# Influence of Biochar and Recycled Gypsum on the Strength and Microstructure of Conventional and Sustainable Cementitious Composites

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## Abstract

The need for sustainable waste management practices to mitigate climate change emphasized discovering alternatives for environmentally unfriendly materials with high carbon footprints such as ordinary Portland cement (OPC). This study examined the effect of biochar and its presoaking on the compressive strength, porosity, and microstructure of sustainable mortars containing fly ash, recycled gypsum drywall, and OPC by manufacturing and testing 270 cubic specimens, 30 mix designs at 7, 28, and 90 days. The main roles of biochar in the matrix were to provide water and an additional surface for internal hydration, boost nucleation, and increase interlock in the cement paste. The water retention potential of biochar particles, however, could contribute to an artificial increase in the early-age compressive strength of cementitious composites (CCs) containing biochar. This strength was lessened over time as the retained water was utilized by cementitious reactions. The low strength of the biochar particles and its effect on the porosity of CCs were the primary sources of strength reduction of specimens containing biochar. The influence of biochar presoaking relied on the initial reactivity of binders. Lower initial reactivity lessened the initial water demand, increasing free water in the mixture, retained by non-soaked biochar. The highly reactive binders, however, needed extra water, which was supplied by

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presoaked biochar. The data also indicated the good compatibility and bond between biochar particles and the cement paste. The effect of adding biochar on the properties of CCs strongly depended on the particle size distributions of the composite constituents and biochar particles.

**Keywords:** Sustainability, Cementitious Composites, Biochar, Recycled Gypsum, Fly Ash.

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## 1. Introduction

In the last decades, global climate change and its negative consequences such as global warming emphasized the significance of a rapid decrease in greenhouse gases by 21-43% by 2030 based on the 2019 benchmark for human and earth survival [1]. Consequently, reducing greenhouse gases comes into the focus of politicians, scholars, and governments. Carbon dioxide ($CO_2$) is the leading greenhouse gas that accounts for over 75% of greenhouse gases [2]. According to this massive contribution, numerous studies have been launched to offset $CO_2$ emissions by approaching sustainable technologies and materials [3–6].

Ordinary Portland cement (OPC) is known as a key ingredient in the construction industry. However, with a 7% contribution to the annual global $CO_2$ emissions, it is one of the major eco-unfriendly construction materials [7]. Maddalena and Roberts [8] showed that optimizing the calcination process could ultimately lower $CO_2$ emissions of OPC manufacturing by up to 50%. This emphasized the significance of lessening the OPC demand in cementitious composites (CCs) such as concrete and mortar. This reduction also conserves the raw materials and cuts down the environmental impacts of their extraction [9]. One of the promising approaches to reduce OPC demand in CCs is its substitution with more sustainable products and by-products [3,4,10,11].

Biochar is one of the potential alternatives to OPC in CCs that has trended in recent studies [12,13]. This carbon-rich porous material is a by-product of a sustainable power-generating technology, which thermally decomposed waste wood (biomass) and other combustible waste in low-level oxygen (pyrolysis technique). In the procedure, 50 and 33% of biomass carbon are captured (stored) inside biochar and released as usable energy, respectively [14]. Accordingly, this sustainable technique can reduce the $CO_2$ emissions of the power plant by at least 67% compared to burning rice straws [15]. Despite this fact, landfilling biochar or using it as a soil amendment may release soluble carbon due to its reactivity and dissolution in water [16]. Subsequently, the carbon will be released into the atmosphere, lessening the sustainability of the power-generating technology. Additionally, the amount of biochar can become one of the major public concerns in the future given the environmental benefits and the increasing prevalence of using the technique. Therefore, a sustainable management practice is required to address this issue.

One of the efficient techniques of reusing such materials is to use them as OPC substitution in CCs [11,12,17]. Using biochar in CCs is a win-win opportunity that can reduce the demand for the most environmentally unfriendly component of CCs while embedding carbon-capture material (carbon sequestration), which can ultimately result in carbon-negative CCs [12,13,18]. Based on this great potential, research on the use of biochar in CCs has gained momentum in the past few years [12,13].

Maljaee et. al [17] represented that replacing up to 4% OPC with biochar improved the strength of CCs. Earlier studies also reported that adding biochar by 2% to the CCs densified its microstructures, improving its durability properties such as freeze-thaw and acid resistance thanks to the structure and chemical compositions of biochar [19–21]. However, it could increase the water absorption of the composites due to its water-retention potential [20]. Moreover, Sikora et. al [20] indicated that up to 5% substitution of OPC with biochar lowered the porosity and thermal conductivity of CCs. Biochar could also promote both hydration and carbonation in CCs, raising the strength of the composites [21–23]. Furthermore, Chen et al. [22] depicted that replacing 5% of OPC with biochar scaled down the greenhouse gas emissions and energy consumption of the CCs by 14.7% and 4.63%, respectively. Despite the mentioned advantages, there are contradictions in the literature on the role of biochar in CCs.

Literature proposed five different roles for biochar in the CCs. The first theory considers biochar as a supplementary cementitious material that can react with the cement paste. Although this theory is in contrast with the stability of the carbon, earlier studies reported pozzolanic reactions between the cement paste and some chemical components of the biochar [24–26]. The Second theory deals with biochar as a filler that occupies the voids and pores of the composites [13,27]. In the third theory, biochar was used to intensify the cementitious reactions (hydration and carbonation). First, biochar has a significant potential to retain water through its pores. This water can be released and used by cement paste to boost internal hydration [11]. Figure 1 schematically shows how the released water from biochar can provide additional water for internal hydration. This extra water is quite beneficial for large-scale concrete casting projects, especially in arid climates. Second, the large surface-to-volume ratio of biochar can provide additional surface for nucleation and hydration growth, intensifying the reactions [27]. Partial dissolution of the biochar in water during curing can also offer extra carbon for the internal carbonation of CCs, improving its strength [22]. The Fourth theory explains that the irregular shape of biochar can provide a strong interfacial transition zone and interlock between biochar and cement paste, reinforcing the CCs [28]. Finally, in the fifth theory, biochar contributes to putting additional materials and organisms into the CCs [29].

Furthermore, in contrast to the promising results of using biochar in CCs, a few studies argued the influence of biochar on the mechanical and microstructural properties of CCs. This argument was raised from the adverse effect of biochar on the mechanical properties of the CCs reported by previous studies [20,30–32]. Microstructural analysis such as scan electron microscopy (SEM) was used to determine the key factors for the strength loss. SEM demonstrated that there was no reaction between biochar particles and the cement paste. According to the analysis, the transition boundary between biochar and cementitious paste was similar to the one between aggregates and the paste [33,34]. Separation in the transition boundaries between biochar and paste was also discovered in some cases [35]. Despite this, an earlier study proposed good compatibility between the cement paste and biochar particles [36].

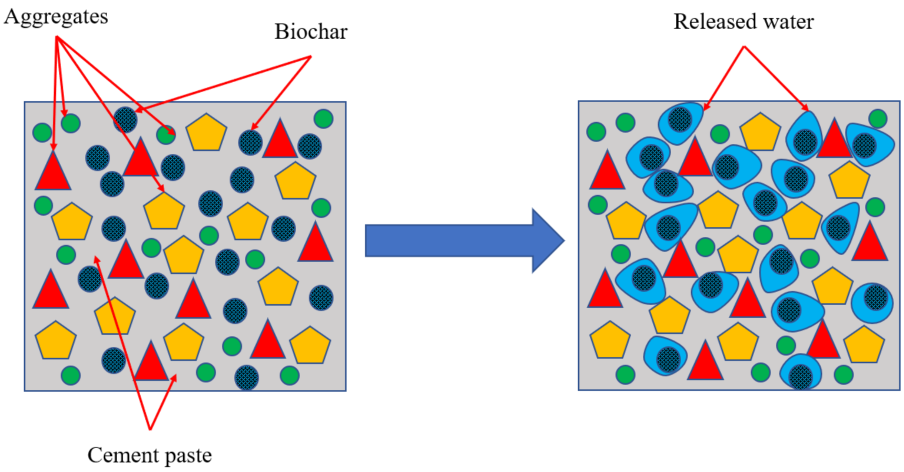


**Figure 1. Internal curing provided by retained water from biochar**.

The biochar drying shrinkage could also significantly impact the mechanical and durability properties of CCs containing presoaked biochar, particularly in dry to semi-dry climates [35,37]. The shrinkage separates the biochar from the cement paste. Accordingly, the biochar could not carry the applied load and acts as a void in the composite. This shrinkage relies on the chemical composition, particle size, porosity, connectivity, and shape and size of pores of biochar [13,38].

The effect of biochar on the properties of sustainable CCs was rarely studied in the literature [39,40]. However, the use of supplementary materials such as fly ash and recycled gypsum drywall (RGD) may significantly affect the influence of biochar and its presoaking on the properties of CCs. This is because of the potential of the reverse effect of biochar as filler in the CCs containing fly ash. The reaction rate of the cementitious materials can also significantly affect the water demand of the mixture during curing since several supplementary cementitious materials have delayed reactions. This reduction in early-age water demand can significantly affect the effect of biochar presoaking on the properties of CCs. Therefore, evaluating the effect of using supplementary materials and their interactions with the biochar and its presoaking could contribute to a better understanding of the role and effect of biochar, and its presoaking, on diverse CCs.

The use of combined fly ash and RGD, which has recently been investigated [4,10], can also compensate for the adverse effect of the potential shrinkage of biochar particles on the mechanical properties of CCs. Fly ash is a potential supplementary material that can significantly reduce the shrinkage of CCs [41]. More importantly, fly ash can lessen OPC demand in the mixture, declining the environmental impacts of the composite [4,42]. Earlier investigations also showed the potential of fly ash to improve the mechanical and durability properties of CCs and its functionality in reducing the voids in CCs [41,43]. Additionally, the fly ash particles may occupy the biochar pores, raising the strength of the biochar. Nonetheless, adding fly ash can disturb the efficiency of the biochar as a filler, which accentuates the importance of evaluating the CCs containing biochar and fly ash.

