

Use of granular coal combustion products as aggregates in structural concrete: Effects on properties and recommendations regarding mix design

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Abstract

This review collates the results obtained in more than 100 scientific publications regarding the performance of concrete with granular coal combustion products (GCCP, commonly referred to as coal bottom ash in literature) as a partial substitute for conventional aggregate. The effects of this by-product on the workability, density and mechanical and durability related properties were analysed. Some conclusions and recommendations are provided to facilitate future research. These recommendations include conducting careful assessments of the specific gravity and water absorption properties of GCCP, replacing conventional aggregate by volume, and compensating the water absorption of GCCP. The use of GCCP as an internal curing water reservoir is proposed as the most promising advanced application.

Keywords: Coal bottom ash, CBA; by-product; valorisation; waste integration; water dosage strategy; workability; durability; lightweight concrete; internal curing; database.

Highlights:

- Granular coal combustion products (GCCP) can be used as aggregates in concrete
- GCCP particles are irregular, rough, light and highly water absorbent
- Compensating for the water absorption by GCCP avoids concrete workability drop
- The strength and durability of concrete decrease if high GCCP contents are used
- GCCP reduces the weight of concrete and works as an internal curing water reservoir

30 1 Introduction

31 Combustion of coal in power stations throughout the world generates large amounts of different
32 residues (coal combustion residues, CCRs), where fly ash (FA), coal bottom ash (CBA), boiler slag
33 and flue gas desulphurisation (FGD) materials are the most abundant. These products have
34 generally been managed as waste materials and disposed of in landfills. However, they can be
35 used in beneficial alternative applications, where they are referred to as coal combustion
36 products (CCPs) instead of CCRs.

37 FGD materials are produced via a process that aims to minimise sulphur dioxide (SO₂) emissions
38 from the exhaust gas system of a coal-fired boiler. FGD materials comprise of Ca-SO_x compounds
39 [1] and have been used in several beneficial encapsulated applications in the construction
40 industry, such as in the manufacture of panels, blocks and plasters [2,3]. Fly ash is the finely
41 divided residue produced by the combustion of ground or powdered coal that is transported by
42 flue gases [4]. The powder-sized particles in fly ash are spherical in shape, and they have hydraulic
43 and/or pozzolanic properties, depending on their chemical composition. Fly ash is already a
44 popular product in the concrete industry and it has been used successfully as a supplementary
45 cementitious material for decades. This beneficial use has contributed significantly to the
46 sustainability of concrete by reducing the utilisation of ordinary Portland cement (OPC) in a wide
47 range of applications. The use of fly ash as a partial substitute of OPC has environmental and
48 economic advantages, and it can also improve the technical properties of concrete, especially
49 those related to durability. CBA comprises agglomerated coal ash particles that are too large to
50 be carried in the flue gas [5] so it remains at the bottom of the boiler. CBA is generally referred
51 to with this term when it comes from traditional pulverised coal furnaces, where it is collected
52 following a dry process. However, other residues that are coarser than fly ash residues are
53 produced in various types of furnaces or collected by different methods, such as boiler slag,
54 fluidised bed combustion ash and pond ash [6]. All of these residues are granular in nature and
55 this is the key characteristic related to their utilisation in the construction sector because they
56 can be employed as aggregates in cement-based materials and other applications. Thus, in this
57 review, the popular term “coal bottom ash” (CBA) is avoided and the more general term
58 “granular coal combustion product” (GCCP) is used instead. This term encompasses all the
59 different types of granular coal combustion by-products referred to in scientific studies of
60 replacements for conventional aggregates regardless of the processes involved in their
61 production or collection.

62 GCCP represents approximately 20% of the total ash produced in coal power stations (the
63 remainder comprises fly ash). GCCP has been used in asphalt paving mixtures and as a base
64 granular material for roads and car parks [2]. The principal beneficial uses of GCCP in the
65 European Union are in reclamation and restoration. However, the encapsulation of GCCP in
66 cement-based materials is rarely practiced. For instance, during 2016, 20% of all the CBA
67 produced in Europe was used to fabricate concrete blocks [3]. In general, the beneficial reuse of
68 CCPs has been increasing in recent years (**Fig. 1**), mainly due to the development of new
69 applications of FGD, but the reused proportion of GCCP has remained constant (**Fig. 2**). This
70 situation has encouraged the development of research works and new applications in the
71 construction field in order to ensure that a higher volume of this by-product is utilised in the
72 future.

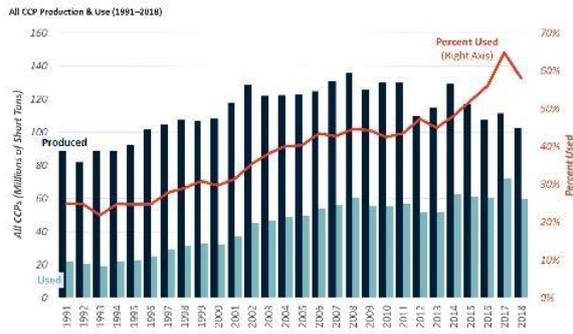


Fig. 1: Changes in the production and use of all CCPs in the USA until 2018 [2].

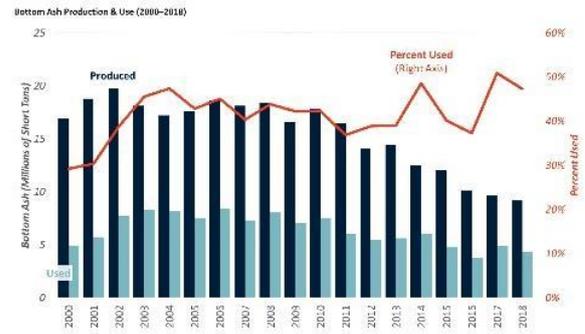


Fig. 2: Changes in the production and use of CBA in the USA until 2018 [2].

73 2 Research significance

74 The use of ground GCCP as a supplementary cementitious material in a similar manner to FA has
 75 been suggested in some studies [7,8,17–26,9–16]. The implementation of a highly efficient
 76 grinding process in power stations has also been proposed [27]. Furthermore, the manufacture
 77 of lightweight aggregates made of pelletised stone dust and ground CBA has been proposed [28].
 78 However, the grinding process consumes energy and the preferred applications would involve
 79 using it in a raw form as an aggregate. Studies have explored the use of GCCP as an aggregate in
 80 concrete together with other recycled aggregates, such as recycled concrete aggregates [29],
 81 granulated blast furnace slag [30,31] and oil palm shell [32].

82 Several previous reviews have investigated the use of GCCP as an alternative aggregate in
 83 concrete [6,33,42–48,34–41]. However, most of these studies mainly considered the pozzolanic
 84 activity of GCCP and they generally ignored important aspects related to the nature of the GCCP
 85 particles themselves, such as their specific gravity, and aspects of the concrete mix design
 86 process, such as the basis on which the conventional aggregate is substituted (by its total volume
 87 or weight) or the strategy selected regarding water dosage. In order to consider these
 88 parameters, the present review analysed results of the workability, porosity, density, mechanical
 89 properties, shrinkage and durability of concrete made with GCCP gathered from more than
 90 previous 100 studies. Furthermore, some conclusions and recommendations are provided based
 91 on this analysis.

92 3 GCCP characteristics

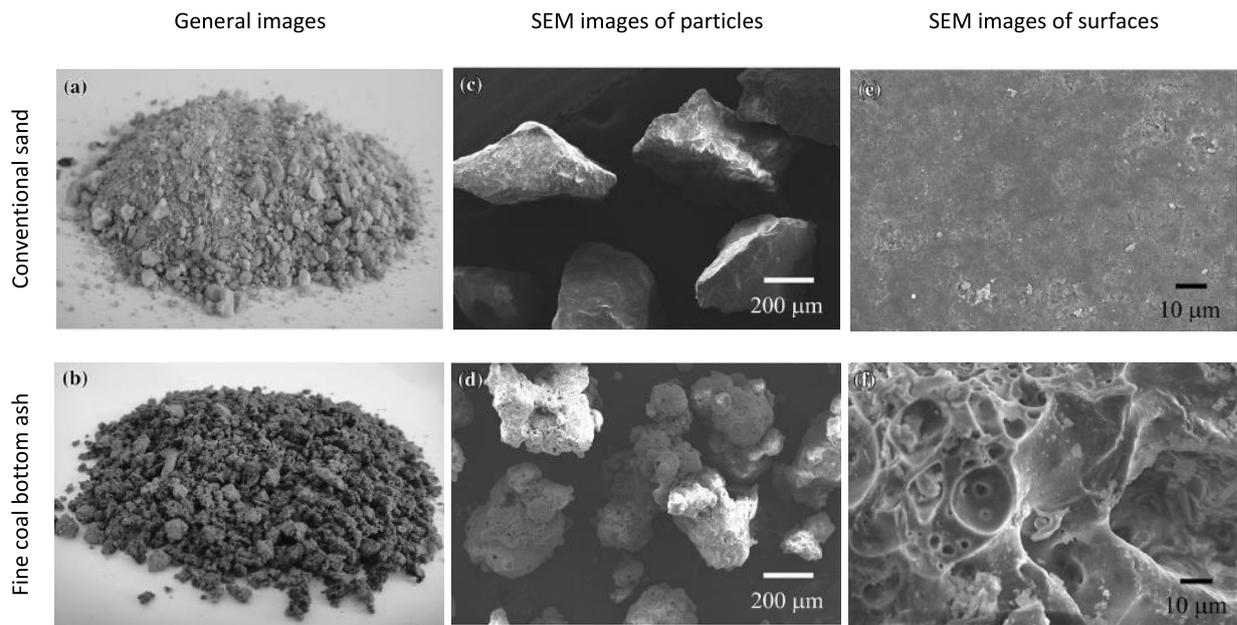
93 The characteristics of GCCP depend on many important variables, such as the coal source,
 94 preparation for burning (degree of pulverisation, etc.), the burning process itself (temperatures
 95 reached, etc.) and the furnace emptying method (dry or wet) [9,49]. The fact that the
 96 characteristics of GCCP depend on many factors during its production make it difficult to quantify
 97 the properties of GCCP as aggregates in a precise and general manner. However, representative
 98 information from different power stations throughout the world has been reported in scientific
 99 articles. The results collated in this review provide broad insights into the ranges, mean,
 100 maximum and minimum values for the physical and chemical properties of GCCP particles.

101 3.1 Physical properties

102 The appropriate physical characterisation of GCCP is required to assess its performance as an
 103 aggregate in any cement-based materials. The size, shape, texture, density and water absorption
 104 are the most commonly studied physical properties.

105 GCCP can contain a wide range of particle sizes, but usually below 20 mm. Particles with a size
106 larger than 4 or 5 mm are discarded when it is used as a fine aggregate, whereas smaller particles
107 are discarded when it is used as a coarse aggregate.

