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Manufacturing belitic calcium sulfoaluminate (BCSA) cement from UK materials: Semi-industrial trial

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Globally over 4 billion tonnes of cement are being produced and consumed annually. Cement is one of the essential materials that form the backbone of modern infrastructure, and its consumption has also been used as an indicator for tracking the growth and development of economies. On the other hand, the manufacturing of cement also exerts a significant environmental burden. The cement manufacturing process is energy intensive and emits approximately 0.8 kg of CO_2 for every kilogram of traditional Portland cement (PC) clinker produced. The combination of the high emissions, coupled with the scale of production contributes to roughly 8% of the total anthropogenic CO_2 emissions. To put this in perspective, if the cement manufacturing industry were a country it would rank as the third-largest emitter of CO_2 only succeeded by China and the United States. With the advent effects of climate change and global warming, industries worldwide are focusing on reducing their carbon footprint. The cement industry also has also developed roadmaps and several strategies to reduce the overall emissions form the industry. As a part of the UKRI Future Leaders Fellowship project, Green, Circular, and Smart Cement Manufacture, we are exploring innovative approaches to manufacture alternative low CO_2 binders using the industrial by-products and waste materials. We aim to enable calcium sulfoaluminate based cements manufacturing in the UK through an innovative process and to create new UK products.

Why calcium sulfoaluminate cements?

Calcium sulfoaluminate (CSA) based cements are a promising alternative low CO_2 binder to the traditional PC. In the cement manufacturing process, about 90% of CO_2 emissions in the cement manufacturing process comes from the calcination of limestone and the fuel burnt to achieve the required kiln temperature. PC binder systems have alite (C_3S) as the primary reactive phase and is a lime/calcium intensive phase whose operational synthesis requires a temperature of ~1450 °C. Unlike PC, CSA binders have ye'elimite ($C_4A_3\$$) as the primary clinker phase (Note: Cement chemistry notations, C: CaO, A: Al_2O_3, S: SiO_2, \$: SO_3). The synthesis of ye'elimite phase requires less limestone and lower kiln temperatures, resulting in process and fuel CO_2 emissions that is lower tha that of C_3S. CSA cements typically have ye'elimite with belite (C_2S) and calcium sulfate ($C\$$) as the other primary clinker phases. Additionally, it is claimed that CSA cements have a porous clinker structure, reducing the energy needed for griding of the clinker. This translates to an overall 25%–35% reduction in CO_2 emissions in the manufacturing of CSA cements from PC, based on their phase assemblage. One of the main advantages of the manufacturing process of CSA is their compatibility with the existing PC manufacturing infrastructure. The production process of CSA is very similar to that of PC thus would not need the development of new facilities (i.e., minimal capital investment). The raw materials used for manufacturing of CSA cements are also similar to PC, though CSA uses lesser limestone and more alumina and gypsum. This makes CSA cement a practical and scalable alternative low CO_2 binder for reducing the carbon footprint.

Although the interest in CSA cements as a low CO_2 binder has increased in the recent times, CSA cements have been commercially used for decades as a special cement. Initially patented by Alexander Klein in 1966 as an expansive binder, CSA cements have evolved into standalone binder overtime. CSA cements based on their phase composition have different terminologies such as the belitic calcium sulfoaluminate (BCSA) and belite-ye'elimite-ferrite (BYF) to name a few. Based on the phase composition, CSA cements can also have varied properties such as being rapid hardening, achieving a high early-age strength, self-levelling, cavity/mine/bulk filling, expansive and self-stressing. The expansive nature (and shrinkage properties) of the CSA cements can be harnessed to construct long span industrial floor slab and bridge decks minimising the expansion joints, making the construction process faster, easier, and enhancing the service life of the structure. The durability performance of CSA based cements after decades of service have also recently shown to be good. CSA cements have also found a wide range of non-structural and structural applications such as a binder in concrete for bridges, infrastructure buildings, runways and taxiways for airport, pavement concrete road-repair, and production of concrete pipes. Despite the numerous advantages, the widespread manufacturing and use of CSA cements have been limited due to the higher manufacturing costs, mainly attributed to the cost and availability of bauxite which was believed to be a necessary source of alumina.

