



Calcium aluminate based cement for concrete to be used as thermal energy storage in solar thermal electricity plants



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ABSTRACT

A concept for thermal energy storage (TES) in concrete as solid media for sensible heat storage is proposed to improve the cost and efficiency of solar thermal electricity (STE) plants. Mortar and concrete mixes were designed with calcium alumina cement (CAC) blended with blast furnace slag (BFS), using aggregates of different sources and size for stability performance after long term at high temperature. Seventy-five thermal cycles of 24 h length, within the temperature range 290 °C to 550 °C, have been used to simulate the expected operating conditions of TES. The dehydration processes at microstructural level have been evaluated and correlated with mechanical properties. Dehydration processes and consecutive heat/cool cycles induce changes in concrete at micro- and macro-level. The stabilization of damage with the charge/discharge heat cycles for thermal fatigue depends significantly on the aggregate type used. CAC is a suitable binder to use in thermal energy storage systems able to maintain its properties under repetitive heat cycles.

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1. Introduction

An increased interest on environmental issues has been rising since the Kyoto Protocol came into force, focusing on electricity production with no CO₂ emissions. One of the main challenges for renewable energy technologies is finding solutions to problems derived from the varying or intermittent nature of most renewable resources. Among these resources, solar energy arises with some major advantages, not only because it is much more abundant than any other renewable source but also because the hours of solar radiation for a certain location are easier to predict and measure. Solar thermal electricity (STE) that uses direct solar radiation for the production of electricity was introduced in the early 1980s in California, USA, and actually are extending all over the world. Compared with other solar energy forms, STE is the only one that allows storage so that extra energy collected can be stored for later usage during the night or on cloudy periods [1]. Energy storage is essential for improving the performance of STE plants and its dispatchability, which is a valuable asset and allows STE plants to be managed like fossil-power plants or hydropower plants [2]. Without thermal energy storage (TES), STE plants would be an intermittent power resource depending exclusively on solar radiation availability, as it is the case of photovoltaic systems. Therefore, the development of an efficient and economical solar energy storage system is of major concern and a key feature of STE plants.

The current state of the art in commercial STE plants with TES facilities is based on sensible heat storage, using 2-Steel-tanks system [1,2]. Binary mixtures of molten salts (MS) are introduced inside the tanks as heat storage material, and ΔT cycles are created for power energy production. However, the high volume of MS needed due to its high cost limits the investments in TES of STE plants. More recently, the thermocline storage system appears as an alternative that reduces the MS content by using only one tank for heat storage [2].

TES using solid materials, like concrete, emerges as an attractive option for storing sensible heat [3,4]. There are several practical reasons that make concrete based materials a promising option: easy handling, workable on site, available anywhere in the world, and relatively low cost. Besides, the technical capability as its high heat capacity has been given priority for research in recent years as a thermal energy storage solid medium. First studies considering concrete as potential material for thermal energy storage were initiated in the 1990s. The high sensible heat storage capabilities of Portland cement pastes hydrated phases as monosulfoaluminates, ettringite, or Friedel salts were analyzed [5,6].

One already developed application of concrete for solar energy storage considers its use as part of a heat exchanger system (a series of pipes inside a concrete module, circulating hot water as heat transfer fluid (HTF)), but the maximum working temperature of this system is limited due to the high steam pressures reached and the working temperature is below 100 °C [7–9], limiting in this way the heat capacity storage of the concrete. More recently, higher temperature storage concrete has been designed and validated up to 400 °C [3,4,10,11] using also a heat exchanger system circulating inside the concrete. This high-

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temperature concrete considers Portland blast furnace slag cement, aggregates with thermal properties (such as iron oxides, basalt...), and also polypropylene fibers (PPF) to reduce the risk of spalling [10]. In the concrete–heat exchanger design, the thermal storage efficiency depends, among other parameters, on concrete conductivity [9]. High thermal conductivity and high thermal capacity of the concrete mix helps to achieve a more uniform heat distribution inside the concrete bulk, but the stability of concrete to support temperatures above 400 °C under heat cycles is crucial.

Concrete has been also considered as solid filling material inside the thermocline tank in direct contact with the MS [12]. Concretes containing ordinary Portland cement (OPC) or calcium alumina cement (CAC) blended with fly ash and silica fume as mineral additions and PPF were evaluated in this case. Limestone was used as coarse aggregate together with siliceous river sand. The concrete mechanical performance was evaluated after 30 heat cycles between 300 °C and 585 °C. The findings show that plain OPC concrete retained only 14% of its initial compressive strength and severe cracking was observed, while the blended OPC concretes and CAC concretes maintained at least 30% of their initial strength. In addition, it was found that coarse aggregates contributed to a higher reduction of the compressive strength.

Future challenge in the STE world is that plants must be designed to operate up to 600 °C for higher efficiency operation; thus, the developing and validation of more optimum concrete mixtures is of significant importance.

Within this context, refractory concretes seem suitable materials for use in structures that have to endure for long periods under temperatures ranging from 500 °C up to almost 2000 °C [13]. As the concrete temperature requirement in future STE heat storage plants has to bear for long periods temperatures up to at least 600 °C, a refractory concrete design can be taken into consideration. Refractory concretes have been used primarily as protective lining so that they are considered as consumable material that needs to be replaced after an appropriate service life. However, attention has been given recently to the possibilities of using refractory concretes for large load-bearing structures, as for nuclear power plants [14].

