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Review

A Review on the Challenges of Using Zeolite 13X as Heat Storage Systems for the Residential Sector

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Abstract: In recent years, several attempts have been made to promote renewable energy in the residential sector to help reducing its CO_2 emissions. Among existing approaches utilizing substances capable of directly storing and transporting thermal energy has recently become a point of interest. Zeolite 13X with exceptional capacity to safely store thermal energy for long periods and release heat due to its unique molecular structure is known to be one of the best options serving this purpose. However, the application of this ceramic as a heat storage material in the residential sector is associated with significant challenges dictated by the limitations of the sector, such as space restrictions and affordability. The current review attempts to explore the extent of these challenges, mainly related to design and efficiency from different perspectives. The main aim here is to provide a clear vision for a better understanding of the state of the art of this technology and to help to identify possible solutions fostering the adaptation of this technology to the residential sector.

Keywords: TES system; heat adsorption; thermal storage system efficiency; residential sector heating

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1. Introduction

One solution to fully exploit renewable energy in residential heating is to develop a technology by which the thermal energy received by the sun could be directly stored during the day and released whenever needed. Doing so would result in a significant reduction in the residential share of fossil fuel consumption and CO_2 emissions while providing residential heating. The key factor to attain such goal lays in finding suitable substances capable of storing high level of thermal energy and efficiently substituting them in a residential thermal system.

In recent years, zeolite 13X has received considerable attention to be used as an efficient heat storage substance in thermal systems. Due to its special molecular structure, which contains well-defined microchannel and cavities, zeolite 13X can store heat by removing humidity and release heat when humidity is introduced to the compound, which gives zeolite 13X a unique heat storage property. The stored heat will be confined as long as no humidity is introduced to the system. This feature provides a simple, safe, and affordable mechanism for portable thermal energy. Despite these unique features, Thermal Energy Storage (TES) systems containing zeolite13X have not yet acquired a significant share in residential heating due to their poor operating performance for the sector.

2. Thermal Energy Storage Systems

To provide a better understanding of TES challenges, in this section, briefly, the underpinning theory, as well their systematic characteristics, are explained. In general, TES systems have been used widely in the industrial sector (with some large-scale residential applications). These systems are divided into three main categories based on their storage methods: Sensible heat, Latent heat, and thermochemical heat, as presented in Figure 1. As can be seen in all of these systems, a high-capacity storage substance is implemented to receive, save, hold, or carry the thermal energy. The performance process of these substances can be characterized in three main steps, charging, storage, and discharging. In Figure 2, these three processes are presented for the different TES categories. As presented in Figure 2a, Sensible heat storage systems use the simple method of storing heat by discharging excess heat from the material without phase change. In Latent heat storage (Figure 2b), the stored heat is the result of a phase change; thus, they can attain a higher thermal density compared to the Sensible method. In Thermochemical systems (Figure 2c), unlike the two other methods, the stored heat is obtained from a reversible thermochemical reaction.

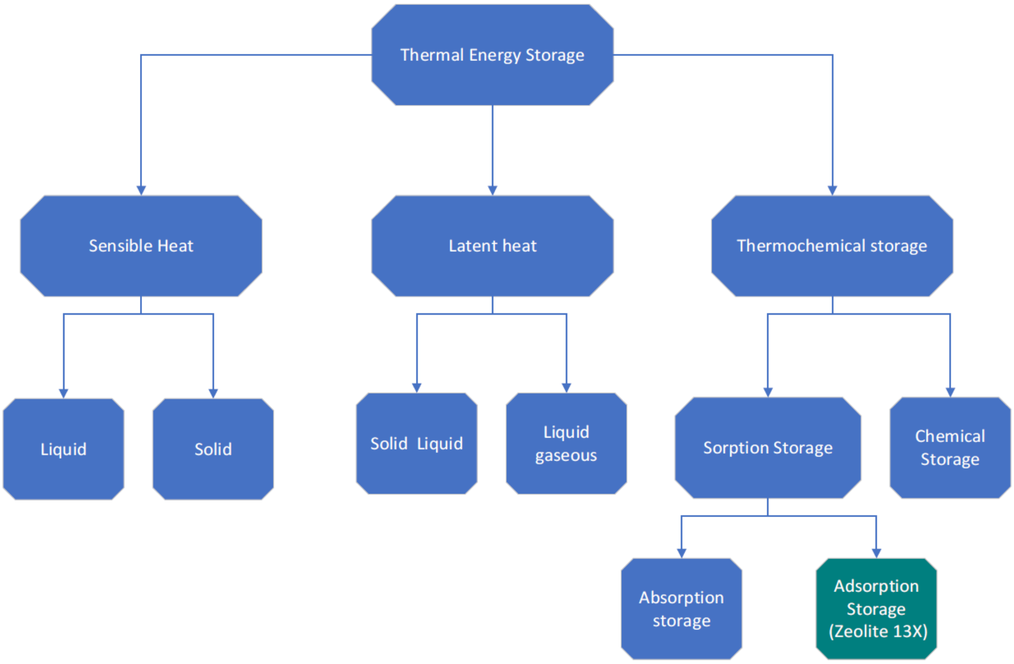


Figure 1. Classification of different thermal storage methods.

