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Belitic calcium sulfoaluminate as mineral admixture in limestone-calcined clay cement and low carbon dioxide binders

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This study investigates blending belitic calcium sulfoaluminate (BCSA) cement with Portland limestone cement (PLC) and calcined clay (CC). Two blends, PLC-BCSA and PLC-BCSA-CC, were tested using different proportions of BCSA cement, PLC and calcined clay (CC). Workability was assessed using a flow test, and mechanical properties were evaluated through compressive strength and shrinkage tests. The hydration process was analysed using thermogravimetric analysis, scanning electron microscopy and X-ray diffraction. The global warming potential (GWP) of the blends was calculated, and the 'carbon intensity' was estimated as carbon dioxide (CO₂) equivalent per unit of compressive strength, a first for BCSA and BCSA blends. Although the PLC-BCSA and PLC-BCSA-CC blends showed slower strength development compared to BCSA, they achieved similar compressive strengths at 28 days while improving the early-age compressive strength of PLC and limestone-calcined clay cement. BCSA blends can significantly reduce carbon dioxide intensity, highlighting their potential as more sustainable concrete alternatives.

Keywords: acceleration/BCSA/carbon dioxide/early-age strength/emissions/ettringite/GWP/rapid-setting concrete/shrinkage/
UN SDG 9: Industry, innovation and infrastructure/UN SDG 13: Climate action

Introduction

Low-embodied carbon dioxide (CO₂) binders have emerged as alternatives to conventional Portland cement (PC) due to the growing awareness of carbon dioxide emissions and their impact on global warming potential (GWP). While PC remains the most widely used binder in concrete production, it is also responsible for approximately 7% of global carbon dioxide emissions (GCCA, 2024). These emissions primarily result from the high limestone content in the raw material feed and the elevated kiln temperatures, reaching up to 1450°C, required during its manufacture (He *et al.*, 2019). In addition to their environmental drawbacks, Portland-based cements are also known for their extended setting times and relatively low early strengths (Ijaz *et al.*, 2024; Sommariva, 2020).

Both industry and academic efforts are ongoing to develop alternative binders with a lower carbon dioxide footprint, including Portland limestone cement (PLC), limestone-calcined clay cement (LC3), calcium sulfoaluminate (CSA) cement and belitic calcium sulfoaluminate (BCSA) cement. Additionally, the use of supplementary cementitious materials (SCMs), such as calcined clay

(CC), fly ash, slag and polymers, has increased as a means of reducing the clinker content in concrete mixtures.

PLC, also known as Type IL, is a low-embodied carbon dioxide binder in which 5% to 15% of the PC is replaced with limestone (Scrivener *et al.*, 2018). It is estimated that the use of PLC can result in a nearly 10% reduction in carbon dioxide emissions per ton of PLC compared to ordinary PC (ACA, 2022).

LC3 is a recently developed low-embodied carbon dioxide binder, composed of approximately 50% Portland clinker, 30% CC, 15% limestone and 5% gypsum by weight (Scrivener *et al.*, 2018). The increased limestone content, which is abundant, and the use of low-grade clays result in reduced carbon dioxide emissions, cost savings and performance similar to that of PC. The use of LC3 in concrete production can reduce carbon dioxide emissions by up to 40% compared to PC (Scrivener *et al.*, 2018).

BCSA is a particular type of CSA cement that offers several benefits, including low embodied carbon dioxide, rapid setting, high early-age strength, low shrinkage, resistance to chemical attacks,

and continued strength gain over time. It is named after its two major phases, belite (C_2S , Ca_2SiO_4) and CSA (ye'elimite, $C_4(AIO_2)_6SO_2$), and does not contain alite (C_3S , Ca_3SiO_5). The combination of 50% belite, 30% ye'elimite by weight, and the absence of alite allows the clinker to be produced at a temperature close to 1250°C, which is around 200°C lower than the production temperature of PC. The amount of calcium oxide (CaO) in the ye'elimite structure is about half that of alite, significantly reducing the limestone requirement for BCSA production (Sommariva, 2020). Together, the reduced limestone content and lower kiln temperature lead to a 30% reduction in carbon dioxide emissions compared to PC (CTS, 2023).

CSA cement hydrates rapidly, producing ettringite (AFt), which contributes to its high early strength. The hydration of ettringite requires significantly more water than the hydration of alite to form calcium silicate hydrate (C-S-H). To fully hydrate PC, a water-to-binder (W/B) ratio of only 0.2 is needed (Mather and Hime, 2002), whereas for BCSA cements, the theoretical minimum for full hydration is around 0.48 (Murray *et al.*, 2019). The higher water demand must be taken into account during mix design to ensure adequate workability (Huang *et al.*, 2019; Trauchessec *et al.*, 2015).

Blends of PC and CSA (PC-CSA) have been shown to achieve compressive strengths comparable to those of pure PC. The hydration products in these blends are a combination of typical phases found in hydrated CSA and PC cements, including C-S-H gel, AFt, AFm (calcium monosulfoaluminate), portlandite and hydrogarnet (Trauchessec *et al.*, 2015; Xie and Qian, 2023). However, long-term performance depends on the continued development of other hydration products (Bianchi *et al.*, 2009; Huang *et al.*, 2019). The addition of CSA as a mineral admixture alters the rheological behaviour of the blends, emphasising the need for proper water and admixture adjustments (Huang *et al.*, 2019). The calcium sulfate content of commercial CSA cement and PC is typically adjusted to optimise performance. Calcium sulfates (gypsum, anhydrite and bassanite) are crucial for controlling dimensional stability and regulating ettringite formation, particularly in blended PC-CSA-anhydrite systems (Chaunsali and Mondal, 2015; Yang *et al.*, 2021). However, excessive ettringite formation can lead to swelling or water depletion, potentially compromising structural integrity (Xie and Qian, 2023). The water-to-cement ratio must be high enough to also provide water for PC hydration that starts after initial ettringite formation.

Research on ternary PC, CSA and CC systems remains limited (Bescher, 2025). It is assumed that CC will contribute to the pozzolanic reaction in ternary systems to form C-S-H. It has been shown that various pozzolans can react with CSA, leading to additional strätlingite and C-S-H formation (Martin *et al.*, 2017; Winnefeld *et al.*, 2017). When PC is replaced with CC alone, it contributes to a lower carbon dioxide footprint; however, this

often comes at the cost of decreased early strength, reduced workability and slower setting time (Zhou *et al.*, 2022).

Given the increasing performance demands for concrete and tightening carbon dioxide emission regulations, the development of lower carbon dioxide alternatives is critical. Due to the reduced Portland clinker factor and the additional limestone and/or CC, PLC and LC3 alone cannot meet the performance requirements for rapid setting low carbon dioxide concrete. Therefore, the primary objective of this study is to introduce the addition of BCSA to PC and LC3 systems and to evaluate its potential to enhance the performance of PLC- and LC3-based cements by improving physical properties of the cements, particularly at an early age (e.g. strength), while also keeping carbon dioxide emissions low.

