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Use of gasification char and recycled carbon fibres for sustainable and durable low-resistivity cement-based composites

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Abstract

In this paper, a newly developed cement and two recycled carbon-based materials, namely gasification char (GCH) and recycled carbon fibres (RCF), were used to manufacture new sustainable and low-resistivity cement-based composites. GCH was added at 0.5% and 1.0%, whereas RCF at 0.2%, on the total mortar volume. Microstructural, mechanical, electrical, and durability properties were studied. Results show that both additions decrease the electrical resistivity of mortars up to 90% without impairing their mechanical properties. Moreover, they slightly reduce drying shrinkage and capillary water absorption of mortars, by decreasing total porosity and the average pore diameter, for the benefit of durability.

Keywords

Carbon-based additions; Recycled carbon fibres; Gasification char; Electrical resistivity; Durability.

28 **1. Introduction**

29 The addition of carbon-based materials, in form of fillers and fibres [1], has been demonstrated to be
30 a useful tool to decrease the electrical resistivity of construction materials [2,3]; therefore, their use
31 is spreading in the field of structural health monitoring (SHM) [4,5].

32 Fibres improve the mechanical properties of cement-based composites [6] and, if electrically
33 conductive, they can supply to mortar/concrete other non-structural functions, such as heating
34 properties for indoor floor heating screeds [7] or de-icing for outdoor pavements [8], electromagnetic
35 wave shielding [9], cathodic protection [10], and self-sensing/piezoresistivity properties [11,12]. The
36 use of carbon fibres in construction materials dates back to the 90s [13]; many studies have shown
37 that they are able to decrease the electrical resistivity, hence potentially improving the self-
38 sensing/piezoresistivity properties of concrete even at low dosages [14]. However, virgin carbon
39 fibres (VCF) are generally used in construction materials, whereas recycled carbon fibres (RCF) have
40 been studied only in the last years. Segura et al. [15] highlighted that RCF are able to improve the
41 signal-to-noise ratio (SNR) during flexural and compressive tests. Belli et al. [16] studied three types
42 of fibres as addition in cement-based mortars, namely VCF, RCF, and steel fibres (SF). They found
43 that at the same 0.2% volume dosage, carbon fibres are much more effective for decreasing the
44 electrical resistivity of mortars than SF and that RCF (95% reduction) are also better than VCF (72%
45 reduction). Moreover, the use of RCF instead of VCF is particularly interesting also for their
46 environmental benefits and lower cost [16].

47 Numerous carbon-based fillers have been used in cement-based materials to decrease their electrical
48 resistivity, such as graphene nanoplatelets (GNP), graphene oxide (GO), carbon nanotubes (CNTs),
49 nano-graphite platelets (NGPs) and carbon black (CB). Le et al. [17] found that when GNP exceeds
50 the percolation value (the limit beyond which the additions touch each other), the electrical
51 conductivity is not influenced by the moisture content; while Du and Pang [18] stated that if GNP is
52 used in a quantity higher than the percolation value, mortars with piezoresistive properties can be
53 obtained and the trend under tensile load is reversible. Liu et al. [19] reported that GNP is better than

graphene oxide nanoplatelets (GONP) to enhance electrical conductivity of mortars, whereas Madbouly et al. [20] found that reduced GO at ratios of 0.04–0.05% increased the compressive strength, reduced the porosity and provide a clear piezoresistive behaviour to cement composites. Zhang et al. [21] reported that multi-walled CNTs increase the flexural strength of mortars and reduce the electrical resistivity up to 40% when used at 0.5 wt% of cement; Jung et al. [22] stated that CNTs considerably decrease the electrical resistivity of ultra-high performance concrete (UHPC) regardless of the free water content. Sun et al. [23] found that NGPs decrease the electrical resistivity and provide piezoresistive properties to cement composites with a percolation threshold of about 2 vol%. Monteiro et al. [24] reported that CB added to 4 wt% of cement enhances the mechanical performance of mortars, whilst between 7% and 10% provides a good piezoresistive behaviour. Çınar et al. [25] exploited CB's ability to conduct electricity to speed up the setting time of the concrete. However, the high cost of above-mentioned commercial fillers makes their use unfeasible at industrial scale. The use of low-cost carbon-based fillers is necessary since mortars/concretes are relatively low-cost materials used at high volume in construction sector [26]. Moreover, from a sustainability point of view, the recycling of carbon-based industrial by-products in mortars/concretes provides environmental benefits, in terms of lower CO₂ emissions, decreased landfill procedures and reduced consumption of not renewable materials and energy [27,28].

Recently, gasification char (GCH) has been introduced to pastes [29] and mortars [30] to decrease their electrical resistivity. GCH is the solid by-product obtained by the thermal treatment of wood chips through the gasification process. Similarly to biochar, which instead is obtained by pyrolysis, it is a stable form of carbon characterized by a low amount of heavy metals, a porous structure, widely used in sustainable agricultural practices [31]. GCH is able to decrease by 65% the electrical resistivity of lime-based pastes at a dosage of 0.25% by hydraulic lime weight [29] and by 42% the electrical resistivity of cement-based mortars at a dosage of 1.0 vol% [30]. Moreover, if GCH is coupled to VCF and RCF, the electrical resistivity becomes 76% and 80% lower than that of a reference mortar without conductive additions, respectively [30].

It is also worthy to consider that, besides being less expensive and more sustainable, construction materials should ensure good mechanical strength and durability in service in order to reduce maintenance activities and repairing costs [20]. Therefore, the research reported in this work aims at characterizing for the first time low-resistivity cement-based mortars prepared with a new sustainable low-clinker cement, developed by HeidelbergCement AG in the frame of the H2020 EnDurCrete project (GA no. 760639) and not yet available in commerce, with the addition of low-cost and sustainable additions, such as GCH and RCF, and investigating their effect on durability. To this aim, mortars have been manufactured with GCH (at 0.5% and 1.0% by volume) and RCF (at 0.2% by volume), both as single additions and combined, and characterized in terms of flexural and compressive strengths, microstructure as well as electrical resistivity. The durability of mortars has been investigated in terms of free and restrained drying shrinkage and capillary water absorption. In fact, the former influences the dimensional stability of the material and the cracking probability under restrained conditions [32], whereas the latter is related to the ability of aggressive agents to enter the construction material, thus impairing its durability.

