

Solar synthesis of calcium aluminates

D. Fernández-González^{a,*}, J. Prazuch^b, I. Ruiz-Bustinza^c, C. González-Gasca^d, J. Piñuela-Noval^a, L.F. Verdeja^a

^a Department of Materials Science and Metallurgical Engineering, School of Mines, Energy and Materials, University of Oviedo, Oviedo, Asturias, Spain

^b Department of Physical Chemistry and Modelling, Faculty of Materials Science and Ceramics, AGH University of Science and Technology, Krakow, Poland

^c Department of Primary Metallurgy and Recycling, National Centre for Metallurgical Research (CENIM-CSIC), Madrid, Spain

^d European University of Madrid-Laureate International Universities, Villaviciosa de Odón, Madrid, Spain

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ABSTRACT

The production of high alumina refractory cements ($> 75\% \text{Al}_2\text{O}_3$) is traditionally carried out in furnaces heated with both electric power (submerged arc furnaces) and fossil fuels (coke, natural gas). This heating system has several disadvantages: electricity price, contamination of the cement or greenhouse gases emissions. Solar energy, when properly concentrated, offers a great potential in high temperature applications, as those required in the manufacture of refractory and ceramic materials. In this paper we propose the synthesis of calcium aluminate compounds using concentrated solar energy. The results obtained prove the feasibility of the solar obtaining of the calcium aluminate compounds and could be useful for future development of this solar process in a larger scale.

1. Introduction

The production of cement is growing because of the increase in the world population but also due to the changes in construction methods in developing countries, and the improvement in infrastructures. In this way, the world production of Portland cement overcame the 4000 Mt in 2015 according to the USGS (United States Geological Survey). The production of cement has several problems: the first problem is that the production of cement is energy intensive (fuel and electricity consumption represent approximately a 40% of the total production costs (González and Flamant, 2014)); and the second problem is that the cement industry is environmentally non-friendly, being responsible for 5% of global anthropogenic CO_2 , of which 50% is derived from the chemical process and 40% from burning fuel (Meier et al., 2005a).

Solar energy, when properly concentrated offers a lot of possible applications in different fields of materials science and metallurgy (Fernández-González et al., 2018). In this way, for instance, concentrated solar energy has been applied in the recovery of valuable components from the ironmaking and steelmaking industry (Ruiz-Bustinza et al., 2013; Mochón et al., 2014), in the production of silicon (Murray et al., 2006), in the carbothermal reduction of ZnO to be used in water splitting to produce H_2 (Osinga et al., 2004), in the obtaining of aluminium foams (García-Cambronero et al., 2008; García-Cambronero et al., 2010), in the sintering of high speed steels (Herranz

et al., 2013; Herranz et al., 2014) or in surface treatment of steels (Llorente and Vázquez, 2009). In the field of non-metallic materials, concentrated solar energy has been applied since at least four centuries. In the 17th, Ehrenfried Walter Von Tschirnaus designed, constructed, and worked with lenses and mirrors with the purpose of concentrating solar energy, and, in this way, melting iron and obtaining ceramics (porcelain) (Gosh, 1991; Newcomb, 2009; McDonald and Hunt, 1982). Felix Trombe demonstrated the possible application of concentrated solar energy in the melting of high refractory ceramics (alumina, chromium oxide, zirconia, hafnia and thoria) after the Second World War (Flamant and Balat-Pichelin, 2010). More recent studies have widened the number of applications of concentrated solar energy in non-metallic materials. The manufacture of silicon carbide using concentrated solar energy was researched by several groups due to the large number of applications of this material (composite materials, combustible cells, electronics, etc.) (Cruz-Fernandes et al., 1998; Gulamova et al., 2009; Ceballos-Mendivil et al., 2015). Silicon nitride (Si_3N_4) with excellent properties (high fractures toughness, high flexural strength, and creep resistance even at high temperatures (Cardarelli, 2008)) was also synthesized in solar furnace (Zhilinska et al., 2003), the same as other materials with high wear resistance such as titanium carbide (Cruz-Fernandes et al., 1999; Cruz-Fernandes et al., 2002) and tungsten carbide (Guerra-Rosa et al., 2002). Other ceramic materials treated under concentrated solar energy were: alumina (Cruz-

* Corresponding author.

