Chapter 29-2. Reutilization of ceramic waste as supplementary cementitious material

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Abstract

La producción de cemento Portland (CP) tiene el mayor impacto ambiental entre los diferentes componentes del hormigón. Asimismo, pese a que los productos cerámicos son consumidos en todo el mundo y constituyen una fracción significativa de los residuos de construcción y demolición, cantidades significativas de residuos cerámicos (RC) son simplemente depositadas en vertederos o utilizadas como sub-bases de carreteras. Este estudio analizó investigaciones previas sobre la reutilización y valorización de RC como material cementante suplementario en conglomerantes de CP, cemento de aluminato de calcio, $Ca(OH)_2$ y yeso. La mayoría de los estudios investigaron el uso de los RC como adición puzolánica en sistemas de CP y, pese a que la reactividad varió en función del tipo de RC utilizado, en general se obtuvieron propiedades satisfactorias (físicas, mecánicas y durabilidad) al sustituir entre un 10-30 % en peso del cemento. Los conglomerantes RC/CP contribuyen a la economía circular y a un desarrollo sostenible, permitiendo reducir tanto las cantidades de residuos depositados en vertederos, como las emisiones de CO_2 y el consumo de recursos naturales y energía asociados a la producción de CP.

Keywords

Ceramic waste; pozzolanic activity; microstructure; strength activity index; durability; sustainability.

List of abbreviations

Ceramic waste (CW); ceramic waste powder (CWP); brick ceramic waste (BCW); ceramic sanitary ware (CSW); tiles ceramic waste (TCW); ceramic waste from polishing tiles (PTCW); construction and demolition waste (CDW); supplementary cementitious material (SCM); Portland cement (PC); calcium aluminate cement (CAC); fly ash (FA); blast furnace slag (BFS); silica fume (SF); metakaolin (MK); strength activity index (SAI); weight percentage (wt.%).

1. Introduction

Rapid global population growth has significantly increased the demand for buildings and infrastructure and led to unsustainable development, due to extremely high demand for natural resources and energy. Concrete is an essential construction material with a worldwide production of approximately 13 billion tons per year [1]. Its high PC content implies using large amounts of raw materials (1.7 tons of rocks per ton of clinker) and energy (3.1–3.8 GJ of heat per ton of clinker), and emitting significant CO_2 emissions to the atmosphere (530-940 kg of CO_2 per ton of clinker) [2–5]. According to Lasseuguette et al. [6], 95% of the concrete CO_2 emissions are attributed to PC and are originated mostly during $CaCO_3$ decomposition (approximately 60%) and the combustion of the fuels required for the clinkerisation process. There are other emissions attributed to transportation and the electricity required to mill raw materials and clinker [5,7]. In fact 5% to 7% of the world's overall CO_2 emissions are attributed to cement production [8,9]. According to the review by Nwankwo et al. [9], world's PC manufacture rose from 0.94 billion tons in 1970 to 4.1 billion tons in 2018, and is expected to rise by 45% by 2050.

In this context, the transition to an efficient circular economy would significantly contribute to sustainable development by diminishing ecological, economic and social impacts [10,11]. The valorisation of CW to develop more sustainable construction binding materials would promote the conservation of natural resources, minimise the waste deposited in landfills and contribute to develop more ecological and sustainable concrete that is more environmental-friendly [3,7,12,13]. As explained by Jaskulski et al. [4], although some alternatives allow the heat used during the clinkering process to lower, limestone is an essential raw material that cannot be reduced. Consequently, using CW as a pozzolanic admixture contributes to minimise the energy use and carbon footprint associated with PC production. Additionally, this implies significant cost benefits [3,12-17] because, as pointed out by Jain et al. [7], although PC occupies only 10-15% volume in concrete, it approximately implies 45% of concrete's cost.

This paper reviews the use of CW as supplementary cementitious material (SCM) in PC, calcium aluminate cement (CAC), $Ca(OH)_2$ and gypsum systems. Figure 1 summarises the bibliographic search, which analysed the works developed between 2010 and 2022. Only CW studies were selected (research using CDW or concrete waste was not included). Of the 193 bibliographic references found, 180 were research articles, two were book chapters and the remaining 11 were reviews. After reading

them all, the most meaningful and significant results on the use of CW as SCM were included.
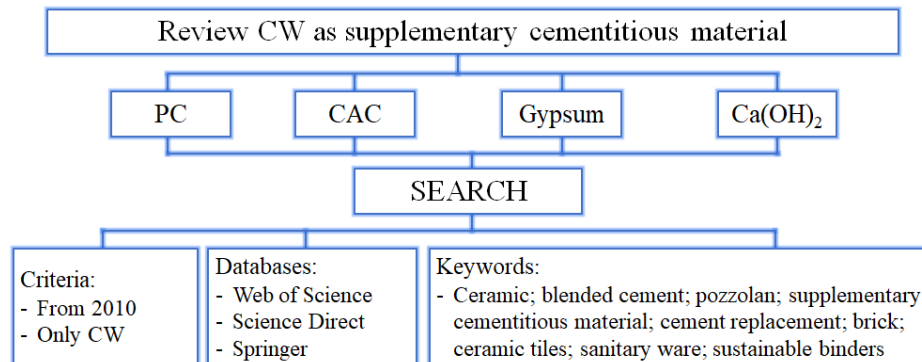

Figure 1 – Review on the use of CW as SCM.

2. CW chemical properties

CW chemical properties depend mainly on the raw materials and production process followed to manufacture the ceramic product, which determines its microstructure and mineralogy [16,18]. Although the pozzolanic activity of the clay minerals is generally low, they may activate while sintering ceramic products due to an amorphisation of their structure [5,9,19]. When sintering ceramic materials [20], clay dehydration (20-200ºC) is followed by a dehydroxylation process, during which OH^- groups in clays are lost, which results in unstable silica and alumina with a disordered structure. This amorphisation process may occur at a wide range of temperatures (350-900ºC) depending on clay minerals' composition [4,5]. Decarbonation of calcites or dolomites generally occurs from 700ºC to 900ºC and, if temperature continues to rise, recrystallisation occurs, leading to the formation of new crystalline stable phases. According to Jaskulski et al. [4], the optimum temperatures to activate clays should be high enough to destroy the structure of clays, but not be excessive to avoid melting minerals and leading to the formation of new chemically inert crystalline stable phases. However according to some studies [17,21], at above 900ºC new amorphous phases may also form due to the partial fusion of crystalline phases. In short, the pozzolanic activity of a particular CW very much depends on raw clays' composition and the sintering process employed to fabricate the ceramic product. As reviewed by Mohammed [5], as the aim of manufacturing ceramic products is to achieve certain properties for a specific use, the sintering process generally differs from the optimal one that should be applied to achieve maximum amorphization of the calcined clay. In line with this, Pavesi et al. [11] investigated the pozzolanic activity of three different clays sintered at temperatures within the 400-1000ºC range. They observed that, although the developed mortars' compressive strength was influenced mainly by the calcination temperature, a general optimum temperature could not be established because it depended on clay's mineralogical