Next generation CSA cements:

The properties of CSA cement are directly linked to its clinker phase composition. By varying the composition of the primary clinker phases ye'elimite, calcium sulfate and belite, we can design to cement to have the desired characteristics. For instance, the compressive strength in CSA cements typically increases with the ye'elimite content, or increasing the gypsum content relative to ye'elimite enhances the expansion characteristics. However, increasing the ye'elimite content would also increase the manufacturing cost due to the higher demand for alumina. The belitic calcium sulfoaluminate cement with the composition of about 20–30% ye'elimite and 50–60% belite, could be an optimal balance, diminishing the requirements of high alumina containing raw materials and enabling the use of high alumina clay instead (with additional alumina supplements where required). BCSA cements can achieve similar strength of PC but in a much faster timeframe. The ye'elimite phase would hydrate rapidly forming a dense microstructure at early ages (within a few hours) contributing to the hardening and early age strength with belite hydration at later age contributing to long-term durability performance.

PC binder often relies on supplementary cementitious materials (SCM) like flyash or ground granulated bast furnace slag to reduce the overall carbon footprint (and improve durability). However, BCSA cements can achieve the desired characteristics without the need of SCMs. Further BCSA cements can also be blended with gypsum (calcium sulfate) to alter/refine the mechanical properties such as the compressive strength and expansion. This distinction is critical when considering the recyclability of the cementitious materials as the hardened cement paste of a blend cannot be readily refired into a clinker. One of the crucial steps in making the cement industry sustainable would be to include circularity in the cement manufacturing process. Researchers are working diligently on the deconstruction and recycling of end-of-life concrete structures. While recycled concrete aggregates (RCA) have been successfully reused, this process always leaves behind the cementitious fines, which are often discarded. These materials (mostly composed of hardened cement paste) could potentially be used as a raw material for manufacturing cement; however, when PC systems are used as PC-SCM blends, this changes significantly the chemistry of the originally manufactured clinker. PC systems depending on C_3S for the hardening and early age strength require a high lime content to synthesise. This also limits the use of alternative materials and lower grade limestone to produce PC, as the raw meal for PC has stringent chemical limits to ensure the required phase assemblage forms during clinkering. Thus, when PC blended hardened cement paste must be reused, this would require additional dose of lime to meet the requirements of the raw meal. BCSA cement on the other hand, at the end of life, can be fully recycled (especially if the sulfate source is produced in the kiln as anhydrite) and incorporated as a raw material for the raw meal. BCSA typically is used directly or blended with gypsum, which is also one of the raw materials used for synthesising, thus does not affect the raw meal. Furthermore, unlike PC, BCSA cement, can also accommodate the use of alternative low-grade materials in the raw meal if used in a diluted manner. When used in applications where time is of the essence, the cost of BCSA could easily be justified, however may never replace PC for all commercial applications. If we can use alternative cheaper sources of alumina, we can potentially make BSCA also economically viable.

Alternative materials for manufacturing BCSA

The main bottlenecks for the wider manufacturing of BCSA cement is its high alumina demand (in particular in the UK where bauxite is scarce), which can be overcome by using alternative industrial by-products. Several researchers have worked on synthesising BCSA in laboratory scales using such alternative materials that are rich in alumina such as flyash (from the coal combustion), granulated blast furnace slag (from steel manufacturing), aluminium sludge (from water treatment), alumina slag and red mud (from aluminium manufacturing). The use of alternative materials not only reduces the dependence on virgin bauxite source but also diverts the waste form landfills (to dispose the industrial by-products). However, the use of such alternative materials comes with its own set of logistic and technical challenge such as the local availability of the material, and the inevitable impurities and minor elements present in them. These minor elements could affect the clinkering process and influence the reactivity of the clinker made with them and we should be careful about the dosage and how we use the material; nonetheless, if used in reasonable (diluted) amounts, these adverse affect can be diminished. In this study, we focus on using one such alternative alumina rich material, understand its implications on the clinker and check the feasibility of using it for manufacturing BCSA cement in a semi-industrial trial using only UK-sourced materials in the raw meal. The by-product from the secondary aluminium production process was identified as an alumina source from within the United Kingdom.

