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## Solar-driven production of lime for ordinary Portland cement formulation

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#### ABSTRACT

Cement production is an energy-intensive manufacturing process with potentially large environmental burdens. Among the others, it is one of the largest industrial sources of CO<sub>2</sub> emission. Limestone calcination is the stage responsible for most of CO<sub>2</sub> emissions and energy requirement. This article aims at supporting the use of solar energy as non-carbogenic renewable source to sustain limestone calcination, with advantages on both the economic and environmental aspects of the process. A directly irradiated Fluidised Bed (FB) reactor was used as limestone precalciner for clinker production. Concentrated solar radiation was simulated with an array of three short-arc Xe-lamps of 4 kWel each, coupled with elliptical reflectors, capable of producing a peak flux of about  $3 \text{ MW m}^{-2}$  at the centre of the reactor. The total irradiated power is of approximately 3.2 kW<sub>th</sub>. Thermocouples and an IR camera were used for the analysis of the FB thermal profiles. Calcination was carried out at a nominal bulk bed temperature of 950 °C, in an atmosphere containing about 70% CO2. The reactivity of lime generated by the solar-driven calcination process has been characterised. Lime produced by the solar-driven process was used together with commercial clay as kiln feed components for the formulation of Portland cement samples. A binary mixture composed by fresh limestone and the same clay as above was employed as a reference. The key focus of the investigation was the assessment of the reactivity of the solar-generated lime toward the main clay components in the clinker production process, as compared to lime from ordinary calcination. An aspect that is specifically scrutinised is whether the different, and possibly more severe, thermal history to which limestone particles undergo during solar-driven calcination in directly irradiated FB reactors may compromise lime reactivity. Portland clinkers were produced by burning the raw meals at 1500 °C for 15 min. Clinkers were mixed with 5% natural gypsum to prepare the related Portland cements, which were then paste hydrated for times ranging from 2 to 28 days (water/cement mass ratio = 0.5, 20 °C, 95% relative humidity). Parameters as lime saturation factor, burnability, phase composition of clinkers and hydration behaviour of cement pastes were taken into consideration. Techniques as X-ray fluorescence and diffraction, and simultaneous differential thermal-thermogravimetry were used to study the materials.

#### 1. Introduction

Concrete is the second most consumed raw material on Earth after water, and is by far the most widely used building material worldwide (Yaphary et al., 2017). It is mainly employed for the construction of buildings and major civil infrastructures. On average, three tons of concrete are consumed by every person on the Planet each year. Concrete has achieved its predominance because of a number of decisive advantages, namely versatility in meeting a wide range of needs and relative inexpensiveness. Cement is the key component of concrete; it is termed "hydraulic binder", an inorganic glue which hardens upon water addition, binding together fine sand and coarse aggregates in concrete. Cement production is a highly energy-intensive manufacturing process (Mikulcic et al., 2016) due to the consumption of high quantities of fuels (mainly fossil fuel and pet coke). Moreover cement production accounts for 26% of the worldwide industrial  $CO_2$  emission, that is 6% of  $CO_2$  globally emitted in atmosphere (industrial and nonindustrial sources) (Telesca et al., 2016; Xu et al., 2015). This derives from a global production equal to about 4.5 Gt in 2015 (Shen et al., 2015; Telesca et al., 2017). Portland cement is a mixture of Portland

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Nomenclature		LSF	Lime Saturation Factor
		PCC	Portland Cement Clinker
BI	Burnability Index	R	reference feed mixture (limestone + clay)
CEM-R	cement obtained from Cli-R	S	solar feed mixture (solar lime + clay)
CEM-S	cement obtained from Cli-S	TES	Thermal Energy Storage
Cli-R	clinker obtained by burning R mixture	$u_{mf}^{t}$	theoretical value of minimum fluidisation velocity
Cli-S	clinker obtained by burning S mixture	X	mass % of the lime source in the feed mixture
CST	Concentrated Solar Technologies	XRD	X-Ray Diffraction
DT–TG	Differential Thermal–Thermo Gravimetric	XRF	X-Ray Fluorescence
FB	Fluidised Bed	Y	mass % of clay in the feed mixture

Cement Clinker (PCC) (Telschow et al., 2012) and a few percent of calcium sulphates (mostly gypsum) finely interground. PCC is obtained by heating an intimate mixture (raw meal) of limestone (about 80%) and clay. To obtain PCC with a calcium oxide content equal to about 67%, a raw meal/clinker mass ratio of 1.5-1.6 is usually employed in the production of Portland cement. The raw meal undergoes a two-step process, namely preheating-calcination and clinkerisation, respectively carried out in a stationary apparatus (up to 800-1000 °C) and in a rotary kiln (up to 1400–1500 °C). It has been estimated that about 0.87 kg of CO<sub>2</sub> are released per each kg of PCC produced (Barcelo et al., 2014). This amount cumulates contributions from limestone thermal decomposition and fuel combustion (Xu et al., 2015). Furthermore, if one assumes that CaO derives entirely from limestone calcination, about 0.53 kg of limestone-related CO<sub>2</sub> are released per kg of PCC produced, thus contributing by 60% to overall CO<sub>2</sub> emission. The theoretical heat requirement for PCC manufacture is equal to about 1.76 GJ per ton of clinker (Gartner, 2004) deriving from the difference: energy requirement of both preheater-calciner (3.06 GJ per ton of clinker) and rotary kiln (0.21 GJ per ton of clinker) minus heat recoverable from clinker cooling (1.51 GJ per ton of clinker). So, the thermal energy demand can be essentially ascribed to the thermal duty associated with limestone calcination (Lea's, 2017; Mehta, 1978). Therefore, in the context of PCC production, limestone calcination is responsible for the majority of both CO<sub>2</sub> emission and heat requirement, this last aspect likely involving the burning of fossil fuels with related environmental impact (both GHG and other pollutants). Effective measures to reduce this impact could result in huge improvements in terms of environmental impact and social acceptance (Dean et al., 2013; Telesca et al., 2014, 2015, 2016, 2017). Altogether, the global energy consumption, sum of the theoretical heat requirement and the total heat losses, lies in the range 3-6 GJ per ton of clinker (Barcelo et al., 2014).