composition. Figure 2 shows milled CW powders that has been previously used as SCM. These particles were usually irregular and angular, and had mean particle sizes close to that of PC.
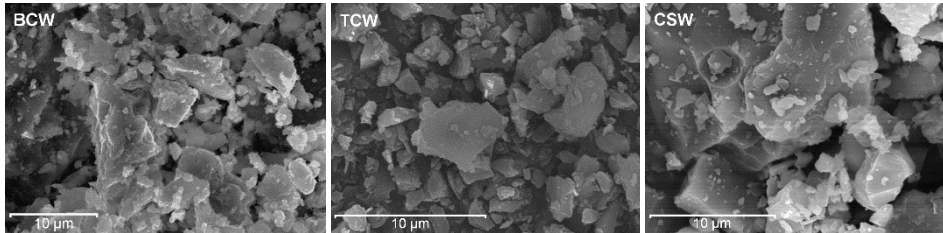

Figure 2. Milled CW particles obtained from bricks, tiles and sanitary ware.

Several studies reported the mineralogical composition of different CW materials used as SCM [1,2,16,20-25]. Brick ceramic waste (BCW), tile ceramic waste (TCW) and ceramic sanitary ware (CSW) were the main employed CW types, and with widely varying chemical and mineralogical compositions depending on clay composition and the sintering process. Although some differences were observed among the X-ray diffraction (XRD) spectra, quartz (SiO_2), mullite ($Al_6Si_2O_{13}$) and feldspars, such as microcline ($KalSi_3O_8$), anhortite ($CaAl_2Si_2O_8$) or albite ($NaAlSi_3O_8$), were the main crystalline phases generally identified in CW materials. The chemical analyses run in diverse CW material types [1,3,9,10,13,14,16,20-23,25,26–35], showed that SiO_2 and Al_2O_3 were the main identified compounds, and significant amounts of CaO, Fe_2O_3, Na_2O or K_2O may exist depending on the specific CW type. Several studies [11,17,19,36] have concluded that the pozzolanic reactivity of CW materials is influenced mainly by particle fineness, the type and amount of the original clays, and the temperature and duration of the sintering process (the last two determine the amount of the available amorphous silica and alumina to react). Of them, only the particle size of CW may be reduced because chemical and mineralogical compositions depend on the ceramic product manufacturing process. The mean particle size when used as a pozzolan generally came close to that of PC particles [2,16,20,21,23]. Regarding chemical composition, all the CW materials included in this review met both specifications ASTM C618-19 and UNE-EN 450-1:2013 for pozzolanic materials, with (SiO_2+Al_2O_3+Fe_2O_3) content higher than 70% and loss on ignition (LOI) generally being lower than 5% (maximum for category A fly ash (FA), according to UNE-EN 450-1:2013) [5,6,37]. This review allowed us to conclude that the relatively large amounts of SiO_2 and Al_2O_3 of the CW materials, together with the amorphous phases that originated when sintering ceramic products (during dehydroxylation or vitrification), confer them pozzolanic activity, especially at later curing ages.

3. Ceramic waste as supplementary cementitious material

Most of the research conducted on using CW as supplementary cementitious material (SCM) has focused on partial PC replacement. Only four studies were found in which CW was used in $Ca(OH)_2$ systems [17,38-40]; one were BCW was employed as pozzolan in gypsum and hydrated lime ternary systems [41]; and another in which CW from thermal insulating bricks was used to replace 5% and 15% calcium aluminate cement (CAC) [42]. Some studies were found in which CW was simultaneously used as both pozzolan and a recycled aggregate [27,43,44], but no studies were observed that combined different CW types as a pozzolanic admixture.

This review focused on the works that used CW to partially replace PC. In these studies, CW was generally obtained from bricks (BCW), tiles (TCW), sanitary ware (CSW) and polishing tiles (PTCW), and up to 30-40 wt.% PC was usually replaced. The commonest properties of PC/CW compounds are summarised in the following sections.

3.1. CW in PC/CW blended systems

Pozzolans are siliceous or silico-aluminous materials that, despite having little or no agglomerating power on their own, when finely ground may react with $Ca(OH)_2$ in the presence of water to form binding compounds [4,9,19,36]. In PC systems, the reaction between SiO_2 and Al_2O_3 in SCM, and the $Ca(OH)_2$ released during primary PC hydration, provide secondary hydration products, such as C-S-H gel, C-A-H, C-A-S-H and, in the presence of CO_2 or $CaCO_3$, carboaluminates [4-6,9]. This densifies the binding matrix by reducing the system's porosity and permeability, and generally enhances its durability and strength.

Different methods are usually combined to assess the pozzolanicity of CW materials [4,5]. These procedures may be: i) chemical, such as Frattini tests [18], pH or electrical conductivity measurements in $Ca(OH)_2$ and pozzolan suspensions [16,36], and saturated lime tests [18]; ii) based on mechanical properties, such as determining the evolution of compressive strength and strength activity indices (SAI) [18]; iii) microstructural studies, performed by field emission scanning electron microscopy (FESEM), XRD, Fourier-Transform Infrared Spectroscopy (FTIR) or thermogravimetric (TG) analyses [12,16,45,46]; iv) calorimetric tests that evaluate the heat released during hydration reactions.

3.2. Properties of PC/CW compounds

3.2.1. Workability and setting time

According to different reviews conducted on using CW as partial PC replacement [8,14,34], workability of pastes, mortars and concrete generally reduce with increasing CW contents [26–28,30,32,47-51]. However, some studies have observed minor variations [2], or even marked improvement with increasing amounts of CW [16,35,52–54]. In this sense, minor variations in consistency were observed by Pitarch et al. [20] when replacing PC with TCW, a slight improvement was recorded with CSW and a significant reduction with BCW, which was attributed to these CW

materials' different water absorption. Similarly, Zito et al. [1] observed that workability diminished when replacing 24 wt.% PC with hollowed bricks, and slightly varied or improved with the same amount of mud bricks or CSW. In the study by Jackiewicz-Rek et al. [50], the lower consistency recorded when increasing CSW contents was attributed to the smaller particle size of CW powder, whose bigger specific surface implied considerable water absorption and higher water demand.

Workability variation can be compensated by superplasticiser addition. In fact some studies have either added superplasticiser or modified the water/cement ratio to keep workability constant, especially in concrete [8,14]. In the study by Ngoc-Tra Lam et al. [27], the amount of water required to keep a constant consistency increased with BCW addition, and mortars' workability (flow spread) reduced when replacing either PC with BCW or natural sand with CW, which was attributed to the greater water absorption and the rough surface and angular shape of CW. A similar trend was observed by Barreto et al. [36] when replacing 10 and 20 wt.% PC with BCW in structural concrete because to keep workability constant, the water/binder ratio increased from 0.49 in the reference sample up to 0.61 with 20 wt.% CW. In SCC, which is placed without vibration and requires high fluidity and resistance to segregation, authors like El-Dieb et al. [55] reported a reduction in flowability and an improvement in resistance to segregation with increasing PTCW contents (up to 60 wt.%).

