



Evaluation and performances comparison of calcium, strontium and barium carbonates during calcination/carbonation reactions for solar thermochemical energy storage



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ABSTRACT

The efficiency and economic competitiveness of thermal storage for concentrating solar power plant can be improved by increasing the operating temperature (above 600 °C). Thermochemical energy storage is an attractive way of efficiently storing high-temperature solar heat, in the form of chemical bonds as a stable and safe solid material, when compared with existing sensible and latent heat storage materials. Among the most interesting materials, BaCO_3 , CaCO_3 and SrCO_3 show high storage temperatures (typically above 800 °C), energy storage densities, and charging and discharging rates. Heat charge corresponds to the calcination (decarbonation) reaction of the carbonates (endothermal step) and heat discharge corresponds to the reverse carbonation of the oxides (exothermal step). A comparative thermodynamic and kinetic study of calcination and carbonation reactions involving commercial and synthesized CaCO_3 , SrCO_3 and BaCO_3 powders was performed for application in thermochemical energy storage. An experimental study based on thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) was conducted to study the decomposition and carbonation reactions and to determine the enthalpy of reaction for each metal carbonate. While complete calcination was achieved regardless of the metal carbonate involved, partial carbonation was observed with loss in CO_2 capture capacity during cycling. The effect of the addition of a promoting agent such as magnesium oxide on thermal stability for improving chemical and structural cyclability of these three candidate carbonates was also investigated. Beneficial effect of MgO addition was demonstrated and noticeable performance stability was obtained in the case of SrCO_3/SrO during successive energy storage cycles.

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1. Introduction

Solar energy is a renewable resource that can be used for the production of electricity, but only during hours with available sunlight. Solar thermal plants use concentrated solar power (CSP) during on-sun hours to produce electricity and they require energy storage in order to keep their production during night or cloudy days, by releasing the previously stored energy. The efficiency and economic competitiveness of thermal storage for CSP can be increased by operating the plant at higher temperatures (>600 °C). However, existing latent and sensible heat storage materials are not stable at these temperatures and energy storage may be a challenge in these next-generation plants.

To this end, thermochemical energy storage (TCES) is proposed to efficiently store solar energy by converting high-temperature heat

into chemical bonds via reversible solid-gas reactions. In the form of chemical bonds within a stable solid material, the energy can be more easily stored, for a long period of time at room temperature, and safely transported. High-temperature TCES in CSP plants is a promising alternative to store solar energy, in order to bridge the gap between solar radiation availability and electricity demand. TCES promises low cost through the use of inexpensive storage materials, making it highly competitive when compared with commercial molten salt storage systems. Reversible reactions are of interest for TCES because their main advantages include high energy densities, benign reactants, and thermal stability in contrast to molten salts. A broad range of candidate materials are being surveyed in order to expand the field of application of thermal energy storage and to help it become more efficient [1,2]. Selected materials for TCES have to meet a number of requirements in order to be suitable for TCES in CSP plants, such as complete reaction reversibility, high phase transition temperature, high energy storage density, fast enough reaction

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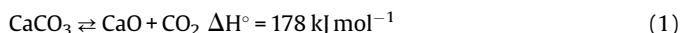
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kinetics for the charge and discharge step, low cost, non-toxic and environmentally-friendly, and no undesirable side reactions [3–5].

Among the possible materials, alkaline-earth metal carbonates are suitable candidates for TCES application. Calcination-carbonation chemical looping is a cyclic process that can be used to attain the desired storage temperature (typically above 800 °C), energy density and charging and discharging rates. Metal oxide produced in the solar-driven endothermic calcination reaction can be cycled to the exothermic carbonation reaction, from which thermal energy is released to drive a power cycle (Fig. 1).

The energy that can be stored and released from carbonates through reversible calcination/carbonation reactions makes metal carbonates potentially attractive for TCES applications [1,6]. The decomposition of several alkaline earth metal carbonates, especially CaCO_3 , SrCO_3 , BaCO_3 and MgCO_3 , has been investigated in order to better understand its mechanisms [7–11], but the reverse carbonation after calcination at high temperature has attracted less attention. Carbonation to form carbonates needs to be complete since the cycling stability of reversible calcination/carbonation reactions is desired to serve applications such as CO_2 capture and TCES. In previous works, carbonates were mainly studied for CO_2 capture applications with an interest in reducing greenhouse gases concentration in the atmosphere. Feng et al. (2007) [12] proposed a screening of CO_2 absorbing materials for CO_2 capture in zero emission power generation systems. Duan et al. (2012) [6] presented a screening scheme for identifying solid sorbents for CO_2 capture, with focus on alkali and alkaline-earth oxides and hydroxides. The availability and low cost of carbonates also makes them attractive in view of future scaled up application. In contrast, carbonates suffer from sintering over multiple calcination/carbonation cycles at high temperature [13–15]. To address this issue, studies have been conducted on the effect of materials morphology, composition and porosity on the reactivity.