108 In general, GCCP particles have irregular, disordered, angular or glassy shapes, and a rough or
109 porous texture. These physical characteristics can be observed with the naked eye and by
110 scanning electron microscopy (SEM) (**Fig. 3**). However, some studies described GCCP particles
111 with appearances that deviated from this trend, where they had rounded, regular or spherical
112 shapes [30,31,50–54]. Attempts were made to minimise the roughness and external porosity
113 GCCP particles in some studies by covering them with cement and fly ash paste using a cold-
114 bonding process [55,56].



115 **Fig. 3:** Appearance of conventional sand and fine coal bottom ash on different scales (based on [57]).

116 The porous surface indicates that the internal structure of GCCP is porous. Coarse particles may
117 have larger pore volumes than fine particles [56], possibly because the latter can be formed by
118 crushing coarse particles and this process opens their inaccessible porosity. Furthermore, the
119 internal porous structure of GCCP was studied by mercury intrusion porosimetry, which
120 demonstrated that more than one half of the pores were shaped like ink bottles [58].

121 The porous structure of GCCP results in a lower specific gravity (down to 1 g/cm³) and higher
122 water absorption capacity (up to 35%) than conventional aggregates. These two properties are
123 illustrated in **Fig. 4** based on several previous studies.

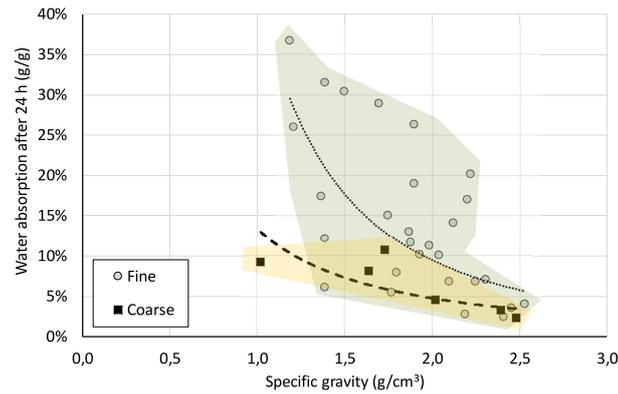


Fig. 4: Relationships between specific gravity and water absorption for GCCP according to several studies [29,30,61–70,31,71–78,50,51,55,57–60].

124 With the same specific gravity, coarse aggregates exhibit lower water absorption than fine
 125 aggregates, probably due to the higher proportion of inaccessible pore volume among the overall
 126 porosity in coarse GCCP. However, some fine aggregates exhibit similar performance to coarse
 127 aggregates, possibly because of the remaining highly inaccessible pore volume or differences in
 128 the characterisation procedure employed. The critical step when determining the specific gravity
 129 and water absorption is drying the particles after wetting to reach a saturated surface dry
 130 condition. Different methods can be used for this purpose but they may lead to substantially
 131 different results, especially when testing lightweight aggregates. For instance, a paper towel-
 132 based method [79] leads to higher water absorption and lower specific gravity values than other
 133 standard methods where the particles are exposed to a gentle current of warm air in order to
 134 reach a saturated surface dry condition [80,81]. The latter methods are considered to lead to
 135 underestimates of these properties [82].

136 The water absorption kinetics is also distinctive for GCCP. They are described by a curve with a
 137 steep slope during the first minutes, before stabilising after 5 min (**Fig. 5**), 10 min [58] or 20 min
 138 [64]. GCCP exhibit more rapid water absorption than conventional aggregates and other highly
 139 absorptive alternative aggregates such as expanded shale (**Fig. 6**).

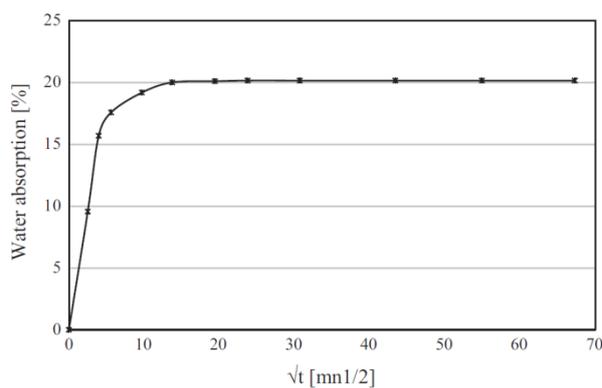


Fig. 5: GCCP water absorption kinetics [63].

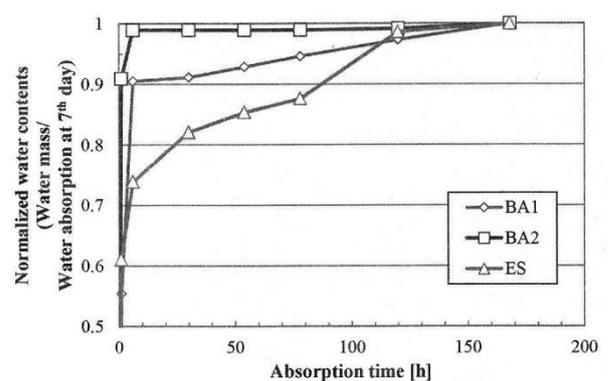


Fig. 6: Normalised water absorption kinetics of GCCP [68].

140 The high porosity and pore size in GCCP also result in a high desorption capacity in the air under
 141 high relative humidity [55,58]. The results obtained by a conventional desorption test in the air
 142 at different levels of humidity are plotted in **Fig. 7**. An alternative test was proposed based on
 143 the desorption of ink in cement paste (**Fig. 8**). This test confirmed that the desorption capacity of
 144 GCCP can also be high in a cement-based material [68].

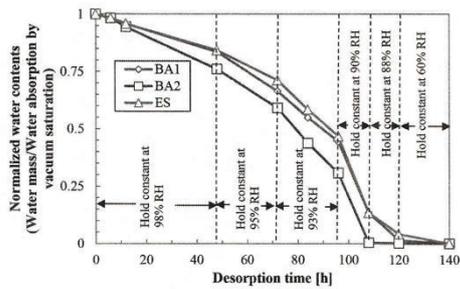


Fig. 7: Normalised water desorption by GCCP during different time periods under various relative humidity levels [68].

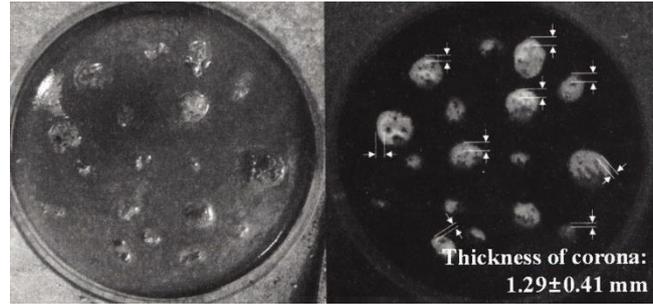


Fig. 8: Desorption of ink in cement paste [68].

145 3.2 Chemical composition

146 The chemical composition of any coal combustion product is critically influenced by the coal
 147 source [50] and by the burning process to a lesser extent [49]. The most common chemical
 148 elements in GCCP are silicon (Si), aluminium (Al), iron (Fe) and calcium (Ca), which coexist with
 149 some minor elements such as magnesium (Mg), sodium (Na) and potassium (K). Their mass
 150 proportions (in oxide form) in the total incombustible material according to different studies are
 151 shown in Fig. 9.

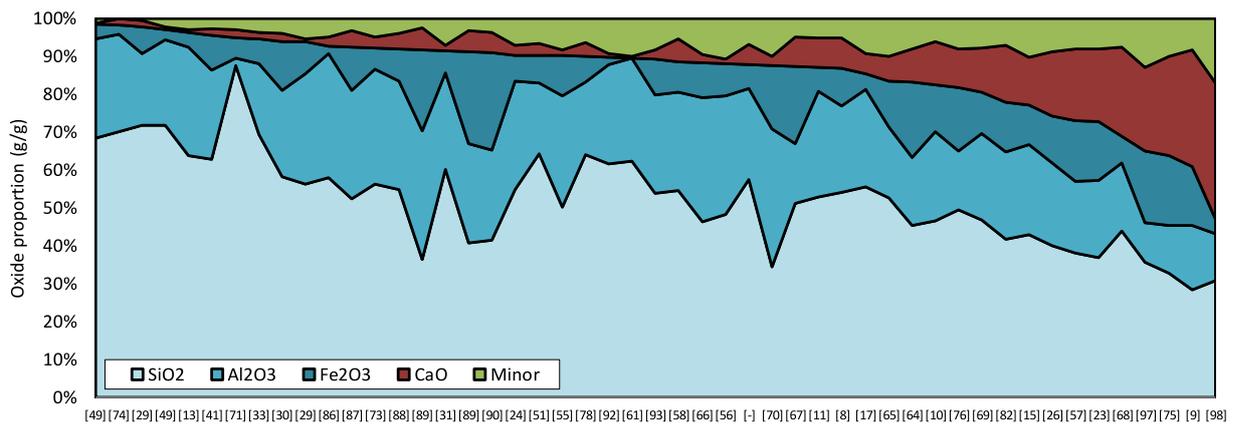


Fig. 9: Chemical composition of GCCP in various studies [8,13,29–31,49,51,56–58,60,63,14,67–71,73,75–78,16,83–92,17,93–98,18,19,21,23,25].

152 The loss on ignition usually accounts for less than 6% of the composition of GCCP, which is the
 153 limit established by American Society for Testing and Materials (ASTM) standard C618 for coal fly
 154 ash. This standard also provides a classification for fly ash based on its chemical composition,
 155 which could be applied to GCCP. For instance, Class F refers to ashes where the sum of SiO₂, Al₂O₃
 156 and Fe₂O₃ exceeds 70% of the total composition, whereas Class C denotes ashes where the sum
 157 ranges between 50% and 70%. Most of the samples considered in this study can be classified as
 158 Class F, but a small minority belong to Class C. In addition, according to ASTM C618, Class F ash
 159 is usually produced by burning anthracite or bituminous coal, whereas Class C ash is generally
 160 produced by burning lignite or sub-bituminous coal [4].

161 Some of the chemical elements present in GCCP are combined to form crystalline compounds.
 162 Various analyses conducted using X-ray diffraction indicate that the most frequent crystalline
 163 compounds are quartz (SiO₂) and mullite (Al₆Si₂O₁₃). Some examples are shown in Fig. 10.