The recycling of aluminium from scrap is done using the secondary aluminium process and has been on the rise globally with the demand projected to be about 97 million tonnes annually. The aluminium production is also accompanied by considerable amounts of by-products. During the recycling, a slat flux is added with the scrap metal to remove the impurities which is finally collected as the slat slag. This slag is further refined to extract the viable aluminium and leaching the salts to be reused in the process, leaving behind the non-metallic (by)product (NMP). NMP is considered a waste material, has found limited commercial applications and typically dumped in landfills bringing economic and environmental pressures to the industry. Although viable aluminium has been extracted, NMP ends up with over 60% of Al_2O_3, making it a potential alternate alumina source. With the UK processing up to 150,000 tonnes of secondary aluminium annually, making use of the NMP would additionally benefit the circularity of materials in our foundation industries. For the first time (to the best of our knowledge), in this study, we demonstrate the use of locally available raw materials in the UK to manufacture BCSA in a rotary kiln.

Production of BCSA clinker: from lab to pilot kiln

Preliminary studies at laboratory scale explored the feasibility of using NMP in the raw meal for synthesising BCSA cement. The raw materials for the study, limestone, gypsum, different sources of clay and NMP were sourced locally within the UK. The raw meal for clinker trials were designed using the different of raw materials for the targeted clinker composition to have approximately 30% ye'elimite and more than 60% belite (as a BCSA cement). Numerous clinker recipes/sample were produced using these raw meal formulations, by varying the two key parameters: maximum clinkering temperature and dwell time (the residence duration at the peak temperature). The clinker samples were characterised and analysed to quantify the clinker phases and understand the role or effect of the minor elements on the clinkering reactions. The results from the study showed the influence of both the raw materials and clinkering conditions on the clinker formation. Based on these findings one recipe and set of clinkering conditions were selected for the pilot scale trial.

The up-scaling of BCSA cement manufacturing, was conducted at IBU-tec Advanced Materials AG using a pilot kiln. The kiln with the dimension of 0.3 m internal diameter and heated length of about 7m uses direct fired natural gas burner to reach the required temperatures. One of the main challenges in BCSA production in kiln is managing the sulphate content of the raw meal. The synthesis of ye'elimite requires a temperature of at ~1250 °C, but the high temperature or prolonged dwell time (which can be caused by process/kiln configuration) can lead to loss of sulfur, altering the chemistry. The environment of burning of natural gas can further create pressures, accelerating the desulfurization from calcium sulfate and ye'elimite. Thus, the key process parameters such as the meal feed rate an form, kiln inclination and rotation and maximum temperature must be optimised to ensure the samples have sufficient time at the sintering temperature for the completion of the clinkering reaction and not cause significant desulfurization.

The pilot trial lasted for over 5 days, using about 3000kg of raw materials and successfully producing more than one tonne of targeted BCSA clinker. The details of the raw meal and composition of synthesised clinker are shown in Fig. 1. The raw materials were characterised on site to check their oxide composition and moisture content, to correct the raw meal proportions. The raw meal was made using the different raw materials in the required proportions in batches of 200 kgs. The raw materials were blended and granulated in a mixer at 1500–3000 rpm (optimised based on preliminary studies done using the raw materials) to obtain granules in the size range of 1–5 mm. The granulated samples were then continuously loaded into the pre-heated rotary kiln in the temperature range of 1260 °C to 1280 °C (based on the findings from the lab scale trial). The kiln was set to a 1.5 ° inclination and rotation speed of 2 - 2.5 rpm with the feed rate of 25 kg/hr for the raw meal granules. These process parameters were selected to ensure the products had about 15 minutes in the sintering zone (totally 60 minutes in the kiln), for the clinkering reactions to be completed (based on the findings from the lab scale).

