Biochar and Recycled Gypsum Drywall in Concrete: Role and Effects on Compressive Behavior, Microstructure, and Carbon Footprint

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Abstract

This research examines the effect of biochar and recycled gypsum drywall (RGD) on the mechanical properties and microstructure of conventional and high-volume fly ash concrete (HVFC) by testing 60 cylinders. Results suggested that although adding biochar increased the porosity of concrete, its effect on concrete's properties relied on its porosity, pore interconnectivity, and water retention capacity, plus characteristics of binders. The biochar-retained water (BRW) accelerated the hydration of C3S, increasing the dosage of calcium carbonate and calcium aluminate silicate hydrate on the top of biochar pores. The accelerated hydration enhanced early-age strength. Besides the hydration acceleration of OPC, BRW primarily promoted the RGD reactions and fly ash activation in HVFC. Nonetheless, the high porosity, inertness, and potential deterioration of biochar weakened the concrete in the long term and lowered its elastic modulus. Biochar also boosted ductile behavior, especially in HVFC with RGD, and lowered the carbon footprint of the concrete.

Keywords: Biochar, Fly ash, Recycled gypsum drywall, Concrete, Compressive behavior

DOI: https://doi.org/10.1080/21650373.2024.2428987

1. Introduction

The swift growth in urbanization increases the demand for new structures and infrastructure and thereby construction materials. Concrete is the most widely used construction material that uses an environmentally unfriendly binder, ordinary Portland cement (OPC). According to earlier evaluations, OPC production accounts for over 7% of annual global carbon dioxide emissions [1,2]. This substantial contribution to emitting the leading greenhouse gas and the urgent need to decelerate global climate change highlights the importance of reducing the OPC demand by replacing it with more eco-friendly binders. The substitution also lowers the demand for natural resources since 1.5 tons of raw materials are required to produce each ton of OPC [3,4]. Accordingly, numerous studies have been launched to replace OPC, either entirely or partially, in concrete with its eco-friendly alternatives, supplementary cementitious materials (SCMs) [3,5–8].

Biochar is a trending potential alternative to OPC, which provides additional water for the internal curing of concrete [9–11]. This carbon-rich porous fine material is a byproduct of a sustainable power generation technique (pyrolysis), which thermally decomposes the combustible biomass waste at a low oxygen level. Prior studies showed that using biochar in concrete significantly lessened its carbon footprint by reducing the OPC demand and its carbon sequestration feature, sequestering 2.52-3.3 kg CO_2-eq/kg [12,13]. Moreover, Park et al. [14] revealed that replacing 8% of cement with biochar could reduce the thermal conductivity of the cementitious mortar by up to 57.6%. Cuthbertson et al. [15] also revealed that substituting 1 to 2% cement with biochar could minimize the thermal conductivity of concrete. This reduction considerably improved the energy efficiency of the buildings, reducing the electricity demand and the need for power generation. The strategy also addresses public concerns about biochar and provides a sustainable waste management practice. It is imperative because more nations are turning to this power-generating technique due to its environmental benefits, including reusing biomass waste and reducing the carbon footprint of power generation plants by up to 67% [16].

Accordingly, numerous investigations examined the effect of using biochar in concrete [13,17,18]. Qing et al. [19] showed that adding biochar up to 1%, with respect to the weight of cement, into conventional concrete improved the 28-day compressive strength (CS) of concrete by 5.3%. Their data also displayed that adding biochar up to 5% enhanced the early-age (1-day) strength of concrete by 30.9%. Dixit et al. [20] assessed the hydration heat of high-performance concrete containing biochar. Their analysis depicted that replacing OPC with biochar up to 5% enhanced the hydration heat evolution of the concrete by up to 10% in the first 7 days. According to the literature [6,17,21,22], the water retention capacity of biochar and its potential to offer nucleation sites for the formation of additional hydration products were the primary characteristics of biochar that accelerated the early strengthening of the biochar-modified concrete.