Similarly to workability evolution, some authors have observed that CW accelerates setting times [27], while others have reported no significant variations in setting times with increasing CW additions [56], and other research works indicated that CW retarded hydration [20,24]. Ngoc-Tra Lam et al. [27] observed a significant reduction in setting times with increasing BCW contents. Indeed the initial setting time (IST) varied from 155 min in the reference paste to 65 min with 40 wt.% BCW, and the final setting time (FST) dropped from 205 min for the reference paste up to 160 min when replacing up to 40 wt.% PC. According to Agrawal et al. [14], hydration in PC/CW blended systems depends on the characteristics of the particular employed CW. Pitarch et al. [20] noted that the IST slightly prolonged with increasing CW contents and the FST was slightly shorter with BCW or TCW (up to 15 min with 50 wt.% PC replacement), but prolonged with CSW (up to 35 min with 50 wt.% CSW). In the study by Pereira and Camarini [32], who developed self-levelling mortars (SLM) by replacing 15, 25 and 50 wt.% PC with electrical porcelain insulators and red ceramic from CDW, the influence on hydration also depended on the type and amount of CW. So 15% red ceramic accelerated hydration reactions, and minor variations were observed with larger amounts of this waste or with up to 25% porcelain waste. Zhao et al. [28] investigated the influence of particle size when replacing 30 wt.% PC with BCW. These authors found that the coarsest CW particles led to similar setting times to the reference paste, and these times were shorter with finer particles.

3.2.2. Compressive strength. The Strength Activity Index

Although some authors have found that compressive strength generally reduces with increasing CW contents, especially for short curing ages [1,11,12,36,33–35,45,57,58], some studies show similar or higher strength values than the reference sample [7,14,15,27,34,53,55,59,60], and other reported optimum replacement percentages, generally within the 5 to 30 wt.% range [7,14,15,27,34,53,55,60]. These variations are attributed mainly to differences in: i) the sintering processes applied when manufacturing ceramic products; ii) the mineralogical compositions and physico-chemical properties of the original clays; iii) the particle size distributions of milled CW powders. This is graphically observed in Figure 3, adapted from Agrawal et al. [14], which shows compressive strength evolution in mortars (4a) and concrete samples (4b) cured for 28 days and prepared by replacing different amounts of PC with CW.
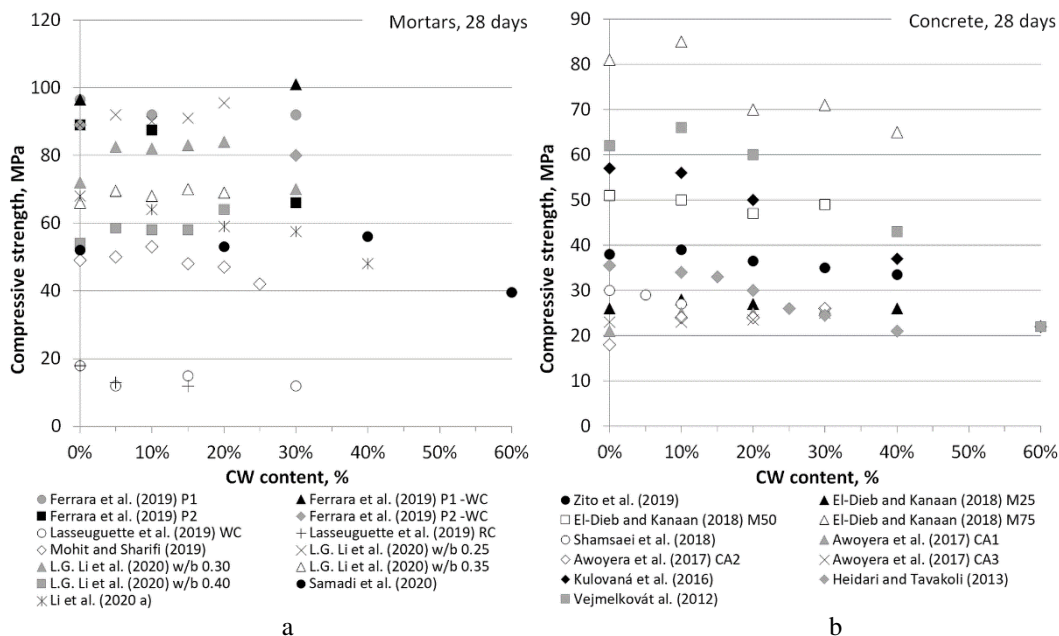

Figure 3 – Compressive strength of PC/CW compounds cured for 28 days: a) Mortars; b) Concrete. Adapted from Agrawal et al. [14].

Although most of the studies that have used CW as a pozzolanic admixture have reported lower strength values than the reference sample for short curing ages, greater strength developments have been generally observed for later curing periods. In their review, Guedes de Paiva et al. [34] attributed the early age compressive strength reduction to the immature pozzolanic reaction of CW materials, which retards strength development. Figure 4, adapted from Pitarch et al. [20], illustrates how the SAIs (SAI:

ratio between the strength of the pozzolanic sample and the reference sample) generally improved with curing time, especially at lower replacement ratios. SAIs close to 100% were obtained in [20] from 28 curing days when replacing up to 20-30 wt.% PC. This trend was similar to that indicated in other studies [2,15,16,20,25-27, 37,52,53,55,60-63], with SAIs even higher than 100% for later curing periods. Consequently, the strength differences between the pozzolanic and reference samples generally reduced with curing time. In line with this, authors like Mas et al. [16] concluded that the requirements set out in UNE EN 450-1:2013 for other pozzolanic materials, such as FAs (SAI higher than 75% and 85% after 28 and 90 curing days, respectively), were met with up to 35 wt.% TCW [16]. As plotted in Figure 4, Pitarch et al. [20] also observed that up to 90 curing days, BCW exhibited slightly better pozzolanic activity than TCW and CSW, so that UNE EN 450-1 requirements were met with up to 25 wt.% CSW or TCW, and with up to 35 wt.% BCW. Further studies [2] concluded that 35 wt.% CSW mortars can be interesting for applications that do not require fast strength development because they give higher SAI values than 85% as of 90 curing days, with a strength reduction of only 10% when cured for 180 days. The strength gain values recorded in all three studies [2,16,20] corroborated that the pozzolanic activity of these CW materials was significant after 28 curing days.
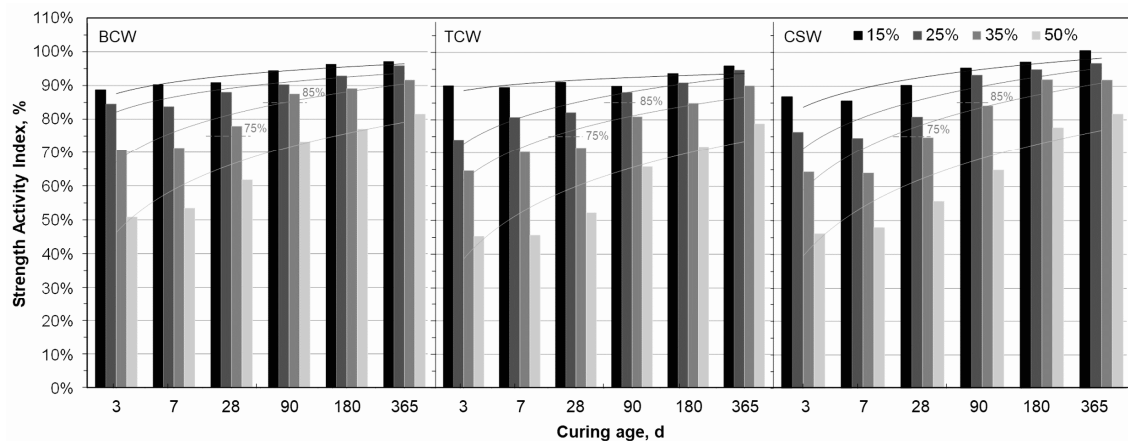