The rotary kiln used is shown in Fig. 2 also showing the hot clinker coming out of the kiln. As a part of the quality assurance for the clinkering process in the kiln, the temperature, air flow rates and pressure in the kiln were monitored in real time. The temperature in the kiln was monitored, using thermocouples attached to the kiln at 6 different locations. Additionally, the temperature of the samples monitored using a manual pyrometer periodically. Fig. 3 shows the sample temperature (T0) and the temperature of the kiln at three locations: near the inlet (T1), mid-way through the kiln at about 4.5 meters from the burner (T2) and near the burner, closer to the sample outlet (T3). Further, the clinker produced was periodically sampled (every 2–3 hours) and characterised throughout the trial using x-ray diffraction. The clinkers were predominantly found to meet the target phase assemblage, with minor fluctuations, proving the robustness of the process. The pilot trial was able to successfully demonstrate the viability of using UK sourced materials to steadily and consistently manufacture over a tonne of BCSA cement.

What's next?

The next phase of the project will focus on demonstrating the performance and applications of the locally manufactured BCSA cement using real-world size members; the quantity produced in the pilot trial allows for a suite of practical tests that would not normally be possible when producing materials in laboratory at gram scale. After detailed studies on the mechanical and durability characteristics of the binder, the BCSA cement will be used for a small-scale demonstration project. As a fast-track project, an access ramp facility is planned to be constructed, with the facility expected to be ready for public use within just 1–2 days of construction. This project would pave way for commercial development and adoption by the UK industry and beyond.

Acknowledgements

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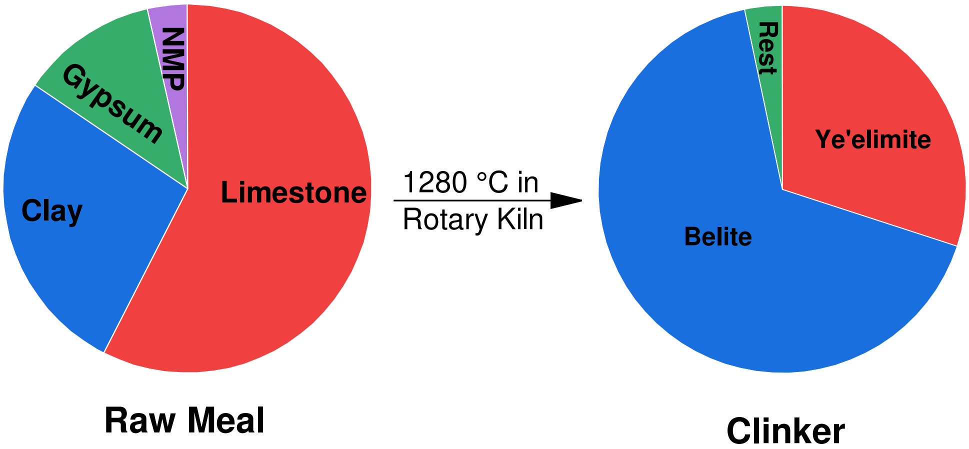

Figure 1:Pie chart illustrating the composition of raw materials and the phase proportions in the BCSA clinker produced

