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Multicycle activity of natural $CaCO_3$ minerals for thermochemical energy storage in Concentrated Solar Power plants

Monica Benitez-Guerrero [a,b], Jose Manuel Valverde [a,*], Pedro E. Sanchez-Jimenez [b], Antonio Perejon [b,c], Luis A. Perez-Maqueda [b]

[a] Facultad de Física, Universidad de Sevilla, Avenida Reina Mercedes s/n, 41012 Sevilla, Spain
[b] Instituto de Ciencia de Materiales de Sevilla, C.S.I.C.-Universidad de Sevilla, C. Américo Vespucio n°49, 41092 Sevilla, Spain
[c] Departamento de Química Inorgánica, Facultad de Química, Universidad de Sevilla, Sevilla 41071, Spain

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ABSTRACT

Thermochemical energy storage in Concentrated Solar Power plants by means of the Calcium-Looping process is a promising novel technology that would allow for a higher share of renewables. A main benefit of this technology is the use of widely available, non-toxic and environmentally friendly calcium carbonate minerals as raw materials to store energy. Efficient integration of the Calcium-Looping process into Concentrated Solar Power plants involves the endothermic calcination of $CaCO_3$ in the solar receiver while the exothermic carbonation of CaO is carried out at high temperature under high CO_2 partial pressure. The heat released by this reaction is carried out by the excess CO_2 and employed for power generation by means of a closed CO_2 cycle. This work explores the multicycle Calcium-Looping performance of naturally occurring $CaCO_3$ minerals such as limestone, chalk and marble for thermochemical energy storage in Concentrated Solar Power plants. Despite their similar composition (almost pure $CaCO_3$), these minerals exhibit a significant difference in their Calcium-Looping multicycle activity, which may be attributed to differences in particle size and microstructure. Pore plugging at the Calcium-Looping conditions for thermochemical energy storage tested in our work is a main limiting mechanism on the multicycle CaO carbonation activity.

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1. Introduction

Solar energy is one of the most promising sources of renewable energy as it is clean, abundant and cost-free. Moreover, Concentrated Solar Power (CSP) plants lend themselves to be integrated into the grid for large scale electricity generation (Hinkley et al., 2013; Siva Reddy et al., 2013). However, to overcome the inherently intermittent nature of direct solar irradiation it is mandatory to develop massive energy storage technologies for power generation during prolonged periods. To this end, a number of *Thermochemical Energy Storage* (TCES) systems are being investigated as a more efficient alternative to energy storage in the form of sensible heat using molten salts as currently employed in commercial systems (Zhang et al., 2013; Fernández et al., 2014). Among the main advantages of TCES systems we can find a potentially high energy density and the possibility of permanent storage without losses (N'Tsoukpoe et al., 2009; Pardo et al., 2014). The high temperatures attainable in CSP plants with tower technology make it possible to achieve highly endothermic chemical decomposition reactions. In this context, the *Calcium-Looping* (CaL) process, which is based on the calcination/carbonation of $CaCO_3$,

$$CaO_{(s)} + CO_{2(g)} \rightleftarrows CaCO_{3_{(s)}} \quad \Delta Hr^0 = -178\ \text{kJ/mol} \tag{1}$$

can be carried out at a fast rate in the temperature range typical of CSP plants with tower technology (above 700 °C). Scalability of the CaL process would be facilitated by its simplicity and the use of cheap (∼10 $/ton), abundant and non-toxic naturally occurring $CaCO_3$ minerals as raw materials (Prieto et al., 2016; Chacartegui et al., 2016).

The idea of integrating the CaL process into CSP plants for energy storage dates back to the 1970s (Barker, 1973, 1974; Flamant et al., 1980), and has gained renewed interest in recent years (Pardo et al., 2014; Chacartegui et al., 2016; Cot-Gores et al., 2012). Briefly, once $CaCO_3$ particles are calcined by concentrated solar irradiation, the products of the reaction (CaO and CO_2) are stored separately. When needed, these byproducts are brought together at carbonation conditions to use the heat from this exothermic reaction to produce electricity. The flow diagram

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* Corresponding author.
E-mail address: jmillan@us.es (J.M. Valverde).

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of a possible CaL-CSP integration, as recently proposed in (Chacartegui et al., 2016), is shown in Fig. 1. The system comprises a heliostat field, a solar calciner, a carbonator, two reservoirs for CaO and $CaCO_3$ storage, a CO_2 compression-storage and a power unit. After calcination, the sensible heat of the reaction byproducts is recovered by heat exchangers before storing them separately. On demand, CaO and CO_2 are recirculated into the carbonator where the heat of the carbonation reaction is recovered and transported to a gas turbine by the excess of CO_2 not intervening in the carbonation reaction. Thus, electricity is generated by means of a CO_2 closed cycle. CaL conditions leading to high global efficiencies in the CaL-CSP integration involve carbonation temperatures above 850 °C under high CO_2 partial pressure (Chacartegui et al., 2016). In addition, fast calcination at moderate temperatures would be desirable to use inexpensive metal based solar receivers (Ávila-Marín, 2011; Behar et al., 2013). As seen in previous works, the use of helium in the calciner atmosphere would allow to achieve fast $CaCO_3$ calcination at temperatures as low as 700 °C (Sarrion et al., 2016) due to the high thermal conductivity of He and the extremely high diffusivity of the released CO_2 in this gas (Berger, 1927; Cussler, 1997). Moreover, the CO_2/He mixture could be separated by using commercial membranes (Taketomo and Fujiura, 1984) for reutilizing both gases in the cycle.

