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Thermodynamic Superiority of Calcium Hydroxide-Based Thermochemical Energy Storage over Ammonia-Based Systems in Concentrated Solar Power

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Abstract - Thermochemical energy storage technology is a promising step in the development of renewable energy and the transition to sustainable energy systems. Thermochemical energy storage systems store thermal energy through reversible chemical reactions and can provide a stable and continuous energy source with appropriate efficiency. This study investigates and compares the performance of two promising thermochemical energy storage systems when integrated with a concentrated solar thermal power plant. Calcium hydroxide- and ammonia-based thermochemical energy storage systems, due to their suitable temperature range and energy density, are coupled with a 20 MW concentrated solar plant. Results show that, in terms of roundtrip efficiency, the calcium hydroxide-based system performs better, achieving 93.6 % thermal-to-thermal efficiency with the capability of generating 2.28 MW of electrical power, while the ammonia-based system achieves 84 % thermal-to-thermal efficiency with a capability of 2.04 MW of electrical power. From a second-law thermodynamics perspective, during the charging phase, the endothermic reactor in the ammonia-based system demonstrates better performance, with approximately 10 % higher exergy efficiency compared to the calcium hydroxide-based system. During the discharging phase, reactors in both systems show similar performance. For the proposed integrated configurations, a heliostat field is designed with an area of 0.055 km^2 and an efficiency of 72.44%.

Keywords**:** Thermochemical energy storage; Calcium hydroxide; calcium oxide; ammonia; solar energy.

1. Introduction

Due to the significant increase in global energy demand, energy supply plants are expanding, and it is essential to develop renewable-based plants to meet this demand, contribute to sustainable energy systems, and reduce greenhouse gas emissions. Renewable energy sources such as solar energy, wind, geothermal, and others can be utilized as energy sources to transition from fossil fuels to renewable energy plants and decarbonized systems [1]. Solar energy is one of the most promising renewable energy sources, which can be used as an energy source for power generation plants or to meet heat demand for other applications. The availability of 173,000 TW of solar energy on Earth, with just one-ten-thousandth of that covering the total energy demand of the world, presents a great potential to maximize the use of this energy source and reduce dependence on fossil fuels and conventional energy sources [2]. Concentrated solar thermal power (CSTP), a promising technology suitable for high-temperature applications, can absorb solar radiation at high temperatures and make it available for various applications or produce indirect electrical power by integrating with power generation cycles. This technology, due to its ability to generate significant amounts of thermal energy at high temperatures for power generation, has been one of the fastest-growing solar-based technologies in recent years [3]. The availability of solar radiation only during the daytime is one of the limitations of solar-based energy systems, as the absorbed heat can only be used for part of the day. To overcome this challenge, integrating these systems with thermal energy storage enables continuous operation by storing the absorbed heat during the day and releasing it steadily throughout the night.

Among thermal energy storage systems, thermochemical energy storage (TCES) is the most promising due to its wide temperature operation range, making it suitable for integration with various heat sources like solar energy. In a TCES system, as long as the reactants remain separate, it is loss-free and leads to high efficiency. Additionally, there is an option for heat upgrade, allowing heat to be released at higher temperatures during the discharging phase compared to the charging phase. This makes TCES superior to other thermal energy storage systems, such as phase change materials (PCM) and sensible heat storage systems. TCES systems operate based on reversible chemical reactions. During the charging phase, heat is absorbed from a thermal source, and an endothermic reaction takes place, with the products stored separately. When heat is needed in the system, the products can be transferred to another reactor where the reverse reaction occurs, discharging the stored energy through the system. Despite most of the research on TCES systems being at the laboratory scale, several studies have explored different TCES systems and integrated them into power generation units or other heat-demanding applications. Depending on the heat source temperature, heat demand, and application the type of system can be different [4].

