specific application is its equilibrium temperature range, which is a unique thermodynamic property to every reaction system. For this reason, a lot of efforts are recently made in the search for new materials within several screening approaches considering a variety of applications.

Dependend on the application, especially with regard to the applied water vapor pressures in case of reaction systems using water as the working fluid, possible melting or deliquescence [**30**] of the discharged storage material may have a strong negative impact on the bulk properties, e.g. heat and mass transport, and should be avoided in case other candidates have a better match to the requirements.

Other criteria will probably be figured out more precisely in the future, which focus on the performance of thermochemical materials in bulk scale. As indicated earlier, some researchers have already figured out obstacles in heat and mass transport of the Ca(OH)<sub>2</sub>/CaO system upon cycling in larger scale by decrease of the particle size below a diameter of 20  $\mu$ m and the accompanying tendency of the material to form agglomerates [**33**]. Within the agglomerates, the microstructure is densified in comparison to the rest of the powder bulk, and thus the rate of conversion upon charging and discharging is decreased. Hence, respective criteria should allow a decision on whether the storage material needs to be modified or not on the basis of experience gained during bulk cycling.

Another obstacle is deliquescence of highly watersoluble compounds during the rehydration of the discharging process. In this case, the storage medium melts in its own crystalline water, forming a solution. When this solution is dehydrated, e.g. charged again, all prior bulk properties are lost, with the material forming a dense bulk. Hence, the next discharging is strongly hindered since the reaction bed is almost impermeable for the gaseous reaction partner. Various works concerning these types compounds present approaches for their persistent stabilization by embedding them in porous carriers as porous carbon [34,35], vermiculite [18], zeolithes [36] or silica gel [37].

# Materials yet considered for thermochemical heat storage and conversion

Although also other systems have been proposed, this section focusses on the major classes of potential thermochemical materials yet discussed in literature. Some rarely considered systems may be found in André *at. al.*, **[28]** or in the work of Yan *et. al.*, **[38]**. For example, metal sulphates (MSO<sub>4</sub>) based reaction systems are left out here, since they are operated with SO<sub>3</sub> or SO<sub>2</sub> as working fluids, which upon contact with only low amounts of water are highly corrosive towards metals due the formation of H<sub>2</sub>SO<sub>4</sub> or H<sub>2</sub>SO<sub>3</sub>.

### Carbonates

Metal carbonate-based reaction systems are operated with  $CO_2$  as the gaseous reaction partner. Most promising candidates are MgCO<sub>3</sub>, CaCO<sub>3</sub>, SrCO<sub>3</sub> and BaCO<sub>3</sub> [2] which have operating temperatures ranging from 300 °C for MgCO<sub>3</sub> up to 1600 °C for BaCO<sub>3</sub> [28]. Due to its beneficial properties as good availability, cost efficiency and recyclability among these materials, the system CaCO<sub>3</sub>/CaO with operating temperatures around 850 °C is one of the most considered for high temperature energy storage [28].

### Metal oxides

Thermochemical conversion of metal oxides is based on reversible reduction and oxidation. Various materials have yet been studied for storage application and are listed in the work of André *at. al.*, **[28]**. In comparison to other classes, these systems have the crucial advantage, that air could



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directly be used without further processing as the working fluid, allowing for cycling in an open loop system. Although some reaction systems are presented which contain costly noble metals copper or platinum, others seem quite reasonable for further investigations, since the operating temperature range is in a reasonable frame from around 205 °C for MgO<sub>2</sub>/MgO up to around 880 °C for BaO<sub>2</sub>/BaO. Some calcium-based reaction systems are also considered for implementation in chemical looping processes [**39**].

#### Hydroxides

Metal hydroxide-based reaction systems contain yet the most discussed systems for thermochemical heat storage in literature. These prominent examples are  $Ca(OH)_2/CaO$ , which has already been tested in 10 kW-scale [40] and Mg(OH)\_2/MgO, which has the disadvantage of a rather slow discharging rate [41]. Hydroxides are in principle susceptible towards carbonation, hence respective systems have to be operated under exclusion of  $CO_2$ .