2. Experimental

2.1. Materials

Mortars were manufactured with a new low-clinker cement with the code CEM II/C-M (S-LL), developed by HeidelbergCement AG in the frame of the H2020 EnDurCrete project and not yet available in commerce. It is produced by blending the cement component with separately ground granulated blast furnace slag (GGBFS) and limestone filler; its detailed composition is reported in [33]. A calcareous sand (0-8 mm) was used as aggregate. Sand was mixed with 2 wt% of water prior to the mixing process in order to reach the saturated surface dry (s.s.d.) condition. To optimize the workability of mortars, an acrylic ether superplasticizer (Dynamon SP1, Mapei) was used as admixture. A char obtained by the gasification of natural wood chips (GCH) in a plant located in central Italy equipped with a Holz-Kraft, Spanner Re² GmbH gasifier was used as carbon-based by-

product filler (with density of 2.0 g/cm^3). GCH was previously ground in a ball mill for 60 minutes and then sieved at $75 \text{ }\mu\text{m}$. Recycled carbon fibres (RCF), obtained by mixing all origins carbon and graphite ex-polyacrylonitrile fibres coming from spools of pure carbon fibres, were supplied by Procotex Belgium SA. The technical data of RCF are provided in [34]. Since the supplier reports a variable density range of RCF, between 1.7 and 2.0 g/cm^3 , an average value of 1.85 g/cm^3 has been used to calculate the volume percentages.

112

113 2.2. Methods

114 2.2.1. Preparation of mortars

115 A mortar without GCH and RCF was prepared as reference mixture (REF). Electrically conductive
116 mortars were prepared with GCH at two dosages, namely 0.5% and 1.0% by mortar volume, whereas
117 RCF were added at 0.2% by mortar volume. GCH and RCF were added both alone and together to
118 evaluate their combined effect on mortar properties. All mortars were prepared with the same
119 water/cement (w/c) and sand/cement (s/c) ratios of 0.45 and 3 by weight, respectively. The mix-
120 design of mortars is reported in Table 1, together with their flow values obtained by means of a flow
121 table according to the UNI EN 1015-3 standard.

122

123 **Table 1.** Mix design of mortars and flow value

Mortar	Cement (kg/m^3)	Sand (kg/m^3)	Water (kg/m^3)	Superplasticizer (kg/m^3)	GCH (kg/m^3)	RCF (kg/m^3)	Flow (mm)
REF	523	1570	235	4.12	-	-	230
0.2RCF	520	1560	234	8.30	-	3.70	225
0.5GCH	520	1559	234	5.56	10.30	-	225
0.5GCH+0.2RCF	517	1550	232	9.70	10.30	3.70	220
1.0GCH	516	1548	232	7.71	20.60	-	250

1.0GCH+0.2RCF	514	1542	231	9.79	20.60	3.70	240
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124

125 Mortars were cured in a climatic chamber at relative humidity (RH) of $(95 \pm 5) \%$ and temperature
126 (T) of $(20 \pm 1) ^\circ\text{C}$ by covering the moulds with plastic sheets for the first week. Then, the plastic
127 sheets were removed and specimens were left at $\text{RH} = (50 \pm 5) \%$ and $T = (20 \pm 1) ^\circ\text{C}$ until 28 days
128 of curing, if not differently specified.

129

130 2.2.2. Mechanical tests

131 In order to investigate the development of strength in time, the flexural (R_f) and compressive (R_c)
132 strengths were measured on 40x40x160 mm mortar specimens after 2, 7, and 28 days of curing.
133 Mechanical tests were carried out on three specimens according to the UNI EN 1015-11 standard and
134 the average results reported.

135 The dynamic modulus of elasticity (E_d) was also measured on three mortar specimens (40x40x160
136 mm) according to the UNI EN 12504-4 after 28 days of curing. The formula used to calculate E_d is
137 given in Eq. (1):

$$138 \quad E_d = \frac{v^2 d [(1 + \gamma_d)(1 - 2\gamma_d)]}{(1 - \gamma_d)} \quad (1)$$

139 where v is the ultrasonic pulse velocity (UPV) (in m/s), d is the hardened density (in kg/m^3) and γ_d is
140 the Poisson's modulus. The Poisson's modulus, equal to 0.20, was estimated as the average value
141 between 0.15 and 0.25 reported in literature for cement-based materials [35].

142

143 2.2.3. Microstructural analysis

144 The pore distribution, the total porosity (V_p), and the average pore diameter (d_p) of mortars were
145 investigated through a mercury intrusion porosimeter (MIP, model Thermo Fisher 240 Pascal) on
146 small mortar fragments of approximately 1 cm^3 after 28 days of curing. The pore size that can be
147 measured with this system ranges between 0.0037 and $7.500 \mu\text{m}$.

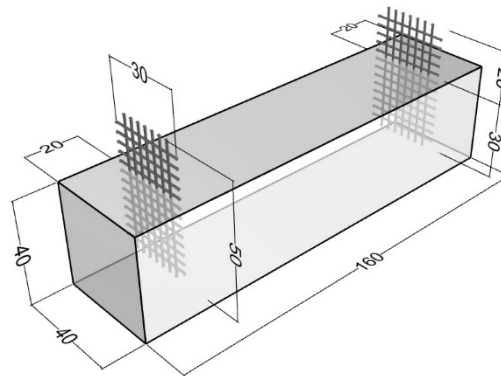
148 The interface between cement matrix and fibres was observed by using a SEM PHILIPS XL20.

149

150 2.2.4. Electrical resistivity tests

151 Electrical resistivity (ρ) of mortars was determined by means of AC two-probes approach on
152 40x40x160 mm specimens after 28 days of curing. The specimen configuration is reported in Figure
153 1 and the complete methodology used for the AC measurements is explained in [16] and in [30].