Figure 4. SAI evolution with the curing age of the PC/CW mortars developed with 0 to 50 wt.% BCW, TCW and CSW. Adapted from reference [20].

Only three studies were identified that have compared the pozzolanic activity of different types of CW materials under the same conditions [6,20,49]. Lasseuguette et al. [6] used two different TCW types (glazed white and unglazed red ceramic) to replace up to 30% PC. These authors observed that, although similar SAIs were obtained when replacing 5% PC, significant differences were found with 15% PC substitutions (the best results were fore the 15 wt.% white ceramics, which provided a SAI close to 100% after 56 curing days). Pereira-de-Oliveira et al. [49] compared the pozzolanic activity of glass waste, BCW and TCW ground to three different particle

sizes. Although no significant strength differences were observed for a given particle size and CW percentage, the TCW mortars presented better strength evolution with curing time than those prepared with BCW, which did not reach the minimum SAI set out in UNE EN 450-1:2013. These results are contrary to those reported by Pitarch et al. [20], who observed slightly better SAI values in BCW than in TCW and CSW. These studies corroborate that CW's pozzolanic activity very much depends on the ceramics production process.

The research conducted by Reiterman et al. [51] compared the pozzolanic activity of CW, FA and blast furnace slag (BFS), and recommended maximum PC replacement percentages of 12.5 wt.%, 37.5 wt.% and 50 wt.%, respectively. This denotes that, compared to other commonly used pozzolanic materials, the CW used in their study exhibited moderate reactivity.

Of the studies that simultaneously used CW as pozzolan and as a recycled aggregate [27,43,44], Samadi et al. [44] obtained similar strength values to the reference mortar (close to 50 MPa after 28 curing days), regardless of curing age, in the samples prepared by replacing 40 wt.% PC and 100 wt.% of natural sand with TCW. In the study by Awoyera et al. [43], compressive strength generally reduced compared to the reference concrete, but the mixture prepared by replacing 30 wt.% PC with TCW and natural gravel with 20 wt.% recycled concrete plus 75% TCW, gave similar strength results (close to 26 MPa after 28 curing days). Ngoc-Tra Lam et al. [27] obtained similar strength values to the reference mortar when combining the replacement of 50 or 100 wt.% river sand with CW aggregates (CWA), plus 10 wt.% PC with BCW. Although up to 28 curing days the mortars developed by replacing PC with BCW generally exhibited lower compressive strength values than the reference (48 MPa), this tendency reversed after 90 curing days, and similar or higher strength values than the reference (55 MPa) were recorded with up to 20 wt.% BCW (72 MPa in the samples that combined 100 wt.% CWA with 10-20 wt.% BCW). It is highlighted that the mortars prepared by replacing natural with CWA always provided better compressive strength results no matter what the curing time (the 7-day values fell within the 20-33 MPa range with natural aggregates, which rose to 28-40 MPa and 33-43 MPa with 50 wt.% and 100 wt.% CWA, respectively). This was attributed to particles' angular shape and rough texture, which improved the interfacial transition zone. Gautam et al. [18] noted a similar trend when replacing up to 30 wt.% PC with bone china CW and 20 to 40 wt.% of sand with granite cutting waste in self-compacting concrete. The optimum amount of CW powder (CWP) when replacing only PC was 10 wt.%, and compressive strength improved when combining CWP with recycled granite sand (optimum with 10 wt.% CWP, plus 30 wt.% recycled fine aggregate).

Some studies also combined CW with other SCM to investigate the synergy between them [3,29]. Heidari et al. [3] compared the pozzolanic activity of TCW and CSW (5 to 15 wt.%) in concrete samples prepared with and without metakaolin (MK, 5% or 10%) and nanosilica (0.5 or 0.8%). These authors observed better compressive strength results with CSW, and the pozzolanic reactions were significant after 28 curing days. Nanosilica and MK improved strength and reduced water absorption. The

effect of nanosilica was significant at early curing ages, as was that of MK at older ages. Karthikeyan and Dhinakaran [29], who combined 5 to 15 wt.% TCW with 1% silica fume (SF), reported the best compressive strength values with 15 wt.% TCW (59.85 MPa), which slightly improved with 1% SF (60.98 MPa).

3.2.3. Density, porosity, water absorption and drying shrinkage

Fresh and hardened densities of CW/PC systems decreased with increasing amounts of CW [14,15,36,54,60,64]. This was attributed to the lower density of ceramic particles compared to PC. Although the porosity evolution varied according to each particular study, pore refinement occurred in them all. Moreover, the pozzolanic reaction between mineral admixtures and the $Ca(OH)_2$ released during PC hydration generally reduced concrete permeability [65]. El-Dieb et al. [55] observed smaller amounts of permeable pores with increasing PTCW contents in samples cured for 90 days, which implies lower water mobility inside concrete. The lower porosity and median pore sizes recorded with increasing amounts of PTCW denoted microstructure densification, with a good correlation with improved durability. Heidari et al. [3] and Zito et al. [25] also found lower water absorption values with increasing CW contents, which is expected to improve durability.

Chen et al. [12] reported that capillary water absorption significantly reduced with 10 wt.% CWP, and progressively increased with further amounts of CW, up to 40 wt.%, which gave similar values to the reference concrete. In the study by Karthikeyan and Dhinakaran [29], although the water absorption of the sample containing 10% TCW was similar to that of the reference concrete (1.25%), it decreased with increasing CW contents (0.88% with 15% TCW) or with the addition of 1% SF (0.72% with 14% TCW, plus 1% SF). This was attributed to the formation of secondary hydration products that densified the microstructure.

