



Chemisorption Heat Pump with High Energy Density Thermal Storage for Space and Water Heating

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Abstract

Space and water heating account for approximately half of the energy consumption in buildings in cold climates, contributing more than 4,000 MtCO₂ of direct and indirect emissions. Electric heat pumps offer high-efficiency heating solutions; however, low ambient conditions present significant challenges to their adoption. Thermally driven adsorption heat pumps provide an energy-efficient, low global warming potential, and robust alternative for such operating conditions. This paper presents a proof-of-concept prototype reactor using magnesium chloride (MgCl₂) and graphite for chemisorption heat pumps. Laboratory experiments show a composite energy density of ~1000 kJ/kg with a heat delivery temperature range of 45–70°C, making it suitable for space and water heating applications. The experimental facility can also be used to evaluate other materials. A four-bed configuration of a compression-assisted resorption heat pump is presented and analyzed using a thermodynamic cycle model. The system utilizes chemisorption reactions of metal halide salts (both low- and high-temperature salts) and ammonia, and the compressor varies the pressure, enabling the utilization of low-grade heat sources for system operation. The developed quasi-steady thermodynamic cycle model of the complete system allows the screening of different materials based on their overall system performance, using the coefficient of performance (COP) for cooling and heating, specific cooling power (SCP), and heating power (SHP), as well as the compression ratio (r_p), as key parameters. Among the different materials evaluated using the model, the salt pair of ZnCl₂-SrCl₂ yields the lowest compressor power and the highest COP of ~0.8, but it has lower specific cooling and heating power. Finally, the electric COP of the proposed system is compared with a standard vapor compression heat pump to assess operational energy savings and economic viability. Ongoing work involves in-depth screening of different salt pairs for realistic operating conditions and development of a lab-scale system prototype.

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1. Introduction

Chemisorption reactions can store and release energy over a wide temperature range, making them highly effective and versatile for heat storage and heat pump applications. For this reason, chemisorption cycles have been applied to refrigeration [1] and heat pumping [2]. Chemisorption reactions have an ab/adsorbed phase (refrigerant) and an adsorbent (activated charcoal, water, salt, etc.). The chemisorption reaction of halide salts with ammonia is promising because of the high energy density of reactions, a suitable operating temperature range (10–150 °C) for heating and cooling, and excellent reaction reversibility [1]. Ammonia has a high vapor density, large enthalpy of vaporization, and a low freezing point (–77°C), making it an excellent refrigerant.

The chemisorption reaction of metal halides with ammonia has an equilibrium state where the rates of adsorption and desorption of ammonia are equal, and it is modeled by the Clausius-Clapeyron equation.

$$\ln \left(\frac{p_{eq}}{p_{ref}} \right) = -\frac{\Delta h}{RT} + \frac{\Delta s}{R} \quad (1)$$

Here, Δh is the enthalpy of the reaction associated with per unit mol of ammonia adsorbed/desorbed, Δs is the entropy change associated with the reaction, R is the universal gas constant (8.3145 J/mol-K), T is the temperature of the adsorbent related to the equilibrium pressure (p_{eq}) of the solid-gas pair, and p_{ref} is 1 Pa. Eq. 1 is used to screen adsorbents based on the pressure and temperature requirements of the application. The methodology for screening adsorbents-adsorbate pairs for a specific end case has been extensively described by Yang et al. [3].

1.1. Prior Art

Chemisorption heat pumps (CSHP) are categorized into two configurations based on design. The first is the adsorption-desorption configuration (also called a single-effect with NH_3 evaporator and condenser [3]), where a single adsorbate bed is connected to an ammonia evaporator/condenser, and it switches between the two modes based on whether it is charging or discharging. The second is the resorption configuration, where two adsorbate beds are connected without any ammonia evaporator or condenser [3]. In this configuration, one bed operates with low-temperature salt (LTS), extracting heat from cold ambient at low vapor pressure. The other bed contains a high-temperature salt (HTS), where adsorption occurs, and useful heat is released at a higher temperature. The LTS is selected from salts whose equilibrium curves are towards the left in the Clapeyron diagram ($\log P$ vs $-1/T$), i.e., near the ammonia vapor line. The HTS must absorb ammonia at low pressure but release heat at high temperatures. As a result, their equilibrium lines are found on the right side of the chart.