The use of a non-carbogenic renewable energy source to sustain limestone calcination could provide substantial advantages on both the economic and environmental aspects of the process, as far as fuel consumption could be extensively reduced. Solar energy is one of the most important among the non-carbogenic renewable sources of energy. Extensive research is ongoing to exploit the huge amount of solar energy falling on Earth. High-temperature thermal energy is effectively provided by Concentrated Solar Technologies (CST): a field of optical sun-tracking mirrors is used to focus and concentrate solar energy onto a receiver, whence it is converted to electricity or industrial process heat.

CST can be easily integrated with Thermal Energy Storage (TES) or hybridised with backup fuels to overcome the intrinsic variability of solar energy (Pardo et al., 2014; Prieto et al., 2016; Yadav and Banerjee, 2016). The CST flexibility of producing both energy and/or chemicals, together with TES integration, is one of the major advantages of these technologies. Due to the high concentration level achieved in CST, the incident fluxes can reach values of several MW m<sup>-2</sup>. As a consequence, the solar receiver has a crucial role not only in collecting solar energy but also in minimising heat losses and local overheating. The use of directly irradiated Fluidised Bed (FB) as solar receivers/reactors has been widely investigated (Almendros-Ibáñez et al., 2018; Bachovchin et al., 1983; Bellan et al., 2018; Flamant, 1982; Flamant and Olalde, 1983; Gokon et al., 2011, 2015; Gomez-Garcia et al., 2017; Koenigsdorff and Kienzle, 1991; von Zedtwitz and Steinfeld, 2005; Tregambi et al., 2016). FB systems feature very favourable thermal properties (bed-to-surface heat transfer and thermal diffusivity) which can be finely tuned under unsteady and uneven fluidisation conditions (Salatino et al., 2016) and also provide an ideal environment for gas–solid chemical reactions. A critical issue in solar FB reactors can be represented by bed surface overheating induced by highly concentrated solar radiation, which can eventually affect the performance of the system due to sintering and/or degradation of bed solids.

Limestone calcination is a chemical reaction that may be conveniently carried out in solar FB reactors. The use of solar energy to sustain limestone calcination has been already investigated in the past (Badie et al., 1980; Flamant et al., 1980; Imhof, 1991, 1997, 2000a; Salman and Khraishi, 1988). Several research groups are also currently studying the integration between CST and the "calcium looping" cycle, a technology essentially based on cyclic limestone calcination and autothermal re-carbonation which can be exploited for thermochemical energy storage applications (Alovisio et al., 2017; Edwards and Materić, 2012; Ortiz et al., 2018) or post-combustion  $CO_2$  capture and sequestration/utilisation (Matthews and Lipiński, 2012; Reich et al., 2014; Tregambi et al., 2015, 2018; Zhai et al., 2016; Zhang and Liu, 2014).

The use of solar irradiated reactors as limestone precalciners upstream of a clinker production kiln was suggested by Imhof (1997, 2000a, 2000b). Meier and co-workers (2004, 2005a, 2006) investigated solar-driven limestone calcination using both a direct and indirect rotary kiln of 10 kWth. The reactivity of solar CaO was evaluated by the  $t_{60}$  parameter, namely the time required by a mixture of quicklime and water to reach the temperature of 60 °C under specified conditions. They successfully developed and tested a multitube rotary kiln for industrial solar production of lime (CaO), obtaining reactor efficiencies of about 30-35% without heat recovery. According to their economic assessment, the cost of a lime material produced in a solar 25  $MW_{th}$ plant would be roughly twice the price of that produced with conventional heating source (Meier et al., 2005b). The incorporation of solar energy in the cement production was also previously suggested by Imhof (2000b). Abanades and André (2018) developed a novel solar reactor concept for the continuous processing of reactive particles involved in high-temperature thermochemical reactions (500-1600 °C), demonstrating the feasibility of continuous solar calcination of limestone particles. The reactor could be, in principle, integrated with cement production process or used for CaO-based sorbent regeneration in a CO2 capture process or for thermochemical energy storage via calcium looping process. González and Flamant (2014) performed a technical and economic feasibility analysis on the use of CST in cement production. They concluded that there are no serious technical barriers to such an integration. From the economic standpoint, it is required that the overall thermal losses of the solar plant be lower than 45% to obtain an acceptable internal rate of return, and also that the coal price does not decrease beyond a threshold value.

The aim of the work was to investigate the possibility of using a limestone pre-calcined in a directly irradiated FB reactor for clinker

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