Drying shrinkage generally reduced in the study by Alsaif [8] with increasing CW contents. Thus the mortars containing up to 20 wt.% CSW obtained lower shrinkage values than the reference sample.

3.2.4. Microstructural analyses by TG, XRD, FTIR or FESEM

Several studies confirmed with microstructural studies the weak pozzolanic activity of CW materials for short curing ages, which improved with curing time and became significant after 28 curing days, compensating for PC dilution [1,25,28]. TG, XRD, FTIR or FESEM analyses were combined to assess the portlandite consumption by pozzolanic reactions and to investigate the evolution of hydration products, which were similar in the reference and blended pastes [1,2,4,6,12,14,16,18,30,44-46,58,61].

$Ca(OH)_2$ dehydration generally falls within the 440-600ºC range on TG curves (variation depending on TG test conditions) [2,5,12,16,18,30,31,45,46], and differences between the mass loss recorded in the reference and the CW blended pastes allowed the lime fixed by pozzolanic reactions to be quantified [2,5,16,20]. So, the negative fixed lime values recorded at short curing ages (generally up to 7 curing days) in studies like those by Mas et al. [16] or Reig et al. [2] indicated accelerated PC hydration due to a particle effect, while the positive values obtained after 28-90 curing

days corroborated the pozzolanic reaction of CW materials [2,16]. As highlighted in [31,58], in those systems containing too much CW, PC dilution is excessive and leads to small amounts of available portlandite to react with the pozzolanic admixture.

Different studies agree that, at early ages, CW mainly acts as a microfiller, and makes a minor contribution to the binder's compressive strength, but provides additional nucleation sites that facilitate PC hydration and help to increase the system's density (particle effect). At larger curing periods, pozzolanic reactions result in new hydration products that contribute to strength development by refining pores, and densifying and homogenising the microstructure [12,26,31,46,48,54,61,62]. In fact Kulovaná et al. [15], who observed no significant reduction in concrete compressive strength with up to 40 wt.% BCW, attributed this behaviour to a filler, plus a pozzolanic effect, which compensates for lower PC contents. Samadi et al. [44], who replaced up to 60 wt.% PC and up to 100 wt.% sand with TCW, also observed how the pozzolanic activity of TCW improved the microstructure and reduced porosity (bigger amounts of C-S-H gel), especially for longer curing periods.

Other studies have corroborated these effects by analysing released accumulative heat. Zito et al. [25] observed that, although PC dilution in pozzolanic systems delayed PC hydration, the nucleation effect provided by ceramic particles partially compensated PC replacement [25]. Consequently, several studies have observed higher PC hydration rates at early ages [26,45,48]. De Matos et al. [48], who investigated the early age behaviour of PC pastes containing up to 30 wt.% TCW, noted how CW particles promoted crystallisation by improving the hydration kinetics and leading to further hydrates formation (i.e. ettringite and portlandite). The heat flow peak and cumulative heat of a 10 wt.% TCW paste was comparable to the reference and reduced with further CW additions. This proved that, up to 7 curing days, the CW particle effect, which provides extra surfaces for the nucleation and growth of hydrated products, was unable to compensate PC dilution. Kulovaná et al. [15] have also reported a progressively reduced heat flow when increasing CW contents, with similar results to the reference sample with up to 20 wt.% CW, plus a significant reduction with 40 wt.%. In the study by Zhao et al. [28], the hydration heat of the pastes containing 30 wt.% BCW also reduced compared to the reference one, and lower ratios with coarser BCW particles were recorded. These studies corroborate that the strength reduction generally observed when replacing more than 30-40 wt.% PC is mainly attributed to PC dilution, with lower $Ca(OH)_2$ available for pozzolanic reactions [30].

3.2.6. Durability: alkali–silica reaction, freeze–thaw cycles, chloride penetration, carbonation and sulphate attack

As stated by Zito et al. [1], extending a structure's service life significantly contributes to sustainable development. In line with this, the reviews conducted on CW reutilisation to partially replace PC [8,9,13,14] agree that durability generally improves when replacing PC with CW because pozzolanic reactions densify the binding matrix and refine the pore structure, which lead to a more discontinuous pore network. Authors like Kulovaná et al. [60] have reported adequate durability

performance (resistance to carbonation, frost, de-icing salts, chemical resistance to $MgCl_2$, NH_4Cl, Na_2SO_4 and HCl) in concrete samples containing up to 20 wt.% BCW. Sánchez-de-Rojas [22], who investigated the durability of PC cements containing 20 wt.% TCW (freeze-thaw and chemical resistance $SO4^{2-}$, Cl^-, and seawater environments), have also concluded that the pozzolanic reaction of TCW, and the consequent formation of secondary hydration products, enhances the chemical resistance and durability of blended pastes. The improved ultrasonic pulse velocity by adding CW recorded by Arif et al. [54] denotes higher concrete density, homogeneity and uniformity. Similarly, Boukhelkhal et al. [52] have also reported an improvement of this property with curing time, especially in pozzolanic mortars and, although values generally lowered with increasing CW contents up to 28 days, after 56 curing days the ultrasonic pulse velocity results obtained with up to 25% CW came close to or exceeded that of the reference mortar. Binding matrix densification also improves protection against corrosion by reducing the mobility of ions [9]. As reviewed by Alsaif [8], the amount of permeable pores generally reduces with increasing CW contents (even with 40 wt.% PC replacement). However, some studies have reported an optimum PC replacement, generally within the 10-20 wt.% range [18,66], and increasing water absorption values with further amounts of CW [14,8].

Chloride ions may diffuse through concrete and oxidise steel reinforcements. However, shallower chloride ion penetration depths have generally been found in the mortars and concrete developed by replacing PC with CW, even with high CW contents (up to 40 wt.%) [1,8,14,18,25,55,62,63,65] More specifically, Chen et al. [12], who replaced up to 40 wt.% PC with CWP and 60 wt.% of natural gravel with CDW aggregates, observed an improvement in chloride penetration resistance with increasing CWP additions, which they attributed to a filling and pozzolanic effect of ceramic powder. In the study by El-Dieb et al. [55], chloride ion penetration also progressively reduced with increasing PTCW contents, and with very low rates compared to the reference concrete. This was attributed to pore refinement and, together with the higher electrical resistivity values generally recorded with increasing amounts of PTCW or for longer curing periods, denotes improved corrosion protection of steel reinforcements. Gautam et al. [18] noted how the combination of CWP and recycled sand from granite provided the shallowest chloride penetration depths, which they attributed to the ceramic powder's pozzolanic behaviour and a better filling ability of recycled fine aggregates. According to the study by Yang and Luo [65], the compressive strength recorded after 2 weeks of immersion in 3.5 wt.% NaCl solutions improved when replacing PC with CW. This behaviour was even better when combining CW with sugarcane bagasse ash (93% better than the reference sample). After 1 month of immersion, polarisation curves also showed a significant improvement in the corrosion resistance of the steel reinforcement in pozzolanic concrete. In line with the review by Alsaif [8], residual strength of the 15 wt.% CW mortars after immersion in hydrochloric acid (HCl) was similar to or better than that of the reference. This was explained by the reaction between HCl and $Ca(OH)_2$ to form $CaCl_2$, a highly soluble salt.