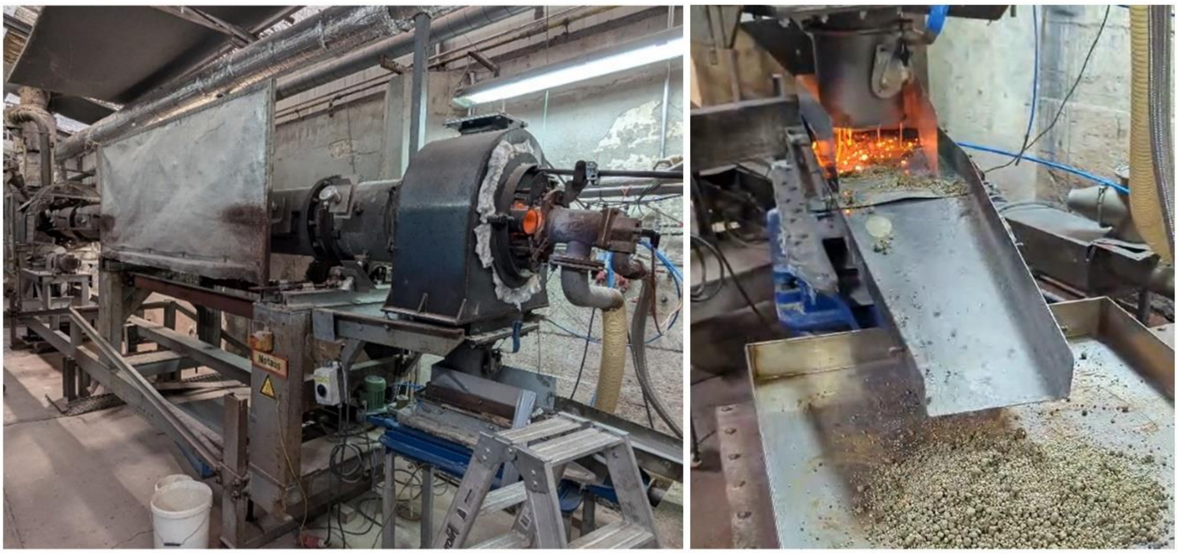

Figure 2:Figure depicting the pilot rotary kiln in operation and the hot clinker exiting the kiln to the vibrating cooling table for sample collection.

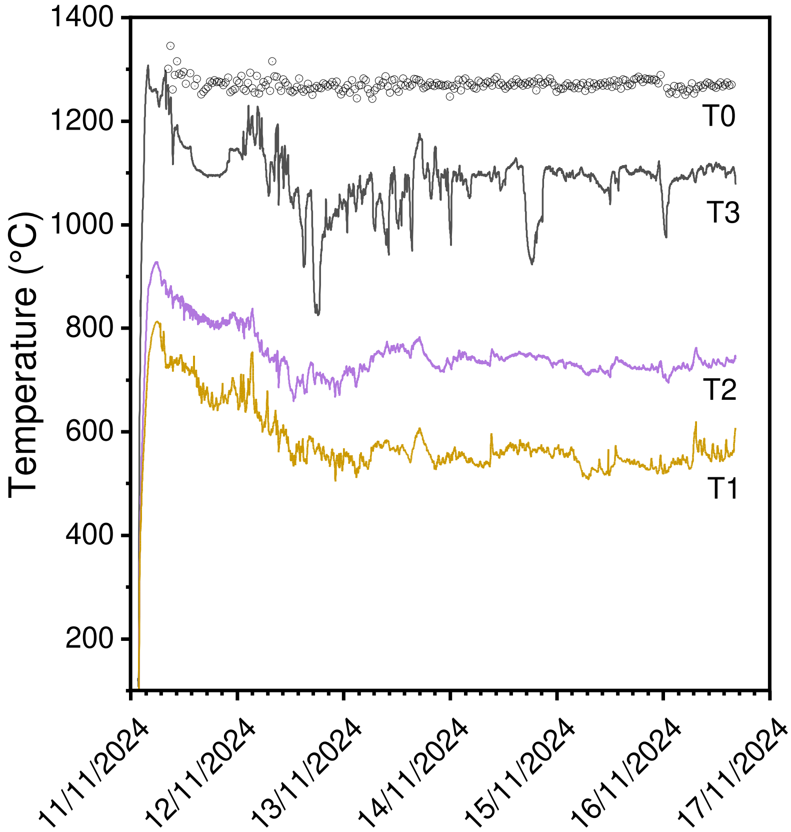

Figure 3:The temperature profile of the kiln, monitored at three locations using thermocouples (lines), with the clinker hottest temperature recorded manually using a pyrometer (symbols) during the 5 days of trial