#### Hydrates

As also in the case of hydroxides, hydrates are operated with water as the gaseous reaction partner. But since water is only bound as water of crystallization, the reaction enthalpies are usually lower than those of hydroxides, oxides or carbonates. However, since their operation temperature ranges are in a scale from roughly 50 °C up to 500 °C, these systems are of special interest for domestic applications or for waste heat recovery [**24,30,32**]. Thus, numerous approaches were recently made in order to identify suitable materials for the various applications [**29,30,38,42-44**]. Thereby, hydrates of SrBr<sub>2</sub>, CaCl<sub>2</sub>, K<sub>2</sub>CO<sub>3</sub>, CuBr<sub>2</sub>, CaC<sub>2</sub>O<sub>4</sub> and CaZn<sub>2</sub>(PO<sub>4</sub>)<sub>2</sub> are identified as promising candidates.



**Fig. 4.** Some examples of the volumetric storage densities and operating temperature ranges of thermochemical materials, classified according to their chemical composition in hydrates, hydroxides, oxides and carbonates.

Within Fig. 4, depicted below, the volumetric storage density of some selected reaction pairs has been calculated using Eq. 3 and plotted in dependency of the respective operating temperature range. These few systems show exemplarily, that carbonatebased systems have in tendency higher operating temperatures than hydroxides, which are in contrast higher than those of hydrated salts. Additionally, it can be seen, that there is a general tendency of increasing storage density with increasing charging temperature, which is quite reasonable from a thermophysical point of view, since chemical bond strength is increasing in the same direction.

#### The scope of potential applications

The scope of future potential applications of thermochemical systems is quite manifold. Probable the first considered application is for ensuring base load capability of concentrated solar power plants by storage of excess heat, which can be later converted into electricity, thereby allowing for a balancing of the generated power [45]. On the other hand, the principle could also be implemented for coupling of the sectors heat and electricity in order to transfer the energy generated by renewables into a utilizable form [4].

A third option for application of respective materials is concerned with the conversion of industrial waste. The amount of generated waste heat is tremendous and could be used to ensure roughly 30% of the space heating demand in Germany. Hence, by converting the temperature level of waste heat into a reutilizable range, the  $CO_2$ -bilance of primary energy production from fossil fuels could be significantly improved and the total energy consumption decreased.

Respective financial savings could be of significant profit for the industry. Hence, these applications could be of specific interest to highly energy intensive processes with high amounts of generated waste heat, as for example the chemical, steel or cement industry. Although there is already some data available concerning the temperature levels of waste heat [46-48], an expansion of such a database could significantly improve future material screenings.

A last important option which has to mentioned here is the utilization of thermochemical systems for domestic heat supply. Especially with regard to the reduction of  $CO_2$  emissions, new solutions for space heating have to be found when considering the fact, that for example around 70% of Europe's total energy consumption is used for domestic heating [49]. Donkers *et. al.*, spent significant efforts during reviewing 563 candidate reaction systems to find a suitable material which fits the defined restrictions for the intended domestic application [30].

All discussed options for future commercial applications are summarized in **Fig. 5**.

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Fig. 5. Classification of possible applications of technical solutions for thermochemical energy storage and heat conversion.

#### Screening strategies for new materials

Most of the recent screening approaches are based on thermodynamic calculations on the storage density and the equilibrium temperature range before defined search algorithms in relation to the criteria defined by certain applications are considered [**29,30,42**]. These calculations were predominantly performed for salt hydrate or hydroxide reaction systems, while Deutsch et. al. also considered other systems as oxides and carbonates. Although respective results allow for a first impression on whether a certain reaction system could be suitable or not, a subsequent experimental evaluation of the reversibility has inevitably to be performed in order to identify possible obstacles which could arise from a large reaction hysteresis between charging and discharging or issues related to the reaction kinetics.