Despite these interesting findings on promoting the early-age strength of cementitious composites, accurate data analysis revealed that the difference in the CS of conventional concrete and those containing biochar shrank when comparing 28- and 7-day data. The strength reduction was also accompanied by a decrease in the optimum biochar dosage at 28-day measurements [10,19]. This raises a serious concern about the possibility of a continuous decrease in the difference of the strengths over time when a considerable amount of biochar-retained water was used in the hydration reactions or cementitious reactions were completed. This is because it may result in a lower strength in the concrete containing biochar compared to conventional concrete in the long term. This concern predominantly stands out when an earlier study [10] revealed that, although the biochar addition improved the early hydration rate, a portion of the increase in the early CS of the cementitious mortar might be an artificial enhancement caused by the load-bearing capacity of the retained water in the biochar pores. Moreover, Xu et al. [23] highlighted the effect of the cement paste environment on the biochar properties. According to their results, exposure of biochar to cement paste could reduce the elastic modulus (by 59%) and hardness (by 81.4%) of biochar particles through their deterioration over time. These possibilities are essential arguments for the effect of biochar on concrete since they can significantly affect the performance and strength of concrete containing biochar in the long term, especially in dry climates. To address this gap, Alice et al. [24] examined the effect of biochar as an additional filler on the long-term mechanical properties of concrete. Their results illustrated that adding biochar up to 2.5% with respect to the weight of OPC boosted the compressive and flexural strengths of concrete at a 2-year measurement. However, the strength rise could be caused by the reduction in free water since they deducted the amount of water used for presoaking biochar from the mix water, which ultimately scaled down the water-to-cement ratio. The demand for further investigation on the long-term effect of biochar was also highlighted in the literature [25,26]. Despite these arguments, the long-term effect of biochar and the effect of biochar-retained water removal on the properties of concrete were not conclusively determined earlier [26].

Given the potential high correlation between the short- and long-term effects of biochar on the concrete properties, determining the optimum range from earlier studies could shed light on optimizing the evaluation. Recent studies [6,20,22,24,27–31] have used substitution dosages ranging from 0 to 5% to examine the effect of biochar on the mechanical properties of cementitious composites. However, a group of studies [10,14,19,32–40] went beyond the dosage and expanded the higher limit up to 30%. Analyzing the data of the group of studies [10,14,19,32–40] showed that the maximum percentage of OPC replacement with biochar that could enhance the 28-day mechanical strength of cementitious composites was 6.5%. However, the majority of the studies reported an optimum dosage in the range of 2 to 5% [14,19,20,27–30,32–34,36–40]. Higher replacement dosages notably reduced the strength of the cementitious composites. Prior research [19] also showed that the optimum replacement ratio could be as low as 1% of cement weight depending on the gradation and chemical compositions of biochar and other concrete components. Therefore, it is reasonable to examine the effect of replacing cement with biochar in the range of 0-6.5%, with a primary focus on 2.5 and 5%. The low substitution dosage also emphasized the importance of using other environmentally friendly cementitious materials in the cementitious composites containing biochar to lower the OPC demand and thereby lessen its detrimental environmental impacts. Unlikely, the effect of biochar dosage on the mechanical strength of eco-friendly cementitious composites was rarely studied in the literature [10,26,27,41]. Nonetheless, the interaction between the biochar and eco-friendly binders could substantially affect the performance of biochar in the composite.

To address this gap, a few studies investigated the effect of biochar dosage on the properties of geopolymer and alkali-activated composites [42]. In the most recent research, Egodagamage et al. [43] examined the effect of two types of biochar, derived from rice husk and sewage sludge, on the properties of alkali-activated concrete. Their results indicated a maximum strength improvement of 12.2% in the specimens containing 2% biochar when the biochar was used as an additive. Despite the promising results of the study and the environmental benefits of the geopolymers and alkali-activated composites [3], their applications in the construction industry were limited because of the safety precautions required for working with alkaline solutions.

Using SCMs is another strategy for reducing the demand for OPC and accordingly the environmental impacts of cementitious composites. Fly ash is a popular SCM used to reduce the OPC demand in concrete. The benefits of reusing this byproduct powder extend beyond concrete.