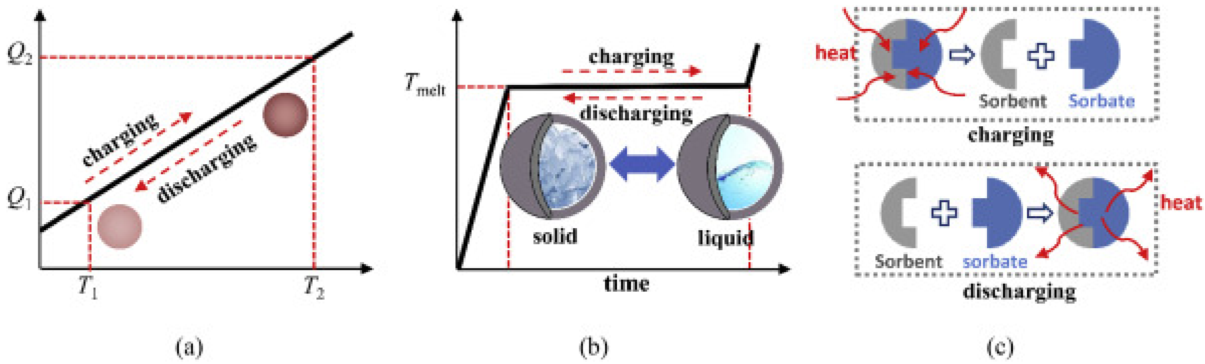


Figure 2. Methods of thermal energy storage (**a**) Sensible heat, (**b**) latent heat, (**c**) thermochemical heat (Sorption heat) [1].

Thermochemical systems are particularly efficient in storing heat for long-term storage applications because heat loss from the system is low.

In addition, Thermochemical storage systems, due to their high-density storage characteristics, provide a spatially efficient and compact storage system compared to the other heat storage. A volumetric comparison between the three main categories is presented in Figure 3; as can be seen, for instance, replacement of latent system by Thermochemical would save up to 10 times the required space, which makes these types of systems advantageous for residential sector application [2].

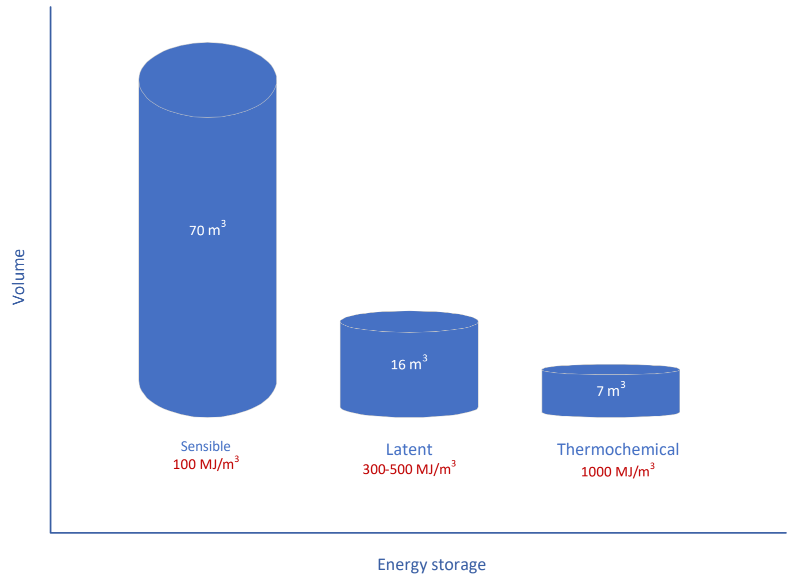


Figure 3. Volume needed to full cover the annual storage need of an energy efficient passive house (6480 MJ) [3].

Given that zeolite 13X stocks heat by adsorption method that belongs to the thermochemical category, in this review, the focus will be on adsorption heat storage.

Adsorption is the movement of atoms or molecules from a bulk phase (which might be solid, liquid, or gas) to a solid or liquid surface. Heat is released as a result of attractive interactions between the surface (adsorbent) and the molecules being adsorbed (adsorbate) [2], as presented in Figure 4.

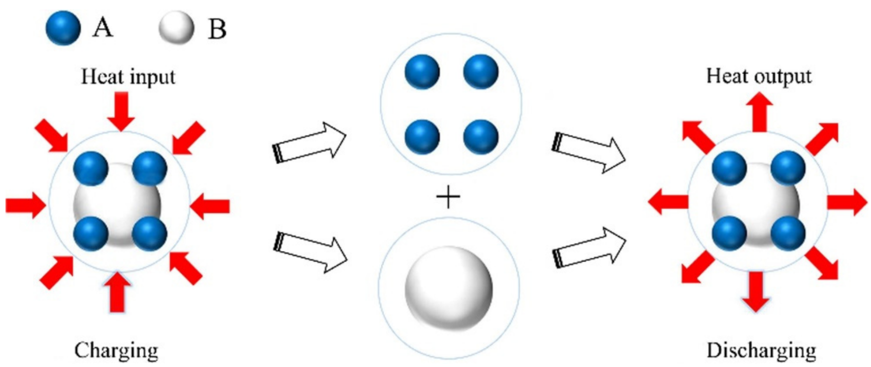


Figure 4. Charging and discharging thermochemical heat by sorption reaction [4].

The attractive potential for this thermochemical reaction can be explained with Lennard-Jones potential (Figure 5) equation as a function of distance:

$$V_{\mathrm{LJ}}(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \quad (1)$$

where σ is the distance at which the potential is zero, and ε is the depth of the potential well. According to Equation (1), higher ε means more heat would be released after adsorption. The extent of this potential varies from 8 to 800 kJ mol^{-1}. The amount of adsorption depends on the inherent properties of the material in reaction, such as specific surface area, and the affinity of the couple adsorbent/adsorbate. The adsorption process also is subject to operating conditions, such as temperature, pressure, and concentrate of the adsorbate. For instance, high specific surface area, high pressure, and low temperature increase adsorption reaction rate.