In this study, the hydration behaviour, mechanical properties and carbon dioxide intensity of BCSA, PLC and CC blends were analysed using thermogravimetric analysis (TGA), X-ray diffraction (XRD), scanning electron microscopy (SEM), flow testing, shrinkage measurements and compressive strength testing. The carbon dioxide emission intensity of the BCSA blends was further assessed using a GWP calculation.

Methods and materials

Methods

A comprehensive study of the physical and chemical properties was conducted to characterise the hydration kinetics, chemical composition, performance properties and carbon dioxide emissions of BCSA, PLC, PLC-BCSA blends and BCSA-PLC-CC blends.

The workability of each mortar was tested using flow test and setting time. The American Society for Testing and Materials C1437 (ASTM, 2020a) flow test was used to measure the flow of the mortars made of the blends listed above. A truncated conical mould was filled with mortar of each blend and then removed. The flow table was raised and dropped 25 times, after which the distance from the centre of the original cone to the outer edge was measured in four directions. The sum of these four measurements represents the flow relative to the original diameter of each mortar. The purpose of the flow test is to meet the criteria of flow $110 \pm 5\%$ that is required by ASTM C109 for a mortar (ASTM, 2020b). The initial and final set times of each mortar were recorded according to ASTM C191 (ASTM, 2021a). The initial set time was determined when the Vicat needle penetrated the specimen to a depth of 25 mm or less. The final set time was recorded when the needle no longer left a visible mark on the surface of the specimen.

The physical properties of mortars were tested through shrinkage (length change) and strength. Length change of the mortar specimens over time was tested using modified ASTM C157 (ASTM, 2017). Modification means that flow values were sometimes outside the range specified by the standard. For modified C157,

specimens were cured in the moulds in a moist cabinet for 24 h and then demoulded. After demoulding, the samples were air-cured and measurements were taken at 1, 4, 7, 14 and 28 days. Two specimens per mix were tested to have an average value. The ASTM C109 test for compressive strength (ASTM, 2020b) was used to determine the compressive strength of 50 mm × 50 mm × 50 mm mortar cubes. Strength measurements were performed at 1, 7, 14 and 28 days, with two replicates tested at each age. Additional testing at 1.5 and 3 h was conducted for fast-setting mixes. The specimens were cured in lime-saturated water. Due to availability of materials, only compressive strength was tested, but future characterisation of concrete samples should be done under compressive and flexural strength.

X-ray fluorescence (XRF) was used to determine the chemical composition of cement powders. Powder samples of PLC, BCSA cement and CC were pressed into pellets and analysed using a Bruker S8 TIGER wavelength-dispersive X-ray spectrometer, which was calibrated specifically for BCSA and PLC compositions.

The hydration kinetics of each blend was studied using isothermal calorimetry. Ten grams of cement and 5 g of water were weighed into dedicated plastic containers and conditioned in the calorimeter at 23°C for 24 h. Subsequently, the samples were mixed using a laboratory vortex mixer for 45 s and transferred to the calorimetry channel. The heat released by the cement paste samples was recorded at 23°C for 24 h using an I-Cal 2000 HPC calorimeter (Calmetrix, Inc.). With the known cement mass both the heat flow (mW/g) and the cumulative heat release (J/g) could be then calculated and reported. XRD and Rietveld analysis was used to identify the major phase contents anhydrous and hydrated cement pastes. A Bruker D2 Phaser benchtop X-ray diffractometer equipped with a Cu-K β radiation source and a scintillator detector was used to identify and quantify crystalline phases in well-ground and homogeneous powder samples. Three hydration stages were analysed to evaluate the evolution of hydration: before hydration (anhydrous sample), at 7 days and at 28 days. Hydration for the 7- and 28-day samples was stopped by immersion in isopropyl alcohol for 24–48 h. Afterward, the samples were dried, crushed and ground until they passed through a 200-mesh sieve. XRD scans were conducted with a step size of 0.02° 2 θ at three seconds per step, ranging from 5° to 55° 2 θ at a scanning speed of 0.384° 2 θ /min. The total scan time per sample was approximately two hours. TGA was used together with XRD to confirm which

hydration products are present in the hydrated pastes. TGA also provides information about amorphous phases. A PerkinElmer TGA 800 thermogravimetric analyser was used to monitor mass loss as a function of temperature. Each sample was weighted into an alumina crucible. The samples were then heated in an inert nitrogen atmosphere from 30°C to 1000°C at a rate of 30°C/min, with a gas flow rate of 20 ml/min. The mass loss percentage (TG curve) and its derivative (DTG curve) were plotted against temperature to identify peaks associated with the decomposition of the different phases present in the samples. The microstructure of 70PLC-30BCSA after 28 days of hydration was analysed using SEM. To stop the hydration at 28 days a piece of hardened cement paste was first submerged in isopropanol for 4 h, then dried and submerged for 24 h and again dried. For SEM, analysis a piece of paste was attached to sample holder using copper tape. An FEI NOVA 230 NanoSEM equipped with an Everhart–Thornley detector was used to examine the microstructure of thin cement pieces under a high vacuum. SEM imaging enabled the observation of features such as ettringite crystals, C-S-H and calcium hydroxide (CH). These observations were used to complement the XRD, TGA and compressive strength results.

Materials

This study used commercially available BCSA cement, PLC and CC (metakaolin). The BCSA cement complies with ASTM C1600 (ASTM, 2019), which specifies a minimum initial set time of 10 min and early strengths in as little as 1.5 h. The PLC complies with ASTM C595 (ASTM, 2008), which specifies a minimum initial set time of 45 min and strength requirements starting at 3 days. Metakaolin was the commercial pozzolan used for this research. In the USA, CCs do not have a dedicated material specification but are accepted as natural pozzolans under ASTM C618 (class N pozzolan) (ASTM, 2025) and are approved for incorporation into blended cements through ASTM C595. Blaine fineness values determined in accordance with ASTM C204 (ASTM, 2018) and reported on the mill certificates for BCSA cement, PLC and CC were 600 m²/kg, 434 m²/kg, and 787 m²/kg, respectively. Only mortar mixes were analysed to better understand the hydration and strength development mechanisms of each cement blend and to establish a basis for future concrete applications. The XRD analysis for both cements is shown in Table 1. Sand complying with the ASTM C778 standard (ASTM, 2021b) was used for the mortar preparation. The binder-to-sand ratio was 1:2.75, and the W/B ratio was maintained at 0.5 throughout the study.