Many researchers have investigated CSHP systems based on the single-effect system with an NH_3 evaporator/condenser and single-effect resorption cycles, and reported a low coefficient of performance (COP) ranging between 0.21-0.35 [4–6]. These two single-effect cycles can incorporate a heat recovery mechanism, allowing heat transfer between two opposite half-cycle adsorbers, to preheat the reactor before desorption, leading to significantly improved COPs ($2 - 4\times$) [7–9]. Babu and Kumar [10] proposed a compressor-operated resorption thermochemical energy storage system designed for heat storage, along with heat upgradation. The system functions as a resorption system during energy storage and utilizes a compressor during energy recovery. They analyzed the system at compression ratios ranging from 5 to 10 and reported high COP values of 4.1 and 4.8 when using sodium bromide and potassium iodide as the LTS, respectively. Meibodi et al. [11] studied the dynamic thermal behavior of a seasonal solar thermal energy storage system utilizing a compressor-assisted thermochemical sorption process, which led to a reduction in the desorption pressure from 8 bar to 4 bar and temperature from 87°C to 67°C in a strontium chloride system.

This paper presents a proof-of-concept experiment and its results, utilizing magnesium chloride (MgCl_2) mixed with expanded natural graphite. The experiments demonstrated that a high-density heat storage reactor is practically feasible and can supply heat at the required temperature. The composite-scale energy density is estimated to be ~ 1000 kJ/kg. Then, a thermodynamic model of a compressor-assisted resorption heat pump using four reactors is presented. The objective is to provide continuous heating and cooling for various applications, utilizing heat from low-temperature sources of 15°C to 45°C . The heat pump performance is analyzed, and key performance indicators, such as COP, compression ratio (r_p), specific heating power (SHP), and specific cooling power (SCP), are presented for different salt pairs. Finally, a comparison of electric COPs with those of a standard vapor compression heat pump is presented to assess the feasibility of the proposed chemisorption heat pump.

2. Proof of concept experiment

To develop a prototype chemisorption heat pump, a proof-of-concept salt- NH_3 reactor using MgCl_2 and graphite was fabricated to store and release heat, thereby evaluating the feasibility of the system under repeated sorption cycles. The experimental setup, shown in Fig. 1(a), features a reactor connected to an ammonia tank, allowing NH_3 to flow during both the adsorption and desorption processes. The reactor is connected to a thermic oil loop through an electric heater and a heat exchanger. During adsorption, the heater remains off, and a known mass of water collects the heat released from the reactor in the sump via the heat exchanger. During desorption, the water loop is disconnected, and the heater is turned on to provide the necessary heat of desorption. The oil temperature at the inlet and outlet of the reactor is measured continuously. During adsorption, the temperature of the water sump is measured and used to estimate the total heat released and the effective energy density of the composite. Thirteen experiments of adsorption and desorption cycles were performed, with the energy density shown in Fig. 1(b). Notably, salt does not exhibit any degradation after all the cycles and maintains an average energy density of ~ 1000 kJ/kg. These proof-of-concept experiments demonstrate the promising potential of chemisorption reactions for heat pumping and energy storage applications. Based on the results, we propose and analyse an advanced chemisorption heat pump system. The configuration is designed to provide continuous heating and cooling using heat from a low-temperature heat source, with the assistance of an ammonia compressor to augment the pressures in the different reactors.