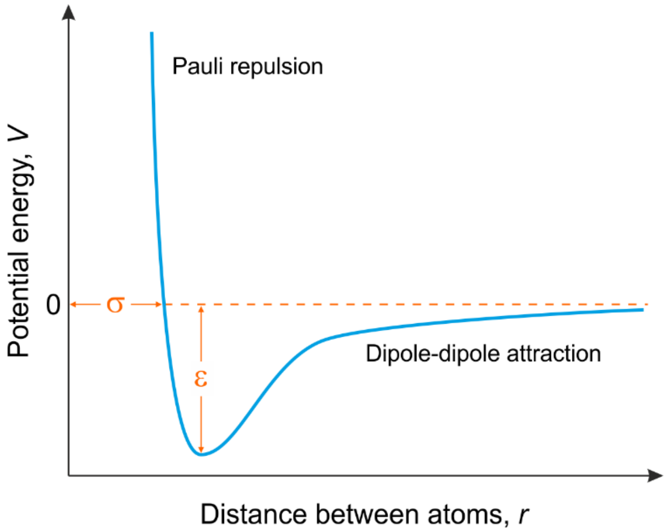


Figure 5. Lennard-Jones potential.

3. Adsorption Heat Materials

Generally, all solids are adsorbents, but only those with a high specific surface area are interesting for sorption applications. In this section, a few adsorbent families are briefly introduced.

In industrial applications, activated carbons are the most often used adsorbents. It is mostly employed in the purification of gases. Non-polar activated carbons are commonly utilized for water treatment. As a result of their poor affinity for water, they are not employed in heat storage applications. Silica gel is a synthetically porous type of sodium silicone dioxide. It may be represented as $SiO_2.nH_2O$ in its chemical compound. Treating synthesis (PH and presence of cations) control carefully permits pores to be controlled. Its large specific surface area (from 600 to 800 $m^2.g^{-1}$) and hydrophilic characteristics make it an excellent desiccant, and it has a very high adsorption capacity at low pressures and temperatures. When silica gel is saturated with water, it loses its capacity to create heat for long periods of time, which is one of its limitations as a heat storage medium.

Zeolites are porous crystalline minerals composed of silicon (Si), aluminum (Al), and oxygen (O) atoms. In tetrahedral configurations, each Si or Al atom is linked to four oxygen atoms (SiO_4 and AlO_4). Each oxygen atom is shared with another tetrahedron that connects Si or Al atoms, as presented in Figure 6. This configuration results in atomic angles remaining the same and, thus, a uniform distribution in pore sizes. On the other hand, since each oxygen atom is shared between two tetrahedral Si or Al atoms, the stoichiometric composition of each tetrahedral unit is SiO_2 or AlO_2 with minimum Si/Al ratio to be 1.0 without any upper limit. Given the fact that rich Aluminium sieves has a high affinity for water (since each Al atom introduces a negative charge in the material and polar molecules, such as water, are sensitive to these charges), it demonstrates a high level of hydrophilic behavior. Conversely, since rich silicon sieves have a hydrophobic behavior, heat induced transition between hydrophilic and hydrophobic behavior (usually occurs at a Si/Al ratio between 8 and 10 [5], plus uniform pore sizes, introduces zeolites as an exceptional adsorbent fit for TES systems.

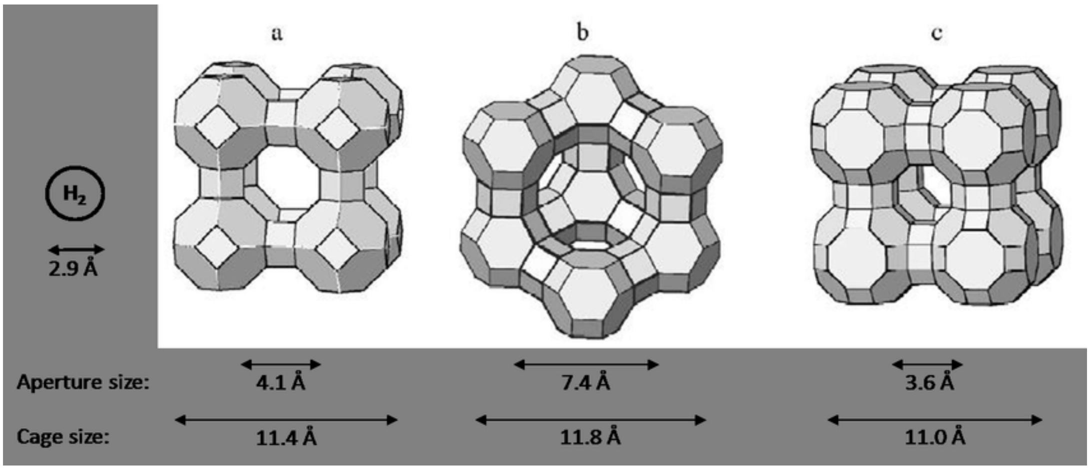


Figure 6. Zeolite framework structures: (**a**) A, (**b**) X and Y, (**c**) Rho. The H_2 molecule is shown to scale for comparison [6].

There are various type of zeolite and their composite used for heat storage [7]. Among the zeolite family, zeolite 13X (Figure 6b) has the largest permeability for water molecules due to its inner hole size, which plays a key role in heat storage efficiency.

In addition, while employing zeolitic composite as heat storage may be more efficient, the use of added materials and the synthesis process raises health concerns that zeolite 13X, in turn, does not.

To make use of this exceptional adsorptive capacity, zeolites must be placed inside reactor beds to undergo the aforesaid three performance processes (charging, storing, and discharging). The efficiency of performance processes is closely related to controlling the reactor bed operational condition, such as temperature, flow rate, and moisture level.