Table 1. Major phase content of belitic calcium sulfoaluminate (BCSA) cement and Portland limestone cement (PLC) through X-ray diffraction analysis (Bruker D2 phaser)

Cement type/phase	Alite (hatrurite, Ca ₃ SiO ₅ , C ₃ S): %	Belite (larnite, Ca ₂ SiO ₄ , C ₂ S): %	Ye'elimite (Ca ₄ (AlO ₂) ₆ SO ₄ , C ₄ A ₃ \$): %	Ferrite (brownmillerite, Ca ₂ (Al,Fe)O ₅ , C ₄ AF): %	Tricalcium aluminate (Ca ₃ Al ₂ O ₆ , C ₃ A): %	Anhydrite (CaSO ₄ , C\$): %	Gypsum (CaSO ₄ *2H ₂ O, C\$H ₂): %	Calcite (CaCO ₃ , C\$): %	Others: %
BCSA	—	47.7	27.0	1.5	—	10.7	0.8	2.8	9.5
PLC	44.4	20.4	—	11.9	3.0	—	9.1	8.9	2.3

Two sets of cementitious blends were prepared: the first set consisted of PLC and BCSA, while the second set included PLC, BCSA and CC. In the first set, 10%, 30% and 50% of the PLC were replaced with BCSA by weight. In the second set, 30% CC was incorporated by weight in all the mixes. The remaining 70% of the cementitious material was distributed as follows for each mix: 50% PLC and 20% BCSA, 30% PLC and 40% BCSA, and 10% PLC and 60%. The BCSA proportions were chosen to provide a wide range of results, allowing for the identification of the ideal proportions for each of the blended systems. The BCSA substitution was limited to a maximum of 60%, and since the PLC + CC system has less clinker, it was hypothesised that a higher replacement rate was needed to provide a similar performance to the PLC systems. Additionally, an LC3 mix was replicated with PLC and CC, with a CC content of 30%. The selection of 30% CC content was done since it is the most typical CC content in LC3. The remaining 70% was PLC, which contains the same constituents – Portland clinker, gypsum and limestone – with slightly different proportions. The chemical composition of the materials used in this study, along with the mix designations and proportions, are shown in Tables 2 and 3.

Results and discussion

Fresh properties

Figure 1 presents the flow test results for all mixes. The BCSA mix exhibited a flow of 85%, while the PLC mix achieved a flow of 103% and was used as the reference control mix. According to ASTM C109 (ASTM, 2020b), the target flow should be approximately 110 ± 5%. Although the mixes showed lower flow values, they remained sufficiently workable for specimen casting. The lower flow in the BCSA mix compared to PLC is attributed to the faster hydration kinetics and higher water demand of BCSA. Both factors are related to rapid ettringite formation, which has been shown through rheology studies to increase the viscosity of the mix (Huang *et al.*, 2019).

The PLC-BCSA blends exhibited higher values than pure PLC, even though it was expected that their flow would fall between those of BCSA and PLC. Previous studies have shown that in PC-CSA blends, flow increases with higher anhydrite content, which,

Table 3. Mix nomenclature and proportions (BCSA, belitic calcium sulfoaluminate; CC, calcined clay; PLC, Portland limestone cement)

Mix designation	PLC: %	BCSA: %	CC: %
PLC	100	0	0
BCSA	0	100	0
90-PLC + 10-BCSA	90	10	0
70-PLC + 30-BCSA	70	30	0
50-PLC + 50-BCSA	50	50	0
50-PLC + 20-BCSA + 30-CC	50	20	30
30-PLC + 40-BCSA + 30-CC	30	40	30
10-PLC + 60-BCSA + 30-CC	10	60	30
LC3	70	0	30

in the case of the 70% PLC-30% BCSA blend, may explain the increased flow due to the additional anhydrite provided by the BCSA component (Huang *et al.*, 2019). In the 50% PLC–50% BCSA blend, however, the flow decreased, likely due to the higher water demand required for ettringite formation. Blends containing BCSA, PLC and CC showed decreased flow values attributed to higher water absorption caused by the finer particle size of the CC (see Blaine in materials section) (Moghul *et al.*, 2025). Despite these lower flow values, all mixes remained workable, and specimens were successfully cast.

Figure 2 shows the initial and final set times for the mortar mixes. The pure BCSA mix exhibited the shortest initial and final set times, while the pure PLC mix exhibited the longest. Increasing the BCSA content in both the PLC and LC3 blends resulted in progressively shorter initial and final set times.

Mechanical properties

The compressive strengths for all mixes are shown in Figure 3. Three specimens were tested at each age, with all mixes evaluated at 1, 7, 14 and 28 days. Early-age strength tests were performed for mortars containing at least 20% BCSA, while the remaining mixes could not be tested at early ages due to slower strength development. Mixes containing 30% or more BCSA were additionally tested at 1.5 and 3 h, and the mix with 20% BCSA was tested at 3 h as well. The hydration of PLC peaked between one and seven days of curing, during which mixes containing 50% or

Table 2. Chemical composition of the raw materials (X-ray fluorescence) and the calculated compositions of the mixes (BCSA, belitic calcium sulfoaluminate; CC, calcined clay; PLC, Portland limestone cement)

Cementitious material	Calcium oxide: %	Silicon dioxide: %	Aluminium oxide: %	Sulfur trioxide: %	Iron (III) oxide: %	Magnesium oxide: %	Others: %
PLC	65.8	17.8	2.9	5.6	3.5	3.0	1.4
BCSA	49.4	14.3	15.4	14.9	0.9	1.4	3.9
CC	0.1	64.8	30.3	1.0	0.9	0.2	2.7
90-PLC + 10-BCSA	64.1	17.5	4.2	6.5	3.2	2.9	1.7
70-PLC + 30-BCSA	60.8	16.8	6.7	8.4	2.7	2.5	2.2
50-PLC + 50-BCSA	57.5	16.0	9.1	10.2	2.2	2.2	2.6
50-PLC + 20-BCSA + 30-CC	44.3	30.0	13.0	6.2	2.2	1.9	2.3
30-PLC + 40-BCSA + 30-CC	40.9	29.2	15.6	8.2	1.7	1.6	2.8
10-PLC + 60-BCSA + 30-CC	37.5	28.5	18.2	10.1	1.2	1.2	3.3

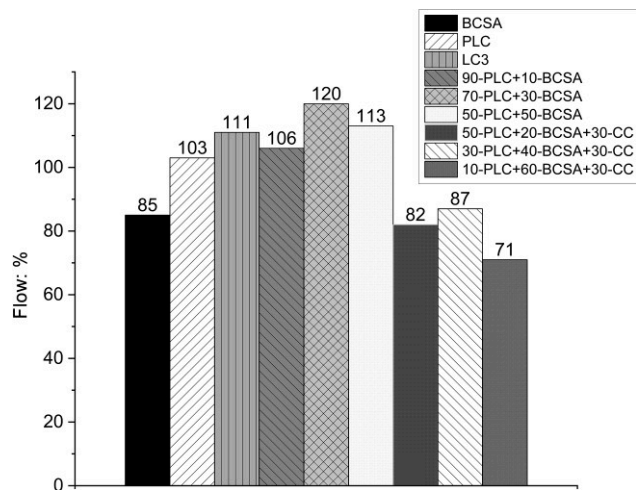


Figure 1. Flow test results for belitic calcium sulfoaluminate (BCSA), Portland limestone cement (PLC), limestone-calcined clay cement (LC3), PLC-BCSA blends and BCSA-PLC-CC blends (CC, calcined clay)