The CaCO_3/CaO system received special attention (Eq. 1), focusing mainly on the improvement of performance upon cycling at high temperature [16].



Many studies focused on the improvement of CaO -based sorbent properties and durability for CO_2 capture, and the obtained results could help in the development of CaO -based materials for TCES, since it also relies on the CO_2 sorption capacity when considering carbonates. CaCO_3 is a promising candidate because it features high energy storage density [1,6], absence of side reactions, non-toxicity and availability at low cost. However, it is known for its loss-in-capacity over several calcination/carbonation cycles mainly due to sintering of CaO grains at high temperature and to a decrease of the reactive surface area with the number of cycles [17–20]. Several techniques have been developed to improve the durability of Ca -based sorbents and minimize their loss in adsorption capacity. In order to increase the active surface area and the stability of the pore structure, the use of rigid porous materials as supports for Ca -based sorbents, the use of additives to improve the sorbent thermal stability, the reduction of the sorbent particle size, and the synthesis of novel sorbents with microporous structure were proposed. Besides enhancing thermal stability, mechanical stability and particle attrition were also identified as other issues affecting the sorbent performance [20]. While performing calcination/carbonation cycles with limestone type material, Grasa et al. (2006) [21] observed the degradation of the material with accelerated loss-in-capacity when using longer calcination time. High calcination temperature (950 °C) and CO_2 concentration (up to 20%) caused a decline in the cycling stability. Crystallographic transformations taking place during the calcination of limestone under CO_2 was described [22,23]. The effect of thermal pretreatment and nanosilica addition to hinder CaO grain sintering was unveiled, and the loss in capacity was assigned to pore plugging [24]. By testing CO_2 sorption capacity and cycling stability of three types of dolomites, Li et al. (2005) [25] showed that CaCO_3 mixed with other compounds presents better stability over calcination/carbonation cycles than pure CaCO_3 . The combination of CaCO_3 and additives is a method with rising interest to counter the loss-in-capacity of carbonates.

The introduction of steam in the CaO/CaCO_3 system was also mentioned to help sustaining material reactivity during cycles [26–28]. The use of calcium hydroxide intermediate in a three-step

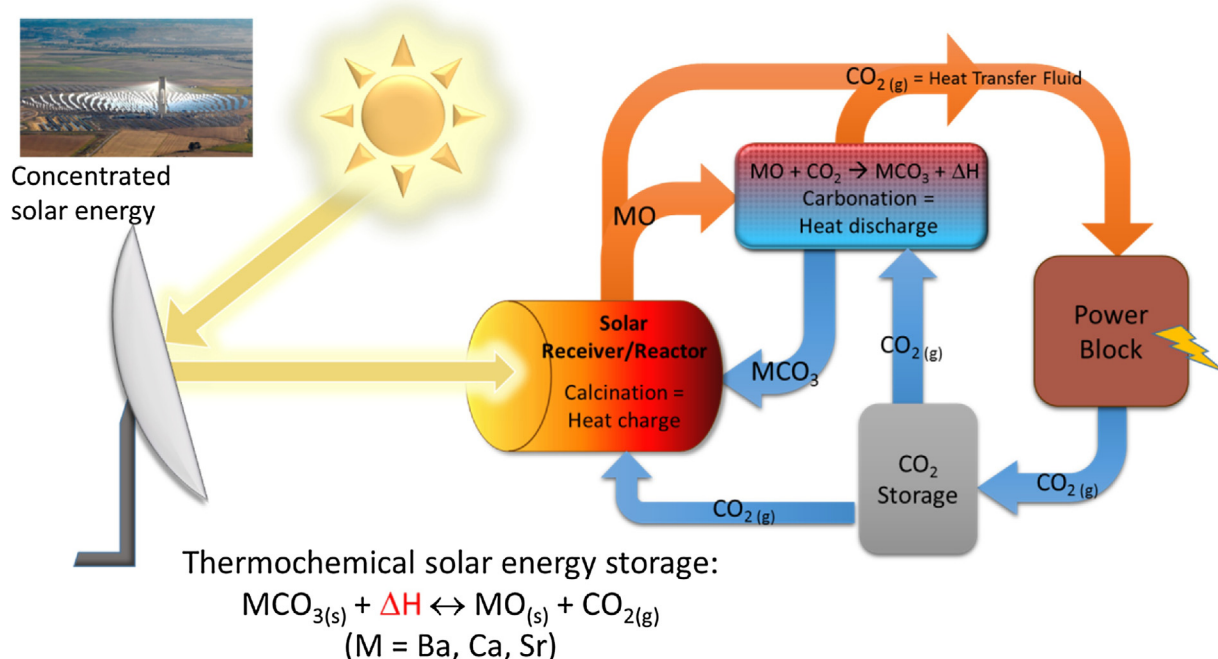


Fig. 1. Operating principle of thermochemical energy storage based on carbonates in CSP plants.

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