There exist varieties of reactor systems that can serve the purpose, among them open adsorption systems with the fixed bed being the most used type. A schematic of these systems is presented in Figure 7. To charge zeolite in this system, a stream of hot air with minimum 120 °C must be injected into the reactor, where zeolite is placed. The injected hot stream into the bed, while charging zeolites, also extracts and transports its trapped moisture.

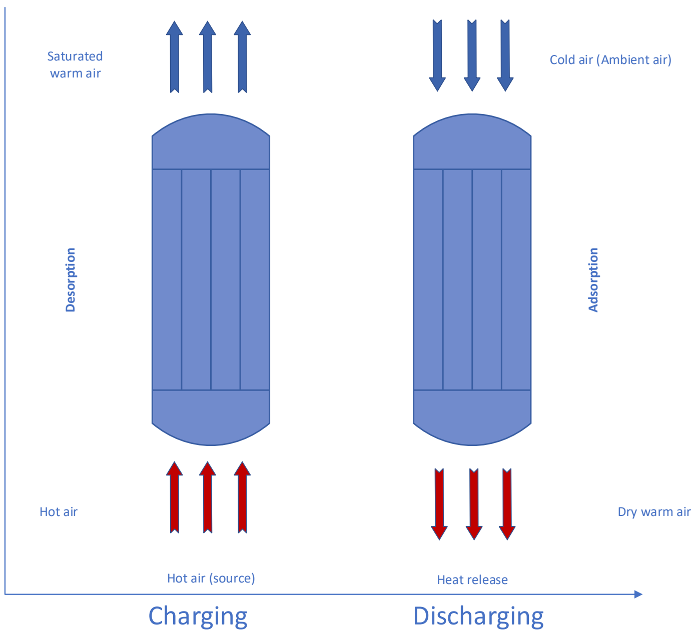


Figure 7. Open adsorption process.

Regarding the storage step, thanks to the unique zeolite structure, the stored heat will remain untouched until moisture re-enters the reactor, which makes this system needless for thermal isolation (a specific advantage compared to other systems). In these systems, heat sorption capacity is defined as:

$$Q \text{ sorbtion} = -V\ |\Delta H|\ \Delta q \quad (2)$$

where V is the bed volume, $|\Delta H|$ is the specific sorption heat, and Δq presents the sorbate uptake capacity between charging and discharging phase [8]. During the discharge (adsorption) process, ambient airflow with precisely controlled humidity enters the reactor. Given that zeolite is hydrophilic at ambient temperature, it absorbs the humidity and releases the stored energy in the form of heat. The outlet air temperature and the discharging duration depend not only on the inherent property of zeolite but also on the effectiveness of reactor system design and its operational conditions. This makes reactors a key player of the heat storage systems, after selecting appropriate materials.

Given that the proper access to zeolite's unique storage capacity and its efficient delivery bonds to reactor parameters, the design and optimization of reactors would then become one of the main challenges in using heat storage systems in the residential sector. Figure 8 presents the reactor most important variables in three main categories: Reactor Structure, Auxiliary Equipment, and Design Parameters, each introducing their challenges.

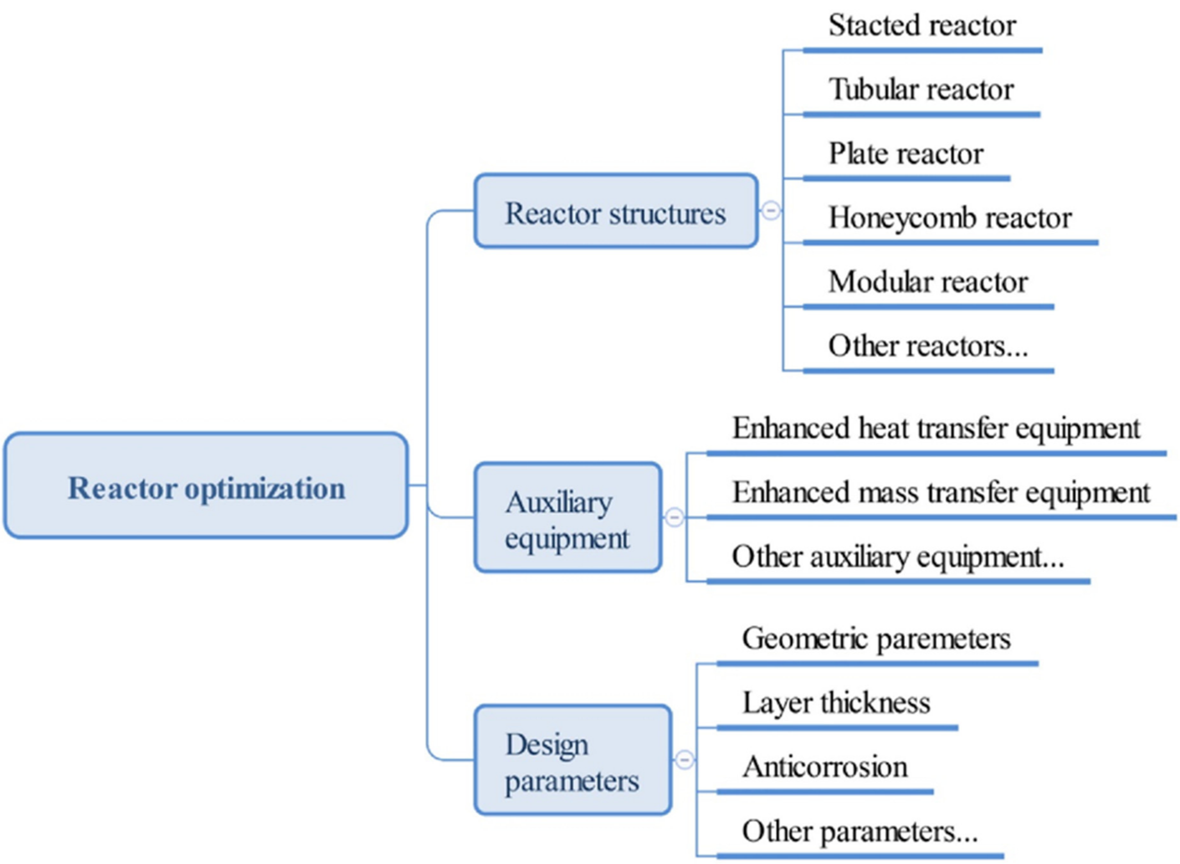


Figure 8. Influence impact on reactor optimization [9].

