



Calcium oxide based materials for thermochemical heat storage in concentrated solar power plants

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Abstract

The present study relates to the preparation of mixed calcium oxide–alumina compositions as candidate materials for a cyclic thermochemical hydration–dehydration scheme at moderate to high temperatures (e.g. 400–600 °C) that can offer the possibility of short and long term energy storage, particularly suitable for concentrated solar power installations. The synthesized materials were assessed in terms of their cyclic hydration–dehydration performance in the temperature range of 200–550 °C. Acknowledging the fact that the particular thermochemical scheme has been identified to result in substantial cycle-to-cycle fragmentation of pure CaO particles which is detrimental to particle reactor bed concepts, one of the main purposes of using Al as additive is related to materials structural enhancement. To this respect, preliminary studies related to macro-structural integrity assessment were also conducted. In addition, the performance of synthesized material is compared to the one of natural lime (benchmark material). The additive content spanned over a wide range of Ca/Al molar ratios, namely from 95/5 to 52/48, while two different calcium oxide precursors, i.e. calcium nitrate and calcium acetate, were employed. Fresh and hydrated compositions were characterized in-detail with respect to their physicochemical properties in order to correlate different behaviors with certain attributes of the materials. Synthetic materials, both calcium nitrate and calcium acetate derived, favored the formation of Ca/Al mixed phases. The latter led to materials with higher surface areas and, for a given Ca/Al ratio, resulted to higher hydration/dehydration performance. Mixed oxides, although capable of being hydrated at ambient temperature, did not participate in the reaction scheme at temperatures ≥ 200 °C and thus presence of such phases resulted in considerable decrease of hydration/dehydration capacity versus the one of natural lime. On the other hand, the presence of such mixed compositions improved, albeit not dramatically, macro-structural integrity. A relatively good combination of hydration–dehydration performance with better-than-natural lime structural integrity was achieved for the mixed materials with a Ca/Al molar ratio equal to 89/11 and 81/19 molar ratio. The Ca-precursor used in these materials slightly affected their cyclic performance with the ex-CaN ones presenting better behavior. © 2015 Elsevier Ltd. All rights reserved.

Keywords: Thermochemical energy storage; Calcium oxide; Aluminum; Composite

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1. Introduction

Concentrated solar thermal power (CSP) technology has gained substantial worldwide interest from both industrial

and research communities, since it can provide a free, endless and fully renewable energy source that can be utilized to cover energy needs of various processes with potentially high efficiencies, mainly due to relatively high temperatures achievable. The employment of CSP technology has been gradually induced by the continuously increasing demand in energy, the depletion of fossil fuels and the serious environmental problems caused by the increase of CO₂ concentration in the atmosphere (Gil et al., 2010; Kuravi et al., 2013).

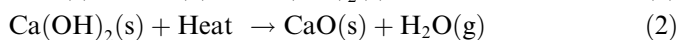
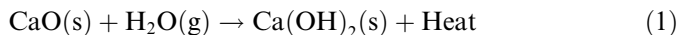
However, the major challenge that CSP technology has to deal with is the intermittence of solar radiation. The development of efficient, simple and high energy density systems, with the ability of storing the excess of solar power during on-sun operation, would allow prolonged or even continuous operation. These are known as Thermal Energy Storage (TES) systems. Recently, they have been employed commercially in CSP plants and their effectiveness relies on their ability to balance the discontinuous energy supply with the continuous energy demand.

The state-of-the-art concepts for TES systems currently utilized in the industrial world are based on the molten salts technology and the use of phase change materials (PCM), that dominate in sensible and latent heat systems (Gil et al., 2010; Mahlia et al., 2014). Although the technology of these systems is based on simple principles and is commercially available, such systems are characterized by low to moderate energy densities (0.2–0.5 GJ/m³) and significant heat losses resulting in large volume of materials required (Abedin and Rosen, 2011; Felderhoff et al., 2013).