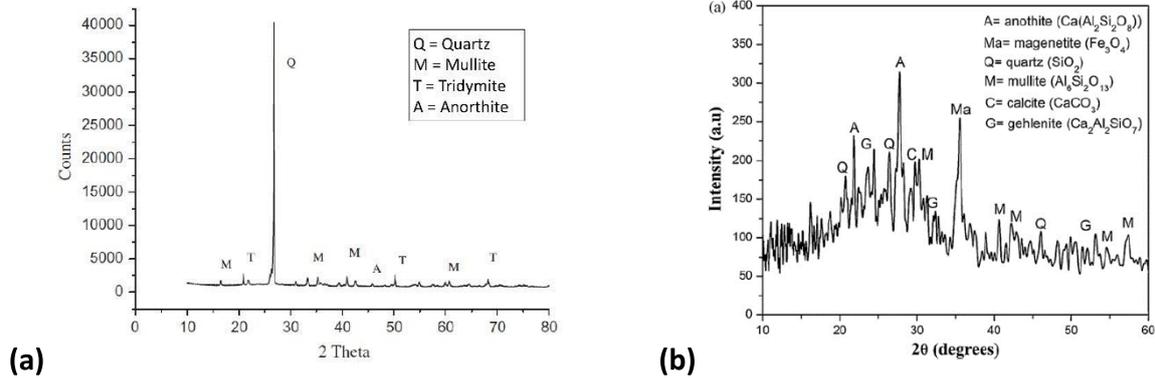


Fig. 10: X-ray diffraction spectra obtained for GCCPs with chemical compositions homologous to (a) Class C [23] and (b) Class F [99].

164 Some of the minor elements present in GCCP can cause environmental problems, particularly
 165 cobalt (Co), manganese (Mn) and other heavy metals. Some works have concluded that the
 166 leaching risk for these elements from concrete containing GCCP is low [66,75,100]. However, it
 167 must be noted that some cases of toxic leaching have been reported in unlined landfills and
 168 surface impoundments [1], but these were non-encapsulated applications with a higher risk.

169 4 Design of structural concrete with GCCP in raw form as an aggregate

170 Structural concrete is the most common cement-based material in which GCCP is integrated in
 171 its raw form. The scope of the present review is limited to this specific application. However,
 172 previous studies have focused on the effects of including GCCP in other construction materials,
 173 such as roller compacted concrete [101,102], pervious concrete [74,75,91], paving blocks
 174 [103,104], masonry units [61,90,105], masonry mortars [73,92,106], grouts [107] and other non-
 175 structural materials [71,93,108,109].

176 Conventional concrete is the most popular subtype of structural concrete considered in previous
 177 studies. Furthermore, different types of high performance concrete, self-compacting concrete
 178 and fibre-reinforced concrete have been investigated. The approximate ranges of the structural
 179 concrete mix proportions reported previously are presented in **Table 1**. Experimental programs
 180 based on the concrete mortar phase obtained conclusions that are valid for concrete. Those
 181 mortars usually contain approximately twice the amount of binder and half the aggregate to
 182 paste ratio compared to the proportions of concrete.

183 **Table 1:** Common ranges for mix proportions of cement-based materials with GCCP as a partial aggregate substitute

| | Conventional Concrete* | High Performance Concrete [30,55,58,72,94,110,111] | Self-Compacting Concrete [83,84,96,112,113] |
|--|------------------------|--|---|
| Binder dosage (kg/m ³) | 400 ±100 | 525** | 525 |
| Water to binder ratio (g/g) | 0.50 ±0.15 | 0.30 | 0.40 |
| Paste to aggregate ratio (m ³ /m ³) | 2.0 ±0.5 | 2.0 | 1.5 |
| Coarse to fine aggregate ratio (m ³ /m ³) | 1.5 ±0.5 | 1.2 | 0.7 |
| Most common supplementary cementitious materials | None/ Fly ash | Fly ash + others | Fly ash |
| Most common admixtures | None | Superplasticizer | Superplasticizer |

| Common particle size substituted | Fine | Fine | Fine |
|---|------|------|------|
| Common maximum fine aggregate substitution rate | 100% | 100% | 50% |

184 *[32,50,65–67,69,70,85,87,88,95,99,51,114–122,52,54,56,57,60,62,63]

185 **In some exceptional cases, the binder content is remarkably higher than the indicated value due to the use of supplementary
186 cementitious materials, such as silica fume and ground granulated blast furnace slag [72].

187 The main conclusion that can be made based on **Table 1** is that no specific proportions have been
188 established for reference concrete in studies of the use of GCCP as an aggregate substitute. It
189 should also be noted that GCCP is commonly used as a fine aggregate, although its use as a coarse
190 aggregate has also been explored [9,72,77].

191 Conventional aggregate can be substituted by volume or by weight, where the latter option may
192 be simpler to apply because the specific gravity of the particles does not need to be known.
193 Testing the specific gravity of GCCP can be challenging, as stated in Section 3.1. However,
194 substitution by weight distorts the paste to aggregate ratios for GCCP with significantly low
195 densities. Therefore, any conclusions must be treated with caution in this situation.

196 The water dosage strategy is another fundamental issue that needs to be considered when GCCP
197 is used as a partial conventional aggregate substitute in a concrete mix design. The most common
198 strategies are as follows.

- 199 • Constant maintenance of the total water content (=W): GCCP is used in a dry state and its
200 water absorption is not compensated.
- 201 • Water absorption compensation (WAC): An amount of water equivalent to the GCCP
202 absorption capacity is added to the mix either with the mixing water or by pre-wetting the
203 GCCP particles.
- 204 • Increasing the water content so constant fluidity is achieved (+W): The amount of mixing
205 water is increased when using GCCP in order to obtain the same fluidity as the reference
206 concrete.

207 **5 Properties of structural concrete with GCCP as an aggregate**

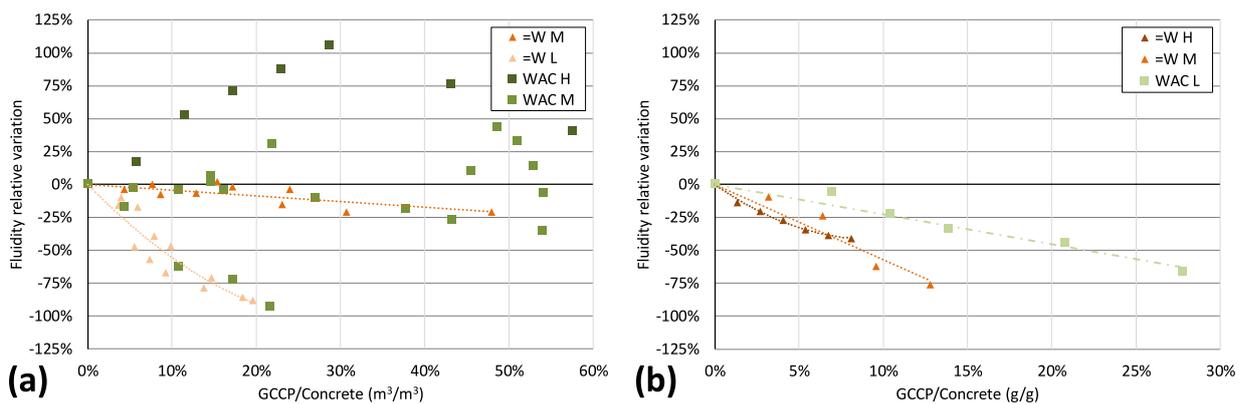
208 The performance of concrete containing GCCP is discussed in this section, where the fresh
209 properties, microstructure and density, mechanical properties, volume stability and durability
210 are analysed.

211 Graphs were prepared by grouping the results obtained from various studies to facilitate a better
212 understanding of the effects of GCCP on concrete. These groupings were based on the water
213 dosage strategy employed and specific gravity of the GCCP. A specific gravity higher than 2.0
214 g/cm³ was considered high (H), that between 1.7 and 2.0 g/cm³ was considered medium (M) and
215 that lower than 1.7 g/cm³ was considered low (L). The groups were then labelled according to
216 the water dosage strategy and specific gravity classifications. For instance, results obtained for
217 concrete where the total water content was maintained constant when integrating GCCP with a
218 specific gravity higher than 2.0 g/cm³ were assigned to the grouping of “=W H”. The results
219 obtained in studies with a different substitution basis criterion (total volume or weight of
220 conventional aggregate) were separated into different graphs when sufficient amount of results
221 were available. For some of the analysed properties, results for all the possible combinations
222 between the different water dosage strategies and the specific gravity ranges could not be found

223 in literature. In those cases, more research should be developed in order to fully understand the
224 influence of these parameters.

225 5.1 Fresh properties

226 The fluidity of concrete decreases when the GCCP content increases and the =W strategy is
227 applied (**Fig. 11**), mainly due to the high water absorption capacity of GCCP. However, the
228 decrease in the workability can be slight when using a highly viscous paste with a low water to
229 binder ratio and high amounts of supplementary cementitious materials [72]. Under these
230 conditions, it is difficult for the mixing water to be absorbed by the GCCP particles. When the
231 WAC strategy is applied, higher fluidity was obtained in some studies, whereas others achieved
232 lower fluidity than the baseline mix (**Fig. 11**). These discrepancies were caused by the difficulty
233 of precisely determining the water absorption by particles (as explained in Section 3.1). The
234 workability may decrease if this characteristic of the particles is underestimated due to ongoing
235 absorption in the fresh state, so the mixing water content is reduced. In contrast, water
236 desorption from pre-wetted particles or insufficient absorption can increase the fluidity when
237 the GCCP water absorption capacity is overestimated. In addition to the effects of the selected
238 water dosage strategy, the irregular shape of GCCP contributes to reducing the workability of
239 concrete. Furthermore, it was reported that the fluidity increased when using rounded GCCP [51]
240 (these results are not included in **Fig. 11**).



241 **Fig. 11:** Relative variations in the fluidity when replacing conventional aggregate with GCCP by volume (a)
242 [51,55,115,56,57,60,63,68,72,85,94] and by weight (b) [62,65,119].

241 The segregation risk also decreases in self-compacting concretes when incorporating GCCP
242 without compensating for its water absorption [96,112]. Bleeding follows a similar trend to
243 fluidity, where it increases when extra mixing water is added to compensate for water absorption
244 by the GCCP [62,70,123], whereas it decreases without compensation for the water absorption
245 by the GCCP [36,115]. Another important property related to the workability of concrete is its
246 setting time, where GCCP can result in slightly longer initial and final setting times compared with
247 the reference concrete [70].

248 5.2 Microstructure and density

249 GCCP is a porous material and it interacts with the paste in different ways depending on the
250 water dosage strategy employed. This interaction can make the porosity of the paste increase or
251 decrease, which usually has a detrimental effect.