The main goal in TES systems is to optimize mass/heat transfer and decrease heat loss. To attain this, several configurations have been examined so far, such as staced, tabular, honeycomb, plate, modular, etc., in all these configurations parameters, such as reactor diameter, length, and connectors, have significant effects on the efficiency of heat storage and have been investigated by many researchers. Anderson et al. [10] found that bed length and heat loss have direct relationship, so, by reducing the length, heat loss can be reduced. Lahmidi et al. [11] used a stacted reactor. In this design, to improve mass transfer a nozzle device is added to the system, which increases the interaction area between solid and water vapor conducive. To further improve mass and heat capacity Stitou et al. [12] developed a pilot plan using high thermal conductivity of ENG. Layer thickness is another key parameter for reactor design, directly affecting the hydration time. Van Essen et al. [13] observed that decreasing the thickness speeds up the hydration process. Oktariani et al. developed system for generation steam by using a zeolite 13X-water system [14]. They found that the flow direction of feeding water from the top of the reactor using nozzle configuration could conform a better result than feeding water from the reactor bottom. Considering the aforementioned facts, it seems simpler reactors, such as a high efficiency staced reactor, could be the better option for the residential sector; however, thanks to the advancement of fabrication technology, modification to reactor design is yet to present future enhancements.

Evaluation of an efficient design can be tested using TES systematic performance parameters. In general, performance parameters can be expressed by three terms: thermal power density with unit (kW·m^{-3}), thermal storage density with unit (kWh·m^{-3}), and the Coefficient of Performance (COP) with the first two being proportional, and can be obtained from various design and condition parameters and the third to be defined as (Figure 9):

$$\text{COP} = \frac{Q_H}{W} \tag{3}$$

where W and Q_H are the power required to run the discharging process and the amount of extracted heat, respectively. As can be seen, the provided COP in the article, although present in the effectiveness of the discharge process, leaves the charging process untouched. To cover the whole performance cycle, it might be better to introduce TES cycle thermal efficiency η as:

$$\eta = \frac{Q_H}{W + Q_{in}} \tag{4}$$

where Q_{in} presents the provided heat to the system. It is now vivid that the thermal efficiency of the TES systems indeed depends on a variety of factors, on top of which the effectiveness of the charging process is present. The multivariable nature of TES ongoing performance process opens several challenges for further optimization and TES systems new applications (e.g., residential applications), to be investigated in the next section.

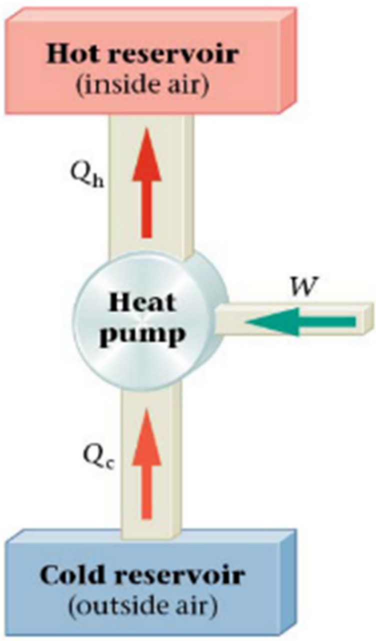


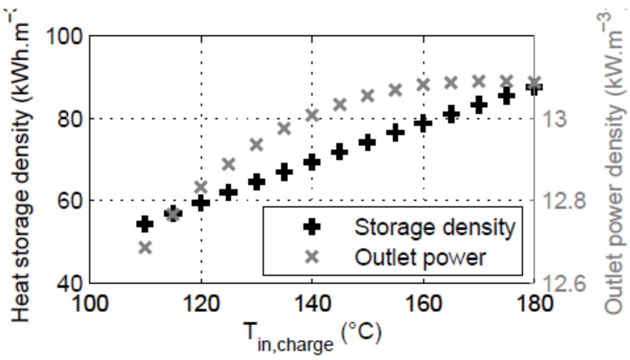
Figure 9. Coefficient of performance [15].

4. Technical Challenge

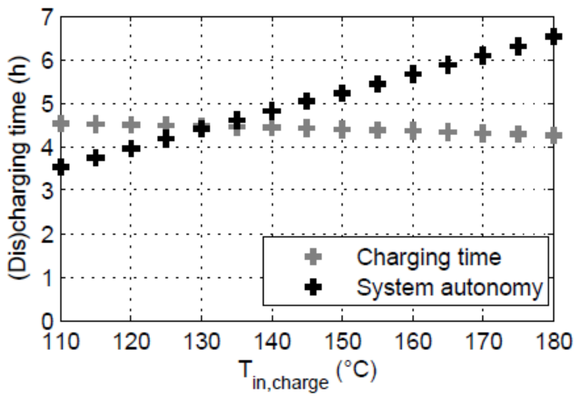
According to the facts mentioned in the previous section, one can easily conclude that to introduce adsorption systems in the residential sector a compact system must be designed due to space restrictions. However, given that providing the required amount of household power is directly proportional to the amount of zeolite, TES sizing would become a significant limiting factor in the development of an adsorption system for residential application. As a result, many researchers are attempting to improve TES system efficiency to make them capable of storing more energy in less space (highly efficient system). This transition introduces several challenges in executing TES performance processes (charge and discharge), such as efficient charging resources, precise humidity control, reactor design, manufacturing, etc. In this section, the main obstacles in achieving such a compact and efficient system will be discussed.