154



155 Figure 1. Specimen and electrodes configuration for AC electrical resistivity measurements
156 (dimensions are in mm)

157

158 The ρ value has been calculated as reported by Mobili et al. [30], by considering a resistive electrical
159 behaviour of the tested specimen. The effect of carbon-based materials with different aspect ratios on
160 the electrical resistivity of cement-based composites depends on the frequency [30], carbon fibres
161 decrease the electrical resistivity of mortars at high frequencies, whereas GCH decrease the electrical
162 resistivity of mortars at medium frequencies. Therefore, the electrical resistivity was determined both
163 at medium frequency ranges ($50 \div 100$ Hz) and high frequency range ($100 \div 10^6$ Hz). Particularly, in
164 REF, 0.5GCH and 1.0GCH specimens the electrical resistivity was obtained at medium frequencies,
165 whereas in specimens containing RCF, namely 0.2RCF, 0.5GCH+0.2RCF, and 1.0GCH+0.2RCF,
166 the electrical resistivity was obtained at high frequencies.

167

168 2.2.5. Capillary water absorption test

169 Capillary water absorption plays a key role in durability and long-term performance of cement-based
170 construction materials [36], since water is the main carrier of aggressive ions (Cl^- , SO_4^{2-} , etc.) [37,38],
171 facilitates the growth of germs and moulds [39,40], and can degrade the material itself. The capillary
172 water absorption of each mortar was tested on three prismatic specimens (40x40x80 mm) after 28
173 days of curing according to the UNI EN 15801 standard. At first, specimens were dried at $T = (60 \pm$
174 $2)^\circ\text{C}$ until a constant weight was reached, and then tested. The average values obtained in terms of
175 water absorbed per unit area (Q_i) are reported.

176

177 *2.2.6. Drying shrinkage tests*

178 Hygrometric shrinkage greatly affects the durability of cement-based materials, since in the presence
179 of constraints it promotes internal stresses which can lead to cracking [41]. In this work, the effect of
180 carbon-based additions on the drying shrinkage of mortars was evaluated both as free and restrained
181 shrinkage. The free drying shrinkage (ϵ_d) was measured according to the standard UNI EN 12617-4
182 on three not reinforced mortar specimens (40x40x160 mm) for 56 days, when also the weight loss
183 caused by water evaporation was measured. The restrained drying shrinkage (ϵ_r) was measured
184 according to the UNI EN 8147 standard on three (50x50x250 mm) reinforced mortar specimens. For
185 both tests, specimens were cured in a climatic chamber at $\text{RH} = (95 \pm 5) \%$ and $T = (20 \pm 1)^\circ\text{C}$ by
186 covering the moulds with plastic sheets for the first day. Then, the plastic sheets were removed and
187 specimens were left at $\text{RH} = (50 \pm 5) \%$ and $T = (20 \pm 1)^\circ\text{C}$ until the end of the tests.

188

189 **3. Results and discussion**

190 *3.1. Workability*

191 The workability of mortars in terms of slump value is reported in Table 1. The obtained results show
192 that according to the UNI EN 1015-6 standard all mortars can be classified as soft mortars, being the
193 flow values higher than 200 mm. It is evident that the addition of RCF and GCH requires a higher
194 amount of superplasticizer to obtain a certain workability. The reduced workability of mortars

195 containing carbon fibres was already found by other authors [14,42], and it is reported to be more
196 noticeable if the w/c ratio is low and aggregates are fine. Moreover, the workability decreases with
197 the same fibre type by increasing the aspect ratio and the percentage of fibres [14,43,44]. In case of
198 chars/biochars, the decrease of workability is related to the high carbon content and to the porous
199 microstructure of the particles, which cause water retention [31,45]. In fact, in the present paper, GCH
200 was not previously soaked with water, but added to mortars in dry condition. Also the loss of
201 workability when chars/biochars are added, increasing with the percentage of addition [46,47], has
202 already been found in previous studies [29–31]. As expected, when mortars are manufactured with
203 both RCF and GCH, a higher amount of superplasticizer compared to that used in mortars with only
204 one addition is necessary to achieve the desired workability.

205

206 *3.2. Mechanical strength*

207 Results of mechanical strength tests are reported in Figure 2; as expected, the mechanical strength
208 increases in time. After 2 days of curing, mortars containing carbon-based additions show a slightly
209 lower flexural strength compared to that of the plain mortar. However, it is worthy to note that after
210 7 days of curing all mortars containing RCF, GCH or both the additions exhibit a comparable or
211 higher R_f value than that of REF (Figure 2a). Those containing 1.0 vol% of GCH have a higher
212 flexural strength (5%) than those containing 0.5 vol% of GCH. The increase of flexural strength of
213 mortars containing gasification char has been found also by Sirico et al. [31] and by the present
214 authors [29]. The reasons are multiple: chars/biochars act as accelerator additives, thanks to the
215 presence of potassium salt, which may speed the hydration process at early age [47,48]; biochar/char
216 particles are characterized by a porous structure that allows the migration of absorbed water to the
217 surrounding paste, promoting the hydration reactions [29] and densifying the interfacial transition
218 zone (ITZ), as reported by Mrad and Chehab [49]. After 28 days of curing, the REF mortar shows a
219 flexural strength of 7.2 MPa, whereas among mortars containing carbon additives best results are
220 obtained when RCF are added at 0.2 vol% alone (0.2RCF mortar) or coupled with 0.5 vol% of GCH

(0.5GCH+0.2RCF mortar), where the R_f value increases by 9% and 17%, respectively. Carbon fibres create a great number of connections, which amplify the adhesion forces, the stitching effect in the presence of micro-cracks which form during loading, and the bridging effect [50]. The bridging effect of RCF is visible also in the SEM images (Figure 3), where some filaments place among the width of micro-cracks. The present authors have found that, at the same volume dosage, RCF are more effective than VCF and brass-plated steel fibres (BSF) [16,30] in increasing the flexural strength of cement mortars. This is because RCF present a high quantity of carbon micro-particles on their surface, which increase the specific surface area of fibres and work as nucleation points for the formation of C-S-H crystals on the filaments [51]. After 28 days of curing, also mortars containing solely GCH exhibit slightly higher R_f values than the REF mortar. The lower flexural strength of 1.0GCH+0.2RCF mortar, even if still comparable with the REF one, could be attributed to the fact that probably the beneficial effect provided by the combination of filler and fibres is masked by the high quantity of superplasticizer incorporated in the mix. It is known that when the quantity of superplasticizer is high, foam is produced during the mix [52] which generates an increased porosity. The compressive strength of mortars after 2, 7, and 28 days of curing is reported in Figure 2b. Results show that after 2 days of curing all mortars exhibit similar R_c values, whereas after 7 days of curing those containing carbon-based additions have even a slightly higher compressive strength than REF. After 28 days of curing, the compressive strength of mortars with carbon-based additions is similar to that of the reference mortar without additions. Probably, the high strength of the binder paste, due to the low w/c ratio adopted, has masked the effect of both fillers and fibres on the compressive strength of mortars at long curing time. In any case, it can be stated that at the selected percentages recycled carbon-based additions do not impair the compressive strength of mortars with high mechanical properties.