The expandable reactions of RGD can also compensate for the biochar's part of the shrinkage. RGD expands during the biochar shrinkage and occupies the spaces left by the biochar shrinkage. However, using gypsum in ordinary CCs adversely affects its properties [4]. Hansen and Sadeghian [4] showed that adding RGD was beneficial in CCs containing OPC and fly ash. Their results revealed that RGD could activate the fly ash and improve its reactivity with cementitious materials [4]. Similar results on improving the reactivity of the slag were reported in the literature, supporting the potential of RGD in the CCs containing fly ash and/or slag [44–46]. The use of RGD in the composite also has environmental advantages. On one side, it can reduce the OPC demand by improving the reactivity of the fly ash. On the other side, it decreases the disposal of gypsum drywall waste in landfills. Landfilling the gypsum contaminates soil and water and emits the $H_2S$ gas into the environment [47,48]. Moreover, previous investigations indicated the closed-loop recycling of gypsum, making RGD a viable alternative to natural gypsum [49]. Using biochar in the CCs containing RGD can further boost the reactivity of the gypsum by providing additional water for gypsum reactions in the composite. The low-speed reactions of RGD can also be beneficial as its late expansion can fill the gradual shrinkage of the biochar particles due to their water drainage.

Based on the above-mentioned, the current study aims to address the four research gaps, i) the contradiction in the role of biochar in CCs, ii) the effect of biochar dosage on CCs' mechanical properties, iii) the compatibility of biochar particles by different type of CCs, ordinary CCs and CCs containing fly ash and RGD, and iv) the influence of reaction rate of cementitious materials on the effect of biochar and its presoaking on the mechanical, microstructural, and porosity of the CCs. These topics were examined by manufacturing and testing 270 cubic mortar specimens via compressive strength, ultrasonic pulse velocity, and SEM. This study also assessed the key parameters that affect the mechanical properties of the CCs containing biochar to provide directions for future studies. The results were analyzed to evaluate the effect of biochar and its combination with fly ash, and RGD on the properties of the composite. The SEM was then used to assess the influence of the fly ash and RGD on the separation between biochar and cementitious paste. The role of biochar and effective parameters on its performance in CCs were then determined based on the data.

## 2. Methodology

### 2.1. Materials

In this study, biochar, general use (GU) Portland cement (CRH Canada Group, ON, Canada), Class F fly ash (Ocean Contractors, Halifax, NS, Canada), RGD (USA Gypsum, Denver, PA, USA), masonry sand (Shaw Resources at Halifax, NS, Canada), and tap water were used to manufacture specimens. Since the RGD contains paper and coarse particles compared to other cementitious materials, the same procedure with Hansen and Sadeghian [4] was used to eliminate the paper and coarse particles. The water absorption of the sand was 2.3% (wt%). The supplied biochar was manufactured by thermally decomposing wood at the pyrolysis temperature of 400℃ for a duration of 210 seconds. The water absorption of the biochar was also tested by the tea bag method as described in the literature [50]. According to the test, the water absorption of the biochar was 100% (wt%). However, according to the observations, much of the water was trapped inside the biochar pores and was not absorbed by biochar shells. The sieve analysis and dry laser diffraction measurement technique were used to determine the grading curves of the masonry sand and cementitious materials (OPC, fly ash, RGD, and biochar), respectively. Figure 2 depicts the particle size distribution of the mix design components. Table 1 also shows the chemical compositions of the biochar determined by X-ray diffraction (XRD).

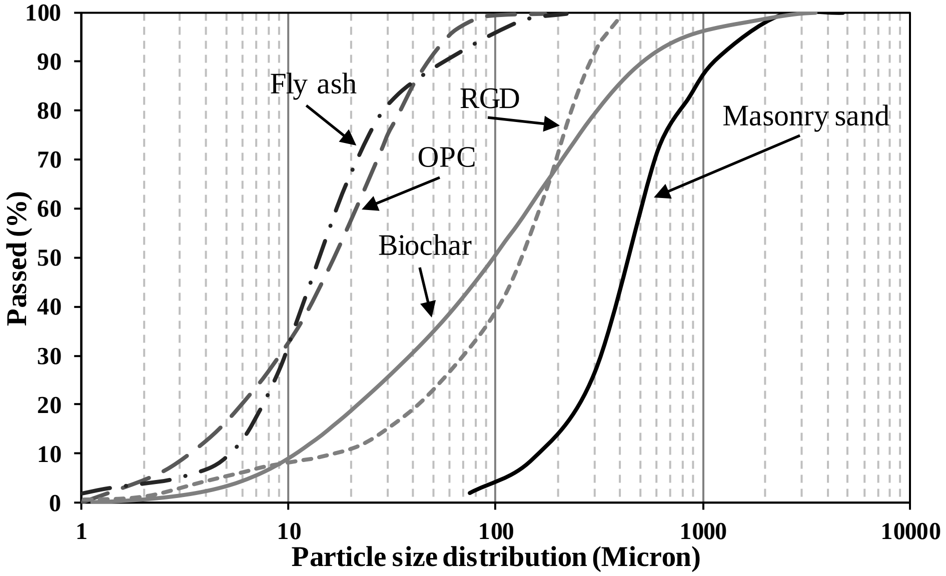


**Figure 2. Size distribution of mix design components (Note: the data of fly ash, OPC, and RGD retrieved from Hansen and Sadeghian [4]. Also, the data of biochar, RGD, OPC, and fly ash were based on volume).**

**Table 1. Chemical composition of biochar**

| Chemical composition | Percentage (wt%) |
|---|---|
| Nitrogen | 0.15 |
| Phosphorus | 0.15 |
| Potassium | 0.22 |
| Calcium | 0.25 |
| Magnesium | 0.06 |
| Sulfur | 0.02 |
| Carbon | 56-76 |
| Ash | 2-4 |
| Water | <5 |
| Others | 36-14 |

### 2.2. Mix Design

In this study, the reference mixture was designed based on the ASTM C109/C109M-21 [51]. Table 2 represents the required materials for manufacturing 9 5×5×5cm cubic specimens with the reference mix design. Table 3 demonstrates the test matrix. Letters A and R at the first of the mix

ID indicated that the biochar was used as an additive and cement replacement in the mixture, respectively. Presoaked biochar was used in manufacturing specimens containing biochar as cement replacement, while the non-soaked biochar was used as an additive in the mixture. For the specimens containing presoaked biochar as the cement substitution, the water content was determined as follows to minimize the amount of water required for achieving consistent workability in the mixture and the effect of water absorption of biochar on the water presented for cementitious reactions in fresh mixture:

a) The weight of dry biochar was calculated based on the replacement ratio,

b) The weight of other cementitious materials, including GU cement, fly ash, and RGD was calculated by deducting the weight of dry biochar from the weight of total cementitious materials in the reference mixture,

c) The required water for biochar saturation was calculated,

d) The amount of water based on the water-to-binder ratio of 0.48 was calculated based on the weight of cementitious materials other than biochar,

e) The total water was equal to the summation of water contents calculated in steps c and d.

Given these calculations, the amount of added water, the total water in each mix minus the required water in the reference mixture, was listed in Table 3 as additional water.

**Table 2. Reference mix design (for 9 cubic specimens)**

| Saturated-surface-dry Masonry sand (g) | Cement (g) | Water (g) |
|---|---|---|
| 2035 | 740 | 359 |

**Table 3. Test matrix**

| Biochar condition | Mix ID | Cement replacement (%wt) | | | Added biochar as an additive (%wt to binder) | Additional water (g) |
|---|---|---|---|---|---|---|
| | | Fly ash | RGD | biochar | | |
| - | Reference | 0 | 0 | 0 | 0 | 0 |
| | B0F50G0 | 50 | 0 | 0 | 0 | 0 |
| | B0F50G10 | 50 | 10 | 0 | 0 | 0 |
| | B0F50G20 | 50 | 20 | 0 | 0 | 0 |
| Non-soaked | AB4F0G0 | 0 | 0 | 0 | 4 | 0 |
| | AB6.75F0G0 | 0 | 0 | 0 | 6.75 | 0 |
| | AB4F50G0 | 50 | 0 | 0 | 4 | 0 |
| | AB6.75F50G0 | 50 | 0 | 0 | 6.75 | 0 |
| | AB4F50G10 | 50 | 10 | 0 | 4 | 0 |
| | AB6.75F50G10 | 50 | 10 | 0 | 6.75 | 0 |
| | AB4F50G20 | 50 | 20 | 0 | 4 | 0 |
| | AB6.75F50G20 | 50 | 20 | 0 | 6.75 | 0 |
| Presoaked | RB2.5F0G0 | 0 | 0 | 2.5 | 0 | 9.5 |
| | RB5F0G0 | 0 | 0 | 5 | 0 | 19.1 |
| | RB6.5F0G0 | 0 | 0 | 6.5 | 0 | 24.8 |
| | RB8F0G0 | 0 | 0 | 8 | 0 | 30.5 |
| | RB10F0G0 | 0 | 0 | 10 | 0 | 38.1 |
| | RB15F0G0 | 0 | 0 | 15 | 0 | 57.2 |
| | RB2.5F50G0 | 50 | 0 | 2.5 | 0 | 9.5 |
| | RB5F50G0 | 50 | 0 | 5 | 0 | 19.1 |
| | RB6.5F50G0 | 50 | 0 | 6.5 | 0 | 24.8 |
| | RB8F50G0 | 50 | 0 | 8 | 0 | 30.5 |
| | RB10F50G0 | 50 | 0 | 10 | 0 | 38.1 |
| | RB15F50G0 | 50 | 0 | 15 | 0 | 57.2 |
| | RB2.5F50G10 | 50 | 10 | 2.5 | 0 | 9.5 |
| | RB5F50G10 | 50 | 10 | 5 | 0 | 19.1 |
| | RB6.5F50G10 | 50 | 10 | 6.5 | 0 | 24.8 |
| | RB8F50G10 | 50 | 10 | 8 | 0 | 30.5 |
| | RB10F50G10 | 50 | 10 | 10 | 0 | 38.1 |
| | RB15F50G10 | 50 | 10 | 15 | 0 | 57.2 |

### 2.3. Specimen Preparation

This study used the same methodology as ASTM C109/109M-21 [51] for mixing, compacting, curing, and testing the cubic specimens. First, biochar (whether presoaked or not) and the saturated-surface dry masonry sand were mixed in the mixer with the minimum speed to avoid the spread of biochar dust. This mixing was continued till the biochar and aggregates were fully mixed together (visually). For the reference mixture only the saturated-surface dry masonry sand was added to the mixture and mixed for a few minutes to ensure the aggregates were combined uniformly. This was because the reference mixture did not contain biochar. Then, the combination of cementitious materials, weighted based on each specific mix design, was added to the mixture and mixed for 2 minutes. Next, the water was gradually added to the mix during the mixing. The mixing was extended till a homogenous mix was obtained. The prepared mortar was then cast into the prepared molds and compacted in accordance with the ASTM C109/C109M-21 standard [51]. Afterward, the specimens were sealed in plastic bags for 24 hours. The molds were removed after 24 hours, and the specimens were placed in the moisture curing room until their testing date.