4.1. Required Temperature Supply

To begin the charging phase in adsorption systems, the inlet air temperature must reach the hydration reaction temperature. Depending on the materials employed, different storage systems require different input temperatures; for zeolite 13X, this temperature must be above 120 °C [16]. As a way to increase efficiency, many researchers investigated the effect of increasing input temperature on system function. Johannes et al., in 2015, used open-source heat storage, including two containers of zeolite 13X. They examined two different temperatures levels for the charging phase and compared their effects on the system charging phase. They discovered increasing temperature from 120 °C to 180 °C can enhance storage density by 40%, while decreasing charging time by 7% (Figure 10).



(**a**)



(**b**)

Figure 10. The effect inlet temperature on the heat storage density (**a**) and the charging time (**b**) [17].

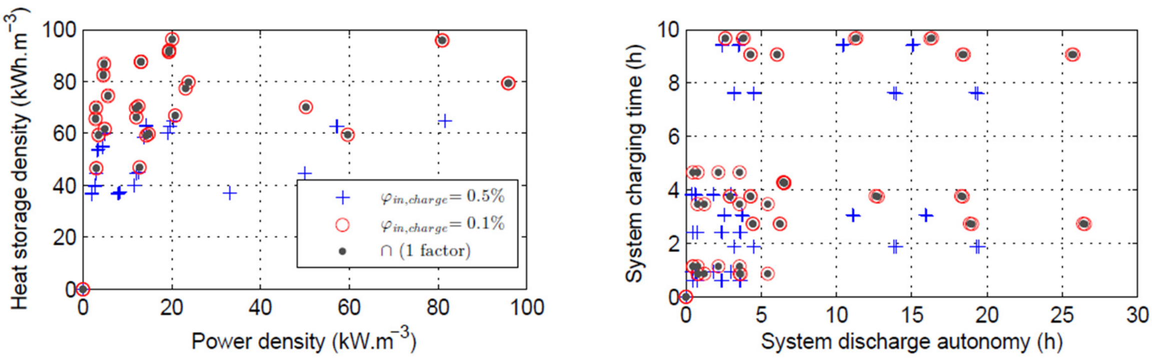
Required high temperature flow can be supplied from various resources. Table 1 illustrates some of the prototypes that were built, as well as the temperatures and heat sources that were used. As can be seen in the highlighted column, electric heaters have been employed to deliver the required heat in almost all instances. However, electric heaters have high consumption due to their low COP as a heating system. With a typical heater (consuming between 2 to 4 kWh), achieving higher temperature input flow seems a less than ideal solution to provide Q_{in}, unless a cost-effective power source can be provided. This necessity makes the intake power supply of compact TES systems a significant challenge to be addressed for residential applications.

Table 1. Used temperature for prototypes.

Setup Name	Year	Inlet Temperature [°C]	Heat Source	Outlet Temperature [°C]	Energy Density [kWh/m^3]	Max Power [kW]
Alebeek [16]	2018	180	Electrical heater	13	54	4.4
STAID [18]	2015	120–180	Electrical heater	20	114	2.25
ASIC [19]	2014	230–180	Electrical heater	25	148	1.5
E-HUB/ECN [20]	2013	185	Oil to air exchanger (Electrical heater)	25–60	58	0.4
MONDESTORE [21]	2008	180	Electrical heater	25	57	
MONOSORP [11]	2006	170	Electrical heater	20	120	1.5

4.2. Relative Humidity Control

The level of humidity entering to reaction area in the collector with open systems has the most influence on power density and storage density [22]. Relative humidity has direct effect on the sorbate uptake (as reflected in Equation (2)), which makes it a key parameter for storage density. Most prototypes employ an electrical humidifier, water tank, and sensor to supply and control the humidity of the system's incoming air [23]. Figure 11 shows how lowering the relative humidity in the charging process from 0.5 to 0.1 percent enhances both heat storage density (a) and charging time (b). It also demonstrates that storage density and power density are very sensitive to humidity changes. A change in humidity of 0.4 percent caused a significant power change, as can be seen in Figure 11. These characteristics must, thus, be considered to build an appropriate power system.



(a) Heat density against power on charge (b) Charging time against autonomy on charge