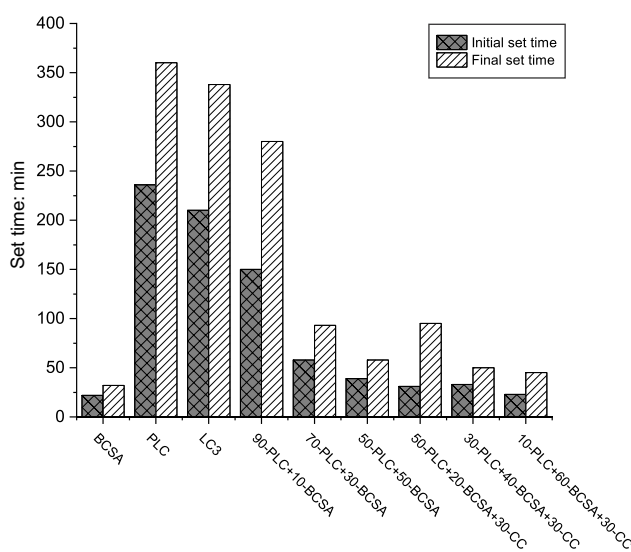


Figure 2. Setting times of belitic calcium sulfoaluminate (BCSA), Portland limestone cement (PLC), limestone-calcined clay cement (LC3), PLC-BCSA blends and BCSA-PLC-CC blends (CC, calcined clay)

more PLC achieved strength increases of 50% or greater, primarily due to the hydration of C₃S. By 28 days, blends containing 50% PLC reached compressive strengths of around 50 MPa, comparable to the strength of the pure BCSA mortar. At 1.5 and 3 h, only blends containing more than 30% BCSA showed strength gain, indicating that the addition of BCSA had a significant impact on improving the early strength of these low-embodied carbon dioxide mixes. Three blends (70-PLC+30-BCSA, 90-PLC+10-BCSA and 10-PLC+60-BCSA+30-CC) exhibited strengths above

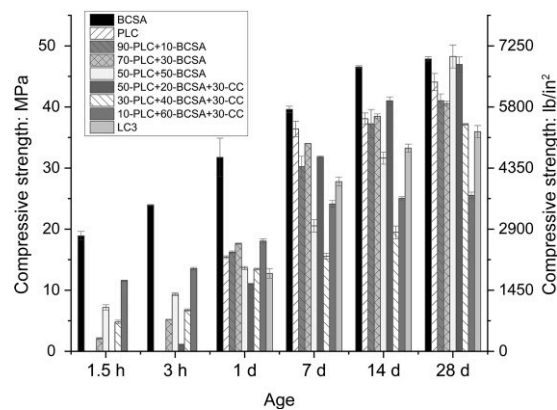


Figure 3. Compressive strength results at six different curing ages

13.8 MPa at one day, although their values were similar in strength to the reference samples. These results demonstrate that BCSA addition is beneficial when early strength development is required within the first 24 h of hydration. In the BCSA and BCSA-containing blends, early strength gain was primarily attributed to ettringite formation. In contrast, the strength development in the PLC resulted from the gradual hydration of C₃S, leading to the formation of C-S-H and calcium hydroxide (CH, Ca(OH)₂) (Hewlett and Liska, 2019). In addition, it is expected that limestone reacts with aluminates from PLC and CC to form monocarboaluminates (Ghazy *et al.*, 2025; Lothenbach *et al.*, 2008). In the PLC-BCSA blends, strength gain was a combination of both mechanisms, as demonstrated later through TGA and XRD analyses.

Shrinkage

Drying shrinkage was measured in mortar prisms at 1, 4, 7, 14 and 28 days. Three specimens were prepared according to modified C157 (ASTM, 2017) and analysed for each of the mixes. The test specimens were cured in the moulds in a moist cabinet for 24 h and then demoulded. After demoulding, the prisms were stored in a room with controlled temperature and humidity conditions. The temperature was maintained at 21°C and the relative humidity was 50%. The main product of early hydration in BCSA cement is ettringite, which incorporates a significant amount of chemically bound water (32 H₂O molecules). In contrast, the primary hydration product of PLC cement is C-S-H, which contains only three water molecules. The theoretical W/B ratio required to fully hydrate PC is approximately 0.2, meaning that any remaining free water is susceptible to evaporation, leading to shrinkage (Mather and Hime, 2002). In the case of BCSA, the water is chemically bound within the structure of ettringite (Aft), which tends to cause expansion or very low shrinkage depending on the amount of ettringite formed during hydration.

Another possible factor contributing to the difference in shrinkage behaviour is porosity (Bianchi *et al.*, 2009); however, a porosity analysis was not included in this study. Figure 4 shows the

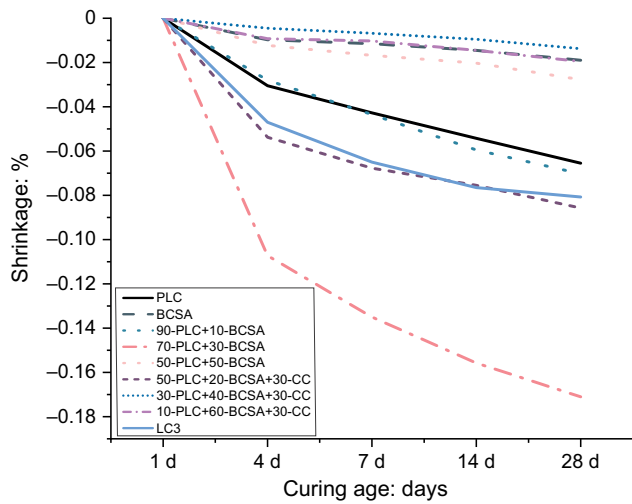


Figure 4. Drying shrinkage per modified ASTM C 157 (ASTM, 2017)

shrinkage for all the mixes analysed, including PLC, BCSA, LC3 and six blended mixes. A trend similar to that observed in the compressive strength results was noted in the shrinkage behaviour. However, the relationship between BCSA content and shrinkage was not linear. Although the BCSA mixes exhibited significantly lower shrinkage (0.019% at 28 days), it was expected that increasing the BCSA content would continuously reduce shrinkage; this trend was not consistently observed across the blends.