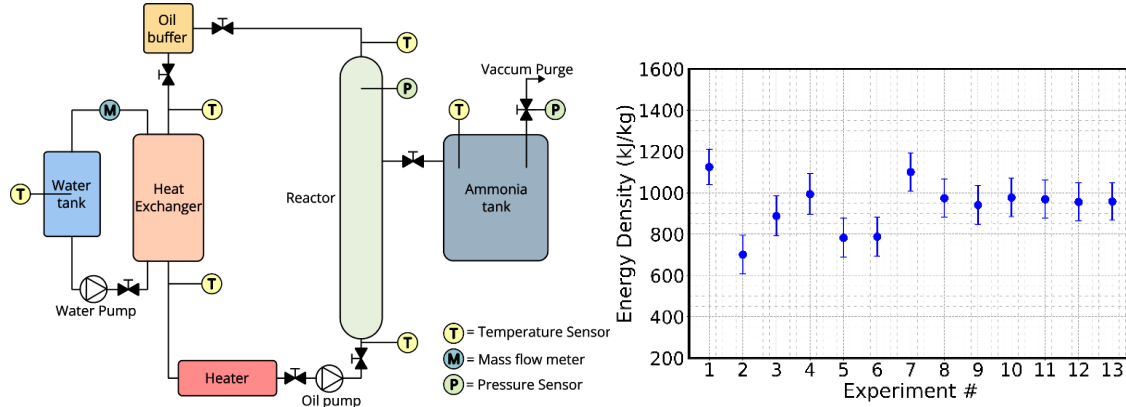


Figure 1: (a) Schematic of the experimental facility for the proof-of-concept chemisorption reactor, and (b) the estimated energy density of the salt (MgCl_2) and graphite coating during different experiments.

3. Thermodynamic Model

3.1. Systems design and description of the model

The proposed compressor-assisted resorption heat pump consists of four reactors and a compressor, as shown in Fig. 2(a). The reactors contain two metal halide salts that act as the LTS and the HTS for the sorption process. The system is designed to operate continuously with desorption on one side of the compressor and adsorption on the other. In Fig. 2(a), the left side HTS and LTS are undergoing desorption, and the right side LTS and HTS are in adsorption mode. Heat transfer fluid (HTF) enters the HTS on the desorption side, where it transfers heat for desorption, and is then routed to the LTS for desorption at a lower temperature. Similarly, on the adsorption side, HTF first enters the LTS to remove its heat of adsorption before moving into the HTS, where the adsorption temperature is higher; the HTF then exits at the desired heating temperature. The refrigerant moves from the desorption side to the adsorption side through the compressor, enabling the HTS and LTS reactors to achieve the appropriate pressures and temperatures.

3.2. Modeling assumptions

To model the system, we have used the following assumptions: a) the system is at a quasi-steady state, b) adsorption and desorption processes in the reactors occur at a constant temperature, c) NH_3 is in thermal equilibrium with salt during adsorption and desorption processes, d) specific heat, density, and the enthalpy of reaction of the salt are constant, e) heats of reaction for adsorption and desorption are assumed to be equal, and f) negligible energy loss from the fluid and the reactors to the surroundings.

3.3. Thermodynamic cycle

The state of ammonia vapor at various phases of the cycle is illustrated in the schematic shown in Fig. 2(a). The thermodynamic cycle is represented by different states of ammonia on the Clausius-Clapeyron diagram

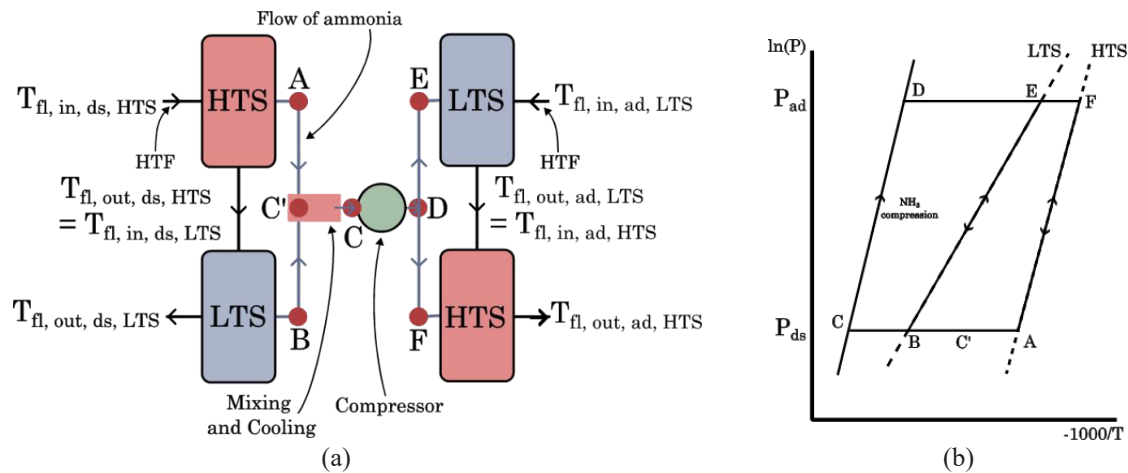


Figure 2: (a) Schematic of compressor-assisted resorption heat pump, showing the flow of ammonia and the heat transfer fluid (HTF). (b) Representation of the thermodynamic cycle on a Clausius-Clapeyron diagram