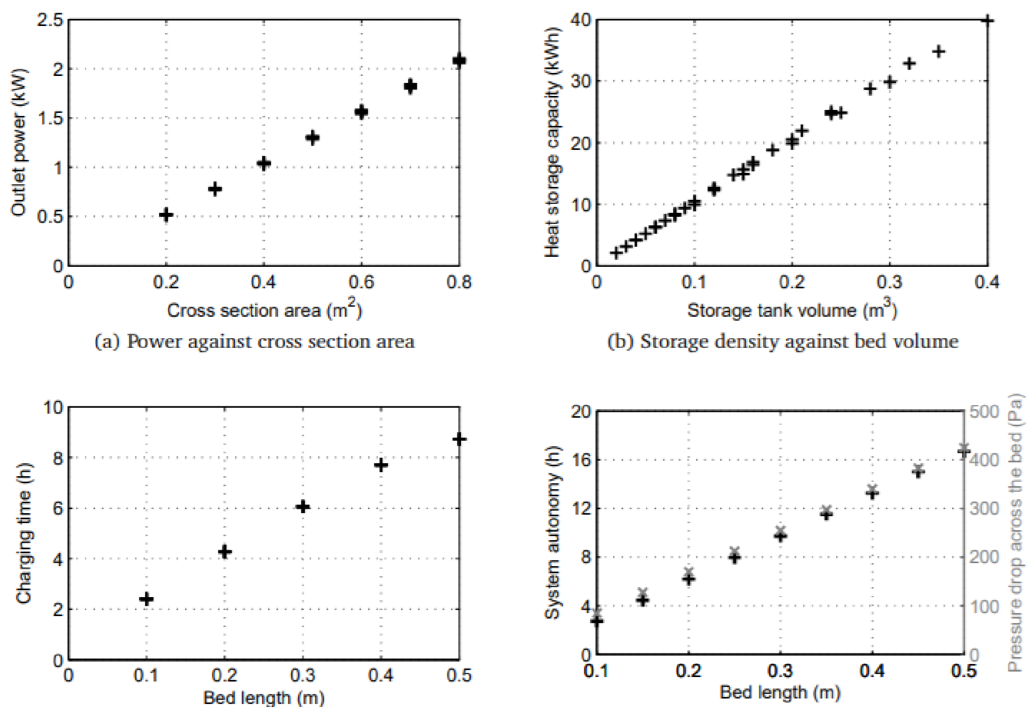
Figure 11. The influence of inlet relative humidity on the heat storage density and power density (**a**) as well as the charging time (**b**) [18].

For household applications, the inlet relative humidity becomes more important as unstable ambient air is used for the charging and discharging process [24]. Due to the strong dependency of stored and released heat on relative humidity, the use of TES system in the residential sector would introduce a significant challenge that necessitates implementing precise humidity monitoring for closed loop control, especially when variable setpoints are desired to adjust variable system's needs.

4.3. Reduce Reactor Size

Reducing the size of the reactor itself can improve their residential development. Smaller reactors occupy less space and are simpler to integrate with an energy source. However, as mentioned earlier, due to the direct relationship between zeolite volume and system power, constructing a smaller reactor faces numerous configuration and manufacturing limitations. Several investigations have been performed to identify and improve effective size criteria, such as bed length, cross section area, and tank volume by Kuznik et al. [22]

and Michel, Mazet, and Neveu [24]. Gondre et al. [17] found that there exists a linear between the outlet power versus cross section area, heat storage capacity versus storage tank volume, and charging time and autonomy versus bed length, as shown in Figure 12.



(a) Power against cross section area (b) Storage density against bed volume (c) Charging time against bed length (d) Autonomy against bed length

Figure 12. (**a**) The influence of cross section area on outlet power; (**b**) The impact of storage tank volume on hear storage capacity; (**c**) The influence of bed length against chrging time; (**d**) The impact of bed length on discharging time [17].

Their experiment presented the following significant outcomes, as illustrated in the Figure 12:

- outlet power is directly proportional to cross section area (a)
- heat storage capacity is directly equivalent to storage tank volume (b)
- charging time and discharging time is directly proportional to bed length (c,d)

Reducing the size of the reactor, as predicted, reduces the power of the system. Therefore, ways to optimize the reactor's dimensions should be investigated if residential applications are in the list.

4.4. System Output Power

Another point of interest in TES system optimization is the output power during its discharge process. Many researchers have been able to enhance TES system output power by selectively adjusting the discharge process parameters. Several prototypes in laboratories were designed for this purpose, presented in Figure 13. Among them, Jahannes et al. developed a high power open sorption system (STAID), which contains two reactors of 80 kg of zeolite13X [18]. The discharge parameter of this system was adjusted to generate heat for the residential sector during peak hours. Input ambient air flow temperature was considered to be 20 °C, and outlet temperature was 57 °C. It was observed that their system delivered 6 h of continuous heating during the discharge phase, equal to a maximum 2.25 kw of thermal power output.

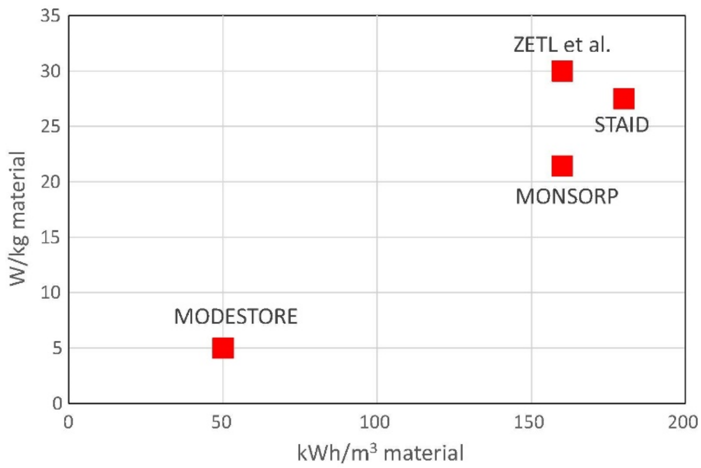


Figure 13. Comparison of mass power obtained from different physical adsorption systems, where STAID is the present work [18].

Zettl et al., of the Austria Solar Innovation Center (ASIC), used different techniques to increase the discharge power. They designed a prototype with a rotating bed that was capable to generate maximum outlet power 1.5 kW only using 50 kg zeolite 4A. The purpose of using rotating bed was to avoid the formation of dead zone to increase the outlet power. The input temperature was 25 °C, and maximum outlet temperature was 60 °C [19].

ADEnergy research Center of the Netherlands developed an open sorption concept using two beds with 150 kg zeolite 13X with the compact bed. This system was designed to supply warm air for the residential sector. The air is humidified with 12 mbar water vapor pressure, and air flow rate is 80 m^3/h. This system generates maximum 0.4 kW, and output temperature is 70 °C [25].