In the PLC and BCSA blends, the mix with the least shrinkage was the one containing 50% BCSA (0.027% at 28 days), while the mix with the highest shrinkage contained 30% BCSA (0.17% at 28 days). In the BCSA, PLC and CC blends, the mix with the least shrinkage contained 40% BCSA (0.013% at 28 days), whereas the mix with the greatest shrinkage contained 20% BCSA (0.086% at 28 days).

When comparing the pure BCSA, PLC and LC3 mixes, the shrinkage observed in the PLC (0.065% at 28 days) and LC3

(0.081% at 28 days) mixes was considerably greater than that of the BCSA mix (0.019% at 28 days). At 28 days, the PLC mix exhibited over three times more shrinkage, and the LC3 mix exhibited over four times more shrinkage, compared to the BCSA mix. The 70-PLC+30-BCSA mix (0.17% at 28 days) showed unusually high shrinkage compared to all other mixes, and this result was confirmed by a second replicate test. This mix also exhibited the highest flow, which may indicate an excess of free water contributing to the increased shrinkage.

Hydration kinetics and phase identification

The strength gain behaviour of each mix can be explained based on the calorimetry, XRD, TGA and SEM data obtained for each material (Essolebe, 2023). The calorimetry results are shown in Figures 5 and 6, revealing differences in hydration kinetics between the cement types and blends. The highest heat release from the pastes occurred within the first 3 h of hydration, during which the formation of ettringite is known to take place. The

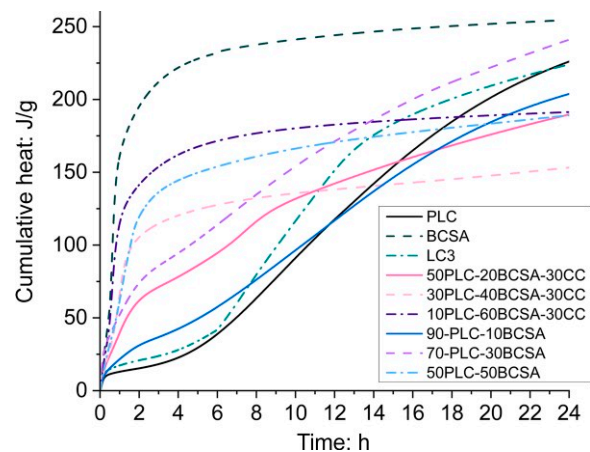


Figure 6. Cumulative heat of Portland limestone cement (PLC), belitic calcium sulfoaluminate (BCSA), limestone-calcined clay cement (LC3) and cement blends during the first 24 h of hydration

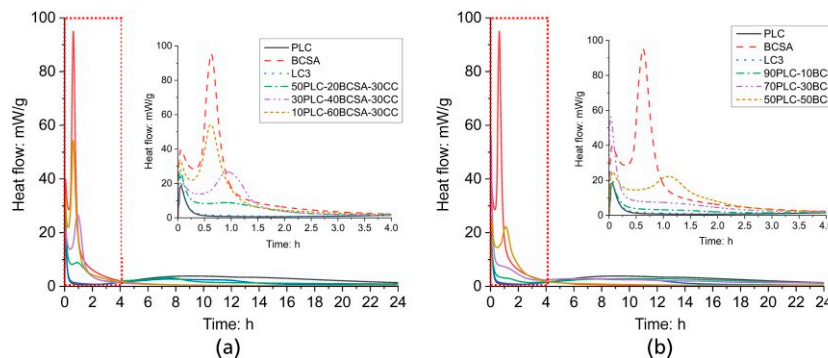


Figure 5. Heat flow of (a) limestone-calcined clay cement (LC3) and (b) Portland limestone cement (PLC) mixes compared to pure PLC and belitic calcium sulfoaluminate (BCSA) during the first 24 h of hydration, with a close-up of the first 4 h to illustrate the formation of ettringite and calcium silicate hydrate (C-S-H)

intensity of the heat release was related to the amount of ettringite formed and was directly correlated with the early strength gain (within 3 h) observed in the mixes containing BCSA. Although part of the heat release during the first 3 h was also attributable to the formation of C-S-H, the heat generated by C-S-H formation was significantly lower than that from ettringite formation. A secondary, relatively flat heat-release event observed in the PLC and LC3 samples between 6 and 20 h was associated with the hydration of calcium silicates to form calcium hydroxide (CH, $\text{Ca}(\text{OH})_2$). As shown in Figure 6, the cumulative heat release within the first 6 h for LC3 and PLC was significantly lower than that observed for the BCSA and BCSA-containing blends.

The XRD analysis is presented in Figure 7. The BCSA content was found to be directly proportional to the increased heat of hydration and strength in the PLC-BCSA blends. This suggests that the higher ettringite content in these blends was beneficial for performance.

In addition to ettringite, monocarboaluminate and hemicarboaluminate phases were observed in the blends. These phases exhibit similar characteristics to monosulfoaluminates (AFm), but with the sulfate (SO_4^{2-}) anion replaced by carbonate (CO_3^{2-}). The formation of carboaluminates indicates that at least some of the carbon dioxide released from the limestone reacted with the PLC, BCSA and CC. It is reported that the formation of monocarboaluminates is typical in modern PC cements with limestone and LC3 (Lothenbach *et al.*, 2008). Another finding was that CH was present in the PLC-BCSA systems but was only detected in the PLC-

BCSA-CC blends with the highest PLC content. It has been reported that in PC-CSA blends, CH is not present because excess aluminium hydroxide (AH_3) reacts with CH to form additional ettringite (Park *et al.*, 2024). In the PLC-BCSA system, it appears that AH_3 , which does not participate in ettringite formation, contributed to the formation of carboaluminates instead of ettringite, while CH remained present in significant quantities. Increased PLC content in the blends promoted the formation of CH. The coexistence of CH, monocarbonates and ettringite indicates that in the PLC-BCSA blend systems, hydration products competed, and their relative amounts were dependent on the PLC-to-BCSA ratio.