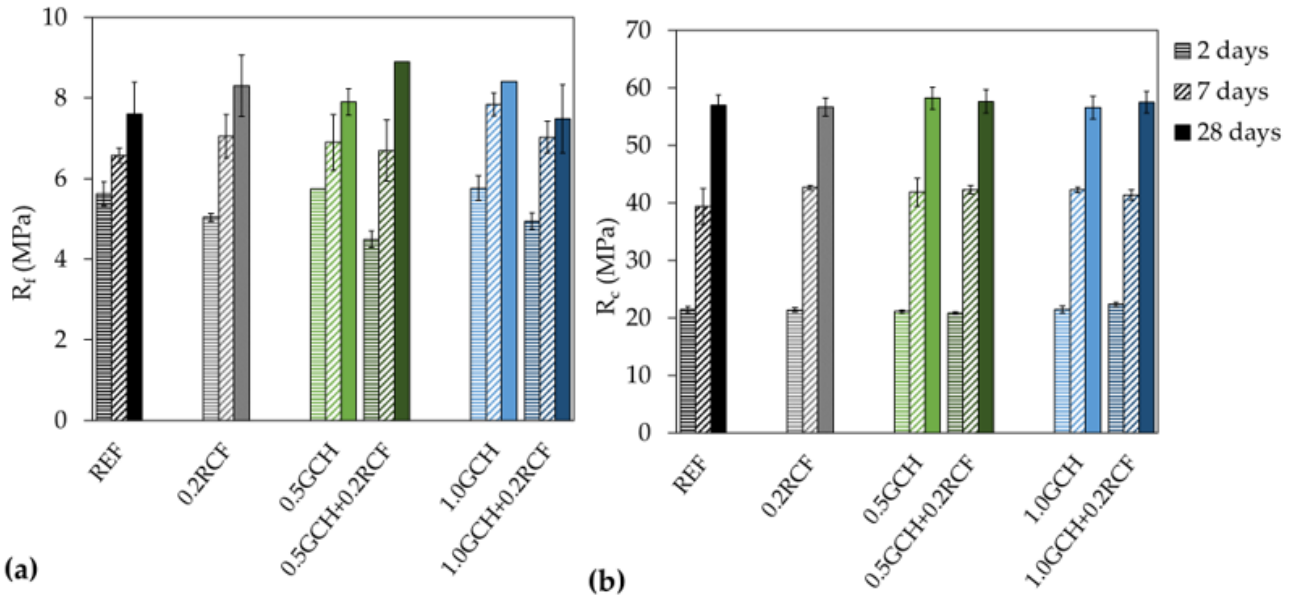


Figure 2. (a) Flexural strength (R_f) and (b) compressive strength (R_c) of mortars after 28 days of curing

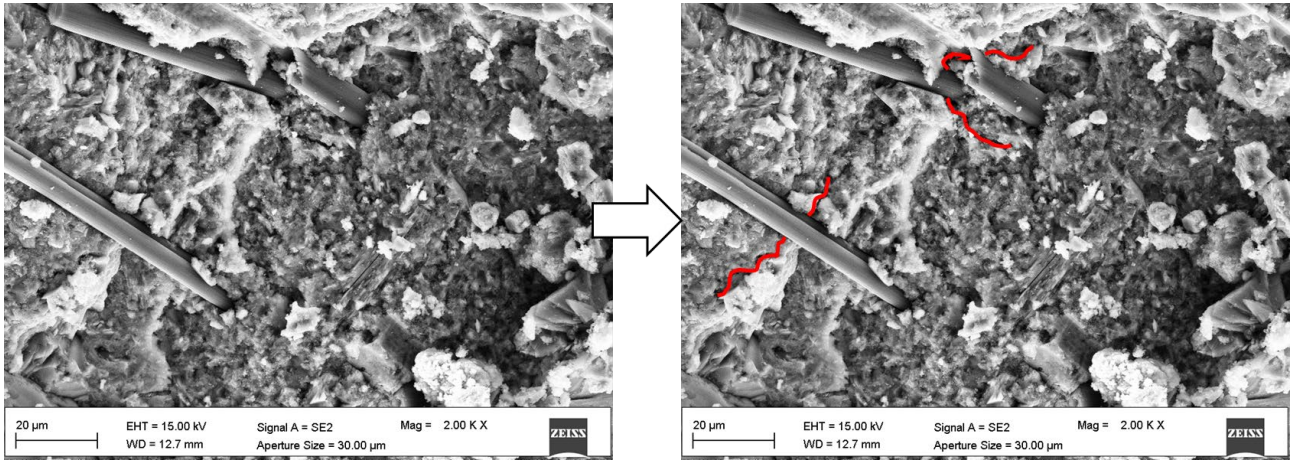


Figure 3. SEM images of RCF bridging micro-cracks (highlighted in red) in mortar

The hardened density, the UPV, and the dynamic modulus of elasticity (E_d) of mortars after 28 days of curing are given in Table 2.

Concerning the addition of fibres, the obtained results show that neither the hardened density nor UPV, and therefore E_d , are influenced significantly. Also Baeza et al. [14] reported that by increasing the concentration of carbon fibres of 3, 6, and 12.5 mm in length from 0% to 1% in cement mortars,

the UPV is not affected, whereas UPV increases with long fibres and higher fibres dosages. On the contrary, Mastali et al. [51] found a UPV decreasing with increasing the fibres volume fractions from 0.5% to 1%, 1.5%, and 2% for both 10, 20, and 30 mm-long recycled carbon fibres. This result was related to the increased porosity generated by increasing the percentage of long fibres addition, which reduced the workability of the mix and incorporated more air into the matrix during the casting activities [53]. In the present paper, fibres are shorter (6-mm length) [54] and the low percentage used (0.2% by volume, which corresponds to 0.7% by cement weight) is not enough to significantly modify density, UPV, and therefore E_d parameters (Table 2).