TCES systems based on dehydration/hydration of calcium hydroxide/calcium oxide ($Ca(OH)_2/CaO$) operate around 500 °C at atmospheric pressure and are suitable for integration with CSTP systems. Storing and releasing heat in this system occur based on the following reversible reaction:

$$Ca(OH)_{2_{(s)}} \leftrightarrow CaO_{(s)} + H_2O_{(g)} \tag{1}$$

The forward reaction, called dehydration, is endothermic and requires a heat source, while the reverse reaction, called hydration, is exothermic and releases heat. Abundant and inexpensive material, along with high energy density and enthalpy of reaction, makes these TCES systems promising TCES systems. Reviewing the literature indicates that the $Ca(OH)_2$-based (CaH) TCES system is a good candidate to use as a TCES system for high-temperature ranges. For instance, a techno-economic study conducted by Bayon et al. [5] investigated different types of solid-gas TCES systems for 100 MW power, and CaH, with a thermal efficiency of 98.5 %, outperformed other technologies. Additionally, the capital cost for the CaH system was calculated to be about 4.78 $/MJ, which is two times less than that of the molten salt energy storage system. Criado et al. [6] proposed a single-stage fluidized bed reactor that operates in a CaH-TCES system. A thermal-to-thermal efficiency of 63 % was achieved for a 100 MW heat capacity hydration reactor. Carro et al. [7] proposed a large-scale CaH system for storing thermal energy through a TCES system. They investigated different conversion ranges in reactors and integrated the CaH system with power blocks. Results showed that a round-trip efficiency of more than 80 % and a thermal-to-electrical efficiency of 40 % are achievable through the proposed system. Angerer et al. [8] set up a large-scale CaH-TCES reactor. Results of the study indicated that a reactor with a 100 m^3 volume can work as a 15 MW thermal reactor. Similar experimental studies can also be found in the literature, which tried to prove the application of CaH-TCES systems in real applications [9], [10], [11].

Ammonia (NH_3)-based TCES systems, due to their temperature range, can be integrated with CSTP. Also, these types of systems, due to their simple cycle and the low cost of ammonia, represent a promising technology for integration with solar power to store solar heat through thermochemical reactions. This system operates based on the dissociation and synthesis of NH_3 a reversible gas-gas reaction:

$$NH_{3_{(g)}} \leftrightarrow \frac{1}{2}N_{2_{(g)}} + \frac{3}{2}H_{2_{(g)}} \tag{2}$$

The forward reaction is endothermic and requires heat to break down NH_3 into H_2 and N_2, while the reverse reaction releases heat during NH_3 synthesis. TCES systems based on NH_3 dissociation/synthesis (NH-TCES) are a mature technology widely used in various processes. Chen et al. [12] proposed an NH-TCES system to produce superheated steam. Results showed that the proposed model produced 350 °C to 650 °C hot steam at 26 MPa, which fits with Rankine cycle temperature levels and suits heat recovery from the synthesis reactor. They also conducted another study to produce supercritical carbon dioxide and steam at 800 °C [13]. Lovegrove et al. [14] developed an experimental closed-loop NH-TCES system, which uses a cavity receiver to supply the required heat for NH_3 decomposition. In addition to the experimental setup, theoretical investigations have been conducted to maximize the system's performance when coupled with power blocks in electrical power generation. Results of the study showed an energy storage efficiency of around 53 %. These are only a few examples of the feasibility of NH-TCES systems in the literature. All studies indicate that NH-TCES systems can be a promising technology to store and release heat through reversible reactions and can be candidates for integration with high-temperature heat sources, specifically CSTP.

This study aims to compare the performance of CaH-TCES and NH-TCES systems when integrated with the same 20 MW heliostat field for storing and releasing heat. Both systems are integrated with a CSTP system, and the overall system performance is compared from a thermodynamic perspective. The advantages and disadvantages of each are analysed based on differences in their reaction types.

2. Methodology and systems description

2.1. Heliostat field

A heliostat field provides the required heat for both TCES systems in the endothermic reaction, and the solar energy can be stored in these systems and then released when needed. Typically, in heliostat solar power systems, mirrors in the field concentrate the solar radiation to a central tower to utilize the solar heat. In this study, this heat supplies the heat demand for endothermic reactions. The heliostat field area A_{helio} [m^2] can be determined using the following equation [15]:

$$A_{helio} = \dot{Q}_{inc}\ DNI \tag{3}$$

where $\dot{Q}_{inc}$[W] is the incident heat flow rate in the heliostat field, and DNI [W/m^2] abbreviates direct normal irradiance.