252 In general, the number of pore spaces increases in the concrete paste as the GCCP content
253 increases (**Fig. 12** & **Fig. 13**). This effect is more remarkable if the +W strategy is used [57,63], as

54 well as when the WAC strategy is employed and the excessively pre-wetted GCCP desorbs water
 55 while mixing [63,94]. The water to binder ratio increases in both cases.

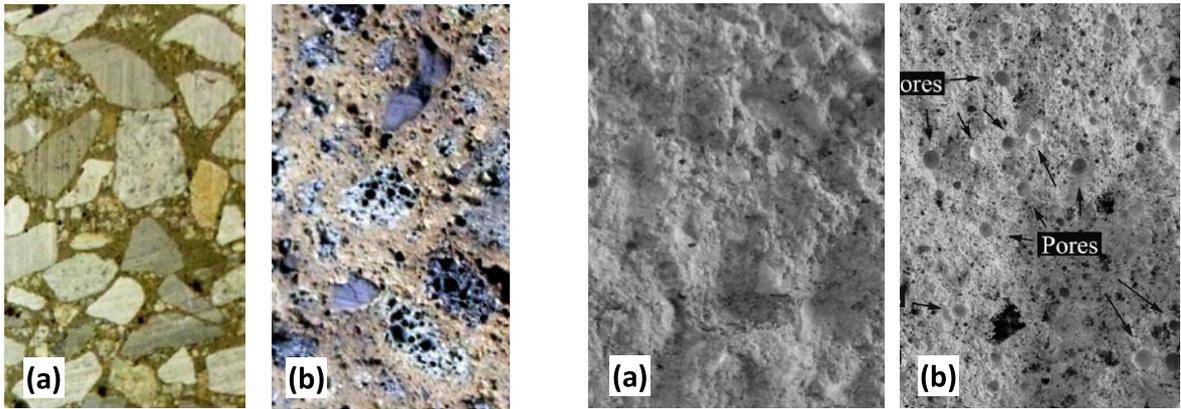


Fig. 12: Cut surface of a geopolymer without GCCP (a) and with all its aggregates substituted by GCCP [77].

Fig. 13: Fracture surface of mortar without GCCP (a) and with all the sand substituted by GCCP (b) [57].

256 By contrast, if the =W strategy is used, the absorption of water by the GCCP reduces the water
 257 to cement ratio in the paste reducing the porosity of the concrete [87]. However, this
 258 phenomenon can also have another effect, as reported in some studies [57,72]. When the GCCP
 259 absorbs water, the pores in its particles release air bubbles. These bubbles remain in the concrete
 260 if the paste fluidity is not sufficiently high or adequate compaction is not applied, and thus they
 261 can migrate to the surface of the material. This process increases the paste porosity but does not
 262 increase the concrete porosity because the pore volume only changes its location from the
 263 aggregate to the paste. Furthermore, the increase in the paste viscosity caused by water
 264 absorption by the GCCP can hinder concrete compaction and promote air entrapment as a
 265 consequence [87]. However, the convenient penetration of the paste into the GCCP macro-pores,
 266 i.e., good interlocking, has been detected when the paste is sufficiently fluid (Fig. 14). It should
 267 be noted that the promotion of interlocking might not occur if the GCCP particles are rounded in
 268 shape [50].

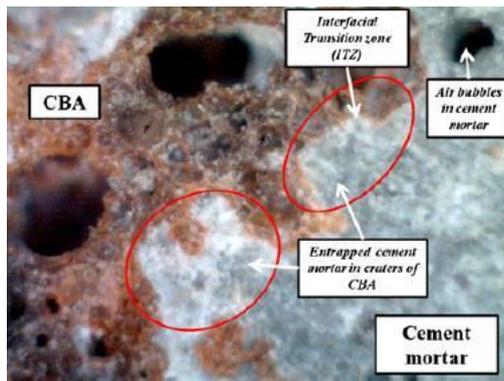


Fig. 14: Cross section of hardened concrete with CBA [72].

269 All of the phenomena mentioned above modify the porosity of concrete via physical interactions,
 270 but chemical interactions have also been found in some studies. For instance, the desorption of
 271 water by GCCP particles after setting can contribute to paste densification due to sustained
 272 cement hydration and pozzolanic reactions, particularly in the interfacial transition zone because
 273 of the presence of reactive silica on the surface of the GCCP (Fig. 15). However, a poor quality
 274 interfacial transition zone has also been detected [62].

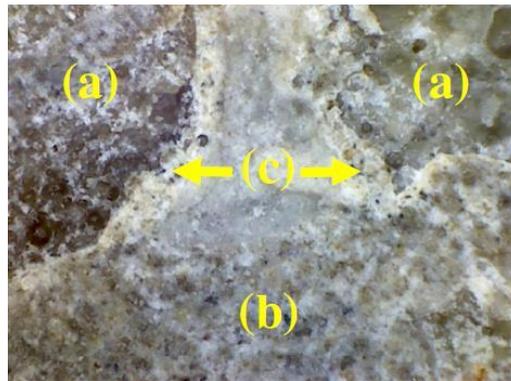


Fig. 15: Microscopic image of the interface between a bottom ash aggregate and cement paste at 200× magnification [114]. The interface between (a) the bottom ash aggregate and (b) cement paste is densified by (c) gel products due to pozzolanic reactions.

275 The density of concrete decreased as the GCCP content increased in all of the studies considered.
 276 The main factor that governs the reduction in the density of concrete appears to be the specific
 277 gravity of GCCP. No significant differences were found between concretes produced with or
 278 without compensating for the absorption of water (**Fig. 16 a**). However, the +W strategy caused
 279 higher reductions in the density (**Fig. 16 b**).

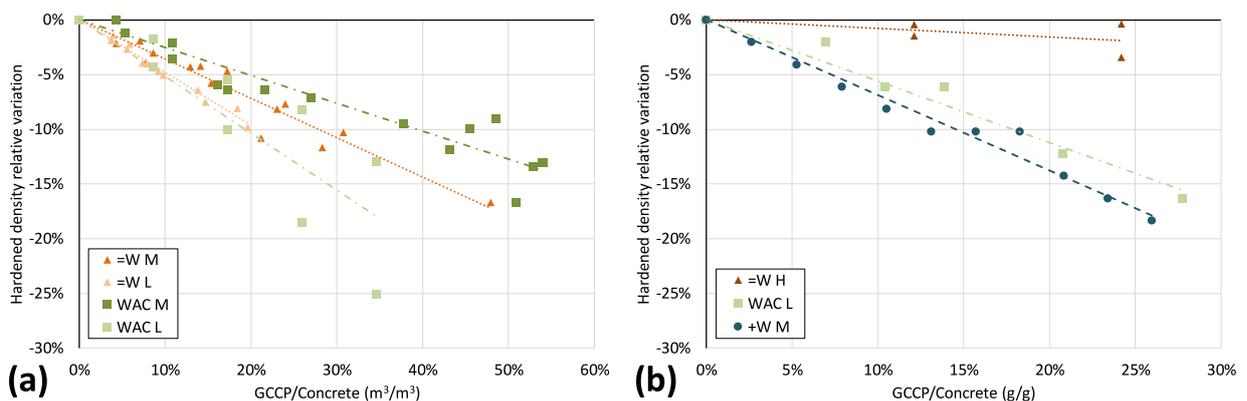


Fig. 16: Relative variations in the density when replacing conventional aggregate with GCCP by volume (a) [57,63,67,72,85,86,94,115] and by weight (b) [9,50,62,87,112].

280 The effects of GCCP on the microstructure, porosity and density of concrete directly influence its
 281 mechanical properties, volume stability and durability, as discussed in Sections 5.3, 5.4 and 5.5.

282 5.3 Mechanical properties

283 The basic mechanical properties of concrete, i.e., compressive, splitting-tensile and flexural
 284 strength and modulus of elasticity, generally decrease as the GCCP content increases. However,
 285 negligible reductions were obtained at low substitution rates (< 25%) and higher values were
 286 even determined in some studies (**Fig. 17a & Fig. 17b**).

287 The decreases in the compressive strength are more pronounced when GCCP with a lower
 288 specific gravity is used because a smaller particle density normally implies lower particle strength
 289 according to many studies [57,62,87,96]. However, the water dosage strategy is also important
 290 because of its influence on the paste properties [62,94]. The application of the +W strategy
 291 clearly aggravates the decrease in the strength, whereas no great differences are found between
 292 concrete produced using the =W or WAC strategies. Substitution by weight boosts any effect on
 293 strength because it implies an increase in the total aggregate content (**Fig. 17b**). The effects of

94 the characteristics of the interfacial transition zone on the mechanical properties are difficult to
 95 evaluate. Some studies have suggested that they contribute to the densification of concrete (see
 96 Section 5.2), but this effect might be minor compared with the other negative effects of GCCP on
 97 concrete.

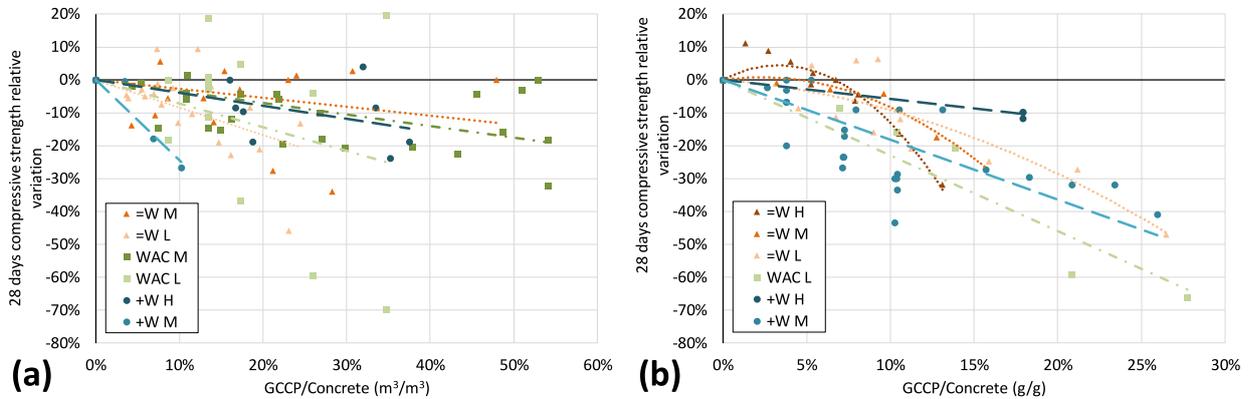


Fig. 17: Relative variations in the 28-day compressive strength when replacing conventional aggregate with GCCP by volume (a) [51,55,85,86,94,95,99,114–116,118,123,57,59,66,67,70,72,78,83] and by weight (b) [9,50,121,124–126,52–54,62,65,88,96,119].