Many different materials can be used for refractory purposes, and some of them are based on the use of hydraulic cements: OPC allows limited refractory applications, and the refractory properties are mainly provided by the use of appropriate aggregates in the mix. Calcium silicate hydrates (CSH) from the hydrated OPC decompose at temperatures above 105 °C and calcium hydroxide (CH) at temperatures around 450 °C, both forming CaO as main dehydrated residue [15]. After cooling, as downtime and maintenance periods in an STE plant, the CaO can be easily rehydrated again by absorbing moisture from the atmosphere with the consequent swelling of the hardened paste, increasing cracking; also it causes powdering of the cement paste after been exposed to temperatures above 500 °C. Therefore, thermal cycling could result in severe disruption and damage [16,17]. Blended cements, using blast furnace slag or natural pozzolans as mineral additions, generate more CSH and less CH than plain OPC so that dehydration follows similarly as in the Portland case except for CH, and then the residual strength can be higher after cooling than in the case of pure OPC [18].

For the reasons mentioned above, attention is then focused on CAC. At normal temperatures and moisture, the first hydrated phases are CAH_{10} and C_2AH_8 and colloidal aluminum hydrate [19–21]. Hydration takes place very rapidly, allowing high mechanical strength after 24 h [20]. The hexagonal CAH_{10} and C_2AH_8 phases evolve with time to more stable cubic phases (C_3AH_6) and AH_3 [19,20]. The density of the C_3AH_6 cubic phase is higher (2.53 g/cm^3) than that of the initial CAH_{10} hexagonal phase (1.72 g/cm^3) [22]. This increase in the density of the hydrated cement paste is accompanied by an increase in its porosity, mainly because of the strength decay after the conversion of hexagonal hydrated CAC phases. Several actions have been considered to retard or completely eliminate the evolution of hexagonal phases to cubic ones [23–28]. More commonly used solutions are based on the incorporation

of mineral blends containing high silica content, as blast furnace slag (BFS), fly ash (FA), or silica fume (SF) to avoid or retard the C_3AH_6 transformation due to the preferential formation of siliceous hydrogarnet phases (type $\text{C}_3\text{AS}_{3-x}\text{H}_{2x}$) or strätlingite (C_2ASH_8) [25,28,29], more thermally stable calcium aluminates containing silica in their structure. Also, promoters of the formation of Afm phases [23,26,27], even hybrid cement mixes OPC/CAC, have been considered to control the conversion process in CAC [30].

There are relatively few studies on CAC dehydration compared with those available for OPC paste. Research conducted [14,28,31] shows that above 105 °C, the chemically combined water of CAC paste is gradually lost from hydrated aluminates. Most important changes occur up to 300 °C: dehydration of CAH_{10} and AH_3 . At temperatures between 400 and 500 °C, the dehydration of aluminate hydrates should be completed [31]. As also occurs with hardened Portland cement paste, the density and the porosity of CAC hydrates change due to dehydration above 100 °C. The porosity of CAC concrete can increase by about 25% up to 500 °C [32]. These changes in porosity and in density affect the mechanical strength of CAC cement paste. From 200 °C to 600 °C, CAC paste strength decays around 50% [14]. At higher temperatures, no significant changes occur until melting above 1300 °C. In addition, the dehydrated CAC is more stable after cooling, reducing swelling risk [14], a critical aspect in OPC.

From the above literature review, the developing of concretes with high thermal stability exhibiting refractory properties is a relevant research topic. The suitability of a refractory based design concrete for TES in STE plants based on CAC stabilized with BFS is considered. The influence of the aggregate composition and size are taken into account, and the mechanical changes observed after repetitive heat cycles are analyzed with regard to the changes in microstructural properties observed; 25, 50, and 75 heat cycles in the range of 290 °C to 550 °C, corresponding roughly to 1, 2, and 3 months of STE plant operation, are considered.

2. Experimental setup

2.1. Samples preparation

A calcium aluminate cement (CAC), electroland from Ciment Molins, containing 40% alumina, has been blended with blast furnace slag (BFS) to control any risk of early conversion from hexagonal to cubic hydrated phases before heating and to improve thermal performance at high temperature. A binder mix of 70% CAC + 30% BFS was selected. The chemical compositions of both components are included in Table 1, in weight %. According to the British Standards Institutions [33], for special refractory concretes, the total alumina content shall be above 32% and the ratio alumina (Al_2O_3) to lime (CaO) shall be between 0.85 and 1.3. The CAC used in the present work (40% Al_2O_3 and $\text{Al}_2\text{O}_3/\text{CaO}$ ratio of 1) meets well with the mentioned basic requirements of hydraulic binding agent in refractory concretes. In addition, the iron content also has a relevant role; contents around 15% Fe_2O_3 limits the maximum temperature of use to 1370 °C, comfortably meeting the requirements in an STE plant with heat storage system.

The particle size distribution of both components is included in Table 2; the mean particle size of BFS is three times lower than that of CAC.

Table 1
Chemical composition of CAC cement and BFS (wt.%).

	%SiO ₂	%Al ₂ O ₃	%Fe ₂ O ₃	%CaO	%MgO	%SO ₃	%Na ₂ O	%K ₂ O
CAC	4.40	40.30	15.20	37.43	0.47	0.06	0.16	0.14
BFS	36.57	10.39	0.29	45.50	7.46	0.05	0.42	0.49

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