The energy balance for the heliostat field can be written as follows [15]:

$$\dot{Q}_{net} = \dot{Q}_{inc} - \dot{Q}_{loss} \tag{4}$$

where $\dot{Q}_{net}$[W] represents the net heat flow rate absorbed by the material to be dehydrated or decomposed in the reactors, and $\dot{Q}_{loss}$ [W] is the heat flow rate lost in the field, respectively. Heat losses in the heliostat field include the reflected heat flow rate from the heliostats ($\dot{Q}_{ref}$[w]) and the heat lost when the non-reflected heat, before being absorbed by the material inside the reactor, is lost during transfer to the material. These losses, including convection ($\dot{Q}_{conv}$[w]) and radiation ($\dot{Q}_{rad}$[w]), can be formulated as:

$$\dot{Q}_{loss} = \dot{Q}_{ref} - \dot{Q}_{rad} - \dot{Q}_{conv} \tag{5}$$

The heliostat field is designed based on the model presented in Ref. [15] and Ref. [16], and the methodology used to specify heat losses in this process can be found in the mentioned studies. Table 1 summarizes the main assumptions in the solar field design:

Table 1: Main assumptions for solar field simulation

Parameter	Value
Sun temperature	4500 °C
Average DNI	500 W/m^2
Field reflection efficiency	0.95
Operation time per day	8 hours

2.2. Calcium hydroxide-based thermochemical energy storage system

Figure 1 shows the schematic of the CaH-TCES system integrated with CSTP. The dehydration reactor absorbs solar heat, causing $Ca(OH)_2$ to dehydrate into CaO and H_2O. The dehydrated material should be stored in a solid tank (green tank) and continuously transferred under steady-state conditions to the hydration reactor, where the reverse reaction occurs to release heat. After the hydration reaction, the hydrated material should be stored in the storage tank (yellow tank) and transferred back to the dehydration reactor only during the daytime when solar energy is available, ensuring a closed TCES loop. These two solid tanks play a vital role in overcoming the limitation of heat source availability, which is restricted to approximately eight hours per day.

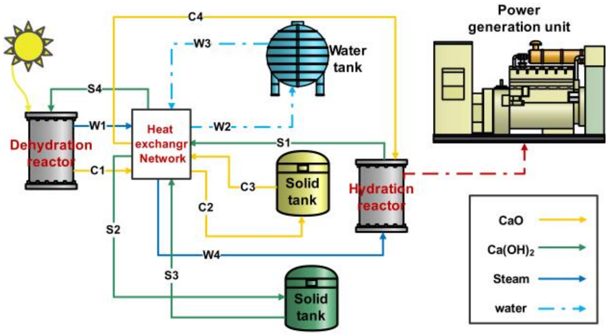

Fig. 1: Schematic of the CaH-TCES system integrated with a solar field.

As shown in Figure 1, a heat exchanger network is integrated into the Ca-TCES system. Since the dehydrated and hydrated materials leave both reactors at high temperatures but must be stored at ambient temperature, they need to be cooled down. Conversely, before entering the reactors, these materials require heating. In the dehydration reactor, heating reduces thermal power output, whereas in the hydration reactor, it increases heat release, facilitates the reaction, and maintains overall system efficiency. To optimize energy use, the heat exchanger network utilizes the heat from materials that need cooling to preheat the materials entering the reactors. This ensures that all preheating and cooling processes are internally managed within the Ca-TCES system.

Table 2: CaH-TCES system operation conditions

Parameter	Value
Dehydration reactor thermal power input	20 MW
Reactors operation pressure	1 bar
Dehydration reaction temperature	569.2 °C
Hydration reaction temperature	469.2 °C
Conversion rate in the reactors	100 %
Ambient temperature	25 °C
Dehydration reactor operating hours	8
Hydration reactor operating hours	24

2.3. Ammonia-based thermochemical energy storage system

NH-TCES systems, as another high-temperature TCES system, were chosen to be integrated with the CSTP system to store solar thermal energy. As shown in Figure 2, the products of this reaction leave the reactor at high temperature and pass through the heat exchanger to heat and evaporate the liquid NH_3 coming from the storage tank. After cooling down, the products are stored at ambient temperature. This charging phase is active only 8 hours daily when the sun shines. For the discharging phase, the stored N_2/H_2 is transferred to the NH_3 synthesis reactor and, before entering the reactor, passes through the exchanger to be preheated and cool down the synthesized NH_3. This reaction is exothermic and releases heat, which can be used in the power block to generate electrical power in steady-state conditions. Table 3 indicates the operation conditions of the NH-TCES system.