Resistance to sulphate attack has also generally improved with CW addition, with

similar or higher residual strength values than the reference sample after Na_2SO_4 immersion [1,8,9,14,44,59,63]. This has been explained by pore refinement with secondary hydration products, and by portlandite consumption during the pozzolanic reaction. $Ca(OH)_2$ reaction with sulphate ions provides gypsum, which may react with calcium aluminates to form expansive ettringite that may, in turn, lead to cracks in hardened concrete [1,8,9,14,59]. In the study by Mohammadhosseini et al. [63], the mortars developed using TCW to replace up to 40 wt.% PC and 100% sand exhibited lower strength reduction than the reference after immersion for 18 months in Na_2SO_4 solutions (16.8% and 41.4% lower than the initial strength for the TCW and reference mortars, respectively). Samadi et al. [44] also observed a significant improvement in sulphate resistance when replacing 40 wt.% PC and 100 wt.% sand with TCW. Conversely in the study by Brekailo et al. [67], up to 20 wt.% PC was replaced with red ceramic, concrete and limestone filler (reference), each separately. The pozzolanic mortars exhibited more expansion after immersion in sodium sulphate solutions. So, the mortar bars containing 12 wt.% or more CW broke or recorded excessive expansion after 70 curing days. The highest porosity values were obtained with 20 wt.% CW (15.48% after immersion for 98 days in sodium sulphate solutions, remarkably higher than for 100% PC mortar, 2.55%). The authors attributed this behaviour of the PC/CW blended mortars to the high aluminium oxide contents of the used CW, which compensated for the C_3A reduction in the reference sample and reacted with the aggressive sulphate ions.

Unlike the PC/CW blended cements' general resistance to sulphate and chloride ions, the carbonation depths recorded when replacing PC with CW generally rose. This was attributed to both PC dilution and the pozzolanic reaction, which reduce the $Ca(OH)_2$ available to react with CO_2 [8,14].

Alkali silica reactions generally reduce in samples developed with CW used as a pozzolanic admixture [1,8,27]. Zito et al. [1] observed that replacing 24 wt.% PC with different CW types (hollow bricks, mud-bricks and CSW, separately) significantly reduced the alkali silica reaction with highly reactive sand compared to the reference mortar. This was attributed to both $Ca(OH)_2$ consumption and the formation of secondary hydration products during pozzolanic reactions. In the study by Bignozzi and Saccani [47], 25 wt.% PC was replaced with CW from polishing porcelain stoneware tiles, and 25 wt.% of natural siliceous sand with three different glass waste types. Expansion of pozzolanic mortars significantly reduced compared to the CEM I samples, which led to lower compressive strength reductions after expansion tests. The expansion percentages of the 25 wt.% CW samples were similar to those recorded with CEM IV/A pozzolanic cement.

According to the review by Agrawal et al. [14], frost resistance also generally improves when CW is used as a pozzolanic admixture. Sánchez-de-Rojas et al. [22] observed no significant damage after 55 freeze-thaw cycles in PC samples containing 20 wt.% TCW. However, in the study by AlArab et al. [68], who combined TCW and BFS to partially replace PC in concrete (constant water/binder ratio of 0.54), the relative dynamic modulus of elasticity, determined after 30 to 150 freeze/thaw cycles, reduced with increasing amounts of TCW (up to 20 wt.%) and with TCW/BFS

combinations. The lower values compared to the reference concrete denoted lower resistance to freeze/thaw cycles. Although the freeze-thaw durability of the mortars developed by Reiterman et al. [51] also progressively diminished with CW addition, in their study it was attributed to the higher water/binder ratios required to keep workability constant.

3.2.7. Thermal conductivity and resistance to high temperatures

The thermal conductivity of samples developed by partially replacing PC with CW generally decreases with increasing waste contents [14]. AlArab et al. [68] observed howt the thermal resistance of concrete samples developed by replacing 15 wt.% PC with TCW (with and without BFS) slightly improved compared to the reference sample (higher R values; as $m^2 \cdot K/W$). Although Kulovaná et al. [60] also noted a reduction in dry thermal conductivity with increasing amounts of CW, these values significantly rose with moisture content, which would influence the thermal insulation of a building façade built with these materials.

For fire resistance, El-Gamal et al. [46] determined the residual strength of 20 wt.% PC/CW pastes cured for 28 days after being exposed to up to 800ºC, followed by gradual cooling or direct immersion in water. Fire resistance improved with up to 10 wt.% CW (highest residual strength) and, although this improvement was more significant with rapid cooling, none of the samples resisted the thermal shock they underwent after heating at 800ºC. The incorporation of 0.05% and 0.1% of carbon nanotubes generally led to similar or better residual strength values. Mohit and Sharifi [69] also found that the mortars containing up to 25 wt.% CWP generally exhibited better compressive strength results after heating at temperatures within the 200-800ºC range. The authors attributed this behaviour to CW pozzolanic activity, which provided additional hydration products that filled existing pores.

4. Sustainability and carbon footprint

PC production has substantially increased in the last two decades: the 1.6 million tons produced worldwide in 2000 were multiplied by 2.5 in 2017 (4.1 million tons), and it is estimated that 4.4 million tons were produced in 2021 [70]. The environmental impact of PC production is the strongest by far of concrete components. This is because of the PC industry's high energy demanding process associated with the clinkerisation (at 1,450-1,500ºC) and grinding steps, together with the CO_2 that is released during limestone decomposition, which occurs at 800-900ºC. The CO_2 emitted by producing 1 kg of Portland clinker is estimated to be 0.94 kg (0.55 kg of chemical CO_2 from limestone decarbonation, plus 0.39 kg from the used fossil fuel) [71]. Commercial PC is a mixture of clinker, gypsum and other SCMs, and it implies that embodied CO_2 depends very much on the processing required by SCM (grinding, drying, calcination, etc.) and the clinker/cement ratio. According to the literature, the clinker/cement ratio is about 0.7 [72] and, since the 1970s, considerable interest has been shown in developing blended cements by mixing Portland clinker with new SCMs or, alternately, by replacing commercial PC with SCMs during concrete

production.