The challenges of the above mentioned state-of-the-art TES systems can be in-principle overcome by an alternative approach based on Thermochemical Heat Storage (THS) concepts. THS is a valid option for TES technology that offers high energy storage density (0.5–3 GJ/m³), low heat losses, easy transfer and the potential for long storage periods (Abedin and Rosen, 2011; Pardo et al., 2014; Solé et al., 2012). Heat storage in THS systems is achieved via reversible chemical reactions, taking place in 2 or more steps. In particular, the surplus energy is stored during on-sun operation through one or more endothermic reactions and transferred to the CSP plant in order to be released through the corresponding exothermic reaction (s) under off-sun conditions (Pagkoura et al., 2014). THS concepts are applied by different reaction categories, such as carbonation–decarbonation of CaO/CaCO₃ energy couple (Abanades et al., 2005; Anthony, 2011; Edwards and Materic, 2012; Grasa and Abanades, 2006; Muller et al., 2011), oxidation via redox materials (Carrillo et al., 2014; Karagiannakis et al., 2014), the decomposition/synthesis of ammonia (Lovegrove et al., 2004; Wentworth and Chen, 1976) and hydration–dehydration of CaO/Ca(OH)₂ (Criado et al., 2014a; Schaube et al., 2012) or MgO/Mg(OH)₂ couple (Ervin, 1977).

Recently, the CaO/Ca(OH)₂ couple has been assessed as a promising candidate for THS applications at moderate-to-high temperatures (400–550 °C). THS technology using

the CaO/Ca(OH)₂ couple is based on the reversible hydration–dehydration reaction presented in the following scheme:



During the CSP plant operation and under surplus of solar irradiation, the endothermic dehydration of Ca(OH)₂ (reaction 2) takes place. CaO is obtained and can be subsequently hydrated (reaction 1), so that previously stored energy is released thereby providing the CSP plant with the ability to operate under off-sun conditions. The energy released through the exothermic reaction can be as high as 104 kJ/mol (or 1.85 MJ/kg). The CaO/Ca(OH)₂ system has been studied both for heat pump applications (Cerkvenik et al., 2002; Ogura and Mujumdar, 2000) and energy storage.

Most of the experimental studies conducted so far relate to the investigation of the CaO/Ca(OH)₂ system at the mg-scale, based on commercial/natural materials. Regarding materials development, some relevant work with interesting preliminary findings has been conducted (Murthy et al., 1986; Yan and Zhao, 2014), however no particular effort has been devoted to the behavior of such materials under multi-cyclic hydration–dehydration conditions. The aforementioned reversible reaction was examined thermodynamically and kinetically by Schaube et al. for partial H₂O pressures up to 95.6 kPa and atmospheric total pressure. They reported full conversion with good stability for over 100 hydration–dehydration cycles (Schaube et al., 2012). The same scientific group also presented experimental data regarding TES with CaO/Ca(OH)₂ in a reactor where agglomeration of the material was noted after 25 cycles, also studying important parameters for the storage process in order to determine a kinetic model for both reaction steps (Schaube et al., 2013). Linder et al. conducted experimental studies with Ca(OH)₂ with the aid of a 10 kW unit and they also employed simulation to determine one possible operation mode for the material (Linder et al., 2014).

In very recent works and in the framework of the EU-funded StoRRe Project, Criado et al. (2014a,b) investigated and proposed a fluidized bed reactor suitable for the hydration–dehydration of CaO/Ca(OH)₂ material couple in order to determine a kinetic model for the hydration and dehydration steps, under realistic conditions (temperatures and atmospheres).

Despite the notable advantages of the CaO/Ca(OH)₂ couple (Pardo et al., 2014), the sensitivity of the material to two different mechanisms, namely recrystallization and sintering, imposed by CaO treatment with water or steam induce challenges for its use in such systems (Azpiazuz et al., 2003). CaO hydration with either water or steam has been considered as a suitable solution to the deactivation of CaO particles through multiple carbonation–decarbonation. The proposed mechanism is fragmentation

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