### 2.4. Tests

In this study, the weight, compressive strength, and ultrasonic pulse velocity of the specimens were measured at 7, 28, and 90 days after demolding the specimens. To achieve consistency in the results of microstructural analysis, the microstructure of 90-day specimens was tested and analyzed. The following subsections explain the compressive strength, ultrasonic pulse velocity, and SEM tests.

#### 2.4.1. Compressive Strength

The compressive strength of the mortar cubic specimens was tested by a compression machine with a loading rate of 0.5 kip/sec. The specimens containing biochar were taken from the curing room a day before testing. This action was taken to avoid the adverse effect of internal water pressure on the test results since the specimens containing biochar were thoroughly wet after the ASTM proposed time [51]. This study represented the average of three compression tests for each mix design for each 7-, 28-, and 90-day compressive strength.

### 2.4.2. Ultrasonic Pulse Velocity

Ultrasonic is a promising non-destructive test method to evaluate the mechanical properties of CCs. The UPV is calculated by dividing the traveling time of the wave through the specimens by the traveling length. Earlier investigations showed that the technique can be used to estimate the air content and porosity of the CCs with reasonable accuracy [52]. The wave velocity changes based on the material solidity, the more solid the composite, the higher the wave velocity. Therefore, the UPV is reduced by increasing the porosity of the composites. Accordingly, the ultrasonic pulse velocity was employed to examine the effect of the biochar on the porosity of the mortar specimens. The ultrasonic wave was transmitted through the specimens perpendicular to the mortar casting director. In this study, honey was used as a couplant to improve the connection between the ultrasonic transducers (probes) and mortar surfaces. The test was conducted two times for each specimen (one time for each direction perpendicular to the casting director). Accordingly, the average of six measurements was reported as a UPV of each mix design in a specific curing time.

### 2.4.3. Scan Electron Microscopy (SEM)

The SEM was used to evaluate the effect of biochar and its presoaking on the microstructure of specimens. In this study, the SEM samples were collected from the core of the specimens after the 90-day compressive strength tests. The samples were then dried in an oven at a temperature of 100±5℃ for 24 hours to stop hydration and pozzolanic reactions. The dried samples were kept in sealed plastic Ziploc bags to avoid their contact with air humidity. After collecting adequate samples, the Leica EM ACE200 machine was used to cover the samples with a 20nm layer of gold-palladium powder via diffusion technique. The covered samples were then tested by HITACHI S-4700 scanning electron microscope.

## 3. Result and Discussion

### 3.1. Unit Weight

Figure 3 displays the effect of biochar dosage on the unit weight of the specimens. The data indicated that increasing the biochar percentage up to 15% decreased the 90-day unit weight of specimens by 14.2, 14 and 13% in the ordinary mortar (OM), mortar with 50% OPC replacement with fly ash, and mortar with 10% OPC substitution with RGD and 50% replacement by fly ash, respectively. Based on the unit weight reduction, it was obvious that adding biochar significantly increased the porosity of the mortar specimens since biochar only accounts for just 2% of the total weight of fresh mixtures in the mixes containing 15% biochar. The finding aligns with the porous structure of the biochar particles. Analyzing the unit weights of the specimens containing presoaked and non-presoaked biochar also revealed that using presoaked biochar intensified the unit weight reduction of the specimens. This is reasonable as the non-presoaked biochar captured a substantial quantity of water during mixing, reducing the amount of free water in the mixture, and increasing the density of the fresh concrete. This water capturing raises the density of the mixture and the specimens, enhancing their unit weight.

Although, at first glance, the intensified unit weight reduction of the specimens containing presoaked biochar could also be due to their higher water content. This higher water content did not affect the amount of free water and porosity of the fresh CCs and their unit weight since the additional water was trapped inside the biochar pores and released during the curing of the CCs.

This assumption was valid only if the biochar particles were not broken during mixing fresh CCs since breaking down the biochar particles could release their captured water and therefore increase the free water in the fresh CCs, increasing the porosity of the composite and reducing its unit weight.

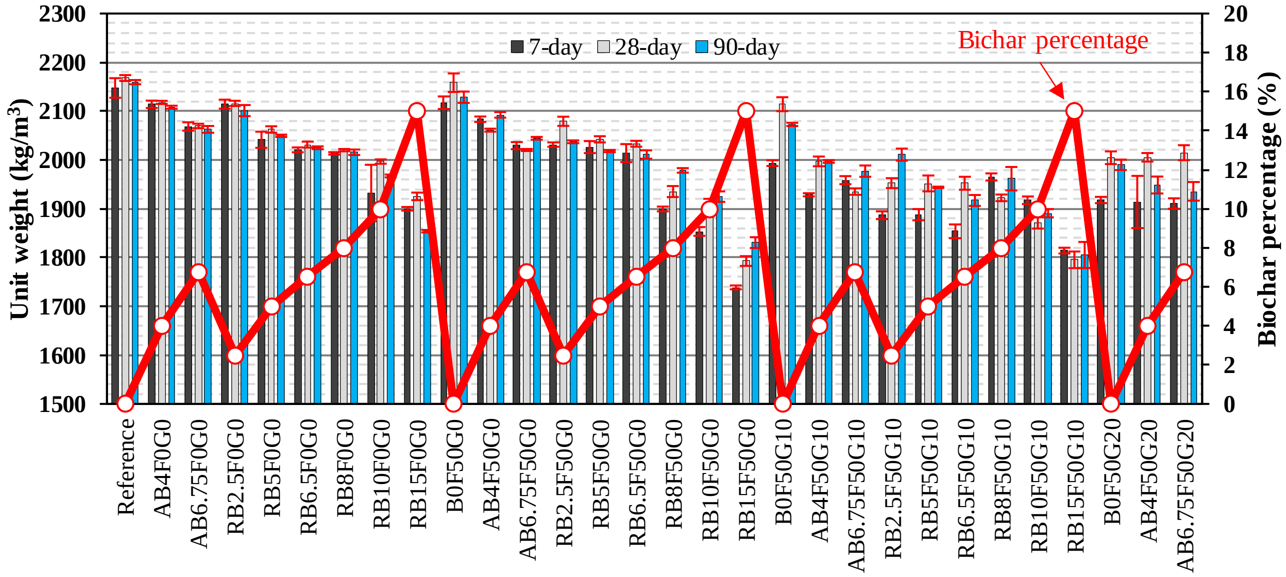


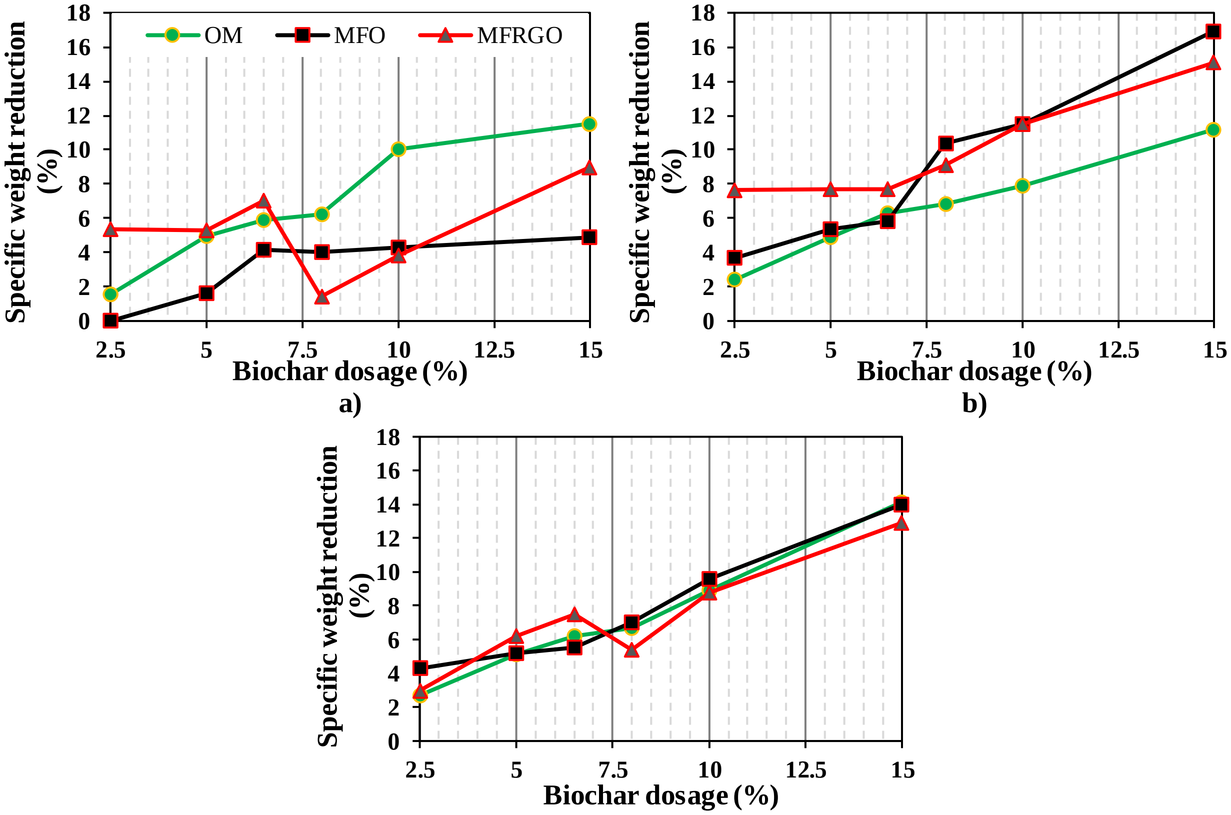
**Figure 3. Effect of biochar dosage on the unit weight**