Ground limestone, ground granulated BFS and FA are the most widely used SCMs in blended cements. However, employing many alternatives as SCM has been investigated, some of which derive from industrial processes (e.g. fluid catalytic cracking residue [73]), agriculture (e.g., sugarcane bagasse [74] or ashes and urban activities (e.g. sewage sludge ash [75]. Of them, CW can be adapted by grinding it to become SCM. The obtained powder possesses pozzolanic activity and a part of clinker can be replaced with this powdered waste to prepare eco-friendly cement. Mortars replacing 10-60% of Type I ordinary PC with tile CW powder (TCWP) were studied in environmental impact terms [76]. The energy use for TCWP was 1.12 GJ/ton, which is a much lower value than that obtained for PC (5.13 GJ/ton). Similarly, greenhouse gas emissions emissions were very different with 45 $kgCO_2$/ton for TCWP and 904 $kgCO_2$/ton for PC. These differences gave 37% CO_2 emissions for the 40% TCWP mortar, which gave higher compressive strengths at 28 and 90 curing days than the 100% PC mortar. Figure 5 (adapted from [76]) shows the CO_2 intensity index values (ratio between the CO_2 emission for m^3 of mortars and their compressive strength) for the mortars cured for 7, 28 and 90 days. The values for the TCWP-containing mortars were significantly lower than those obtained for the PC mortar, and for all the curing times. The lowest values were obtained with the 40% and 60% replacements cured for 90 days.
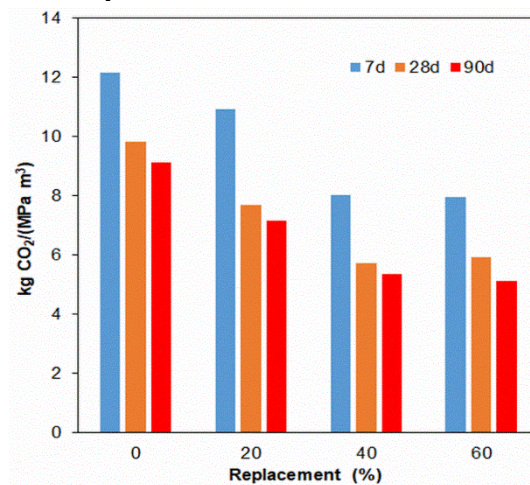

Figure 5 – CO_2 intensity index for the mortars containing ground TCW replacing PC (data calculated from [76]).

Chen et al. [12] analysed the behaviour of white TCWP by replacing 0-40% PC. The prepared concrete also had recycled coarse aggregates (RCA) from demolished concrete replacing NCA. As the TCWP cost was null and RCA is cheaper than NCA, the concrete containing TCWP and RCA offered economic benefits. Specifically after 56 curing days, a 10% cost saving and a 22%compressive strength reduction were

obtained for the concrete sample that replaced NCA with RCA. However, better economic savings (15-20%) and lesser compressive strength reductions (only 12-17%) were recorded when replacing 10% and 20% PC with TCWP. Different environmental impacts were also calculated (global warming, GWP; acidification, AP; eutrophication, EP; primary input-non-renewable, PIEInon-re; and photochemical ozone creation, POCP). The concrete with RCA and 20% TCWP reduced GWP by 21.65%, AP by 18.94%, EP by 20.57%, by 6.6% in PIEI-non-re and by 25.76% in POCP.

   The red ceramic waste obtained from damaged bricks (BCW) also offers environmental advantages [45]. The CO_2 emissions related to BCW were calculated: 327 $kgCO_2$/ton of BCW (brick production, crushing/grinding and transport were considered for this calculation). For the concrete mixed with a 0.35 water/binder ratio, the sample that contained 30% BCW had 94 $kgCO_2/m^3$ from waste and 486 $kgCO_2/m^3$ from PC. The total CO_2 emissions were 580 $kgCO_2/m^3$, with 695 $kgCO_2/m^3$ for the control concrete. The reductions in the CO_2 intensity values ($kgCO_2/MPa \cdot m^3$) for the concretes mixed with a 0.35 water/binder ratio are depicted in Figure 6. The 12.6 % and 9.9% reductions after 28 and 182 curing days, respectively, are noteworthy.
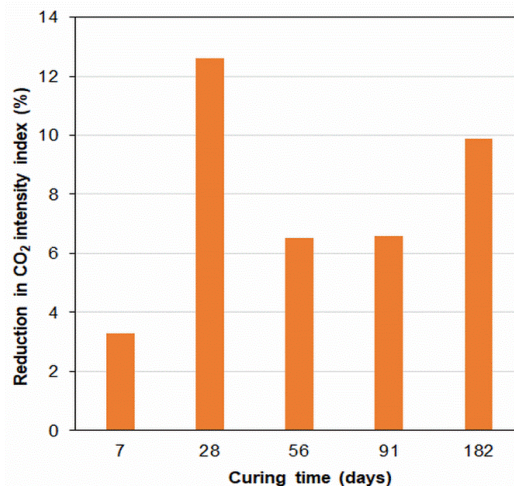

Figure 6 – Reduction (%) of the CO_2 intensity index for the mortars containing red ceramic waste (replacing 30% PC) at different curing ages (data calculated from [45]).

   A similar trend was demonstrated by Pavesi et al. [11], who analysed different concrete series in which the water/binder ratio ranged from 0.35 to 0.55. In almost all cases, the CO_2 intensity index was lower than that obtained for the control concrete. Specifically of the concretes prepared with a water/binder ratio of 0.35 cured for 90 days, the control sample presented 6.50 $kgCO_2/MPa \cdot m^3$, and this value lowered to 6.37 and 5.92 $kgCO_2/MPa \cdot m^3$ when replacing 10% and 20% of PC with red CW.

   Environmental saving was also assessed in SCC containing bone China CW powder (BCCWP) [18]. In addition to replacing part of PC with BCCWP (10%, 20% and 30%), part of the fine aggregates was also substituted (20%, 30% and 40%) for

granite cutting waste (GCW). The embodied energy, which is linked to CO_2 emissions, of the different concretes containing BCCWP and GCW is represented in Figure 7a. These values were calculated after including all the concrete components: binder, water, aggregates and superplasticiser ($\approx$700 mm slump flow). The strong dependence of these parameters on BCCWP content is noteworthy, and the contribution of GCW content was very poor. In relative cost terms (Figure 7b, in Indian rupee/MPa·m³), the best economic conditions were obtained when combining 20% BCCWP and 30% GCW: 156.06 INR/MPa·m³, 17.6% lower than the reference concrete, without any replacement. The concretes with BCCPW-GCW proportions of 10-30, 10-40, 20-20 and 20-40 were also cheaper than the reference one (157-161 INR/MPa·m³).

a)   b)
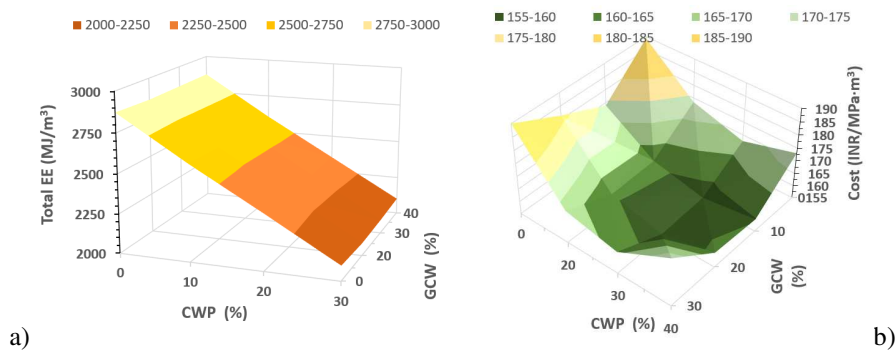
Figure 7 – Parameters for the concretes made by partially replacing PC with BCCWP and NCA with granite cutting waste (GCW): a) Total embodied energy; b) cost (data calculated from [18]).