E-mail address: fernandezgdaniel@uniovi.es (D. Fernández-González).

Fernandes et al., 2000; Román et al., 2008) and cordierite (Costa-Oliveira et al., 2009).

The production of refractory and ceramic materials requires high temperatures (Verdeja et al., 2008; Verdeja et al., 2014; Pero-Sanz et al., 2017). Conventional technologies used in reaching such high temperatures require the utilization of electricity and/or the combustion of fossil fuels (Verdeja et al., 2008; Verdeja et al., 2014), causing both economic and environmental problems. Economically, the problems are mainly associated to the price of either the electricity or the fossil fuels, but also to the costs of releasing CO₂ (taxes associated to CO₂ emissions). Environmentally, the problems are linked to the emissions of CO₂ associated to the combustion of fuels used to reach the high temperatures (in the case of electric furnaces, the emissions associated to the production of electricity should be considered), but also other emissions such as SO_x and NO_x.

Calcium aluminate cements are the most important kind of non-Portland cements due to their properties (rapid hardening, resistance to high temperatures, resistance to temperature changes, resistance to chemical attack and resistance to impact and abrasion) (Scrivener, 2003). However, calcium aluminate cements are expensive and are used only in special applications. There are different types of calcium aluminate cements depending on the amount of the three constituents of the ternary diagram (CaO-SiO₂-Al₂O₃, see in Fig. 1 where calcium aluminate cements are in this ternary diagram), and according to Kurdowski, 2014 (page 604) these types of calcium aluminate cements are:

- Type 1: 37–40% Al₂O₃; 11–17% Fe₂O₃; 3–8% SiO₂; 36–40% CaO.
- Type 2: 48–51% Al₂O₃; 1–1.5% Fe₂O₃; 5–8% SiO₂; 39–40% CaO.
- Type 3: 51–60% Al₂O₃; 1–2.5% Fe₂O₃; 3–6% SiO₂; 30–40% CaO.
- Type 4: 78–80% Al₂O₃; 0–0.5% Fe₂O₃; 0–0.5% SiO₂; 17–27% CaO.

For instance, Almatix commercializes calcium aluminate cements with two qualities: 70% Al₂O₃ (25–30% CaO) and 80% Al₂O₃ (17–19% CaO), while the maximum levels for impurities is in both qualities of 0.3% Na₂O, 0.3% SiO₂, 0.2% Fe₂O₃ and 0.4% MgO. It is possible to establish two groups of calcium aluminate cements: normal product, dark grey or black in color, used in a broad temperature range; white varieties, characterized by the high alumina contents, used for refractory purposes (Bensted, 2008). The manufacture of normal product quality is carried out by fusing a mixture of calcium carbonate (limestone) and ferruginous bauxite in reverberatory furnaces at

1500–1600 °C (Bensted, 2008). The heating of these furnaces is performed with pulverized coal, oil, natural gas, or a mixture of these fuels (Bensted, 2008). The production of white quality cement is carried out in rotary kilns by sintering or in electric arc furnaces also at high temperatures (Scrivener, 2003; Bensted, 2008). The main requirement is the purity of the raw materials (limestone and bauxite), which are expensive (Scrivener, 2003), and the heating should avoid the contamination of the cement by using specific combustibles with low level of impurities (Bensted, 2008). High alumina calcium aluminate refractory cements are for those reasons expensive materials.

Solar energy has been used in obtaining calcium aluminates (Abdurakhmanov et al., 2012). They proposed the utilization of concentrated solar energy in the synthesis of calcium aluminates with impurities of rare earth elements (Nd and Sr). The objective of Abdurakhmanov et al., 2012 was synthesizing light-generating material from calcium aluminate. First, they melted mixtures of calcium and aluminum oxides, and then after fast cooling down, the product was milled in water and Nd₂O₃ and SrO were added to the powders. The powders were compacted and sintered to obtain calcium aluminates. The research proposed by Abdurakhmanov et al., 2012, as opposed to our research, had as objective synthesizing light-generating material (absorb solar energy and reemit it in different ranges of visible light).