(Fig. 2(b)). The equilibrium temperature of the salts can also be inferred from Fig. 2(b), specifically at points A, B, E, and F. Since the system operates in a cycle, each salt bed alternates between adsorption and desorption. They remain at their equilibrium temperature during a process and are either sensibly cooled or heated to the equilibrium temperature required for the following process.

3.4. Governing equations

The thermodynamic model is governed by the Clausius-Clapeyron equation (Eq. 1). The salt releases or absorbs heat, which is transferred to or from the heat transfer fluid flowing through the reactor. The reactor is a heat exchanger that must be optimized for both the heat transfer rate and total energy requirements. The modeling methodology of the reactors is described in the following section.

3.4.1. Reactor model

During operation, both the LTS and HTS initiate the desorption process upon completion of the last adsorption phase. The reactors must achieve the equilibrium desorption temperature for the corresponding low-pressure. Therefore, sensible cooling of the reactor from the adsorption temperature is required, which is:

$$Q_{s, \text{salt}, \text{ds}} = n_{\text{salt}} \cdot c_{\text{salt}} \cdot (T_{\text{ds}} - T_{\text{ad}}) \quad (2)$$

Along with the salt, ammonia exists in the reactor and is also bonded to the salt. Its enthalpy is calculated as a function of the temperature and pressure of the ammonia in the reactor. This ammonia is also sensibly cooled to the desorption temperature and pressure. The sensible cooling is modeled as:

$$Q_{s, \text{NH}_3, \text{ds}} = m_{\text{NH}_3} \cdot [h_{\text{NH}_3}(T_{\text{ds}}, P_{\text{ds}}) - h_{\text{NH}_3}(T_{\text{ad}}, P_{\text{ad}})] \quad (3)$$

Finally, for the desorption to proceed, the heat of reaction is given by:

$$Q_{\text{ds}} = n_{\text{NH}_3} \cdot \Delta h_{\text{salt}} \quad (4)$$

Δh_{salt} is the enthalpy of the reaction associated with per unit mol of ammonia change (adsorbed/desorbed), same as in Eq. 1. Finally, the total heat that needs to be supplied to the LTS or HTS for completion of desorption is the summation of the absolute values of Eq. 2-4.

$$Q_{\text{total}, \text{ds}} = |Q_{s, \text{salt}, \text{ds}}| + |Q_{s, \text{NH}_3, \text{ds}}| + |Q_{\text{ds}}| \quad (5)$$

The adsorption process in the LTS and HTS is modeled identically, as described above.

$$Q_{s, \text{salt}, \text{ad}} = n_{\text{salt}} \cdot c_{\text{salt}} \cdot (T_{\text{ad}} - T_{\text{ds}}) \quad (6)$$

$$Q_{s, \text{NH}_3, \text{ad}} = m_{\text{NH}_3} \cdot [h_{\text{NH}_3}(T_{\text{ad}}, P_{\text{ad}}) - h_{\text{NH}_3}(T_{\text{c,out}}, P_{\text{ad}})] \quad (7)$$

$$Q_{\text{ad}} = n_{\text{NH}_3} \cdot \Delta h_{\text{salt}} \quad (8)$$

$$Q_{\text{total}, \text{ad}} = |Q_{\text{ad}}| + |Q_{s, \text{salt}, \text{ad}}| + |Q_{s, \text{NH}_3, \text{ad}}| \quad (9)$$