Table 3: NH-TCES system operation conditions

Parameter	Value
Dissociation reactor thermal power input	20 MW
Reactors operation pressure	30 bars
Dissociation reactor temperature	510 °C
Synthesis reactor temperature	469.2 °C
Dissociation reactor operating hours	8
Synthesis reactor operating hours	24

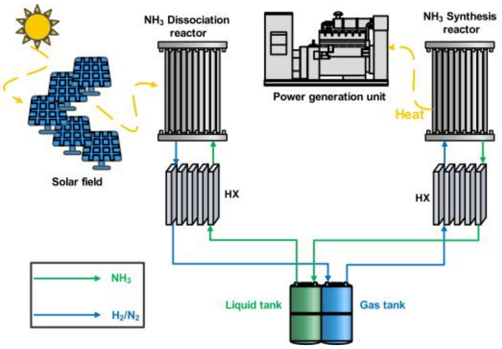

Fig. 2: Schematic of the NH-TCES system integrated with a solar field.

2.4. Performance evaluation

To evaluate the thermodynamic analysis of the proposed systems, the following mass and energy balance equations are used:

$$\sum\nolimits_{in} \dot{m} = \sum\nolimits_{out} \dot{m} \tag{6}$$

$$\sum\nolimits_{out} \dot{m}h - \sum\nolimits_{in} \dot{m}h = \dot{Q} - \dot{W} \tag{7}$$

where $\dot{m}$ [kg/s] represents the mass flow rate and h [J/kg] stands for the energy rate of each stream while $\dot{Q}$ [W], and $\dot{W}$ [W] represent the heat and work rates of the control volume, respectively. Also, the enthalpy change of the reaction can be calculated using the following equation, which specifies the released and absorbed heat in the discharging and charging phases.

$$\dot{Q} = \sum \dot{H}_{Products} - \sum \dot{H}_{reactants} \tag{8}$$

where $\dot{Q}$ is the heat of the reaction [W], $\dot{H}$ is enthalpy rate for products and reactants [W], and the summation refers to all reactants and products.

To evaluate the efficiency of each system, performance indicators have been defined to indicate the performance of each system and compare them. Using equations 6 and 7, the energy balance can be applied to all components shown in Figures 1 and 2 to determine the enthalpy and mass flow rate of each stream at the inlet and outlet of the components. Once the characteristics of each stream are calculated using these equations, the heat released through the reactors can be determined from equation 8. Equations 9 and 10 show the roundtrip efficiency of the integrated TCES system with the solar field in terms of thermal-to-thermal η_{th-th} [%] and thermal-to-electric conversion η_{th-el} [%], respectively. Thermal-to-thermal efficiency specifies how much of the absorbed heat from the CSTP system is converted to thermal power in the discharging phase, while thermal-to-electric efficiency indicates how much of the absorbed heat converts to electrical power if this integrated system is coupled with a power block.

$$\eta_{th-th} = \frac{\dot{Q}_{HY}}{\dot{Q}_{CSTP}} \tag{9}$$

$$\eta_{th-el} = \frac{\dot{W}_{el}}{\dot{Q}_{CSTP}} \tag{10}$$

To evaluate the reactors' efficiency based on the second law of thermodynamics, the exergy balance for reactors in both systems is conducted using the following equation:

$$\sum_{in} \dot{Ex} = \sum_{Out} \dot{Ex} + \dot{I} \tag{11}$$

where $\dot{Ex}_{in}$ and $\dot{Ex}_{out}$ are the input and output exergy flow rates, respectively, including both physical and chemical exergy, $\dot{I}$ represents the exergy destruction rate, And the summation refers to all input and output streams of the reactors. Exergy efficiency for each reactor can be defined as:

$$\eta_{ex} = 1 - \left(\frac{\dot{I}}{\dot{Ex}_{in}}\right) \tag{12}$$

4. Results and discussion

4.1. Heliostat field

The designed heliostat field supplies 20 MW of net thermal power to the endothermic reactors, and to provide this amount of heat, a 0.055 km^2 area is surrounded by heliostat mirrors. About 27.56 % of the incident heat in the solar field is lost, while the material absorbs the rest to decompose or dehydrate in the reactors. Table 4 summarizes the heliostat field performance.

Table 4: Solar field performance

Parameter	Value
CSTP system efficiency	72.44 %
Field area	0.055 km^2
Incident heat flow rate	27.6 MW

4.2. Thermochemical energy storage systems performance

As both CaH-TCES and NH-TCES systems are coupled with the same heat source, the performance of each system in converting the absorbed heat from the endothermic reaction to the released heat in the discharging phase during the exothermic reaction, as well as the exergy destruction in the corresponding reactions, reveals each TCES system's advantages and disadvantages when integrated with high-temperature sources, specifically the CSTP system.

The CaH-TCES system, with its appropriate internal heat exchanger network design in the proposed configuration of this study, enables the recovery of all available heat streams to preheat the reactants before they enter the reactors and increase the heat output of the hydration reactor. From the solar field, 20 MW of net heat power is input to the CaH-TCES system, and this heat is available for only 8 hours. The designed system stores this heat and releases 6.24 MW of heat under steady-state conditions. In other words, 160 MWh of thermal energy per day is converted to 149.76 MWh of thermal energy, which can be used in the power block under steady-state conditions. This means a 93.6 % thermal-to-thermal round-trip efficiency for the integrated TCES system. On the other hand, in the NH-TCES system, the same amount of input heat as in the CaH-TCES system results in the release of 5.6 MW of heat through the NH_3 synthesis reactor. This means that 160 MWh of thermal energy per day is converted into 134.4 MWh, achieving an 84 % round-trip thermal-to-thermal efficiency.

Considering the capability of integrating a power block with the mentioned TCES systems and comparing their performance in electrical power generation, both exothermic reactors operate at the same temperature and release heat at the same temperature. A Rankine cycle that matches the temperature of these TCES systems can be coupled. Such a power block was proposed in previous work by the authors of this study, where its electrical efficiency was reported as 36.58 % [17]. Assuming integration with the proposed TCES systems in this study, it can be determined that 2.28 MW of electrical power can be generated from the CaH-TCES system and 2.04 MW from the NH-TCES system, respectively.

Table 5 shows the performance of each proposed system from an energy analysis perspective. As shown in Table 5, the CaH-TCES system has better performance in terms of roundtrip efficiency, and a greater amount of absorbed heat can be released under the same conditions. This leads to higher electrical power generation. This can be justified by the higher enthalpy of reaction in the CaH-TCES system compared to the NH system, resulting in greater heat production. Apart from the energy efficiency perspective, the CaH-TCES system is a solid-based TCES system, and as seen in its configuration, it appears more complex than the NH system, with the design of the heat exchanger network playing a vital role in thermal efficiency. Additionally, system control and material transfer can be more challenging compared to the NH-TCES system.

Table 5: Energy analysis results for proposed TCES systems

CaH-TCES system	
Parameter	Value
Heat input through the dehydration reactor	160 MWh (daily basis)
Heat released through the hydration reactor	149.76 MWh (daily basis)
Electrical energy generation in the coupled Rankine	54.72 MWh (daily basis)
Thermal-to-thermal efficiency	93.6 %
Thermal-to-electricity efficiency	34.2 %
NH-TCES system	
Parameter	Value
Heat input through the dehydration reactor	160 MWh (daily basis)
Heat released through the hydration reactor	134.4 MWh (daily basis)
Electrical energy generation in the coupled Rankine	48.96 MWh (daily basis)
Thermal-to-thermal efficiency	84 %
Thermal-to-electricity efficiency	30.6 %

Figure 3 shows the exergy efficiency of each reactor in proposed TCES systems. It clearly demonstrates how much of the input exergy is destroyed through the chemical reactions and indicates the performance of each system from the second law of thermodynamics point of view. The exergy efficiency of each reactor represents only the specific reactor under the design conditions of this system and specifies the exergy destruction ratio in the corresponding reactions.