As we have mentioned, the production of cement is energy intensive, and is also responsible of significant CO₂ emissions. In this way, by using concentrated solar thermal (CST), CO₂ emissions could be reduced by 40% and savings could also reach the 40% as fuels and electricity are replaced with solar energy (González and Flamant, 2014; Meier et al., 2005a). Similar savings and reductions in CO₂ emissions are expected in the event of applying concentrated solar thermal to the synthesis of calcium aluminate cements. In this way, we propose the synthesis of calcium aluminates (main phases of the calcium aluminate cements) by using concentrated solar energy.

2. Experimental methodology

Experiments were carried out in a vertical axis 1.5 kW solar furnace (see Fig. 2) in the facilities of the PROMES-CNRS located in Odeillo (France). The functioning of the vertical axis solar furnace is described as follows: a solar tracking heliostat reflects the rays towards a 2.0 m in diameter parabolic concentrator (see Fig. 2), which makes converging the sun radiation at a focal point with a dimension of around 15 mm in diameter. The maximum concentration for this solar furnace is approximately 15,000 times the incident radiation. The average incident radiation took values of around 900 W/m² in all experiments, although during each experiment the values fluctuated in the range ± 10% of the average value for the incident radiation. The control of the power

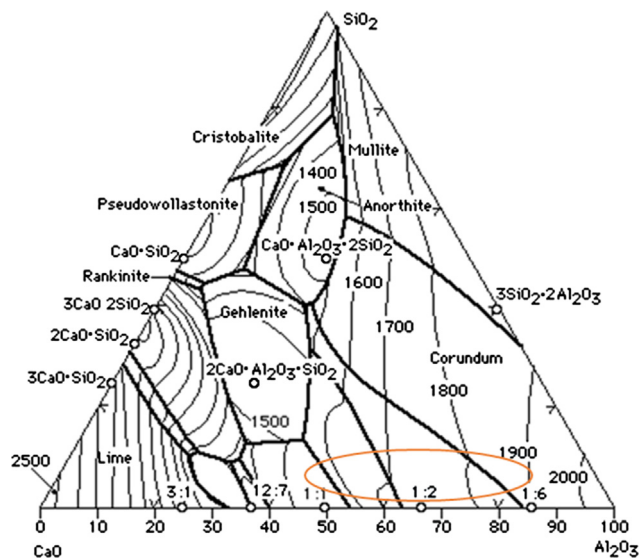


Fig. 1. Location of calcium aluminate cements in the diagram Al₂O₃-CaO-SiO₂ (TAPP 2.2; Levin et al., 1964).

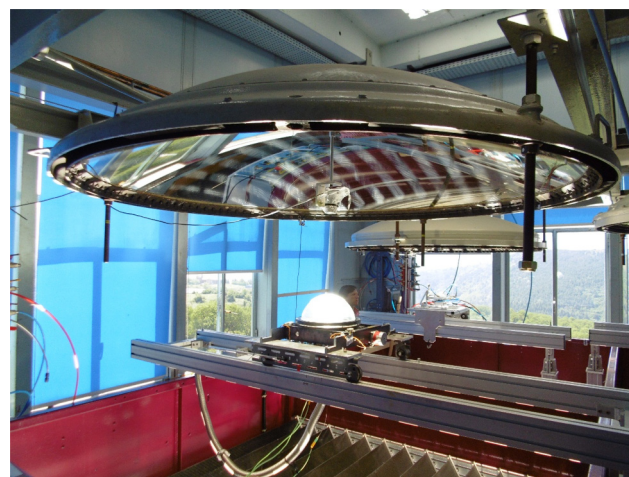


Fig. 2. 1.5 kW vertical axis solar furnace at Odeillo.

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