Considering the addition of the carbon-based filler, no effect was found for GCH added at 0.5 vol%, whereas a slight increase of UPV and E_d , (from 36 to 38 GPa) is found for 1 vol% addition. Gupta et al. [39] reported that when biochar is added to cement mortars a negligible E_d increase from 29.8 to 31.0 GPa is found for 1 wt% addition, no effect is found for 2 wt% addition, whereas for contents from 5% to 8% by cement weight E_d decreases by 16% and 29%, respectively. This effect was attributed to the higher content of biochar particles with a lower modulus of elasticity (4 - 10 GPa [55]), compared to that of plain cement mortar (28 GPa at w/c = 0.40). Rodier et al. [56] found that by increasing the content of biochar from 2% to 6% by cement weight E_d decreases, in line with other authors [53] using agricultural waste fibres in cement mortars. In this paper, by increasing the volume content of GCH from 0.5 vol% to 1 vol% (which correspond to 2% and 4% by cement weight) only a negligible E_d increase was detected, as found by Gupta et al. [39].

When RCF and GCH are added together, the E_d value remains equal to 38 GPa. However, all the manufactured mortars have high E_d values (≥ 36 GPa), meaning that they are very stiff. The modulus of elasticity reflects the ability of composites to deform elastically: the more flexible the material, the lower the modulus of elasticity [57]. The modulus of elasticity reflects also the mechanical strength of mortars and, at the same binder type, the variation of water/binder (w/b) ratio results in a variation of E_d value [58]. In this work, all the mortars have been manufactured with the same w/b ratio of 0.45 and exhibit approximately the same compressive strength; the difference between mixtures are related

to the type and quantity of carbon-based materials used. In this research, mortars with the highest quantity of additions, namely 0.5GCH+0.2RCF, 1.0GCH, and 1.0GCH+0.2RCF, are those with the highest value of E_d , thanks to the densification of the matrix related to the formation of more C-S-H products as shown by the hardened density values reported in Table 2. However, the increase in E_d is so low (+6%) that it can be stated that RCF and GCH, both alone and together, do not impair the stiffness of mortars.

Table 2. Hardened density, ultrasonic pulse velocity (UPV), dynamic modulus of elasticity (E_d), total porosity (V_p) and average pore diameter (d_p) of mortars after 28 days of curing

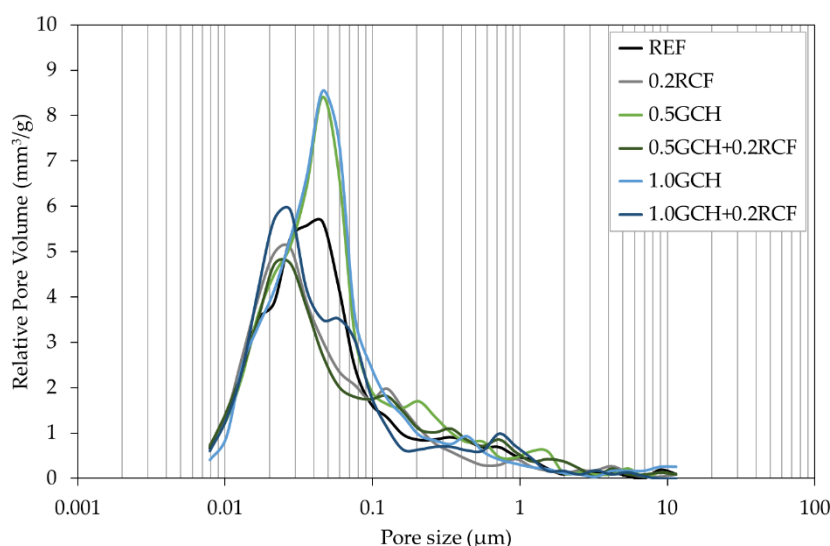
Mortar	Density (kg/m³)	UPV (m/s)	E_d (GPa)	V_p (%)	d_p (μm)
REF	2256	4223	36	10	0.033
0.2RCF	2258	4232	36	9	0.030
0.5GCH	2257	4203	36	12	0.036
0.5GCH+0.2RCF	2291	4311	38	9	0.032
1.0GCH	2265	4305	38	12	0.036
1.0GCH+0.2RCF	2285	4293	38	10	0.031

3.3. Microstructural analysis

Results of the microstructural analysis of mortars after 28 days of curing are reported in Table 2 and Figure 4. The REF mortar has a V_p of 10% and an average pore diameter (d_p) of 0.033 μm . Mortars containing GCH alone at both percentages show a total porosity 20% higher than REF, with also a slightly bigger d_p value (0.036 μm). On the contrary, mortars manufactured with RCF, both alone and together with GCH have a V_p similar to REF but slightly lower d_p values. However, all mortars show very low total porosity values, ranging from 9% to 12%, and d_p from 0.030 to 0.040 μm , meaning

300 that the carbon-based additions do not affect the compactness of microstructure, in line to what found
 301 by compressive strength test and E_d values.

302



303

304 **Figure 4.** Pore distribution of mortars after 28 days of curing

305

306 3.4. Electrical resistivity

307 The electrical resistivity of mortars during 56 days of curing is reported in Figure 5. All mortars with
 308 carbon-based additions register a lower electrical resistivity than REF. In particular, when GCH is
 309 added to cement mortars at 0.5 vol%, only a slight reduction of ρ is registered, whereas the doubling
 310 of the GCH percentage decreases the ultimate ρ value of approximately 13%. The lower electrical
 311 resistivity of pastes/mortars containing gasification char can be attributed to its elevated carbon
 312 content (77%) [29,30].