Endothermic reactions in both systems play a role in the charging phase. As shown in Figure 3, the CaH-TCES system experiences greater exergy destruction in the charging reaction compared to the NH-TCES system. The dehydration of $Ca(OH)_2$ is a solid-gas reaction, where a solid reactant decomposes into solid and gas products. Since the reactant and products exist in different phases, this reaction has a higher exergy destruction ratio compared to a gas-gas reaction. For the discharging phase, where exothermic reactions occur, the NH system performs better from a second-law thermodynamic perspective. However, this difference is not as significant as in the endothermic reactions.

Comparing the energy and exergy results for the proposed TCES systems, the CaH-TCES system, despite its better performance in round-trip efficiency and energy conversion rate, has a higher exergy destruction ratio in the reactors, particularly during the charging phase, due to the reaction characteristics, especially the dehydration reaction. The second law of thermodynamics could therefore be one of the improvement objectives for these types of systems.

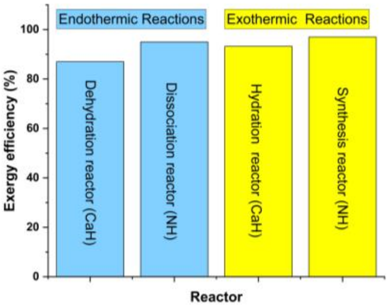

Fig. 3: Exergy efficiency of reactors in proposed TCES systems

5. Conclusions

This study investigated two different types of TCES systems integrated with a 20 MW CSTP system and compared their performance in storing solar thermal energy through reversible chemical reactions and their capability in releasing stored heat. Since thermal energy storage in CSTP systems operates at high temperatures, two of the most promising high-temperature TCES systems were selected for integration. The CaH-TCES system relies on the dehydration and hydration of $Ca(OH)_2/CaO$, a solid-gas reaction, while the NH-TCES system operates based on NH_3 dissociation and synthesis, a gas-gas reaction. Results showed that, from a thermal conversion perspective, the CaH-TCES system performs better than the NH-TCES system. It can release 93.6 % of the absorbed heat, whereas the NH-TCES system releases 84 %. Both systems demonstrate promising thermal-to-thermal efficiency, highlighting the role of thermochemical reactions in thermal energy storage. Integrating these systems with a power block can enhance their potential for electrical power generation. When coupled with a Rankine cycle, the CaH-TCES system generates 2.28 MW of electrical power, while the NH-TCES system generates 2.04 MW. Thermal analysis further indicates the superior performance of the CaH-TCES system, attributed to its reaction characteristics and the nature of the dehydration/hydration process. From an exergy efficiency perspective, results reveal that both reactors in the NH-TCES system achieve higher efficiency compared to the CaH-TCES system. This difference is particularly significant in the endothermic process and can be explained by the solid-gas reaction characteristics and the specific conditions required for the decomposition of solid $Ca(OH)_2$ into solid and gas products. From a thermodynamic standpoint, the following advantages and disadvantages can be highlighted for the CaH-TCES and NH-TCES systems.

- CaH-TCES system showed better performance in terms of roundtrip efficiency compared to NH-TCES system.
- CaH-TCES system has a more complex design compared to the NH-TCES system.
- The endothermic reactor in the CaH-TCES system has a higher exergy destruction ratio compared to the NH-TCES system.
- The exothermic reactors in both systems have similar performance from a second-law thermodynamics standpoint.

Despite the proposed systems' performance from a thermodynamic point of view, highlighting each one's advantages and disadvantages, it should be noted that several factors affect the efficiency of such systems. These factors include variations in the temperature and pressure of the reactor, instability in weather data and solar radiation, and reaction kinetics. Additionally, practical considerations, such as system maintenance, cyclic stability of the material, and control of the system's operation, can impact performance. When it comes to real applications, these factors should be considered as significant influences.

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