3.4.2. Modeling of the compressor

The ammonia desorbed from both the HTS and the LTS during the desorption process is mixed and cooled before it is introduced into the compressor. Since the pressure of both ammonia streams is the same, the temperature can be calculated assuming adiabatic mixing of desorbed ammonia from the LTS and HTS. If the temperature of ammonia after mixing is too high, it is cooled to a temperature approaching the ambient before being introduced into the compressor. The compression process is modeled as an isentropic process with a constant efficiency of 75%. The work input to the compressor is given as:

$$W_{\text{comp}} = m_{\text{NH}_3, \text{total}} \cdot (h_{\text{out}} - h_{\text{in}}) \quad (10)$$

3.4.3. Coefficient of performance (COP)

The work input of the compressor is converted to its equivalent electrical input, using the source-to-site electrical energy factor of 3.4 [12]. The equivalent electrical input is then defined as (where $P_{\text{ec}} = 3.4$):

$$Q_e = P_{\text{ec}} \cdot W_{\text{comp}} \quad (11)$$

The COP_{heat} is the ratio of the total heat output to the total heat supplied during a complete cycle.

$$\text{COP}_{\text{heat}} = \frac{Q_{\text{total}, \text{HTS}, \text{ad}} + Q_{\text{total}, \text{LTS}, \text{ad}}}{Q_{\text{total}, \text{HTS}, \text{ds}} + Q_{\text{total}, \text{LTS}, \text{ds}} + Q_e} \quad (12)$$

For cooling COP, the total input desorption heat is divided by the total heat supplied.

$$\text{COP}_{\text{cool}} = \frac{Q_{\text{total}, \text{HTS}, \text{ds}} + Q_{\text{total}, \text{LTS}, \text{ds}}}{Q_{\text{total}, \text{HTS}, \text{ds}} + Q_{\text{total}, \text{LTS}, \text{ds}} + Q_e} \quad (13)$$

3.5. Model validation

The system model is developed using Engineering Equation Solver (EES) [13] and is validated against the published experimental results of Xu et al. [6]. They presented a resorption system from simultaneous heating and cooling using MnCl_2 and NH_4Cl as the HTS and the LTS, respectively. They estimated the COP at various operating conditions, and the maximum deviation of the present model with respect to their data is $\pm 3.13\%$. The model is also validated against the theoretical COP estimated for a NaBr and MnCl_2 system studied by

Atkinson et al. [14]. Their model predicted an ideal COP of 1.64 and a COP considering the thermal mass of the system as 1.35. The current model predicts these two COPs as 1.636 and 1.35, respectively. Therefore, the proposed modeling method is suitable for analyzing these systems.

4. Results and discussion

Various salt pairs were analyzed as HTS and LTS, considering specified inlet and outlet temperatures of the HTF for heating and cooling applications in buildings. In this analysis, HTF is a 50% concentrated ethylene glycol solution mixed with water. The closest approach temperature (*CAT*) for all heat exchangers is assumed to be 5°C. The inlet temperature of the HTF is considered to be from a low-temperature heat source. The exit temperatures for adsorption and desorption are selected to provide realistic and usable cooling and heating in buildings. The HTF input temperature of 45°C from a heat source is sufficient for desorption of the HTS at a lower equilibrium pressure. If this fluid can be cooled to a temperature range of 10–15°C (the temperature at the outlet of the LTS), it can be used for space cooling. An input HTF temperature of 15–30°C can be heated to 50–60°C (the temperature at the outlet of the HTS) during adsorption for heating purposes.

The following salt pairs were evaluated for the proposed heat pump: BaCl₂ and NH₄Cl, CaCl₂ and BaCl₂, and ZnCl₂ and SrCl₂, as the HTS and LTS, respectively. The required compression ratios, along with the COP for heating and cooling for these salt combinations, are shown in Fig. 3. While the COP for heating and cooling across all three pairs are almost comparable, the compression ratios differ significantly, which affects the electrical power requirement and overall feasibility of operation. The lowest compression ratio is found for the ZnCl₂ and SrCl₂ pair, when the inlet temperature for adsorption is 15°C and the outlet temperature is set to 50°C. The inlet temperature on the desorption side is 45°C, with an outlet temperature of 15°C (shown in Fig. 3(g)). Due to this low compression ratio, less work is required from the compressor, which is reflected in the higher COPs for both heating (Fig. 3(i)) and cooling (Fig. 3(h)) compared to the scenarios where the outlet desorption temperatures are 10°C and 12°C. The COP for heating is the highest with ZnCl₂ and SrCl₂, reaching