313 On the other hand, the addition of RCF at 0.2 vol% reduces the electrical resistivity of mortars up to
 314 83% at the end of the test. The great decrease of electrical resistivity of mortars containing RCF is
 315 due to their high carbon content (94%) and to the volume dosage near to their percolation threshold.
 316 In fact, as reported in [16], the percolation zone of RCF (and also VCF) with 6-mm length is between
 317 0.1 vol% and 0.2 vol%. The combination of GCH and RCF gives a further decrease of ρ value, which
 318 becomes 90% lower than that of REF at the end of the test. As already found by Wen and Chung [59]

319 and the present authors [30], the combined use of conductive fillers and fibres is able to decrease the
320 electrical resistivity of the material compared to that of fibres and fillers used alone at the same
321 dosages, since the filler locates in the spaces between fibres, improving the electrically conductive
322 path.

323 In order to verify if the electrical resistivity was higher in mortars containing less water, the water
324 content of mortars was measured (Table 3). In particular, the difference between the weight of mortars
325 after 7 days of curing and after drying in oven 28-day cured mortars was calculated. Results obtained
326 show that the reference mortar has the lowest moisture content. Mortars manufactured with GCH
327 have the highest water content (approximately 25-30% more than REF); however, their electrical
328 resistivity is slightly lower than that of the reference mortar. On the other hand, mortars containing
329 RCF both alone and coupled with GCH have a lower water content than those manufactured with
330 GCH alone (around 15-20% higher than REF), but they have the lowest electrical resistivity.

331 Considering the obtained results, it is evident that even if water is a good electrical conductor, its
332 effect on the electrical conductivity of mortars is limited, whereas the great contribution in the
333 reduction of the electrical resistivity is given by the nature of carbon-based additions. However, the
334 electrical resistivity of all mortars increases in time, because of the continuous hydration of the
335 cement paste and the gradual water evaporation which reduce the free water content [60]. Anyhow,
336 the increase is much more evident in REF and GCH mortars whereas in mortars containing RCF alone
337 or with GCH it is very low. As reported in [16], this effect is due to the electrical conduction between
338 the fibres, which overpowers the ionic conduction of the solution present in the mortar pores. RCF
339 contribute to highly decrease the electrical resistivity of mortars already at early ages, thus making
340 the slope of the curve flatter compared to the others.

341

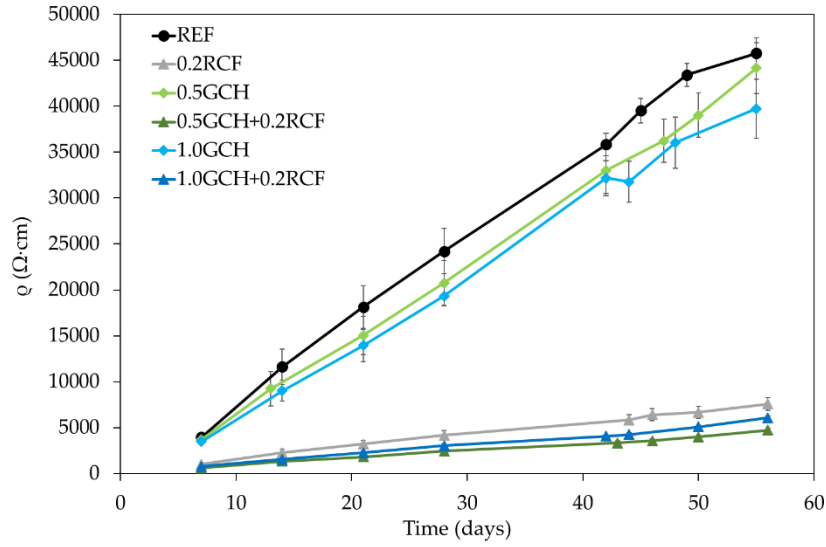


Figure 5. Electrical resistivity (ρ) of mortars up to 56 days of curing

Table 3. Weight of mortars after 7 days of curing and after drying and total water content

Mortar	Weight after 7 days (g)	Dry weight (g)	Water content (g)
REF	590.3	554.1	36.2
0.2RCF	595.6	552.0	43.6
0.5GCH	590.8	545.2	45.6
0.5GCH+0.2RCF	606.6	564.8	41.8
1.0GCH	594.2	547.7	46.5
1.0GCH+0.2RCF	604.6	560.7	43.9

3.5. Capillary water absorption

Results of capillary water absorption of mortars after 28 days of curing are reported in Figure 6. In general, all the mortars seem to have not reached the saturation at the end of the test, since the curves do not show a plateau. From the obtained results, REF mortar absorbs most water. In particular, the addition of GCH at 0.5 vol% and 1.0 vol% reduces the Q_i value of 15% compared to REF, whereas

the use of RCF alone decreases the capillary water absorption by 27%. Literature already reports many investigations concerning the effect of char/biochar addition on the water sorptivity of cement composites [61] and the decrease of water capillary absorption of mortars manufactured with pyrolytic biochar at 1 and 2 wt% [39,62,63] or gasification char at 4 wt% [30]. This is due to the hydrophilic nature of biochar, which reduces the free water in mortars by absorption [39], and to the fact that biochar particles lead to the formation of a thin layer of water on their surroundings, referred to as hydraulic boundary layer [64]. These characteristics retard the water front resulting in overall less water absorption [63]. Gasification char used in the present experimentation, even if slightly increases the mortars total porosity (from 10% to 12%, Table 2), provides the same properties to the mortars. Also the decrease of Q_i value in cement mortars containing RCF at 0.2 vol% has already been found [16]. In this case, the reduced water absorption could be related to the slightly lower total porosity and average pore diameter of mortars (Table 2) since the lower the total porosity, the lower the capillary water absorption [58]. The fibres improve the microstructure of the composite, thanks to the optimal adhesion with the cement paste and their ability to act as nucleation points for the formation of hydration products, which reduce the micropores amount [65]. Mortars manufactured with GCH at 0.5% and 1.0% coupled to RCF show similar capillary water absorption values to those containing GCH alone (12% lower than REF). In this case, the lower total porosity and average pore diameter (Table 2) together with the characteristic of char to act as a “barrier” for the water ingress have lowered the water absorption capacity of mortars.