XRD analysis of the PLC-BCSA-CC blends revealed that the CC had not fully transformed into metakaolin during its manufacturing process (calcination) and therefore clay minerals were still present in the anhydrous blends. It also was seen that a major proportion of the clay minerals did not hydrate even after 28 days of hydration. In contrast to the PLC-BCSA blends, the PLC-BCSA-CC blends showed that the highest BCSA addition resulted in the lowest strength. Although ettringite formation in the PLC-BCSA-CC blends increased linearly with BCSA content, this did not correspond to a proportional increase in strength. A possible explanation for why the 30PLC+40BCSA+30CC and 10PLC+60BCSA+30CC mixes did not show considerable strength gains at 7 and 28 days is the lack of water available for the PLC to react with the CC, as most of the water may have been consumed by the BCSA at early ages during ettringite formation. Another factor is the high water adsorption capacity and specific surface area of CCs (Hou *et al.*, 2021; Nair *et al.*, 2020), which consume water and have

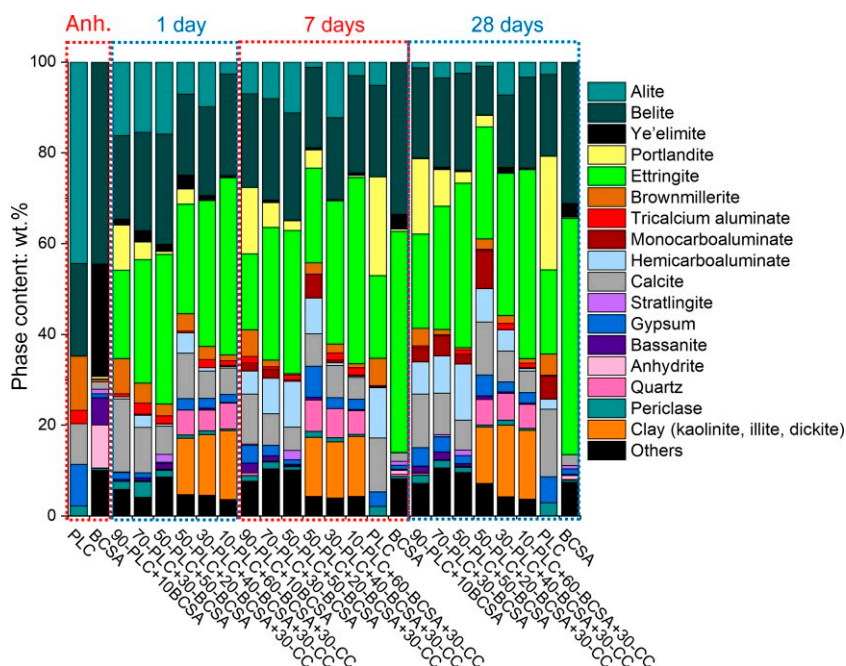


Figure 7. X-ray diffraction analysis of anhydrous Portland limestone cement (PLC) and belitic calcium sulfoaluminate (BCSA) and of hydrated mixes after 1, 7 and 28 days of hydration

been reported to cause a loss of flow in PLC-CC blends (Moghul *et al.*, 2025).

TGA was performed at 7 and 28 days, and the results are shown in Figure 8. As observed in the XRD analysis, the PLC-BCSA blend (Figures 8(a) and 8(b)) with higher BCSA content exhibited the highest amounts of ettringite. The ettringite peak appeared at around 120–140°C but partially overlapped with the C-S-H peak, which typically appears at around 110°C in PC-type cements (Trauchessec *et al.*, 2015). Monocarboaluminates identified by the XRD analysis were also detected in the TGA, appearing at around 150°C. The results for the BCSA, PLC, and CC blends are shown in Figures 8(c) and 8(d).

AH₃ was observed after seven days of hydration in the pure BCSA sample and the 60BCSA+10PLC+30CC blend. After 28 days of hydration, the 40BCSA+30PLC+30CC blend exhibited the presence of both AH₃ and carboaluminates. This suggests

that there may be a threshold CSA content below which carboaluminates and CH disappear, while ettringite and AH₃ become the dominant hydration products.

The SEM imaging analysis of a 70PLC-30BCSA sample is presented in Figure 9. The analysis was performed for hydration-stopped samples at 28 days of age. The results showed that unreacted limestone (calcium carbonate, CaCO₃) and cement phases (alite, C₃S and belite, C₂S) were present. The brightest crystals were identified as calcium carbonate, while the grey, well-defined crystals were identified as unreacted calcium silicates. In Figures 9(c) and 9(d), typical hydration products for PLC, such as plate-like calcium hydroxide, are observed along with needle-shaped ettringite originating from the BCSA.

Carbon dioxide emissions and global warming potential

Carbon dioxide emissions are generally expressed as kg of carbon dioxide equivalent per tonne. The GWP is a sustainability

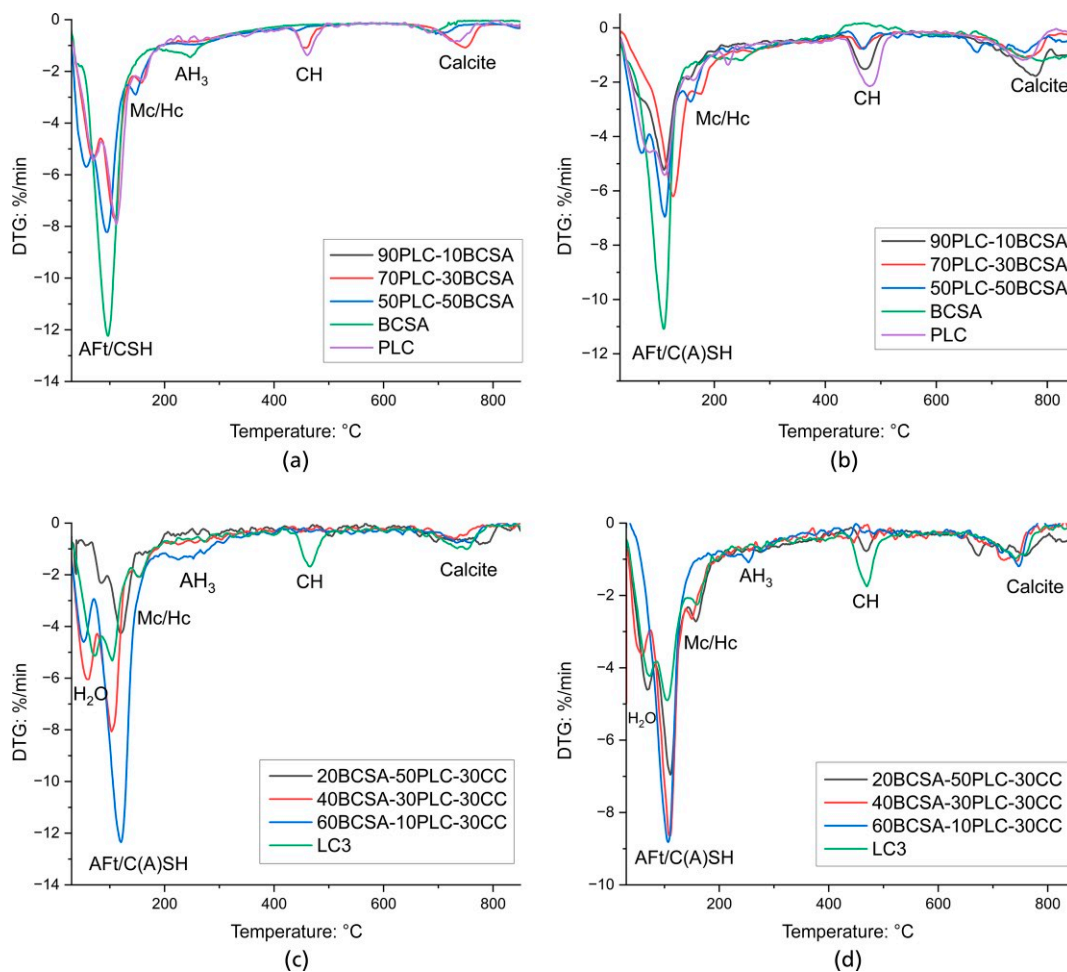


Figure 8. Thermogravimetric analysis of limestone-calcined clay cement (LC3) and Portland limestone cement (PLC) blends: (a) PLC at 7 days, (b) PLC at 28 days, (c) LC3 at 7 days and (d) LC3 at 28 days (Aft, ettringite; AH₃, aluminium hydroxide; BCSA, belitic calcium aluminosulfate; CC, calcined clay; CH, calcium hydroxide; C(A)SH, calcium (aluminium) silicate hydrate; CSH, calcium silicate hydrate; DTG, derivative mass loss percentage; H₂O, water; Mc/Hc, monocarboaluminate/hemicarboaluminate)