MonoSorp prototype was designed as heat storage system with opened bed for space heating. The input temperature is around 20 °C, and the maximum outlet temperature is approximately 42 °C. They used zeolite as extruded honeycomb structures to avoid pressure loss. This system was able to deliver maximum thermal power of 1.5 kW [13].

Figure 13 collectively present these TES systems specific power output concerning their involved substance mass and volume The research process, as shown in Figure 13, is aimed at improving extractable power based on material mass (W/kg material) and improving heat storage density (kwh/m^3), which, in addition to designing smaller systems, enables these systems to be used for a longer period of time, allowing them to deliver the required heat not for several hours, but for many days.

Although the current attempt has already proven the feasibility of utilizing such systems for residential use, it seems that further improvement of system output power is required market justifications. This would become a significant challenge as power optimization would directly point toward zeolite's physical limitations.

4.5. Efficiency

As mentioned before, zeolites have the unique capacity to store heat, and, due to their high structural endurance, they can tolerate numerous thermal cycles. These characteristics make zeolites known for their high thermal efficiency; however, utilizing them in TES systems has yet to reveal anything near to their optimum capacity. In this regard, in the literature, one can find two different efficiencies reported by scientists and engineers which indicates zeolite material efficiency and its system efficiency. Figure 14 shows these efficiencies for TES system containing zeolite in an open bed reactor. In this prototype, developed by Kuznik, the reported material efficiency was about 70%. However, one can clearly see that, when it is placed in the storage systems, this efficiency drops to around 36%.

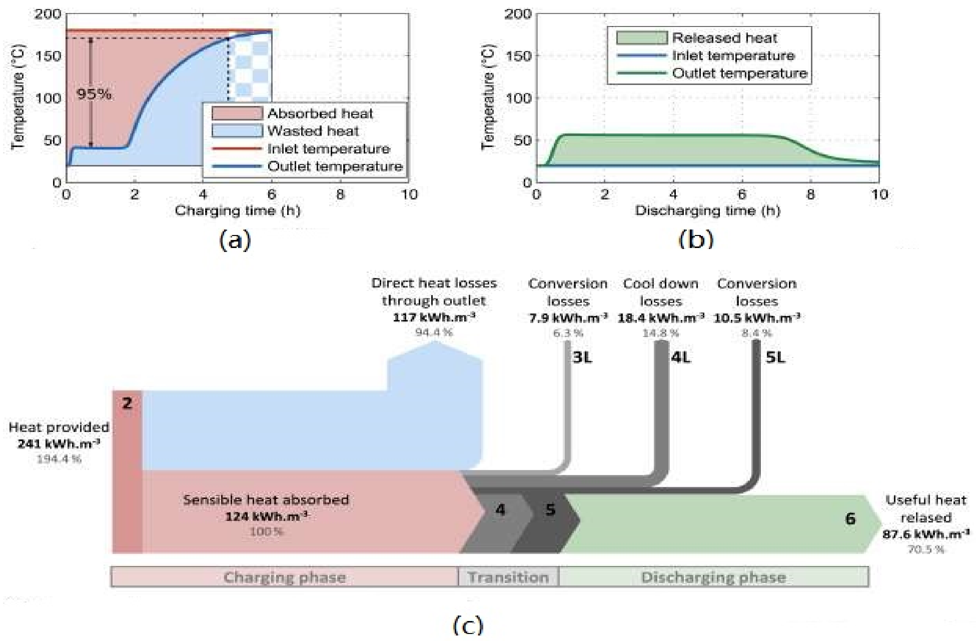


Figure 14. (**a**) The influence of charging time on the temperature profile (**b**) The impact of discharging time on the temperature profile; (**c**) Heat losses in the TES system [22].

As can be seen in Figure 14c, there exist several losses associated with all the three TES main processes which drop the engineering efficiency significantly. Nearly half of the injected heat is directly discharged through the outlet in the charging process.

The type of reactor and the inherent feature of zeolite can be responsible for the large amount of energy wasted during the charging phase. Zeolites, despite their high energy storage capacity, have poor thermal conductivity. Thermal conductivity is very important for increasing the internal temperature of zeolites to a level where the zeolite's internal moisture can release as a gas. Figure 14a shows the consequences of low thermal conductivity, as the reactor outlet temperature begins to increase after two hours of the charging process begins.

During the storing period, there exist charge conversion losses, cool down losses, and, finally, discharge conversion losses for the discharge process. There is no energy loss during the discharging phase, and the outlet temperature remains constant for six hours Figure 14b. All these losses, on the other hand, provide a window of opportunity to be addressed by engineers which, of course, brings many new challenges to the table.

5. Conclusions

Different aspects of utilizing zeolite 13X as a heat storage medium have been briefly discussed, and the main challenges have been summarized. It was found that zeolite 13X has high potential to be used in the residential sector; however, there exist several challenges that should be addressed prior for this to happen. This review demonstrated and discussed the variety of challenges from different perspectives, such as compact system design, charging supply, humidity management, system output power, and system efficiency. It was concluded that, to bring this technology to the residential sector, compact efficient designs are required, including techniques that lead to increasing the contact area while reducing the size of the system, a contradiction to be solved. In addition, it was concluded that, to make system energy intake justified, more affordable energy resources should be identified and implemented. Overall, the current review reveals that, although the implication of the zeolites TES system as a heating system seems feasible, they are, by far, set to become a serious competitor for current heating appliances in the residential market. This outcome, although it may negate the business side of the technology, offers several nitce opportunities for potential experts of the field.

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