371

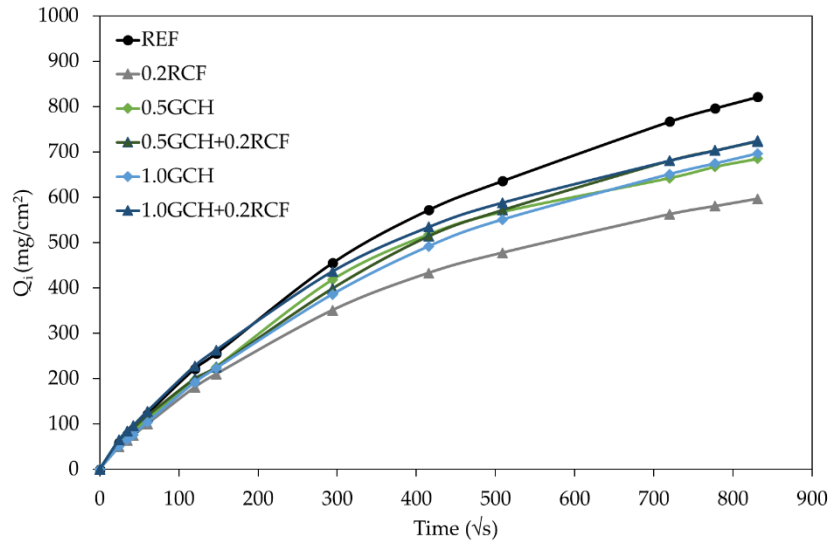


Figure 6. Water absorbed per unit area (Q_i) of mortars

3.6. Drying shrinkage

The free drying shrinkage (ϵ_d) and the weight loss of mortars during 56 days of test are given in Figure 7. The obtained results show that at the end of the test the REF mortar has an ultimate ϵ_d value of 0.85 mm/m (Figure 7a) and a weight loss of 6% (Figure 7b). The two mortars 0.5GCH and 1.0GCH shrink approximately 15% more than REF, even though the water lost during the test is nearly the same measured in REF. Also Gupta et al. [39] found that adding biochar at 2 wt% the drying shrinkage of mortars increases since biochar has a lower modulus of elasticity than cement paste, hence it has no significant restraining effect on shrinkage of hardened paste [39]. Moreover, 0.5GCH and 1.0GCH mortars are those exhibiting the highest volume of capillary pores (Figure 4). Capillary pores (mesopores with average radius ranging from 5 to 5000 nm [66]) are those most responsible for drying deformation [67] since, according to Laplace's equation, water evaporating from smaller pores causes a higher stress than from larger pores, thus leading to a higher shrinkage strain [32]. On the contrary, the 0.2RCF mortar is the one which shrinks the least, showing an ultimate ϵ_d value of 0.60 mm/m, 29% lower than REF, since it has the lowest water evaporation. The reduced drying shrinkage of cement-based materials containing carbon fibres has been already reported in the literature. Chen and Chung [68] found that the addition of 0.19 vol% of 5 mm-long carbon fibres decreases the drying

shrinkage of concrete by approximately 40% due to their bridging effect. Song et al. [69] found that 0.1 vol% and 0.2 vol% of carbon fibres reduced the drying shrinkage of concrete by 20% and 29%, respectively. Also Belli et al. [16] found that drying shrinkage of cement mortars decreases by increasing the carbon fibres content but only at concentrations higher than 0.4 vol%. As reported by Chen and Chung [68], the decrease of drying shrinkage in mixtures containing carbon fibres is much more evident if the material contains big aggregates. In the present paper, the aggregate maximum diameter is 8 mm, while in the paper by Belli et al. [16] is lower than 1 mm. This is the reason why the decrease of drying shrinkage is much more appreciable for the mortars containing 0.2 vol% of RCF of the present paper compared to those studied in the previous one [16].

Combining GCH and RCF in mortars, in the case of 0.5GCH+0.2RCF mortar, both free drying shrinkage and weight loss are comparable to those of REF, whereas in the case of 1.0GCH+0.2RCF, the shrinkage increases of approximately 32% ($\epsilon_d = 1.12$ mm/m) due to the highest water evaporation (6.6% after 56 days). In the case of the mortar with the highest addition of carbon-based material, namely 1.0GCH+0.2RCF, the high water absorbed by GCH and RCF during the casting is subsequently released during the test.

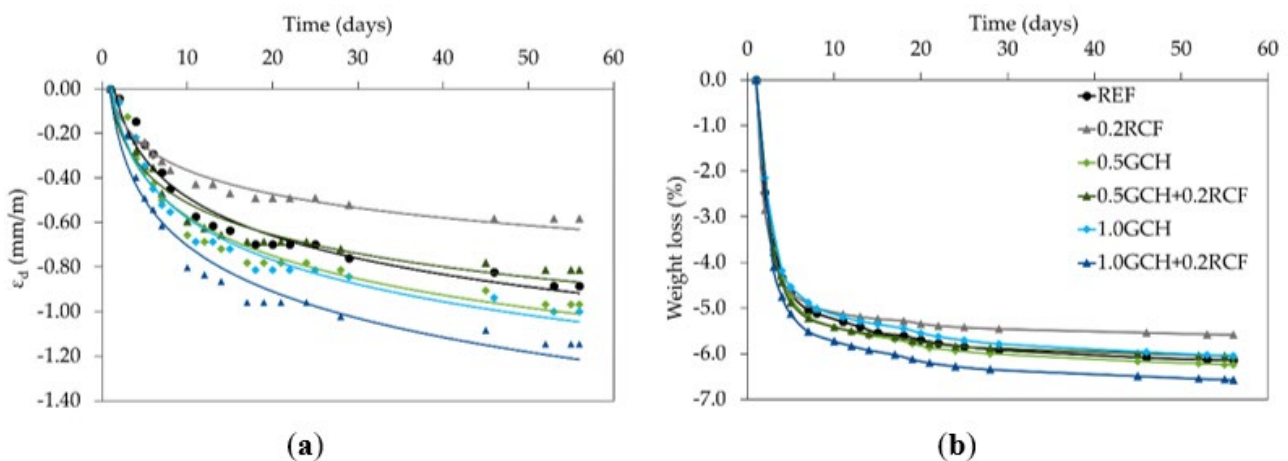
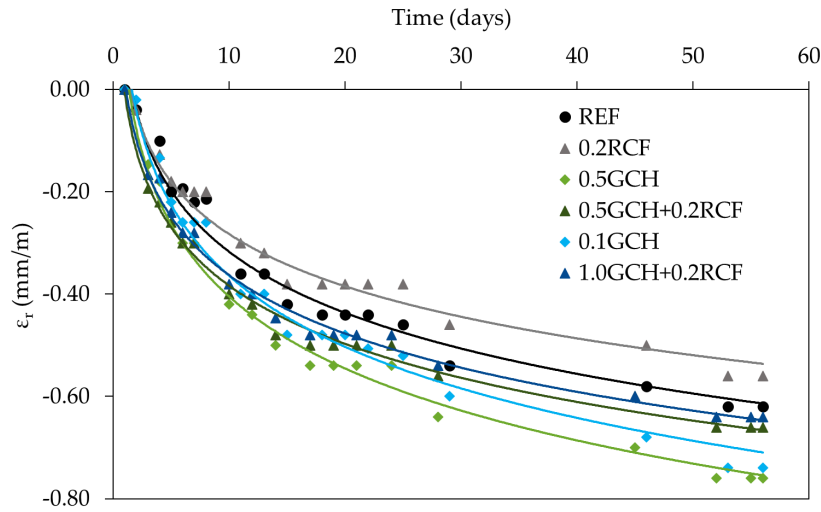