Figure 4 shows the unit weight reduction of the specimens containing presoaked biochar on the testing days based on their benchmarks (specimens containing 0% biochar: B0F0G0, B0F50G0, and B0F50G10). The results demonstrated that, however, the effect of adding biochar on the unit weight of the specimens varied at 7-day measurements, the trends began to converge into an approximately linear trend at 90-day measurements. The addition of presoaked biochar had a lower impact on the unit weight reduction in the specimens containing fly ash compared to those containing OPC as a sole binder at 7-day measurements. However, the unit weight reductions of the specimens converged to a linear trend similar to that of ordinary mortars (OM) by 90 days when the delayed reactions of fly ash and gypsum were completed [4]. The 90-day pattern could also be supported by the porous structure of biochar particles and its effect on the porosity of the specimens. This emphasized that the biochar released the retained water when it was needed for the cementitious reactions. Therefore, it could be stated that the influence of biochar on the unit weight reduction of the mortars relied on their hydration level, however, further research on the hydration reactions and hydration heat of the CCs is required to validate this statement.

According to the data, the optimum percentage of cement replacement with biochar also changed depending on the supplied cementitious materials, which reflected the effect of the particle size distribution of the mortar components on the influence of biochar. For the ordinary mortar (OM) and mortar containing fly ash and OPC (MFO) increasing the substitution from 6.5% and 8% significantly raised the unit weight reduction rate. However, the data represented that the optimal percentage of OPC substitution with biochar in the mortar containing fly ash, RGD, and OPC (MFRGO) was 8%. Equations 1 can be used to estimate the 90-day unit weight reduction of the mortars containing presoaked biochar as cement replacement.

$$UWR(x) = \begin{cases} 0.9237 \times x & OM \\ 0.9392 \times x & MFO \\ 0.8905 \times x & MFRGO \end{cases} \qquad 0 \leq x \leq 15 \tag{1}$$

where UWR and x are the percentages of unit weight reduction and OPC substitution with biochar with respect to the benchmarks. Table 4 displays the coefficient of correlation ($R^2$) and root mean square error (RMSE) of Equation 1 for different CCs. The values of the factors showed that the linear functions could be used to predict the output with reasonable accuracy.



a) b) c)

Figure 4. Effect of biochar dosage on the unit weight reduction with respect to benchmarks in a) 7-day, b) 28-day, and c) 90-day specimens. OM: Ordinary mortar (BxF0G0), MFO: mortar containing 50% fly ash and OPC (BxF50G0), MFRGO: Mortar containing fly ash, recycled gypsum, and OPC (BxF50G10)

Table 4. The R factor and RMSE of the proposed equations for estimating the unit weight reduction

| Type of mortar | Benchmark mixture | $R^2$ | RMSE (%) |
|---|---|---|---|
| OM | B0F0G0 | 0.9906 | 0.4394 |
| MFO | B0F50G0 | 0.9601 | 0.8771 |
| MFRGO | B0F50G10 | 0.9028 | 1.2909 |

### 3.2. Compressive Strength

#### 3.2.1. Ordinary Mortars (OM)

Figure 5 exhibits the results of compression tests. The data demonstrated that the use of presoaked biochar in the mortar reduced the 90-day compressive strength of the specimens (from 5.7 to 50.1% in the specimens containing 2.5 to 15% presoaked biochar). However, findings indicated that replacing OPC with presoaked biochar up to 2.5% in the ordinary mortar increased the 7-day compressive strength of the specimens by 3%. This might be because the water provided by the presoaked biochar could accelerate the hydration reactions of the specimens at the early age [23,32]. Earlier studies stated that the ability of biochar particles to retain water and lessen voids in concrete due to their micro-filler features could account for this strength improvement [13,53].

The 7-day compressive strength enhancement was not detected in the 28 or 90-day measurements. The duration of water retention by biochar and its porous structure may account for this variation. According to previous studies, the retained water inside the biochar pores was released during the curing time and used by internal hydration reactions [12,28]. The authors' observations revealed that inside of the specimens was still wet after the 7-day tests, representing that the biochar pores were moistened during the tests. This could display that the retained water inside the biochar pores carried the applied loads to the biochar by increasing internal pressure in its pores in the 7-day measurements, which enhanced the compressive strength of the biochar and consequently the specimens. The retained water was gradually released and used by hydration reactions. Accordingly, there was less amount of water inside the biochar pores in the 28 and 90-day tests, which increased the air voids in the mortar and reduced the capability of biochar particles to carry the applied loads. Therefore, this strength loss could be explained by the low strength of biochar particles due to the lower amount of trapped water, internal pressure, in their pores in the

28 and 90-day specimens compared to the 7-day ones. Accordingly, this study would suggest additional air curing for the specimens containing biochar after the moist curing, which could reduce the effect of retained water on the properties of the CCs.

Results also evaluated the effect of presoaked and non-soaked biochar on the mechanical properties of the mortar. The data revealed that the compressive strength of the specimens containing up to 8% presoaked biochar was approximately time-invariant from 7 and 90-day measurements. This may be because presoaked biochar supplied extra water for internal hydration in the early ages which accelerated the strengthening reactions [32,54]. This could also show a drawback of using presoaked biochar in the mortar since this was an abnormal stop in the strengthening of the CCs. There were two possible reasons for this phenomenon 1) the hydration reactions were completed at the early age and 2) the fact that over time the biochar released the retained water, reducing its strength due to the internal pressure loss. The first theory was refuted as an increase in the strength of the specimens was observed from 28 to 90 days. Thus, this halt in the strengthening of the specimens was attributed to the reduction in the strength of the biochar, an increase in the porosity of the mortar, and the contribution of trapped water in the load-bearing capacity of the specimens containing presoaked biochar at early ages.

Despite the specimens containing presoaked biochar, the data showed a significant strength improvement in the specimens containing non-soaked biochar between 7 and 28 days (31.18 and 26.34% in the AB4F0G0 and AB6.75F0G0, respectively). This showed that adding non-soaked biochar increased the density of the specimens by capturing the water from the fresh mixture. The gradual discharge of the retained water during the curing time enhanced the internal hydration and the compressive strength of the mortar. However, it decreased the early age strength and possibly initial reactions of the mortar by reducing the available water for the reactions. The low early age strength of the specimens containing non-soaked biochar also increased the potential of the theory that the low strength of biochar accounted for the lower strengthening rate of the specimens containing presoaked biochar between 7- and 90-day measurements compared to the reference CC. This is because the biochar could not be fully saturated in the specimens containing non-soaked biochar and the majority of its pores were occupied by air. This reduced the water internal pressure in the pores compared to the presoaked biochar particles during loading and limited the strengthening of the specimens. Less OPC in the specimens containing presoaked biochar as binder replacement could also account for the lower strengthening rate of the specimens. However, the lower amount of OPC in the specimens containing presoaked biochar could also account for their lower strengthening rate. This theory challenged the use of biochar as a supplementary cementitious material since even a 2.5% substitution reduced the compressive strength of the specimens.

Finally, the comparison between the compressive strength specimens containing presoaked and non-soaked biochar showed that using non-soaked biochar had less impact on the long-term mechanical properties of CCs due to its ability to retain water and increase the density of the specimens. However, it could be expected that the use of non-soaked biochar might significantly reduce the workability of the fresh mortar in higher dosages. Moreover, the higher amount of water in the specimens containing presoaked biochar could increase the porosity of the CCs, weakening the composite. However, this may be a potential factor for the strength reduction, free water in the fresh mix containing presoaked biochar was less than the reference mixture since the extra water was retained inside the biochar pores and did not generate pores during the hardening of the composite. Nonetheless, breaking the biochar particles during mixing could result in releasing the retained water, raising the free water and porosity of fresh and hardened CC, respectively.

### 3.2.2. Mortar Containing Fly ash and OPC (MFO)

Results revealed that in contrast to ordinary mortars, adding non-soaked biochar into the mixture increased the 7-day strength of the MFO specimens. This might be caused by the slow reactions of fly ash at the early age with respect to OPC and the ability of non-soaked biochar to boost the density of the mixture by capturing free water in the mixture. In this case, the water retained by non-soaked biochar was beneficial for the mortar as the reduction in the early-age reactions (initial reactions) decreased the water demand in early ages. Therefore, it could be suggested that the type of cementitious materials and their level of reactivity at the early age played a major role in how the presoaking of the biochar affects the early age strength of the mortar specimens. This also accentuates the importance of determining an optimum saturation degree of the biochar for different cementitious mortars.

The data also represented that adding non-soaked biochar by up to 4% with respect to the weight of cementitious materials did not significantly influence the 90-day compressive strength of the mortar containing fly ash. However, adding the presoaked biochar into the mixture from 2.5 to 15% reduced the 90-day compressive strength of specimens from 18.48 to 69.63% compared to the B0F50G0. The lower demand for the retained water for strengthening reactions could cause this notable strength reduction in comparison with those of ordinary mortar specimens. The adverse impact of adding 6.75% non-soaked biochar could also emphasize the potential to disrupt the performance of fly ash and biochar in higher replacement ratios. Although each of the fly ash and biochar particles could act as fillers in the mixture, combining them adversely affected the porosity of the specimens since exceeding the amount of filler from an optimum in the mixture reduced their efficiency and increased the number of pores between the fillers. The lower reactivity of fly ash particles compared to the OPC lessened the required water for internal hydration in the matrix resulting in the extra water that could dissolve some of the hydration products such as ettringite. This dissolving could be considered a source of strength reduction since it increases the porosity of the mortar and weakens the bond between biochar and cement matrix. However, it could be expected that the strength of the composite will be gained slightly over time by using the water in the long-term strengthening reactions. These results highlighted the importance of examining the effect of biochar in CCs with various supplementary materials as the particle size distribution of mortar components could significantly affect the properties of CCs containing biochar.