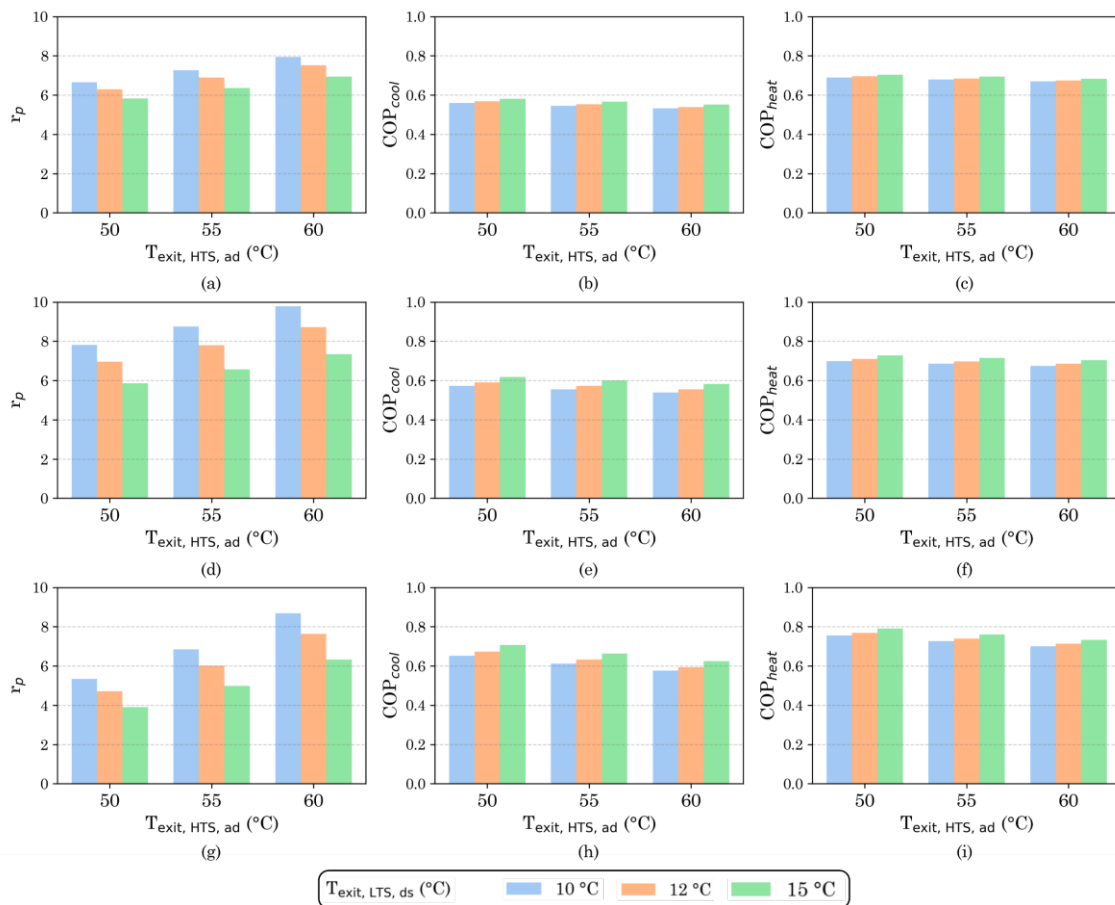


Figure 3: Parametric study for different salts as HTS and LTS, with different HTF outlet conditions. The compression ratio and the COP are compared. (a)–(c) is for BaCl₂ and NH₄Cl as the HTS and LTS. (d)–(f) is for CaCl₂ and BaCl₂ as HTS and LTS. (g)–(i) is for ZnCl₂ and SrCl₂ as the HTS and LTS. For (a)–(f), the inlet temperature for the desorption is constant at 45°C, and the inlet temperature for adsorption is maintained at 30°C. For (g)–(i), the inlet temperature for the desorption is maintained at 45°C, and the inlet temperature for adsorption is maintained at 15°C.