Figure 7. (a) Free drying shrinkage (ϵ_d) and (b) weight loss of mortars up to 56 days of curing

410 The restrained drying shrinkage (ϵ_r) of mortars during 56 days of test is reported in Figure 8. Results
 411 show that at the end of the test the REF mortar shrinks approximately 0.62 mm/m. GCH increases
 412 the restrained shrinkage of mortars (23% higher than REF) at both 0.5 vol% and 1.0 vol%
 413 percentages. On the contrary, mortars containing RCF shrink less with similar ϵ_r values if
 414 manufactured with GCH and even lower (10%) if they are added alone. The slightly higher restrained
 415 shrinkage of mortars with only GCH is related to the high free drying shrinkage of the mortars
 416 containing this carbonaceous addition (Figure 7a). On the contrary, RCF reduce the restrained
 417 shrinkage of mortars containing GCH at both percentages. The reduced shrinkage of the 0.2RCF
 418 mortar compared to REF is related to its low drying shrinkage (Figure 7a) at the same E_d (Table 2).
 419 Conversely, the slightly higher restrained shrinkage of 0.5GCH+0.2RCF and 1.0GCH+0.2RCF
 420 mortars compared to REF is due to their slightly higher E_d (Table 2). In fact, a low restrained
 421 shrinkage is related to a low modulus of elasticity and/or large creep relaxation [70].



423
 424 **Figure 8.** Restrained drying shrinkage (ϵ_r) of mortars up to 56 days of curing

425 4. Conclusions

427 In this experimentation, the effect of adding GCH and RCF, both alone and coupled, to decrease the
 428 electrical resistivity of mortar composites manufactured with a new lo-clinker cement, CEM II/C-M

429 (S-LL), not yet available in the market, has been evaluated. GCH was added to mortars in two
430 percentages, namely 0.5 vol% and 1.0 vol%, whereas RCF at 0.2 vol%. Besides mechanical and
431 electrical properties, also the effect of the two sustainable additions on the durability of mortars has
432 been evaluated in terms of capillary water absorption and drying shrinkage.

433 The obtained results have shown that:

- 434 • GCH and RCF increase the flexural and compressive strengths of mortars already after 7 days
435 of curing, since they act as nucleation sites for C-S-H products. After 28 days, the highest
436 flexural strength is found for mortars containing 0.2 vol% of RCF both alone and coupled to
437 GCH at 0.5 vol%. On the other hand, the addition of GCH and RCF does not modify R_c and
438 E_d values after 28 days of curing.
- 439 • The total porosity and average pore diameters of mortars are slightly increased when GCH is
440 added alone, but if it is combined with RCF the two parameters return similar to those of REF.
- 441 • GCH decreases the electrical resistivity of mortars only by 3% if added at 0.5 vol%, and by
442 13% at 1.0 vol%. On the other hand, RCF decrease the electrical resistivity of mortars by
443 83%, if used alone, and by 90% if coupled to GCH at both the two dosages, highlighting their
444 marked capability to decrease the material's electrical resistivity. The decreased electrical
445 resistivity of mortars containing carbon-based additions is not related to the moisture content
446 of mortars themselves, but it is mainly related to the nature of the carbonaceous addition.
- 447 • All the carbon-based additions reduce the capillary water absorption, in particular by 27%
448 when RCF are used alone and by 12% when GCH is coupled to RCF.
- 449 • GCH slightly increases (whereas RCF slightly decreases) the free drying shrinkage of mortars.
450 Also the restrained shrinkage of mortars is slightly increased when GCH is added to mortars
451 and decreased if RCF are used alone, whereas it remains similar to that of the REF mortar in
452 the other cases.

453 The obtained results demonstrate that the addition of GCH and RCF does not impair the compressive
454 strength of mortars both if they added alone or in combination. On the other hand, while GCH alone

455 is not sufficient to significantly decrease the electrical resistivity of mortars, the addition of RCF is
456 able to provide mortars an electrical resistivity even 90% lower than that of the plain mortar. The
457 RCF addition to mortars containing GCH is also beneficial to slightly decrease their total porosity
458 and to increase their durability in terms of lower drying shrinkage and lower propension to capillary
459 water absorption, especially when GCH is added at 0.5 vol% and RCF are added at 0.2 vol%.
460 Thanks to the addition of low-cost and sustainable carbon-based fillers and fibres to low-clinker
461 cement-based mortars, new sustainable materials with low electrical resistivity and improved
462 durability can be obtained.

463

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470

471 **CRedit authorship contribution statement**

472 **Alessandra Mobili:** Conceptualization, Methodology, Software, Validation, Formal analysis,
473 Investigation, Data curation, Writing – original draft. **Gloria Cosoli:** Software, Validation, Data
474 curation, Writing - review & editing. **Tiziano Bellezze:** Conceptualization, Data curation, Resources,
475 Writing - review & editing, Supervision. **Gian Marco Revel:** Resources, Writing – review & editing,
476 Supervision, Funding acquisition. **Francesca Tittarelli:** Conceptualization, Resources, Writing -
477 review & editing, Supervision.

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