The lower OPC content of the specimens containing presoaked biochar compared to those containing non-soaked biochar could also be used to explain the inferior compressive strength of the CCs containing presoaked biochar in high dosages. The lower the OPC content, the lower the hydration heat. The higher hydration heat increased the temperature of the fresh mixture, which could raise the pozzolanic reactivity of fly ash and improve the compressive strength of CCs [3]. Nonetheless, since the literature [28,54] showed that using biochar could intensify the hydration of CCs, further research is required to validate this theory by evaluating the cumulative hydration heat of the specimens containing biochar as OPC replacement with reference, i.e., biochar-free, mixtures.

### 3.2.2. Mortar Containing Fly ash, RGD, and OPC (MFRGO)

The data showed the positive effect of replacing OPC with presoaked biochar up to 2.5% on the 7-day compressive strength of the specimens. However, the 90-day compressive strength of the specimens containing 10% RGD was reduced between 26.29 to 74.75% by increasing the dosage of presoaked biochar from 2.5 to 15% with respect to the weight of cementitious materials. This strength reduction was more than even those of the specimens containing fly ash. This might be due to the lack of sufficient OPC, hydration heat, and the adverse effect of using multiple fillers with almost the same particle distribution size, i.e., RGD and biochar. The reactivity of the RGD and fly ash was significantly affected by the amount of OPC in the cement matrix since the hydration reactions of OPC were the source of heat in the matrix, promoting the reactivity of RGD and fly ash [4]. The similar size distribution of the biochar and RGD could potentially increase the porosity of specimens, reducing the strength of the composite.

The data showed that the addition of non-soaked biochar had less of an impact on the compressive strength of the mortar when compared to presoaked biochar. This could be because of the low reactivity of the fly ash and RGD at the early age, scaling down the demand for water in the initial cementitious reactions compared to the ordinary mortars. Accordingly, there was extra water in the mixture retained by the non-soaked biochar particles, which gradually released over time. In this case, the water capturing of the non-soaked biochar was beneficial because the initial reactions required less amount of water and the water retention increased the density of the specimens, which was reflected by the higher compressive strength of the specimens compared to the presoaked ones. Moreover, the increase in the dosage of presoaked biochar further reduced the cement content in the fresh mixture, which decreased the hydration heat of the CCs, reducing the reactivity of fly ash. The potential of releasing the retained water inside the biochar pores due to breaking down the presoaked biochar particles during mixing could also be responsible for the strength reduction since it increased the free water in the fresh mixture, boosting the porosity of the composite.

Findings indicated that replacing OPC with 10% RGD in the specimens containing fly ash was more beneficial for the long-term, i.e., 90-day, compressive strength of the mortar since the strength did not significantly drop with respect to the B0F50G0 in the case. The data also indicated that the low early-age strength of the mortar was one of the main barriers to using CCs containing fly ash and RGD in the construction industry. However, the 90-day compressive strength of the specimens containing 10% RGD was almost equal to the strength of the specimens containing fly ash. This showed the potential for using RGD in the mortar containing fly ash and OPC.

The data also demonstrated that adding the non-soaked biochar to the mix containing 20% RGD caused a 15.45% strength reduction in 90-day measurements. However, this strength loss was slightly less than that of specimens containing 10% RGD (16.75%). The noticeable strength gain was observed from 28 to 90 days in the specimens, which not only showed the significance of biochar and its retained water for internal hydration of the specimens but also highlighted the potential of occupying the biochar pores by the RGD-cement reaction expandable products. However, this theory needs proof from the microstructural analysis of the specimens discussed in Section 3.4. The data also represented that replacing 20% of OPC with RGD in the mortar containing 50% fly ash had less of an impact on the strength of the mortar than substituting 5% of OPC with presoaked biochar in the mixture, revealing the feasibility of using RGD in the mortar compared to biochar. This difference in the effect of biochar and RGD could refer to the reactivity of RGD with cement paste and the poor structure of biochar particles.

Figure 6 shows the influence of partially substituting OPC with presoaked biochar on the 90-day compressive strength of the specimens. Findings presented that the effects of biochar on the 90-day compressive strength of the MFO and MFRGO were comparable and could be modeled linearly. Nonetheless, the influence of biochar content on the compressive strength of OM was more complex. According to the data, the behavior could be simulated by a second-order polynomial function, which resulted in an $R^2$ of 0.95. Equation 2 illustrates the formula that can be used to estimate the 90-day compressive strength of the specimens.

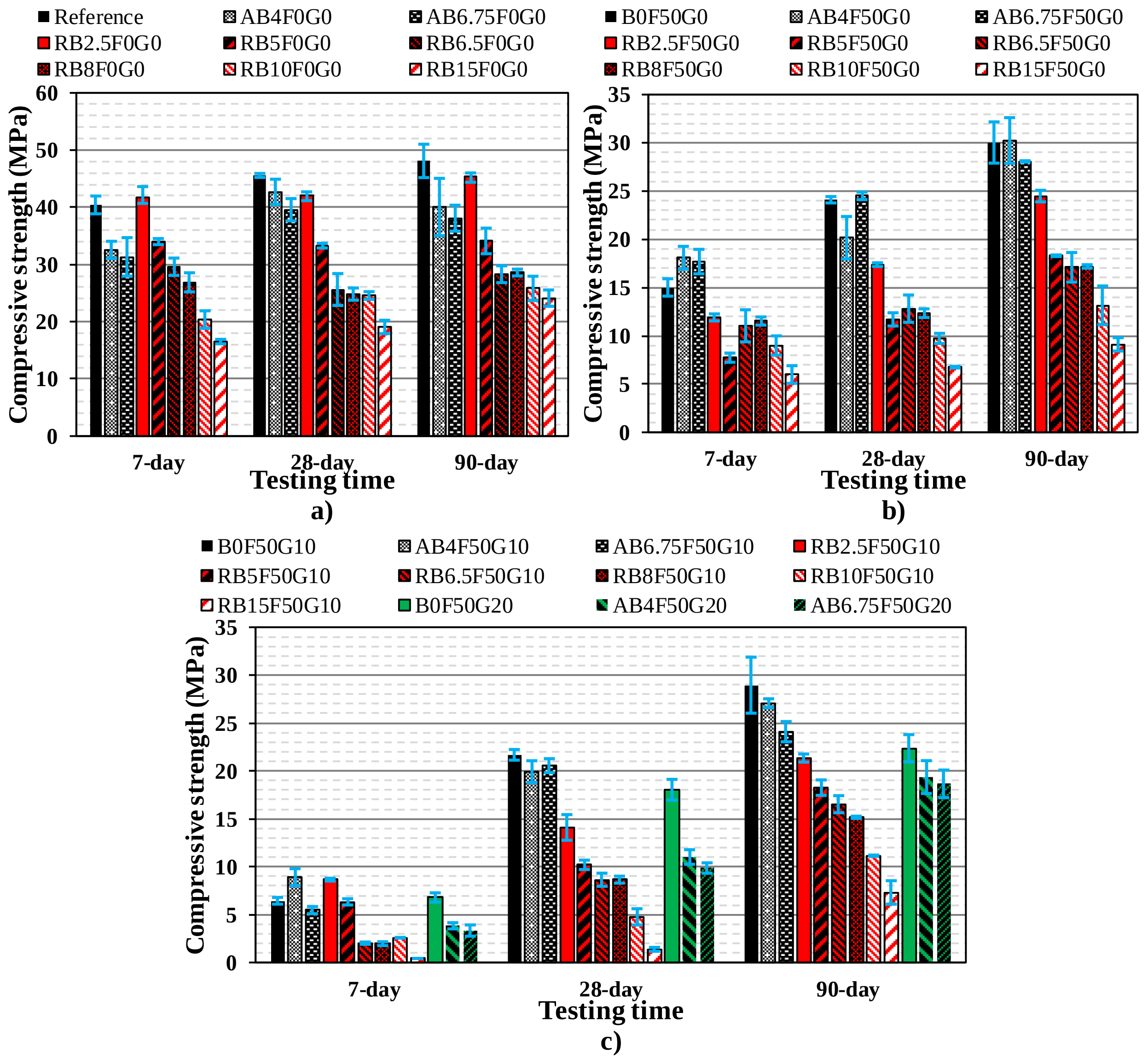


**Figure 5. The effect of biochar content on the compressive strength of a) OM, b) MFO, and c) MFRGO**

$$CS(x) = \begin{cases} 0.1406x^2 - 3.8537x + 50.073 & OM \\ -1.3541x + 27.576 & MFO \\ -1.3778x + 26.207 & MFRGO \end{cases} \quad 0 \le x \le 15 \qquad (2)$$

where CS and x are the 90-day compressive strength and the percentage of cementitious materials, including OPC, fly ash, and RGD, replacement with biochar with respect to the benchmarks.