The CaL process has been widely investigated in the last years for its application to capture the CO_2 released by the combustion of fossil fuels (Stanmore and Gilot, 2005; Blamey et al., 2010; Cormos and Simon, 2015). The standard CaL process to capture CO_2, which has been successfully demonstrated at the pilot-scale level (Perejón et al., 2016), employs lime (CaO) in a bed reactor (carbonator) at temperatures close to 650 °C fluidized by the post-combustion flue gas at atmospheric pressure and loaded with a relatively low CO_2 concentration (15% vol). After carbonation of CaO particles, they are circulated into a second fluidized bed reactor (calciner), where CaO is regenerated at high calcination temperatures (930–950 °C) under a high CO_2 concentration environment. Thus, CO_2 at high concentration is recovered from the calciner to be compressed and stored, and the regenerated CaO particles are recirculated to the carbonator to be used in a new cycle. Under these harsh calcination conditions, CaO particles are severely sintered, which causes CaO conversion to drop dramatically in just a few cycles. Remarkably, these CaL-CO_2 capture conditions are diverse from those specific of CaL-CSP storage above described.

Particle size is a relevant physical property for industrial applications, such as the CaL process, based on circulating fluidized beds (CFBs), where open cyclone exchangers are employed to recover the fine particles elutriated in the gas stream (Chacartegui et al., 2016; Ebrahimi and Rahmani, 2016; Lu et al., 2008). According to Barker (Barker, 1974), the carbonation/calcination reaction would be only reversible in short residence times for particles of size around 10 nm while the optimum size of particles to be used in CFBs is in the rage 100–500 μm (Dieter et al., 2013; Arias et al., 2013; Ströhle et al., 2014; Hanak et al., 2015). For particles of size in this range, physical mechanisms such as intraparticle pore diffusion and pore plugging might limit significantly the reaction kinetics thus hampering the CaO multicycle activity as depending on reaction conditions (Grasa et al., 2008; Alvarez and Abanades, 2005).

In general, most of the studies focused on the effect of particle size on CaO conversion have been carried out under CO_2 capture conditions. According to Abanades and Alvarez (2003); carbonation at 650 °C under 20% $CO_{2;}$ calcination at 900 °C in air) the multicycle CaO conversion decay is similar for particles of size in the range between 100 and 800 μm for limestone samples obtained from different sources. On the other hand, the study carried out by Grasa et al. (2008); carbonation at 650 °C under P_{CO2} = 0.01 -

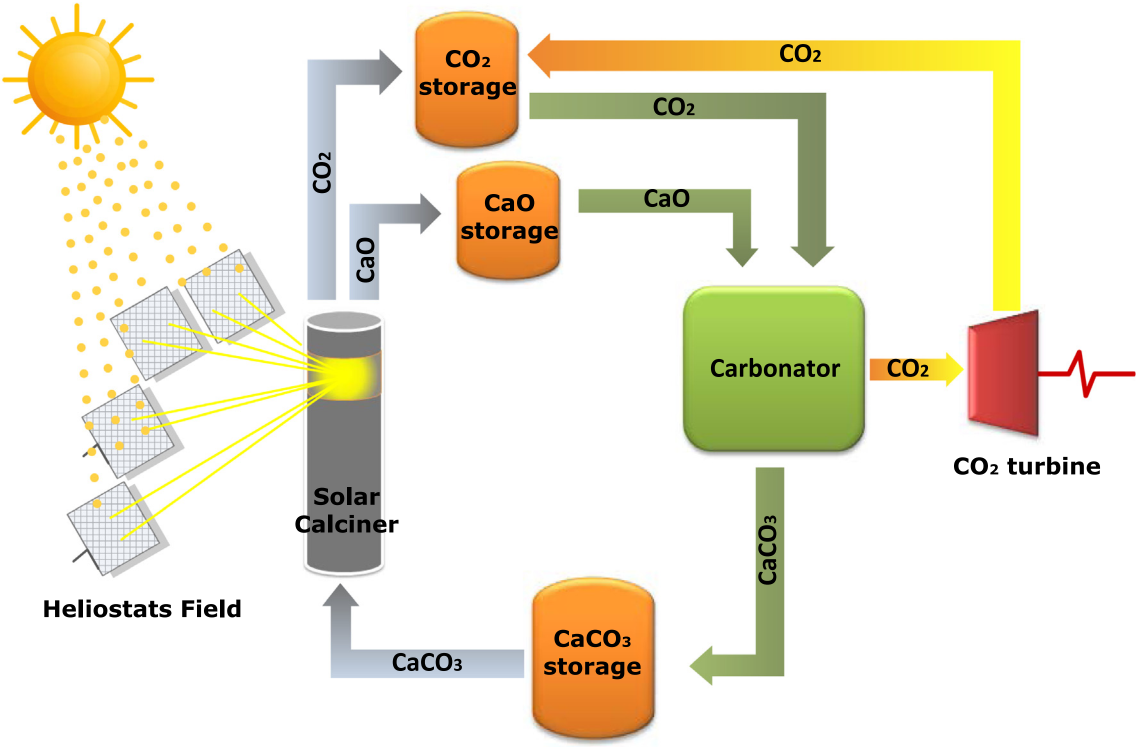

Fig. 1. Flow diagram of the Calcium-Looping thermochemical energy storage system for Concentrated Solar Power plants. A detailed description is found in (Chacartegui et al., 2016).

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