Microceramic powder (MCP) is also interesting for developing eco-friendly concrete. Li et al. [61] studied the influence of finely powdered waste from decorative vases and flowerpots. Very fine material was obtained after a jaw crushing operation and an air jet mill process, with a mean particle diameter of 3.5 µm and most particles < 10 µm. The 10% and 20% PC replacements with MCP yielded mortars with higher compressive strengths than the PC control mortar after 90 and 180 curing days. Energy saving was calculated by considering the energy used to condition the MCP, which was 79 kWh/ton, significantly higher than that required to adapt the particle size of PC (45 kWh/ton, which is 38% of the total embodied energy associated with PC production). Notwithstanding, the energy saving of the mortars containing 10% and 20% MCP was still 3.31% and 6.62%, respectively (compared to the reference sample). Additionally, despite the high energy demand for grinding MCP, the mortar containing 20% MCP was 9.6% cheaper than the control mortar, whose cost was 49.045 USD/m³.

Pitarch et al. [20] ran a gate-to-gate life cycle analysis, which considered only the CO_2 emissions associated with binder production. Several CW materials were analysed: red clay brick (RCB), ceramic tiles (TCW) and sanitary ware waste (CSW). Replacements within the 0-50% range were assessed, and the CO_2 intensive index

values were calculated for one ton of all these mortars cured for 365 days. The control mortar (with CEM I 42.5R cement) presented 15.62 kgCO$_2$/MPa·ton, and this value fell with the 13.51-15.10 kgCO$_2$/MPa·ton range for the CW-containing mortars. The highest CO$_2$ intensive index reductions were obtained for the samples prepared with RCB.

Studies about UHPC containing TCWP have also been reported [31]. In these concretes, 15-55% PC replacements were tested, and a constant water/binder ratio of 0.18 and a steel fibre content of 2% were maintained in all the prepared mixtures. After 28 curing days, the UHPC containing 25% TCWP gave a 6.4% higher compressive strength than that presented by the control UHPC. The CO$_2$ emissions of this sample lowered from 0.93 to 0.79 kgCO$_2$/kg concrete. Cost also dropped from 680 Chinese yuan (RMB)/ton of concrete to 600 RMB/ton.

Finally, Zhao et al. [28] assessed the influence of the milling process on the environmental enhancement of mortars containing clay brick powder (CBP). The original waste was ground for different times, and four samples were obtained with differing finenesses: 27.1, 15.8, 10.5 and 3.4 µm mean particle diameters (MPD). These powders replaced 30% PC in pastes and mortars. Figure 8 shows the following values: energy intensity, CO$_2$ emissions and cost per kg of blended cement. It is noteworthy that the replacement of 30% PC with the coarsest CBP (27.1 µm-MPD) brought about a marked reduction in all three parameters. However, the situation was not as advantageous when CBP fineness was increased.
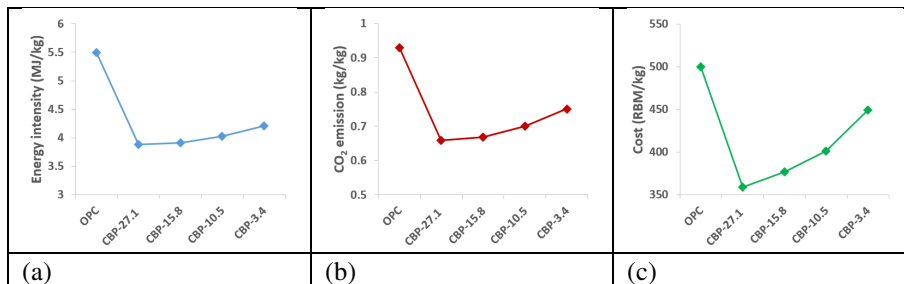

(a)   (b)   (c)

Figure 8 – Environmental and economic parameters for blended cements containing 30% ceramic brick powders (CBP) of different finenesses: a) Energy intensity; b) CO$_2$ emission; c) Cost in Chinese yuan (data taken from [28]).

When SCMs are selected, several aspects must be carefully considered: not only the emissions associated with drying and crushing/grinding, but also the cost and energy required for their transportation [9]. SCMs are not available in every geographic location and are not, consequently, 'environmentally-free'. In some cases, especially for agricultural waste, it is possible to obtain ash with negative CO$_2$ emissions: if a given quantity of energy is recovered because biomass combustion is done under controlled conditions; if the CO$_2$ generated during the combustion process is not considered because the raw material previously fixed it from the atmosphere [77].

5. Conclusions

This paper reviewed the use of CW as SCM to develop more sustainable binders, and reached the following conclusions:

- Several studies have successfully proved that CW valorisation as a supplementary cementitious material in PC systems is viable. Using PC/CW blended cements with smaller amounts of clinker contributes to sustainable development by promoting the use of more environmental-friendly construction materials and minimising CW dumping problems.
- Although CW's pozzolanic activity very much depends on the sintering treatment applied when manufacturing a ceramic product, most CW materials exhibit moderate pozzolanic activity.
- The main drawback when replacing PC with CW is slow strength development. Although no significant pozzolanic reactions occur at early curing ages, filler and nucleation effects accelerate PC hydration. Slow reactions at early ages may be advantageous when large volumes of concrete are used because hydration heat lowers, which helps to prevent temperature cracking.
- For larger curing ages, pozzolanic reactions densify the microstructure, refine pores and reduce their interconnectivity, which generally enhances durability properties.
- Although the optimum CW content to replace PC depends on performance criteria, the mechanical requirements established for other pozzolanic materials, such as FA, are generally accomplished by replacing up to 20 wt.% PC. However, further CW contents can be used depending on the CW type and particle size distribution.
- No studies have been found to have combined different CW types to partially replace PC.
- Although optimum PC replacements with CW usually fall within the 10-30 wt.% range, further reductions in natural materials and used energy can be achieved by combining CW to replace PC and natural aggregates.

This review has summarised the behaviour of CW when used as supplementary cementitious material. Although ceramic products generally contain crystalline stable phases that remain after pozzolanic reactions, CW has been successfully used as pozzolanic admixture to develop binding materials with similar properties to traditional ones. This is a promising strategy towards circular economy and one that may significantly contribute to reduce the CO_2 emissions associated with the construction industry.

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