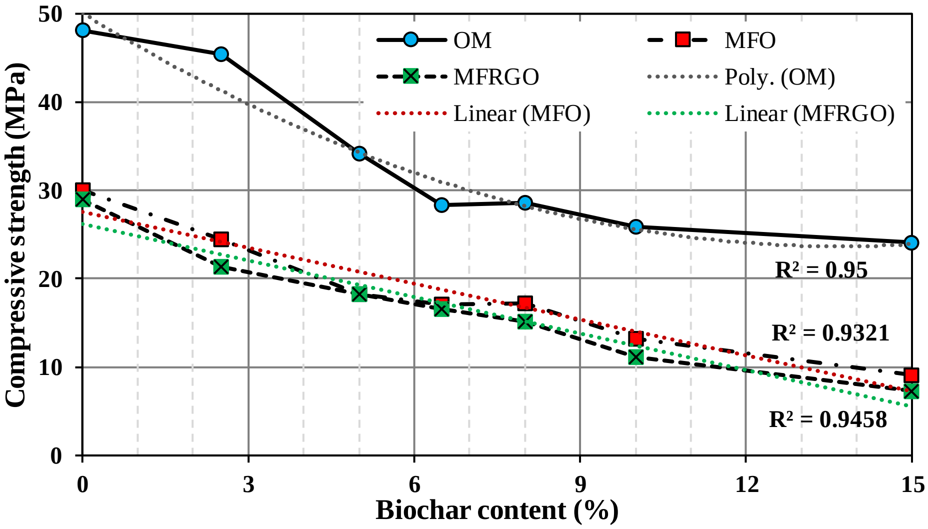


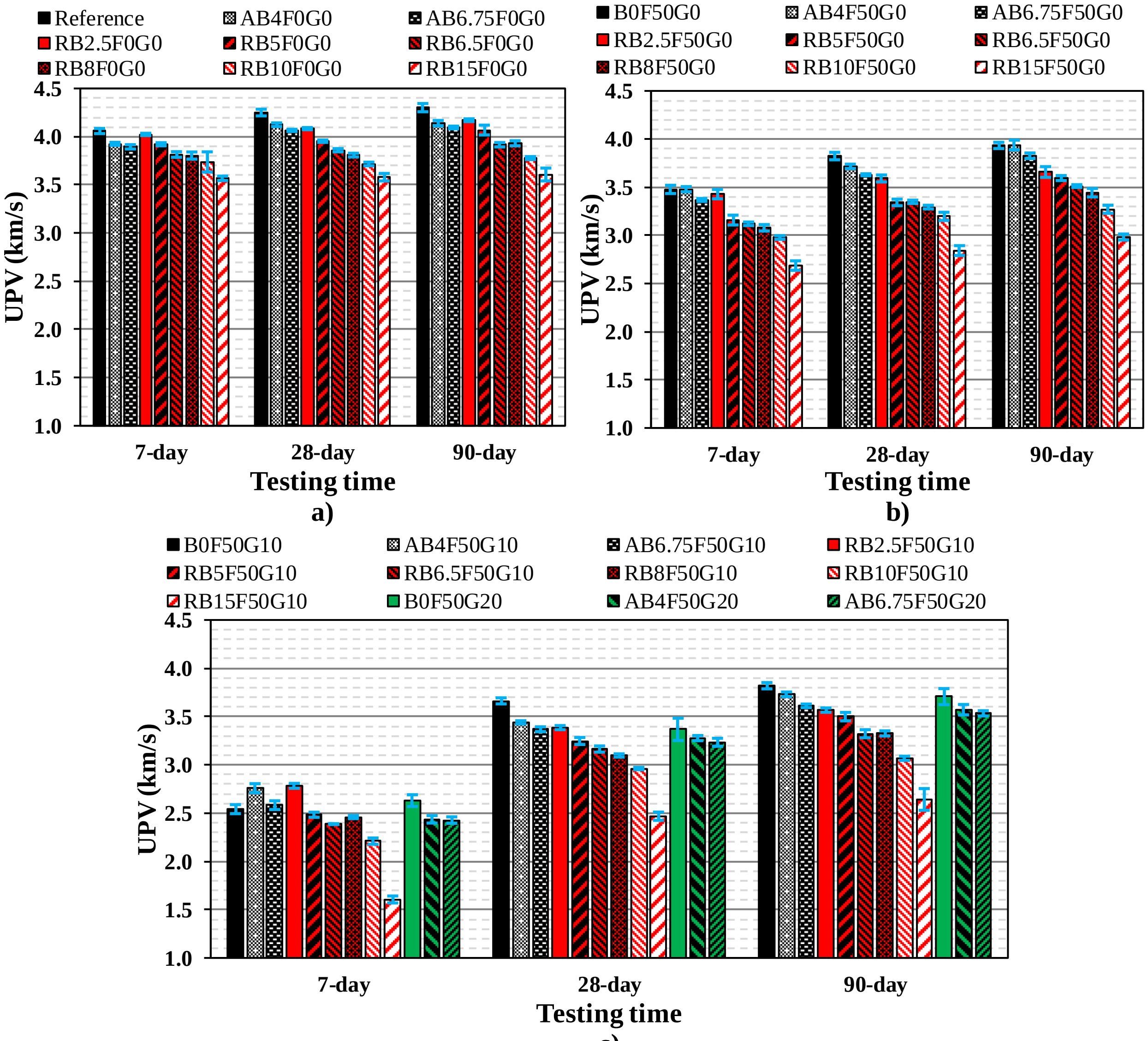
**Figure 6. The influence of biochar dosage on the 90-day compressive strength**

### 3.3. Ultrasonic Pulse Velocity

Figure 7 displays that increasing biochar dosage reduced the UPV of the specimens, which pointed to an increase in the porosity of the specimens. This could be explained by the poor structure of the biochar and the fact that adding biochar increases the number of pores in the CCs. However, it modified the size of the pores according to the size of biochar pores, which was beneficial for some of the durability characteristics such as resistance to the freeze-thaw cycles [21]. The data also indicated that adding non-soaked biochar had a less adverse effect on the porosity of the composite compared to the use of presoaked biochar in the MFO and MFRGO. This supported the fact that the non-soaked biochar increased the density of the matrix compared to the presoaked biochar by trapping the free water in the mixture. Reducing cement content and a possible increase in the free water due to the release of retained water from broken biochar particles might also have a significant impact on the porosity and compressive strength of the composite. The significant increase in the porosity of the specimens containing biochar compared to the benchmarks also emphasized the importance of developing a technique to fill the biochar pores during the curing time while it has a minimum effect on the water retention of the biochar. This is because the biochar loses its ability to provide water for internal hydration if the pores are occupied with solid materials before mixing.

The results also demonstrated that the 90-day UPV reduced by 3.02, 6.87, and 6.54 to 16.05, 24.17, and 30.89 when the OPC substitution was increased from 2.5 to 15% in the OM, MFO, and MFRGO, respectively. This could identify the reverse impact of using two fillers (biochar and fly ash) on the porosity of the mortar and emphasize the importance of research on determining the desirable content of the combination of the fillers and their optimum ratios in the mixture. Analyzing these data and the effect of biochar dosage on the compressive strength of the specimens also revealed two points. First, the UPV did not accurately evaluate the porosity of the mortar due to the water captured inside the biochar pores, affecting the speed of wave transition inside the specimens. Second, while the UPV considered some parts of the biochar particles as solid materials, they were not able to carry the load. This accentuates the significance of improving the mechanical properties of biochar particles in the mixture.

UPV is also well-known as one of the non-destructive methods that can be used to evaluate the mechanical strength of the CCs. Figure 8 shows the scatter plot of the 90-day compressive strength of specimens with respect to the UPV results, which demonstrates the correlation between these two parameters. The data revealed a high coefficient of correlation between the parameters which depicted the potential of using UPV in simulating the compressive strength of the CCs. Equation 3 also represents the proposed formula that can be used to estimate the 90-day compressive strength of the CCs based on the UPV evaluations.



a)

b)

c)

**Figure 7. The effect of biochar content on the UPV of a) OM, b) MFO, and c) MFRGO specimens**

$$CS = \begin{cases} 37.981 \times UPV - 117.2 & 3.5 < UPV < 4.5 \quad OM \\ 22.168 \times UPV - 58.779 & 3.0 < UPV < 4.0 \quad FMO \\ 17.663 \times UPV - 41.685 & 2.5 < UPV < 4.0 \quad MFRGO \end{cases} \tag{3}$$

where CS and UPV are the compressive strength and UPV results of the specimens at 90-day measurements, respectively.

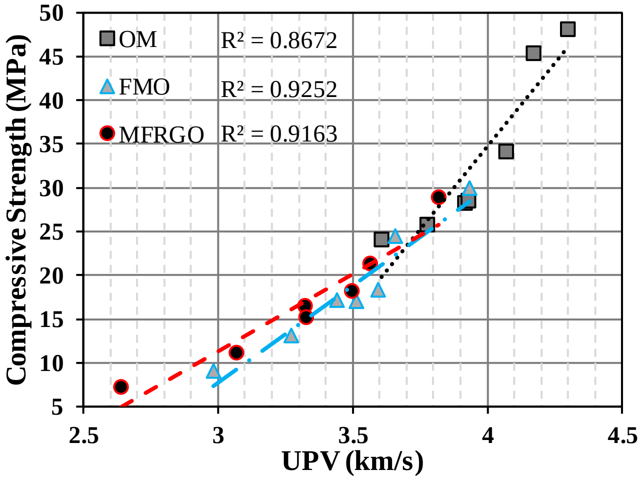


**Figure 8. Scatter plot of compressive strength of CCs vs UPV**

### 3.4. Microstructural Analysis

This section evaluates the microstructure of the specimens at 90-day measurements. Figures 9 to 11 represent the scan electron microscopy (SEM) of the OM, MFO, and MFRGO specimens. The SEM results depicted that the biochar particles were bound to the cement paste since no discernible separation could not be observed. The finding showed no debonding between biochar particles and cement paste at the 90-day measurements. Additionally, Results indicated a notable interlock between cement paste and biochar particles, supporting the theory that the strengthening effects of adding biochar to the CCs include an increase in interlock. These results indicated that the biochar has a good compatibility with the CCs. However, the increase in the interlock pointed out another major cause for the significant effect of biochar on the workability of the fresh CCs.

Examining the surface of the biochar particles also illustrated the fact that there was no reaction on the surface of the biochar particles. This emphasized the fact that the biochar particles did not react with the cement paste. This also aligns with the chemical compositions of the biochar as carbon, one of the most stable chemical elements, constitutes more than 56% of the chemical compositions of the biochar. Additionally, the presence of ettringite crystals next to biochar particles in some cases, even in the mixture without RGD, can cause serious concerns about the durability of the CCs containing biochar. This is because ettringites can dissolve in the water and produce air voids in the CCs, impacting the durability of the CCs in marine and coastal environments. Nonetheless, this could support the capability of biochar to provide nucleation sites with higher crystallization rates (See Figures 9-f, 10-f, and 11-f). According to this effect, adding biochar can increase the nucleation rate and size of crystals in the composites, which can affect the durability properties of CCs [55].