298 Early strength development, i.e., the $\frac{CS7}{CS28}$ ratio, is lower in concrete containing GCCP (**Fig. 18a**),
 299 which has been attributed to the existence of late pozzolanic reactions [69,70,121] and hydration
 300 sustainment via water desorption (internal curing) [55,127]. The higher water content of mixes
 301 produced using the +W strategy also leads to lower strength development during the first 7 days
 302 because the higher water to cement ratio delays paste setting and early hardening [128,129].
 303 When a conventional aggregate is substituted by weight, the trends are more difficult to establish
 304 because the aggregate to paste ratio is modified (**Fig. 18b**).

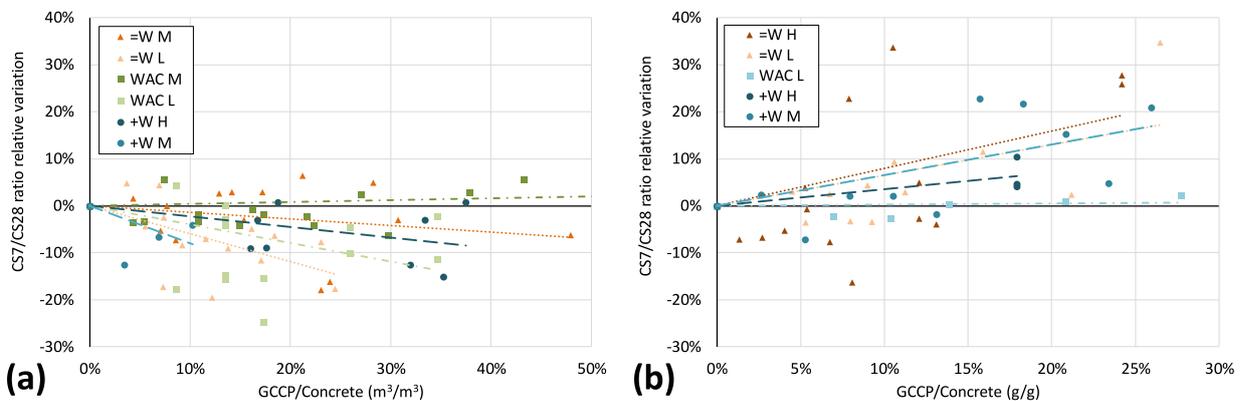


Fig. 18: Relative variations in the 7-day to 28-day compressive strength ratio when replacing conventional aggregate with GCCP by volume (a) [51,57,95,99,115,118,123,130,66,67,70,72,78,83,85,86] and by weight (b) [9,50,125,52,54,62,65,88,96,121,124].

305 The modulus of elasticity follows a similar trend to the compressive strength, where the modulus
 306 of elasticity is lower when the specific gravity of the GCCP is lower (**Fig. 19a**). This effect is
 307 explained by the usually greater deformability of less dense aggregates. Moreover, the +W
 308 strategy has a clear detrimental effect. These trends are less pronounced when the substitution
 309 is applied by weight (**Fig. 19b**). In this case, the stiff conventional aggregates are replaced by weak
 310 GCCP, but part of the paste is normally more deformable than the aggregates, and even more
 311 than GCCP. Thus, the effect of substitution on the paste counteracts the effect of substitution of
 312 the conventional aggregate, thereby resulting in less drastic decreases in elasticity.

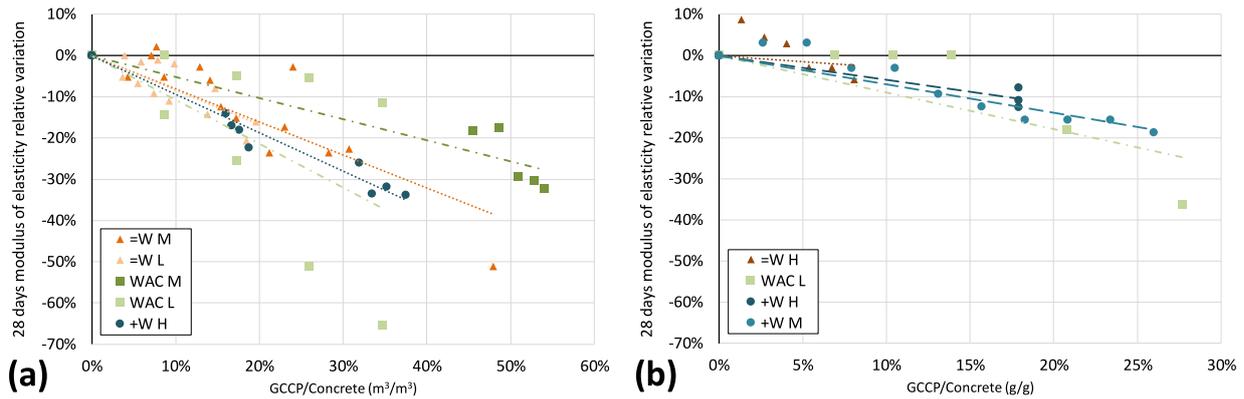


Fig. 19: Relative variations in modulus of elasticity when replacing conventional aggregate with GCCP by volume (a) [59,67,118,70,72,85,86,94,99,115,116] and by weight (b) [50,62,65,121,124].

313 In general, the splitting-tensile and flexural strength of concrete decrease when incorporating
 314 GCCP (**Fig. 20** and **Fig. 21**). However, some studies found performance improvements, possibly
 315 due to better paste quality because of the occurrence of pozzolanic reactions [65] or to the
 316 blockage of cracks because of the presence of pores in the paste of bottom ash concrete [115].
 317 GCCP particles with a lower specific gravity yield worse results. Furthermore, the effect of the
 318 water dosage strategy might not be the same as that on the compressive strength. In particular,
 319 it seems that the +W strategy leads to higher flexural strength than the =W strategy.

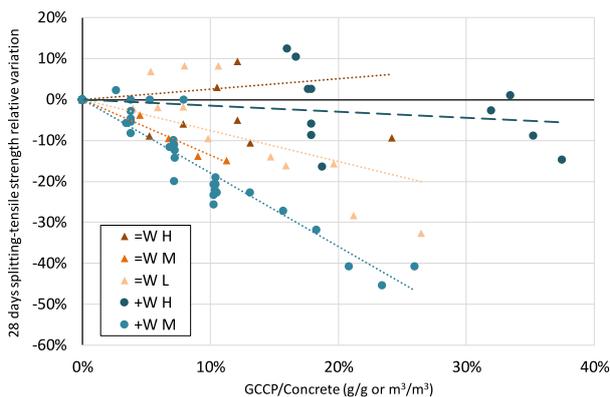


Fig. 20: Relative variations in 28-day splitting-tensile strength when replacing conventional aggregate by GCCP [9,50,125,126,52,54,69,70,83,85,120,121].

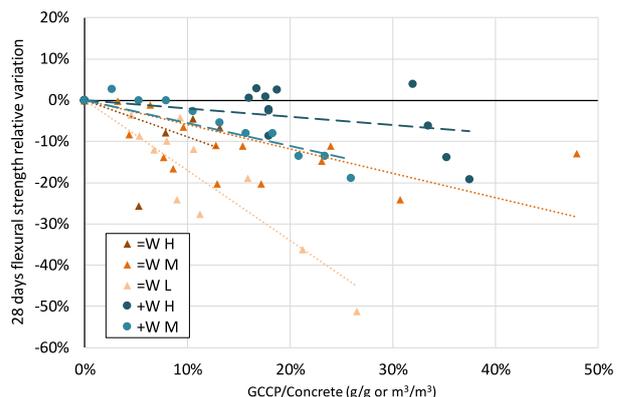


Fig. 21: Relative variations in 28-day flexural strength when replacing conventional aggregate by GCCP [50,52,54,69,70,72,119,121,124,125].

320 The wear resistance decreases when GCCP is integrated in concrete (**Fig. 22** & **Fig. 23**) because
 321 of its lower strength compared with conventional aggregates and the effects of water absorption
 322 or desorption on the paste quality [70,85,111,116].

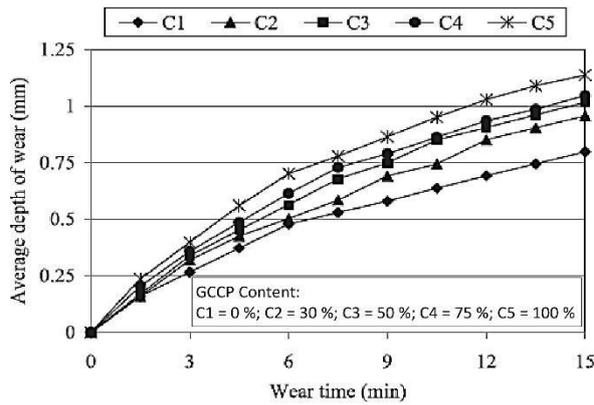


Fig. 22: Depth of wear (mm) [116].

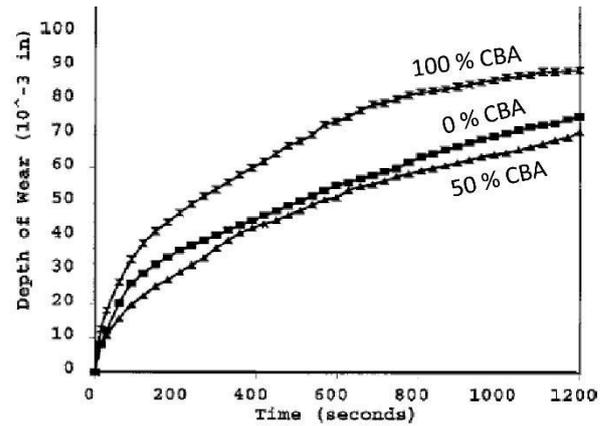


Fig. 23: Depth of wear (10^{-3} in) [70].

323 5.4 Volume stability

324 Different types of shrinkage can be affected when GCCP is integrated in concrete. Any
 325 deformation might be boosted because of the presumably lower elasticity of GCCP particles, but
 326 some reductions can occur depending on the water dosage strategy selected. For instance,
 327 autogenous shrinkage decreases if compensation for water absorption is implemented [55,78],
 328 as it can be seen in Fig. 24. Furthermore, a higher water loss is registered in air-drying conditions
 329 if the selected water dosage strategy increases the total amount of water in the concrete. This
 330 water loss leads to drying shrinkage. Therefore, greater shrinkage is generally found in air-drying
 331 conditions when using GCCP [70,78,96,115,118,123]. However, autogenous and drying shrinkage
 332 can coexist in air-drying conditions depending on the concrete water to cement ratio,
 333 environmental humidity and size and shape of the specimens. Therefore, compensation effects
 334 have led to decreased shrinkage in some studies [58,60,95,99,123]. Some examples of the
 335 different shrinkage results obtained in air-drying conditions are shown in Fig. 25 and Fig. 26.
 336 Finally, concrete absorbs more water when the GCCP content is higher (see Section 5.5.1),
 337 thereby leading to more swelling in water immersion conditions [69,70,78], as shown in Fig. 27.