a value of 0.79 when the desorption inlet and outlet temperatures are set at 45°C and 15°C, respectively, and the adsorption inlet and outlet temperatures are set at 15°C and 50°C, respectively. However, the specific power for the $\text{ZnCl}_2/\text{SrCl}_2$ pair is considerably lower than that of the other two pairs. This is due to the combined effect of the molar masses of the salts and the number of moles of ammonia that a single mole of salt can react with. For instance, ZnCl_2 has a high molar mass and can only react with two moles of ammonia before reaching saturation. However, SrCl_2 and BaCl_2 have comparable molar masses and can react with 7 and 8 moles of ammonia, respectively. Moreover, NH_4Cl has a much lower molar mass. As a result, the salt pair of ZnCl_2 and SrCl_2 exhibits lower specific cooling and heating power than the BaCl_2 and NH_4Cl pair. Therefore, the careful assessment of salt pairs is crucial to determine the feasibility of the proposed chemisorption heat pump.

4.1. Performance comparison with a standard vapor compression system

To assess the potential performance enhancement offered by the proposed chemisorption heat pump, the COPs are compared with those of a standard vapor compression system (VCS), utilizing a Carnot factor of 50% [15]. The salt pair of ZnCl_2 and SrCl_2 is selected for this assessment, and ambient temperatures representative of both hot and cold days are used to evaluate the COP. The hot-day ambient temperature is typical of a humid subtropical climate during the summer (Austin, USA), while the cold-day ambient temperature is characteristic of a humid continental climate during the winter (Chicago, USA). For the VCS, the supply temperatures are set as 60°C for heating and 10°C for cooling. These temperatures are used to calculate the Carnot COP, employing an appropriate closest approach temperature (5°C) for both the evaporator and the condenser. These temperatures are then multiplied by the Carnot factor to obtain the COP of actual systems operating under the ambient conditions of the representative days. To compare the electricity usage, the electric COPs of the proposed system are defined as:

$$\text{COP}_{\text{heat}, e} = \frac{Q_{\text{total, HTS, ad}} + Q_{\text{total, LTS, ad}}}{W_{\text{comp}}} \quad (14)$$

$$\text{COP}_{\text{cool}, e} = \frac{Q_{\text{total, HTS, ds}} + Q_{\text{total, LTS, ds}}}{W_{\text{comp}}} \quad (15)$$

The predicted COP values for a full-day cycle, along with the COPs of the VCS and ambient temperature, are shown in Fig. 4. As seen in Fig. 4(a), on a hot summer day in Austin, USA, the proposed heat pump performs well during the higher-temperature hours when cooling is needed. As the ambient temperature rises, the difference between the ambient and the constant supply temperature needed also increases, leading to a decrease in the COP of the VCS. However, the COP of the CSHP remains constant throughout the day with negligible variation, which is attributed to the performance of the intercooler heat exchanger before the compressor. A constant and high COP of the chemisorption system with a decoupling from ambient conditions makes it well-suited for such cooling applications. Assuming a constant cooling capacity of 5 kW, which is representative of the normalized design cooling load of a small office building, the electricity consumption of the proposed heat pump is estimated to be 18.17 kWh/day. For the same conditions, a standard VCS requires 21.14 kWh/day of electricity.

Similarly, on a cold day representative of Chicago, USA, the electric COP of the proposed chemisorption heat pump is significantly higher than that of the VCS heat pump. The low COP of the VCS (Fig. 4(b)) is