Accordingly, it could also be stated that the reduction in the compressive strength of the specimens was caused by two main factors: an increase in the porosity of the CCs, and the low strength of biochar particles. The boost in the porosity of the CCs by adding biochar was already noted in the literature. However, examining the SEM pictures could reveal that the biochar particles were smashed into pieces in the tested specimens under compression test. This claim was confirmed by the fact that there was cement paste on the remained biochar-cement boundaries in the pictures (see Figure 9-c). However, the broken side of the biochar was completely cement paste-free. Therefore, this study proposes the use of filled biochar particles to lessen their effect on the porosity and increase their strength. This fulfillment may also intensify the cementitious reactions if the fulfilments choose from the reactive materials with cement paste. The results could also support the hypothesis that retained water in the biochar pores carried the applied load to the specimens containing biochar at early ages since there was no vivid separation in the cement paste-biochar interface. This accentuates the need to examine the effect of dry climate on the long-term properties of CCs containing biochar.

The SEM analysis of the specimens containing fly ash and RGD also discovered the potential of fly ash activation by RGD because several fly ash particles with non-smooth surfaces were observed in the SEM pictures (see Figures 11-b and d). The results also indicated that the use of RGD boosted the formation of ettringite crystals in the hardened mixture, pointing out the significance of examining the long-term properties of the CCs containing RGD in aggressive and marine conditions.

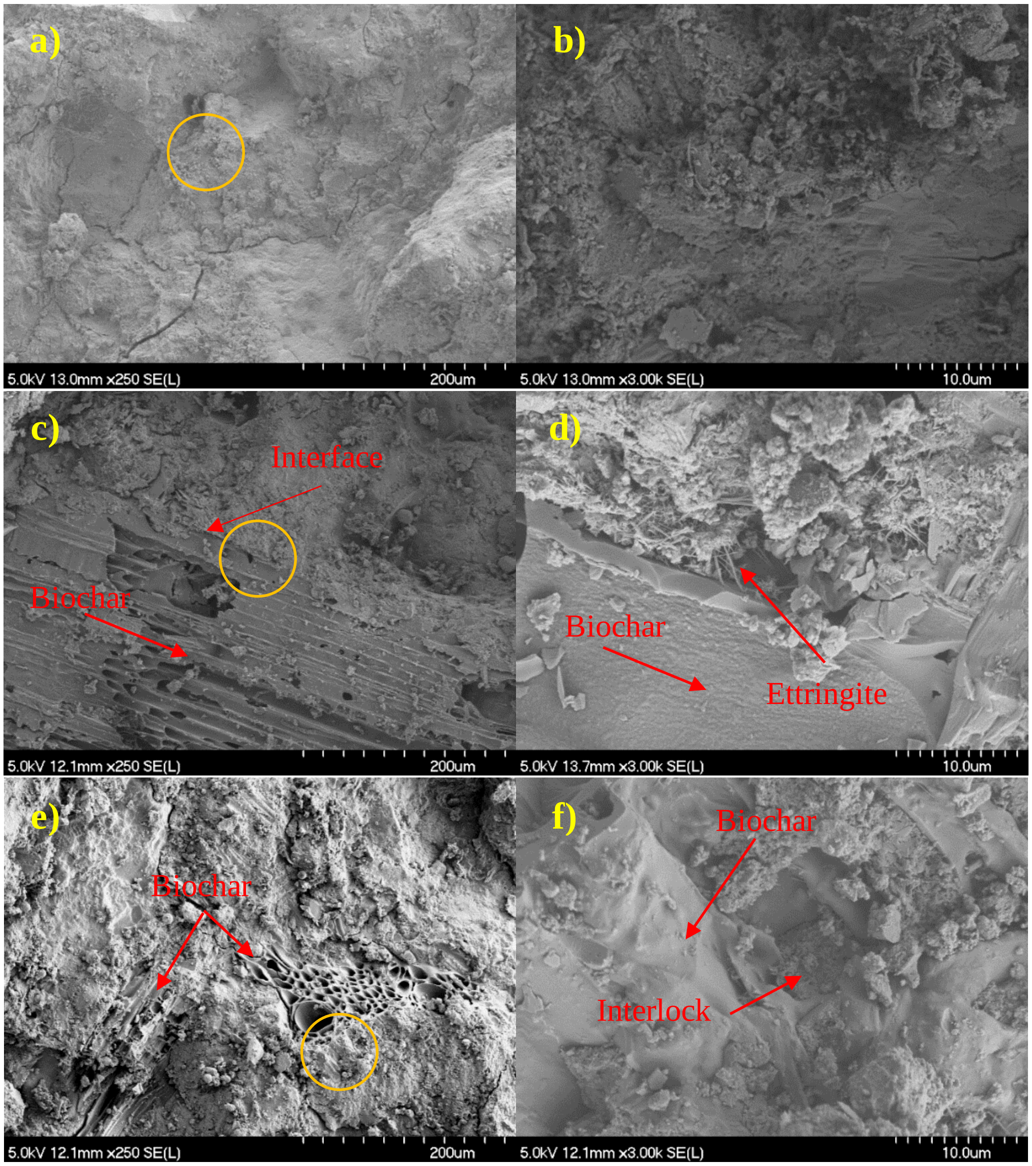


**Figure 9. SEM images of a & b) B0F0G0, c & d) RB2.5F0G0, e & f) RB5F0G0, and g & h) AB4F0G0**

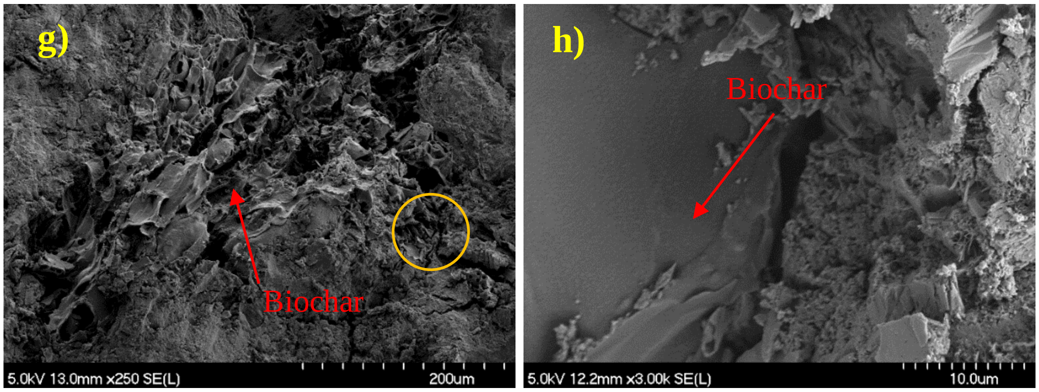


Figure 9. SEM images of a & b) B0F0G0, c & d) RB2.5F0G0, e & f) RB5F0G0, and g & h) AB4F0G0 (cont.)