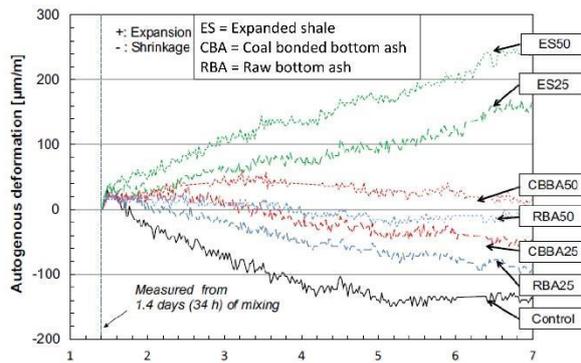


Fig. 24: Autogenous shrinkage [55].

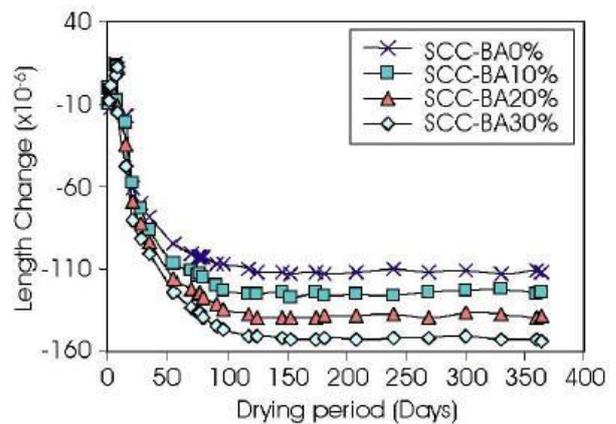


Fig. 25: Shrinkage in air-drying curing conditions [96].

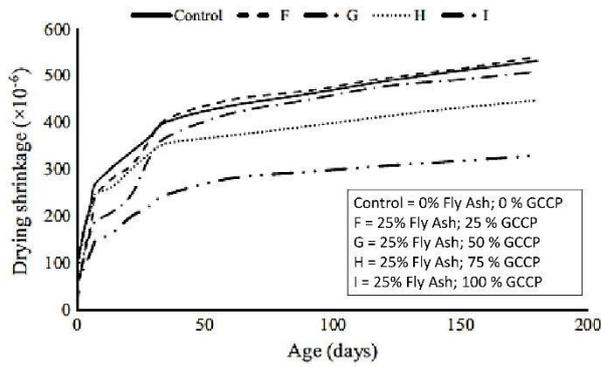


Fig. 26: Shrinkage in air-drying curing conditions [95].

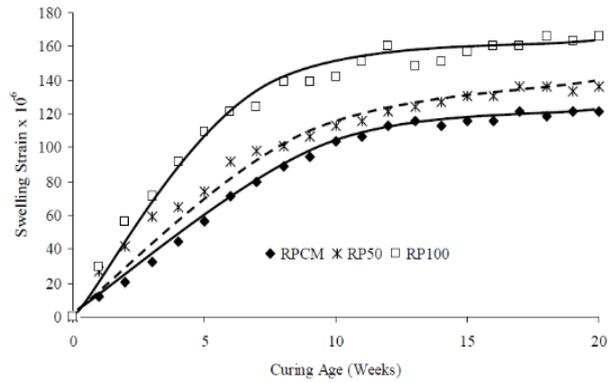


Fig. 27: Swelling [69].

338 **5.5 Durability**

339 Previous studies have focused on durability to obtain important information about the long-term
 340 performance of concrete containing GCCP. The permeability and resistance to different harmful
 341 external agents are the properties investigated most frequently.

342 **5.5.1 Permeability and absorption**

343 The higher porosity of GCCP concrete leads to greater permeability to any external agent. For
 344 instance, the use of GCCP results in higher gas permeability [63] and this can lead to a higher
 345 carbonation depth (Fig. 28). The increased porosity due to the inclusion of GCCP also enhances
 346 the capillary water uptake [85,111,115,116] and water penetration [50]. However, results similar
 347 to the reference are obtained with low substitution rates (Fig. 29 & Fig. 30).

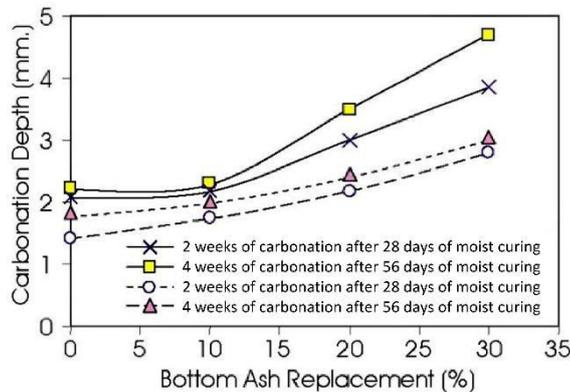


Fig. 28: Carbonation depth [96].

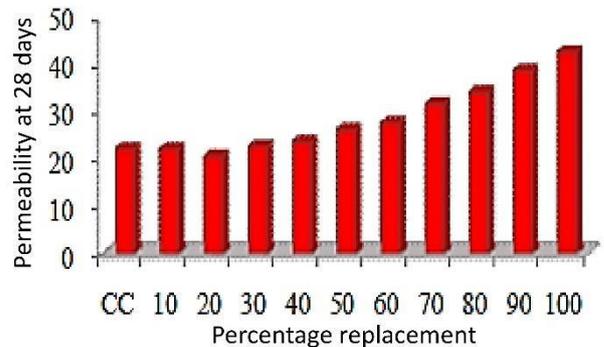
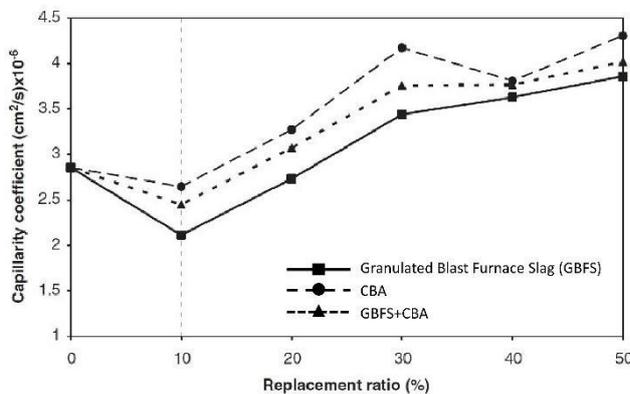


Fig. 29: Water absorption by capillary action [111].

Fig. 30: Water penetration [50].

48 The water absorption of concrete with GCCP is higher than the absorption of the conventional
 49 concrete, especially when the specific gravity of the GCCP employed is low (Fig. 31). If the =W
 50 strategy is applied, the lower water to cement ratio results in a denser paste and lower water
 51 absorption. Imperfect aggregate to paste interlocking can contribute to greater porosity and
 52 water absorption. Furthermore, a potential relationship can be established between a cement-
 53 based material and its water absorption, which might differ depending on the paste content (Fig.
 54 32).

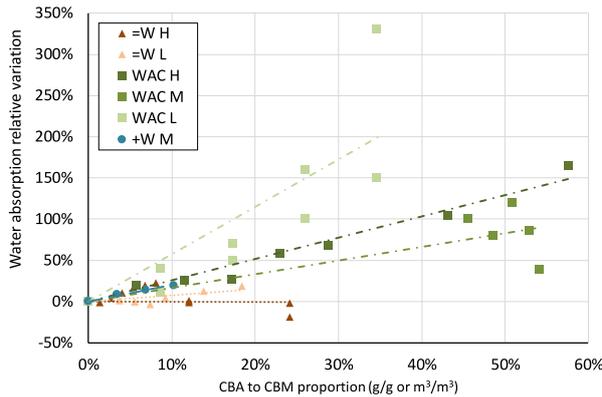


Fig. 31: Relative variations in water absorption when replacing conventional aggregate by GCCP [9,63,65,83,86,94,99,115,116].

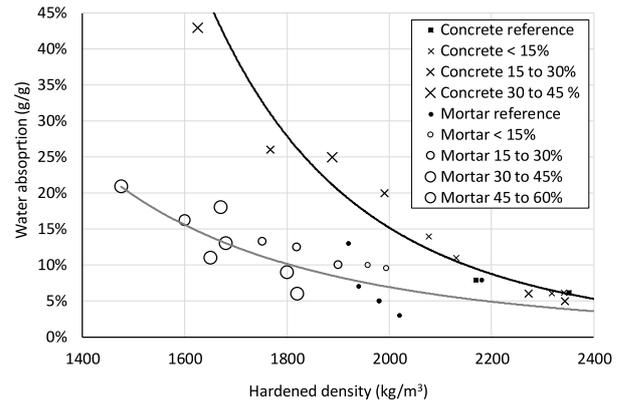


Fig. 32: Relationship between hardened density and water absorption for cement-based materials with different GCCP contents [9,63,86,94].

355 Chloride ion permeability is another durability-related property that is strongly linked with
 356 porosity. At early ages, the use of GCCP leads to a significant increase in permeability. However,
 357 the differences with various substitution rates tend to decrease over time, and better
 358 performance may be obtained using concrete containing GCCP in some cases (Fig. 33 & Fig. 34).
 359 These late improvements are explained by paste densification due to internal curing and
 360 pozzolanic reactions [96,114].

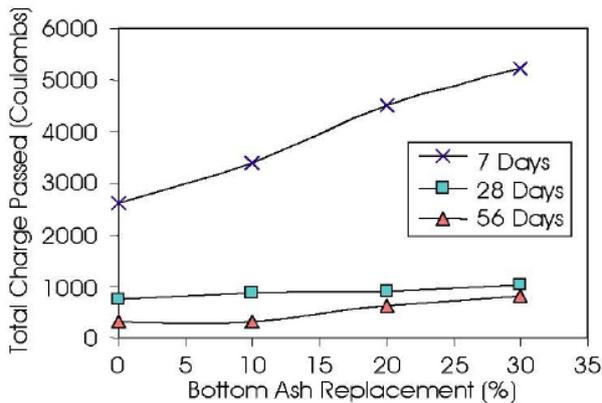


Fig. 33: Chloride ion permeability [96].