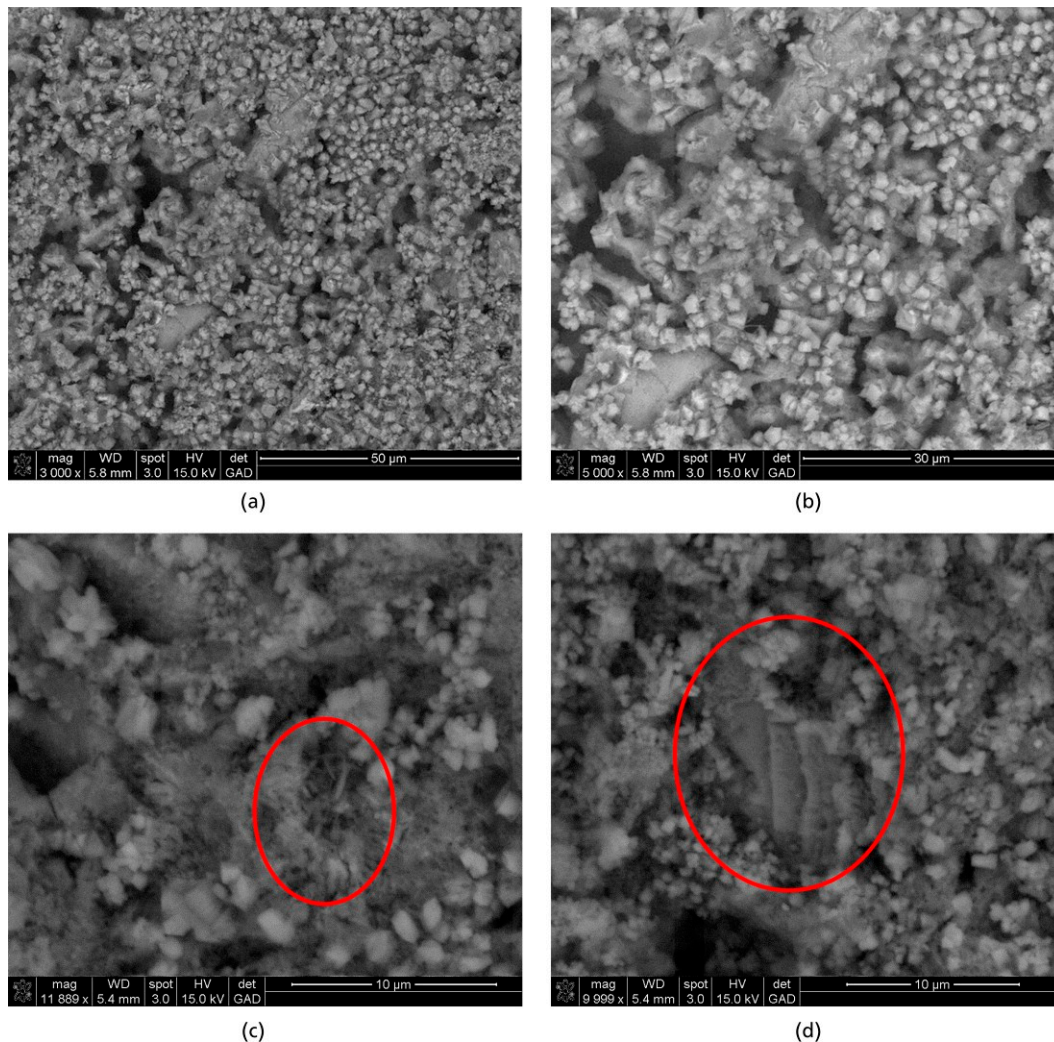


Figure 9. Backscattered electron (BSE) images of the 70PLC-30BCSA blend: (a) overall view at 3000 × magnification; (b) detailed view at 5000 × magnification; (c) ettringite needles (circled) and calcium silicate hydrate (C-S-H); and (d) plate-like calcium hydroxide (Ca(OH)₂) (circled)

metric that accounts for greenhouse gas emissions. A lower GWP means that the mix is more sustainable and has reduced carbon dioxide emissions. The environmental product declaration (EPD) of each mix component was used to calculate the total carbon dioxide emissions for each mix. Only the production stage (A1–A3) was considered. There is a published EPD for BCSA, and the industry average was used for the PLC. According to these published records, PLC releases 844 kg of carbon dioxide per tonne of cement produced (ACA, 2022), BCSA releases 673 kg of carbon dioxide per tonne produced (CTS, 2023) and CC is estimated to release 300 kg of carbon dioxide per tonne produced according to initial life-cycle analysis calculations provided by the manufacturer of the material used. Using a concrete mix design of 362 kg/m³ of cementitious material, carbon dioxide emissions were calculated for each mix. The calculated emissions for the mixes are summarised in Table 4.

Table 4. Carbon dioxide emissions per mix (BCSA, belitic calcium sulfoaluminate; CC, calcined clay; PLC, Portland limestone cement)

Mix designation	Carbon dioxide emissions: kg/m ³
BCSA	244
PLC	307
90-PLC + 10-BCSA	300
70-PLC + 30-BCSA	288
50-PLC + 50-BCSA	275
50-PLC + 20-BCSA + 30-CC	202
30-PLC + 40-BCSA + 30-CC	190
10-PLC + 60-BCSA + 30-CC	177
LC3	200

Carbon dioxide emissions are typically reduced by increasing the percentage of SCM in concrete, but it is still essential to achieve high strengths, in some cases, at an early age. Carbon dioxide emission per unit compressive strength (carbon dioxide intensity)

is a new sustainability metric reflecting cement or concrete performance. The carbon dioxide intensity can be expressed in kg/m³ of carbon dioxide equivalent per MPa of the cement blend at a given age, as shown in the equation below.

$$\text{Carbon dioxide intensity} = \frac{\text{carbon dioxide emission (kg/m}^3\text{)}}{\text{compressive strength (MPa)}}$$

The calculated carbon dioxide emissions for each mix are shown in Table 4. The carbon dioxide intensities were calculated as the carbon dioxide emissions divided by the compressive strength value for any time of interest, since it is not only important to have low carbon dioxide emissions but also equally important to provide adequate early strengths that will result in savings due to reduced construction windows. This concept is particularly important if the primary reason these binders are used is their early strength, since the carbon dioxide footprint is an economic incentive. Figure 10 shows a summary of the carbon dioxide intensities for all mixes at 1 day and 28 days. It is important to note that the emissions used correspond to an equivalent concrete mix while the strength used to calculate the carbon intensity were obtained from the mortar specimens.