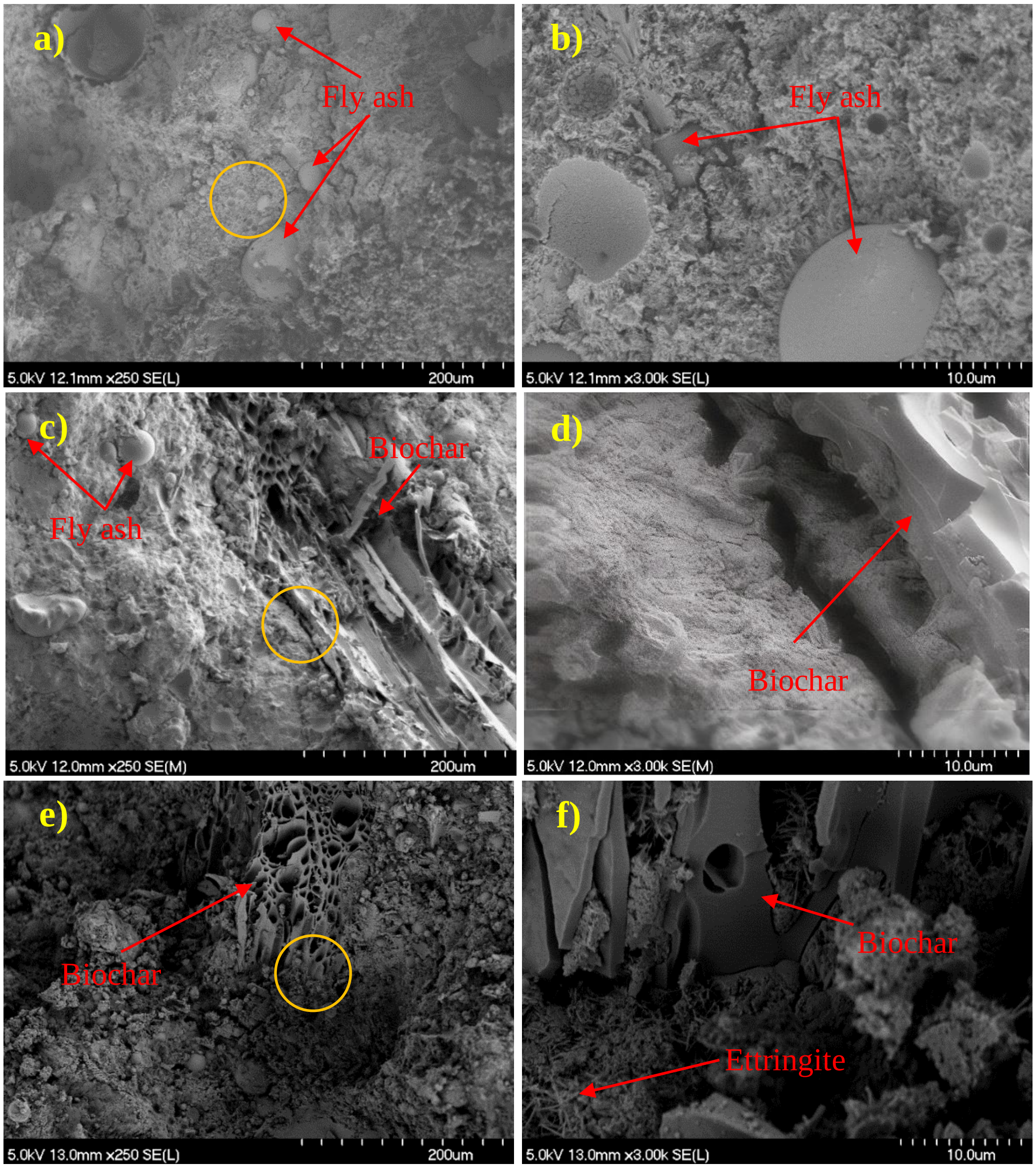


Figure 10. SEM images of a & b) B0F50G0, c & d) RB5F50G0 and, e & f) AB4F50G0

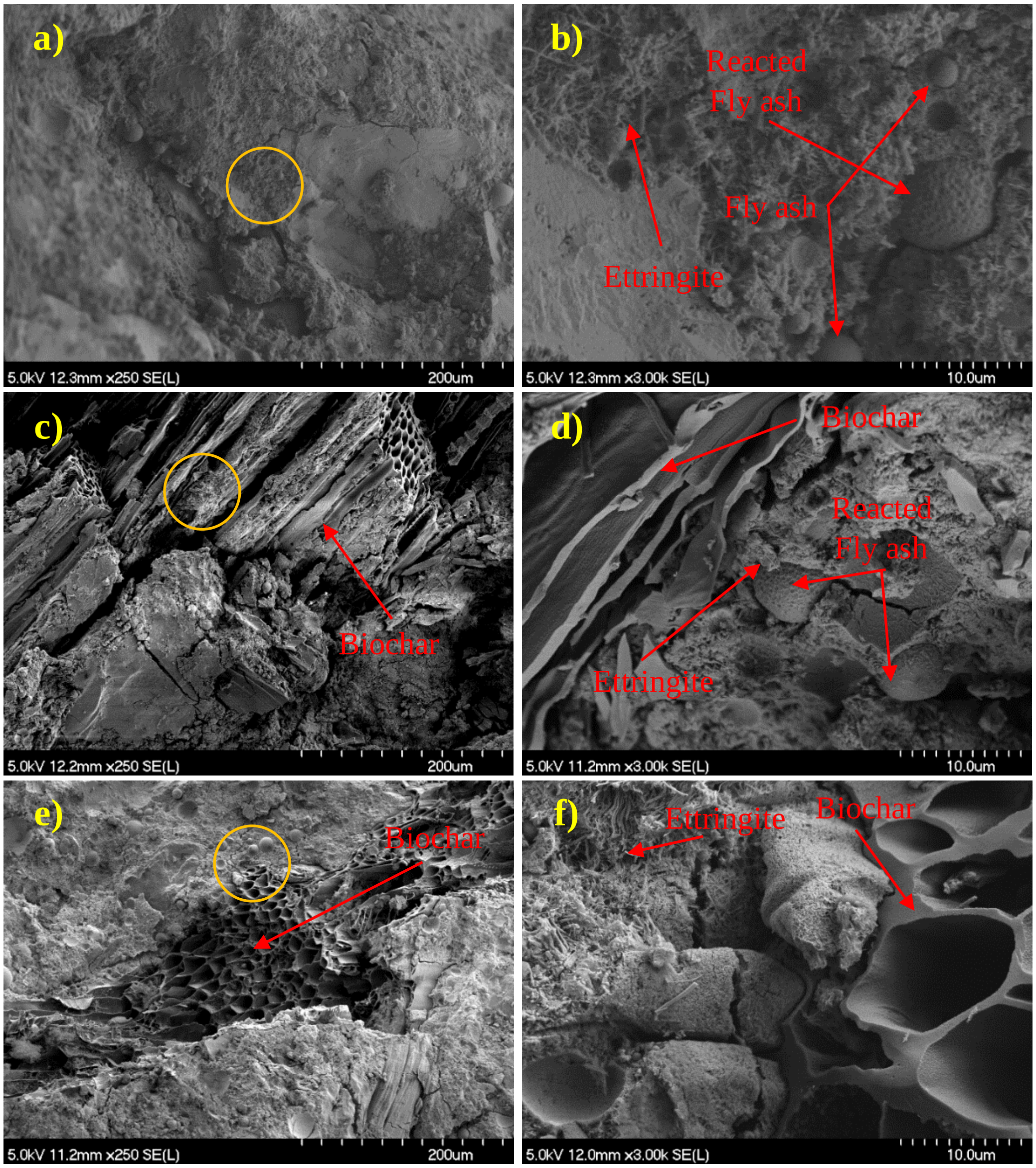


**Figure 11. SEM images of a & b) B0F50G10, c & d) RB5F50G10 and, e & f) AB4F50G10**

## 4. Conclusions

This article studies the effect of biochar on the compressive strength, UPV, and microstructure of ordinary mortar, mortar containing fly ash and OPC, as well as mortar containing fly ash, RGD, and OPC to address four research gaps in the literature. 270 specimens were manufactured and tested at 7, 28, and 90 days. This study also investigates the influence of presoaking on the properties of the mortar specimens. The main outcomes of the study can be pointed out as follows:

- Adding biochar to the mixture reduced the compressive strength of the specimens due to the low strength of the biochar particles after releasing their retained water and their influence on the porosity of the mortar. Therefore, developing a technique that can gradually occupy the biochar pores during cementitious reactions could significantly increase the strength of the specimens by increasing the load capacity of the biochar particles.
- Since releasing the retained water from the biochar pores considerably affected the mechanical properties of the composite, the long-term strength of the specimens containing biochar in dry conditions should be examined to evaluate the service life and final strength of the mortar containing biochar in arid climates.
- The effect of presoaking significantly depended on the initial reactivity of cementitious materials in the mixture. The presoaked biochar was beneficial for the mortar containing cementitious materials with a higher level of initial reactivity since it promoted the initial reactions by providing additional water. However, using the non-soaked biochar was proposed for the mortar containing cementitious materials with low early age reactivity. It is because the non-soaked biochar retained the extra free water that was not used in the initial reactions, which increased the density of the specimens, improving the mechanical strength of the specimens compared to those contained presoaked biochar.
- The influence of the biochar addition to the properties of the specimens significantly depends on the particle size distribution of the cementitious materials in the mixture. The reverse effect of using multiple types of fillers (biochar, RGD, and fly ash) was determined.

This highlighted the necessity of examining the effect of biochar on the specimens containing RGD and fly ash to figure out their optimum proportions to minimize their reverse impact on the properties of mortar.

- Adding biochar to the matrix increased the porosity of the specimens, though it adjusted the size of voids in the mortar. However, this adversely impacted the strength of the specimens, it could enhance the durability characteristics of the specimens.
- A good compatibility between biochar particles and cement paste was detected. However, since the interfacial transition zone was mostly filled with ettringite crystals, the long-term durability properties of the CCs containing biochar should be evaluated to address the serious concerns about dissolving the formed ettringites in marine and coastal environments.
- The effect of biochar on the mechanical properties of CCs can be summarized in i) increasing interlock in the fresh CCs, ii) reducing the water-to-binder ratio in the mixture containing non-soaked biochar and thereby increasing the density of fresh CCs, iii) boosting the early age hydration by providing additional surface and water, iv) enhancing the nucleation rate, and v) providing water for internal curing.
- Due to the high-water retention capacity of biochar particles, the specimens need to be in the air condition for more than the required time in ASTM, since the water inside the biochar pores could result in overestimating the compressive strength of the CCs as it carried the significant part of the applied load to the biochar particles in the CCs.
- The potential of using RGD in the mortar containing fly ash to develop a sustainable cementitious composite was detected. RGD activated the fly ash and had a less negative impact on the strength of the specimens compared to biochar. However, it was revealed

that one of the essential drawbacks of using RGD was its delayed reactions, lowering the early-age strength of the specimens containing RGD.

## 5. Future Studies

This study examines the effect of biochar dosage and its presoaking on the compressive strength, porosity, and microstructure of mortar specimens. The examination highlighted multiple theories that needed determined validations. Moreover, the following research directions could be proposed for future studies with respect to the obtained results.

a) Since the compressive strength of the specimens containing biochar artificially increased due to the internal water pressure inside the biochar pores, investigations on the effect of curing type (immersion or spraying) and the long-term properties of CCs containing biochar in arid climates are needed. It is because the provided water for curing the concrete was different in each technique and the retained water in the biochar particles may not be used in the immersion method. Thus, the retained water could still carry the applied load and the internal pressure was not significantly changed during curing. The arid climate could also accelerate the evaporation of the retained water in the biochar, producing micro to macrocracks in CCs and reducing the artificial strength of biochar particles. Simulations such as drying specimens in the oven could also be used to evaluate the effect of arid environments on the properties of the CCs. However, the temperature of the oven should adjust based on the temperature of the studied climate.

b) The low strength of biochar particles also emphasized developing a method to increase their strength without affecting their performance in providing additional water for internal curing. This can be done by using pretreatment biochar particles such as the used biochar in water and wastewater treatment. This pretreatment can also improve the bond between the biochar and cement paste since the treated biochar particles are covered with heavy metals that can react with cement paste in an alkaline environment. The products of the reactions can also gradually occupy the biochar pores and strengthen the particles, while reducing the ettringite crystals in the nucleation zone.

c) The correlation between the saturation level of biochar and the reactivity of the cementitious materials also needs further research. The research can ultimately result in a diagram that provides the saturation levels of biochar based on the used cementitious materials in a specific CC. This not only could maximize the effect of biochar in CCs by optimizing porosity and hydration levels in the hardened CC.

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