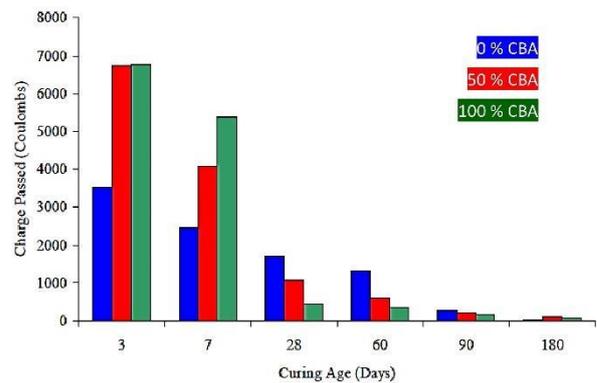


Fig. 34: Chloride ion permeability [69].

361 5.5.2 Resistance to chemical attacks

362 In addition to carbon dioxide, water or chloride ions, other external chemical agents can
 363 penetrate into concrete and damage it. Contradictory results have been obtained in various
 364 studies regarding the effect of the GCCP content on sulphate attack resistance. Higher expansions
 365 were detected in some studies [69,70,99], thereby indicating worse performance, whereas

66 others found lower expansions [96]. Examples of both possibilities are shown in Fig. 35 and Fig. 36.

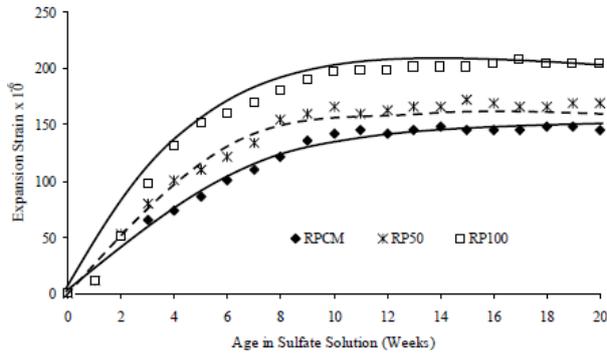


Fig. 35: Expansion of concrete when submerged in a sulphate solution [69].

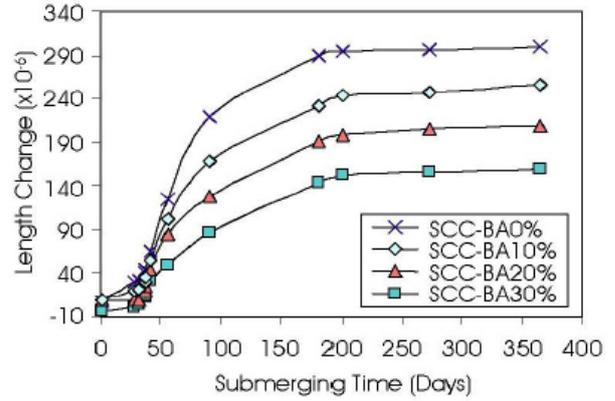


Fig. 36: Expansion of concrete when submerged in a sulphate solution [96].

368 Similar acid attack resistance performance (Fig. 37) or even better (Fig. 38) can be obtained by
369 incorporating GCCP in concrete [66][99].

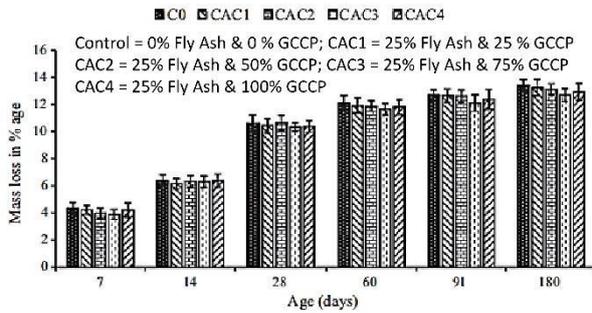


Fig. 37: Mass loss of concrete when submerged in an acid solution [66].

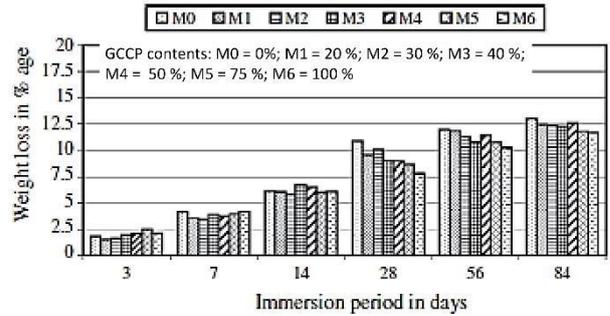


Fig. 38: Mass loss of concrete when submerged in an acid solution [99].

370 5.5.3 Resistance to exposure to high temperature

371 GCCP can contain a certain amount of unburnt coal, so concrete produced using this sub-product
372 may respond poorly when subjected to high temperatures (> 800°C), as confirmed by several
373 studies (Fig. 39). However, better performance was obtained at low substitution rates in other
374 studies (Fig. 40).

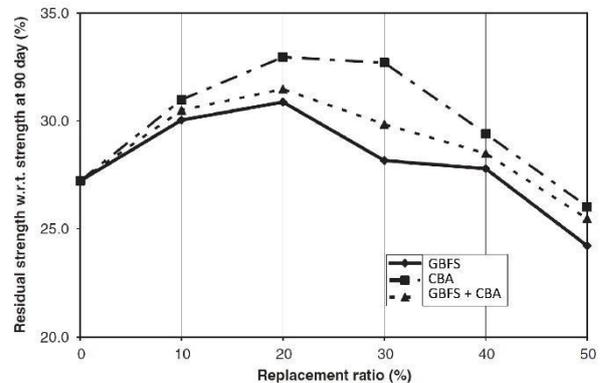
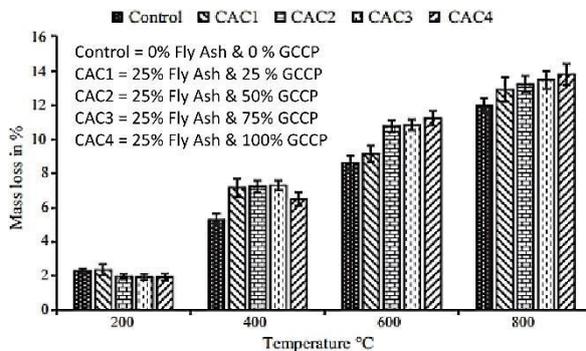


Fig. 39: Mass loss of concrete after exposure to high temperature [66].

Fig. 40: Mass loss of concrete after exposure to high temperature [111].

75 **5.5.4 Resistance to freezing and thawing**

76 The resistance to freezing and thawing decreases when incorporating GCCP [30,69,121].
 77 However, similar results to the reference or acceptable values can be obtained with low
 78 substitution rates (Fig. 41 & Fig. 42).

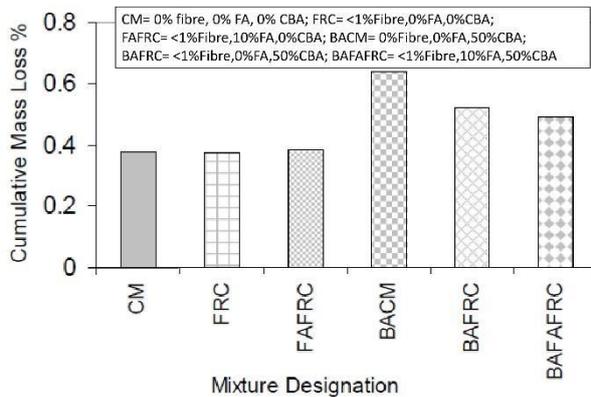


Fig. 41: Cumulative mass loss after 50 freezing and thawing cycles [121].

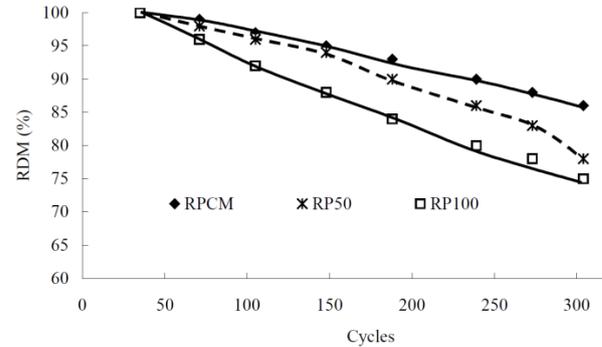


Fig. 42: Decrease in the relative dynamic modulus (RDM) after freezing and thawing cycles [69].

379 **6 Discussion of the applications of concrete with GCCP**

380 The use of GCCP as an alternative aggregate in concrete has important environmental and
 381 economic advantages. First, it helps to reduce the consumption of natural and quarry sand.
 382 Therefore, the impacts related to the exploitation of these resources are decreased. Less virgin
 383 resources are depleted and mining is reduced, which is an activity with high social, environmental
 384 and economic costs. Second, the beneficial use of GCCP instead of its management as a waste
 385 material reduces the use of landfill space, as well as the risks and costs associated with the
 386 disposal process [5]. In addition to these advantages, the integration of GCCP in cement-based
 387 materials can potentially improve their technical performance because of its special
 388 characteristics. In particular, low specific gravity and a high capacity for water absorption are the
 389 most distinctive properties of GCCP, and they allow researchers to propose advanced
 390 applications such as lightweight concrete and internally cured high performance concrete.

391 The low specific gravity of GCCP particles (1.2–2.3 g/cm³) might be useful for decreasing the
 392 weight of concrete by substituting for normal weight conventional aggregates. However, studies
 393 suggest that integrating this waste material is rarely sufficient to satisfy the density and
 394 compressive strength requirements for lightweight structural concrete (< 2000 kg/m³ and > 15
 395 MPa, respectively [131]). The requisite conditions can only be satisfied by mixes with the highest
 396 GCCP contents or those that include other lightweight aggregates (Fig. 43).

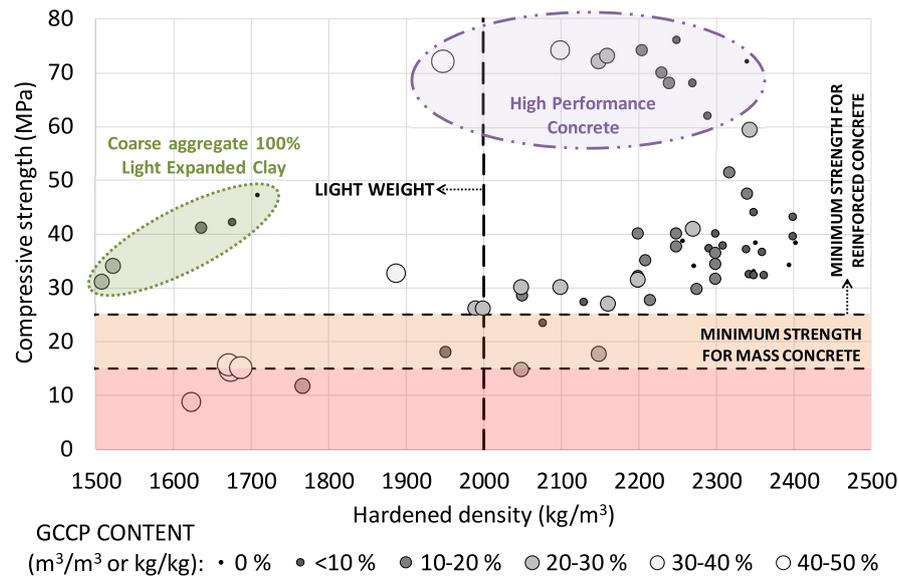


Fig. 43: Relationship between hardened density and compressive strength of concrete with different GCCP contents [9,50,57,62,67,72,77,85,86,115].