The BCSA mix had the lowest carbon dioxide intensity at 1 day, but three alternative blends provided an equal or lower intensity at 28 days. The 50-PLC+50-BCSA, 50-PLC+20-BCSA+30-CC and 30-PLC+40-BCSA+30-CC mixes had intensities under 5.7 kg/m³ of carbon dioxide equivalent/MPa. This means that there are four concrete mixes with low embodied carbon dioxide that can potentially be used in the road-paving industry and that provide high early-age strength, which is beneficial when dealing with shorter construction and closure times.

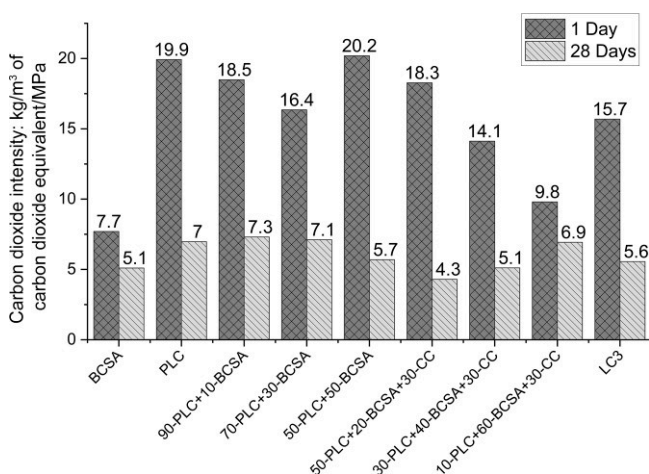


Figure 10. Carbon dioxide intensities for concrete mixes based on compressive strengths at 1 day and 28 days (BCSA, belitic calcium sulfoaluminate; CC, calcined clay; LC3, limestone-calcined clay cement; PLC, Portland limestone cement)

Figure 11 shows the carbon dioxide intensities of the mixes for which compressive strength was tested at 1.5 and 3 h, as it is important to evaluate the carbon dioxide intensity of concrete mixes that develop high early-age strength. Having the ability to provide high early-age strength while also having low embodied carbon dioxide allows several of the concrete mixes to be considered as strong alternatives to the traditional concrete mixes used in the road-paving industry. In particular, it was observed that the BCSA concrete mix and the 10-PLC+60-BCSA+30-CC exhibited a lower carbon dioxide intensity when analysed for compressive strength at 1.5 h, 3 h and 1 day. Both of these mixes presented a carbon dioxide intensity lower than 15.3 kg/m³ of carbon dioxide equivalent/MPa as early as 1.5 h. These values further decreased when performing the analysis at three hours and one day.

Conclusions

Blending BCSA with PLC and LC3 can enhance the physical properties of these low carbon dioxide binders while simultaneously reducing their carbon dioxide emissions. The addition of PLC and CC resulted in decreased early heat of hydration and slower strength development compared to pure BCSA. However, the 50PLC-50BCSA and 50PLC-20BCSA-30CC mixes achieved compressive strengths after 28 days of curing comparable to those of the pure BCSA and PLC mortar samples. TGA and XRD analyses showed that calcium hydroxide was favoured in blends with higher PLC content, while AH₃ was more prevalent in blends with higher BCSA content. SEM analysis revealed that both ettringite and calcium hydroxide were present in the 70% PLC-30% BCSA blend after 28 days of hydration. Unreacted limestone was clearly observed; however, the TGA and XRD results indicated that at least some of the limestone reacted in both the BCSA-PLC and BCSA-LC3 blends, leading to the formation of carboaluminates.

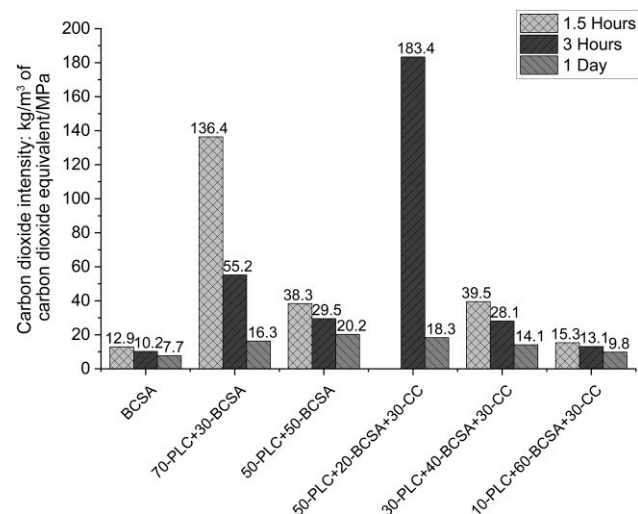


Figure 11. Carbon dioxide intensities for concrete mixes based on compressive strengths at 1.5 h, 3 h and 1 day (BCSA, belitic calcium sulfoaluminate; CC, calcined clay; LC3, limestone-calcined clay cement; PLC, Portland limestone cement)

This study demonstrated that carbon dioxide intensity, measured as emissions per unit of compressive strength, can be significantly reduced with BCSA blends, presenting them as sustainable alternatives for concrete production. Mixes exhibiting early strength development and carbon dioxide equivalent intensities per MPa at 28 days comparable to BCSA ($\leq 5.15 \text{ kg/m}^3$) included 50PLC-50BCSA, 50PLC-20BCSA-30CC and 30PLC-40BCSA-30CC. These three mixes show strong potential for application in the road-paving industry.

However, the deviation in shrinkage behaviour observed in the 70PLC-30BCSA mix could not be fully explained based on the results of this study. This highlights the need for further investigation into the non-linear behaviour of compressive strength and shrinkage in these new cementitious blends, as well as their performance under real-world conditions through field testing. Notwithstanding this non-linearity, the addition of BCSA to LC3 or PLC improves early strength and decreases GWP and carbon dioxide intensity.

In the future there are aspects that still need to be studied, such as the causes of the non-linear behaviour in compressive strength and shrinkage, which seem to be related to the ettringite content; strength testing of concrete specimens; the GWP of the concrete mixes; and, lastly, testing these alternatives in a pilot project to assess the durability and field performance of the new blends.

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