This information is necessary for a sophisticated decision and could lead to an exclusion of a respective candidate. An experimental study on salt hydrates for low temperature thermochemical energy storage was performed by N'Tsoukpoe et. al., who evaluated 45 systems and identified SrBr<sub>2</sub>·6H<sub>2</sub>O, LaCl<sub>3</sub>·7H<sub>2</sub>O and MgSO<sub>4</sub>·6H<sub>2</sub>O as suitable candidates [43]. Richter et. al., Also Richter et. al., applied an experimental approach to identify salt hydrates as materials for a thermochemical heat transformer by combining the most frequent cations and anions to binary compounds and then searching for possible hydrates. Similar to N'Tsoukpoe, after a first discrimination due to criteria as toxicity, melting or possible side reactions, a number of 32 salts was characterized regarding their reversibility of charging and discharging. Finally, SrBr<sub>2</sub>·H<sub>2</sub>O was matching the defined requirements [32].

In order to also gain information on ternary or higher compounds, the author performed an experimental screening among mineral species, from which  $CaZn_2(PO_4)_2 \cdot 2H_2O$  was identified as possible candidate for the low temperature range around 200 °C [**50**]. Although this material is a rare mineral and not available commercially, it can easily be synthesized [**51**] and has corrosion protective properties. Apart from the various screening approaches, there are also efforts made in adapting the thermophysical boundary conditions of thermochemical reaction systems by chemical modification. For example, Shkatulov et. al. reported on the doping of magnesium hydroxide with sodium nitrate and observed a reduction of the decomposition temperature by 25 °C at a content of 2 % (w/w) of the additive accompanied by a faster decomposition rate upon charging [**52**].

Similar approaches have been investigated on other reaction systems as  $Ca(OH)_2/CaO$ ,  $MnO/Mn_2O_3$  or  $CaCO_3/CaO$  [53-55]. It is reasonable, that by induction of lattice distortions and other crystalline defects, the onset of the respective charging or discharging reaction can be shifted. Hence the strategy of doping, or more general, modification of the chemical composition, could allow for an adjustment of the working temperature range, at least to a certain extend.

#### **Conclusion & future prospective**

The discussions contained in this work reveal the variety of potential future applications of thermochemical storage units as for example in CSP plants, for the reutilization of industrial waste heat for process heat generation as well as for housing purposes.

Corresponding to the applications, numerous reaction systems principally matching the respective thermophysical boundaries have been identified yet, but the search is still going on. Thereby, focus is set on one hand on the identification and thermal investigation of new reaction systems. On the other hand, efforts are made to influence charging and discharging temperatures by modification of the chemical composition the material design. In order to promote technical development of systems for thermochemical energy conversion or storage, which in any case consist of a reaction pair cycled under certain conditions inside a reactor, matches of applications and materials have to be identified.

In some cases, respective matches have already been found.

However, it could make sense to establish a database containing the boundary conditions of potential use-cases in the future. This could not only direct future screenings for thermochemical materials into industrially useful directions, it could also help to identify suitable options for the amendment of respective thermophysical material properties of reaction systems which are already known.

In all cases, the characterization and adjustment of the bulk performance of thermochemical reaction systems under realistic conditions seems to be a next step towards technical maturity. Of course, such investigations cannot be included in materials screenings, which are by themselves quite challenging. But they would allow for a mutual adaption of thermochemical reactors to respective materials, which is an essential part of the upscaling necessary for industrial implementation.

#### **Conflicts of interest**

The authors declare no competing personal or financial interests.

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#### **Authors Biography**



Dr. Sandra Afflerbach studied Chemistry at the University of Siegen, Germany, where she obtained her PhD in 2018 in the group of Building- and Materials Chemistry, headed by Prof. Dr. Reinhard Trettin. Since 2019 she is working as a senior engineer in the group of Environmental and Process Engineering headed by Prof. Dr.-Ing. Wolfgang Krumm. Her field of research is thermal energy storage with special regard on mutual material and reactor development for thermochemical heat storage and conversion.

#### Graphical Abstract





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