397 In addition, GCCP can be used to improve high performance concrete, which is usually
 398 characterised by a low water to binder ratio and it can be affected adversely by considerable
 399 autogenous shrinkage. This deformation is the self-created bulk strain generated in cement-
 400 based materials during hardening at constant temperature [132] and the driving force
 401 responsible is paste self-desiccation [133,134]. One strategy for mitigating paste self-desiccation
 402 and reducing autogenous shrinkage is internal curing. ASTM C1761 defines internal curing as the
 403 supply of water within a cementitious mixture using pre-wetted lightweight aggregate or other
 404 materials that readily release water from within the particles, thereby mitigating self-desiccation
 405 and sustaining hydration [135]. The particles that can release water inside concrete are known
 406 as internal curing water reservoirs and they can be wet aggregates with high water absorption
 407 and desorption capacities. These requirements can generally be satisfied by GCCP (water
 408 absorption varies from 5% to 35%), so its use as an internal curing reservoir in high performance
 409 concrete is considered to be a promising advanced application [68]. Furthermore, low GCCP
 410 contents are sufficient to achieve effective internal curing due to the remarkably high water
 411 absorption capacity of GCCP, thereby reducing any detrimental effects on the mechanical
 412 properties. Similar to any other internal curing reservoir, GCCP can be wetted before mixing or
 413 added dry to the mix so it can absorb water from the concrete paste in the fresh state and then
 414 release it after setting. Some studies suggest that these two procedures lead to similar concrete
 415 properties [58] but the first method is preferred. The water absorption process is more efficient
 416 when the aggregate particles are exposed to water, e.g., by water immersion, compared with
 417 that when they are integrated in concrete to absorb water from the paste. Moreover, pre-wetting
 418 GCCP allows the wetting process to be extended for a long time so conditions closer to saturation
 419 can be achieved. In addition, specific procedures such as vacuum saturation can be applied in a
 420 pre-wetting scenario.

421 **Fig. 44** presents the coal combustion products, their distinctive properties, the processes applied
 422 for them to be used in different applications, the most common mix design strategies and their
 423 effects on the performance of concrete.

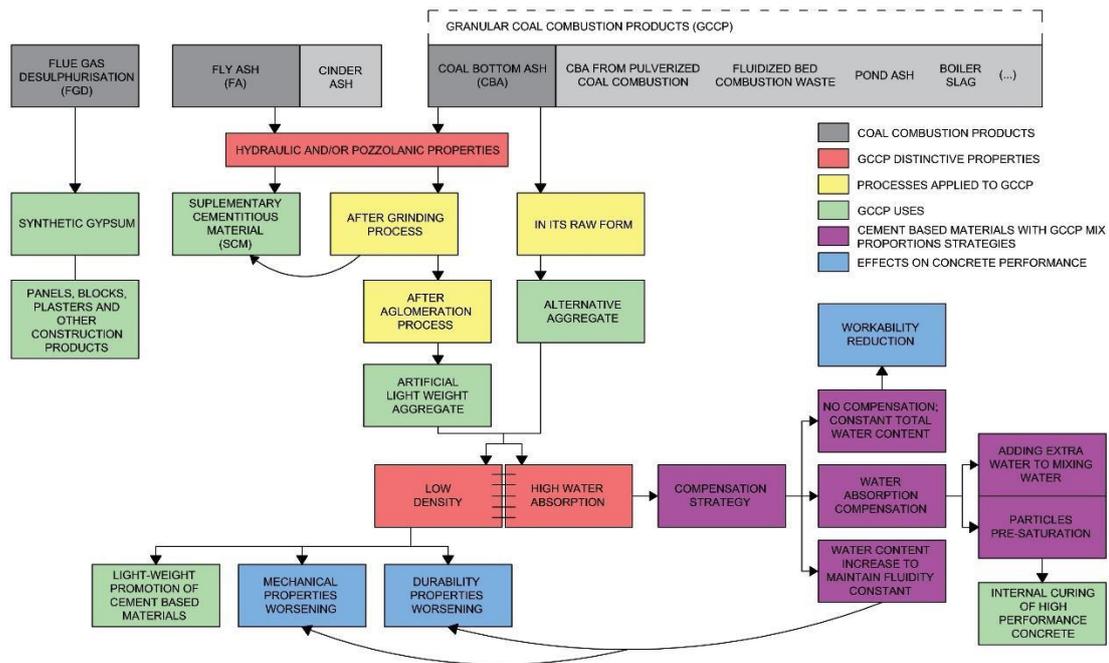


Fig. 44: GCCP in construction and cement-based materials.

424 To sum up, the comprehensive review of the scientific literature supports the feasibility of
 425 integrating GCCP in concrete. The authors-created graphs can help to understand the influence
 426 of GCCP in different properties, depending on the water dosage strategy used and the specific
 427 gravity of the by-product. The analysis of the collated findings permits to conclude that this by-
 428 product can be used to decrease the weight of concrete and also to work as an internal curing
 429 water reservoir therefore reducing the self-desiccation of mixes with low water to binder ratios.

430 7 Conclusions and recommendations

431 The following conclusions can be drawn based on this analysis of the results obtained in more
 432 than 100 studies of the use of GCCP as aggregates in concrete. The general recommendations
 433 based on these conclusions may provide useful insights to facilitate future research or field
 434 studies.

- 435 1. In general, GCCP particles are physically characterised by an irregular shape and rough
 436 texture. Moreover, the specific gravity of GCCP, i.e., approximately 1.2 to 2.3 g/cm^3 , is lower
 437 than that of conventional aggregates and its water absorption capacity (from 5% to 35%) is
 438 higher than that of conventional aggregates, with rapid kinetics. These key characteristics
 439 must be carefully evaluated before using GCCP as an aggregate in any cement-based
 440 material. However, their correct determination is challenging and different procedures can
 441 lead to inaccurate results.
- 442 2. The chemical composition of GCCP is highly dependent on the coal source, where it mainly
 443 comprises silicon (Si), aluminium (Al) and iron (Fe), which can be combined in the forms of
 444 quartz and mullite. The presence of reactive silica on the surface of GCCP results in
 445 potentially pozzolanic activity. Some minor potentially hazardous elements are also present.
- 446 3. GCCP has been used as an aggregate substitute in many different cement-based materials.
 447 Conventional concrete is the most popular material but the integration of GCCP is also
 448 feasible in different types of high performance and self-compacting concrete. GCCP has
 449 mostly been employed as a fine aggregate but descriptions of the use of GCCP as a coarse
 450 aggregate have also been reported.

- 451 4. The amount of GCCP used in order to replace conventional aggregate should be established
452 based on the volumetric proportion, i.e., by considering its specific gravity. If this method is
453 not used and aggregates are substituted by weight, the concrete aggregate to paste ratio
454 can increase, thereby resulting in reduced workability, strength and durability.
- 455 5. The water absorption capacity of GCCP should be considered when designing a concrete
456 mix. Three different water dosage strategies can be employed comprising constant total
457 water content (=W), water absorption compensation (WAC) and water increase so fluidity
458 is maintained constant (+W). Each one has different effects on the properties of concrete,
459 but the second is generally preferred because it does not greatly affect the workability and
460 it avoids modifying the water to cement ratio.
- 461 6. If the =W strategy is applied, the workability of concrete decreases because of the ongoing
462 absorption of water by GCCP. If the WAC strategy is applied, the workability can decrease
463 or increase due to underestimation or overestimation of the water absorption capacity. The
464 irregular shape of GCCP also contributes to decreases in workability under any strategy.
- 465 7. The porosity of concrete increases with the GCCP content because of the porosity of the
466 particles themselves and the irregularity of the particles that enhances air entrapment.
- 467 8. The mechanical properties of concrete containing GCCP are generally worse than those of
468 conventional mixes due to the weakness of the waste particles and detrimental effects on
469 the paste. Worse results are obtained with less dense GCCP and the +W strategy. Low
470 substitution rates lead to slight decreases in strength. Greater late strength development is
471 usually attributed to pozzolanic activity.
- 472 9. The water absorption capacity and permeability are greater when the GCCP content is
473 higher. GCCP content also increases drying shrinkage and swelling, but decreases
474 autogenous shrinkage. There is no general agreement regarding other durability-related
475 properties due to the lack of reported results, but some studies indicate that GCCP has only
476 slight negative effects on these properties.

477 The low density of GCCP may be advantageous for decreasing the weight of any cement-based
478 material. However, lightweight concrete with acceptable strength is difficult to obtain if other
479 conventional lightweight aggregates are not used in combination with GCCP. Nevertheless, the
480 high water absorption and desorption capacities of GCCP mean that it can satisfy the basic
481 requirements for use as an internal curing water reservoir in high performance concrete.
482 Therefore, this is considered one of most promising advanced applications of GCCP in the field of
483 concrete production. Thus, future research efforts should focus on the less well characterised
484 phenomena related to GCCP. In particular, the following issues should be investigated: suitable
485 procedures for determining the specific gravity and water absorption capacity of GCCP (which
486 could be applicable to other lightweight aggregates), further studies of the leaching risk for the
487 minor potentially hazardous elements present in GCCP, and experimental studies of the influence
488 of GCCP on the durability of concrete.

489 **Declaration of Competing Interest**

490 The authors declare that they have no known competing financial interests or personal
491 relationships that could have appeared to influence the work reported in this paper.

492 **Credit authorship contribution statement**

493 **Roberto Rodríguez-Álvarez:** Conceptualization, Formal analysis, Investigation, Writing – original
494 draft, Visualization. **Sindy Seara-Paz:** Formal analysis, Investigation, Writing - review & editing,

495 Supervision. **Belén González-Fontebo**: Formal analysis, Investigation, Writing - review & editing,
496 Supervision. **Fernando Martínez-Abella**: Formal analysis, Investigation, Supervision.

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502 Technical Research and Innovation 2013-2016, Call 2017.
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504 sustainable materials and products for the eco-construction in the building and civil
505 engineering field (Cenicienta), funded by the Innovation Galician Agency, Galician
506 Government (Xunta de Galicia), FEDER 2014-2020, Call 2016.

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