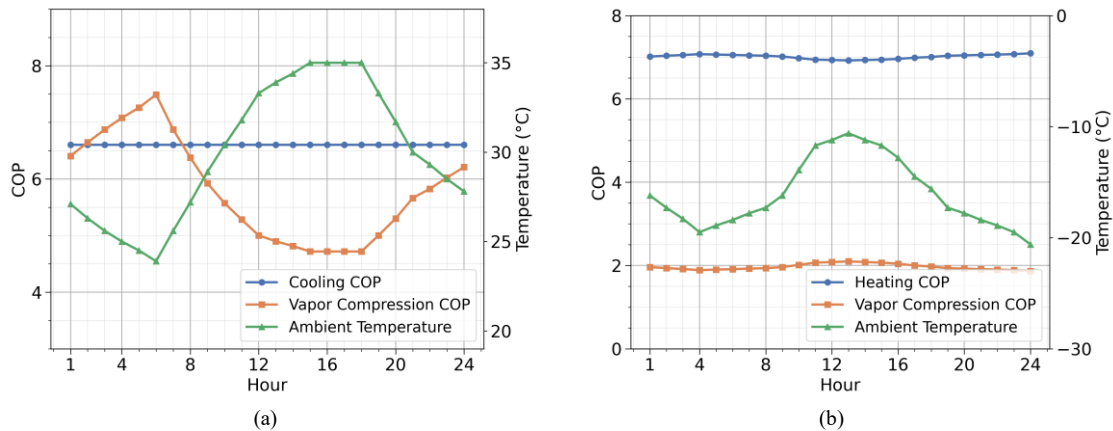


Figure 4: (a) Comparison of the electric cooling COP of the CSHP, with a standard vacuum compression system for a representative hot day. The COP of the CSHP remains constant, performing better during the time of the day when the temperatures are relatively higher. (b) Comparison of the electric heating COP of the CSHP with a standard heat pump system for a representative cold day. The CSHP performs more efficiently throughout the entire daily cycle.

primarily due to its close coupling with ambient conditions, which makes heat pumping an energy-intensive process, especially at very low ambient temperatures. For the same constant and normalized design heat load of 5 kW, the VCS consumes 61.09 kWh/day, whereas the proposed heat pump requires only 17.11 kWh/day of electricity. The proposed CSHP demonstrates enhanced effectiveness for heating in cold climates, outperforming the standard VCS heat pump. Therefore, the proposed chemisorption heat pump can provide both cooling and heating in a cost-effective manner, making it a viable option for wider adoption in buildings.

5. Conclusion

The proposed thermodynamic model of a chemisorption heat pump (CSHP) can be used to evaluate the efficiency and viability of various salt pairs for heating and cooling applications at different temperatures. This study presents the coefficients of performance (COPs) and the specific heating and cooling powers for three salt pairs, taking into account the heat input from the low-grade source in performance calculations. The proposed system configuration operates without requiring a condenser or evaporator, thereby eliminating the need to store liquid or gaseous ammonia in pressure vessels, which improves overall system safety. An energy usage analysis is also presented, comparing the COPs of the proposed CSHP and its energy requirements with those of an existing vapor compression system. Proof-of-concept experiments were conducted to evaluate the viability of salt-ammonia chemisorption reactions for thermal energy storage and heat pumping. With an effective energy density of ~ 1000 kJ/kg obtained for MgCl_2 , the results are promising, especially since the salt shows no performance degradation after multiple adsorption and desorption cycles. Further analysis of additional salt pairs is necessary, and the configuration must be optimized to minimize the compression ratio in the heat pump. Additionally, there is a requirement to enhance the effective energy density of the reactor in the experimental study, which is crucial for making these types of systems compact and commercially viable for HVAC systems integrated with energy storage. This can help facilitate the broader adoption of renewable power and improve the resilience of buildings.

Nomenclature

<i>amb</i>	Ambient	<i>fl</i>	Fluid	<i>R</i>	Universal Gas Constant
<i>ad</i>	Adsorption	<i>h</i>	Specific Enthalpy	<i>ref</i>	Reference
<i>c</i>	Specific Heat Capacity	HTF	Heat Transfer Fluid	<i>s</i>	Sensible (Heating/Cooling)
<i>CAT</i>	Closest Approach Temperature	HTS	High-Temperature Salt	SCP	Specific Cooling Power
<i>comp</i>	Compressor	LTS	Low-Temperature Salt	SHP	Specific Heating Power
COP	Coefficient of Performance	LMTD	Log Mean Temp. Difference	<i>T</i>	Temperature
CSHP	Chemisorption Heat Pumps	<i>m</i>	Mass	<i>W</i>	Compressor Work
<i>ds</i>	Desorption	<i>n</i>	Moles	Δh	Enthalpy of reaction per unit mole of ammonia
<i>e</i>	Electric	<i>P</i>	Pressure	Δs	Change in entropy
<i>eq</i>	Equilibrium	<i>Q</i>	Change in Energy		

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