The approach offers a sustainable waste management practice for the byproduct and mitigates its negative environmental influences such as contamination of nearby soil and water resources by heavy metals [44]. Moreover, earlier studies denoted that using fly ash in conventional concrete significantly improved its mechanical strength and durability characteristics by reducing air voids and drying shrinkage [45,46]. These positive effects encouraged researchers to investigate the influence of using fly ash in higher percentages, between 30 to 50% of the weight of cement. Their results revealed that although the CS of concrete could be significantly reduced in the presence of higher dosages, it could meet the structural requirements for concrete while notably decreasing the OPC demand and thereby the carbon footprint of concrete [7].

Regarding these benefits, Gupta and Kashani [28] investigated the influence of biochar from unwashed peanut shells as cement replacement to improve the early-age strength of mortar containing 20% fly ash. Their data showed that adding biochar by 3% improved the 7-day strength of the specimens by 19%. According to these promising results, Mishra et al. [27] evaluated the effect of substituting cement with biochar in mortar specimens containing 10, 25, and 40% fly ash. Their data revealed that replacing 5% of cement with biochar could minimize the adverse impact of fly ash addition on the CS of specimens. Praneeth et al. [32] also studied the effect of replacing cement with biochar from corn stover in mortar containing 20 to 50% fly ash, when biochar dosage ranged from 0 to 8%. The data showed that in all mixes, the 28-day CS of the specimens maximized at 4% replacement. Their findings also demonstrated that the effect of biochar in the CS enhancement was increased when the dosage of fly ash rose. The result of these studies promoted the use of biochar in the range of 0 to 5% in composites containing fly ash. Their outcomes also proposed that the effect of biochar on the strength improvement of the specimens maximized when the fly ash dosage changed from 40 to 50%. Regarding the environmental benefits of using fly ash and biochar together, Mishra et al. [47] reported that replacing cement with biochar by 1% in mortar containing 10% fly ash could increase carbon dioxide uptake by 92%. Accordingly, the contribution of fly ash and biochar in the mix could be an efficient step toward carbon-negative concrete.

Recent studies discovered that the use of recycled gypsum (RG) powder significantly improved the CS of cementitious composites containing fly ash and/or slag [7,10,48–51]. RG activated the fly ash and slag in the composites by producing SO_4^{2-} alkaline environment in the mixture [52]. The addition of RG also modified the microstructure of the hardened concrete by forming a denser ettringite structure (AFt) [10,53]. Accordingly, using RG as SCM increased the 90-day CS of concrete containing fly ash/slag, reduced the OPC demand, and provided an eco-friendly waste management practice for this hazardous waste. Currently, the majority of gypsum waste is disposed of in landfills, emitting H_2S gas and producing contaminated leachate in the landfills, thereby threatening the nearby ecosystem [10].

In the last decades, the use of gypsum drywall in the construction industry has rapidly boosted due to its perfect noise and heat insulation. This spike in supply increased drywall waste such that it accounts for almost 9% of the annual construction and demolition waste in Canada [54]. This significant contribution raises public concerns about the environmental impacts of disposing of the waste and accentuates the importance of developing a sustainable waste management practice for gypsum drywall waste. Given this and the close-recycling-loop of the gypsum drywall [55], few researchers [7,10,48] studied the influence of fine recycled gypsum drywall (RGD) as SCM in cementitious composites. Hansen and Sadeghian [7] examined the effect of replacing OPC with fine RGD in concrete. Their results showed that replacing OPC with RGD by 5 and 10% with respect to the weight of binders in concrete containing 25 and 50% fly ash enhanced the mechanical strength of the concrete by 14.4 and 7.4%, respectively. Another study [10] also supported the potential of RGD as a cement replacement when 10% of cement was replaced by fine RGD in a mix containing 50% fly ash. These results emphasized the potential of reusing fine RGD as SCM in cementitious composites. Nonetheless, their data was derived from using fine RGD powder that passed sieve #50 and had compatible particle size distribution with OPC. Despite the advantages of reusing fine RGD as SCM, this approach returns almost 70% of the recycled gypsum drywall to landfills [7], which can be considered one of the drawbacks of these studies. Therefore, further research on the effect of reusing whole RGD products on the concrete characteristics is required to minimize the return of coarse RGD products to landfills. Given the above-mentioned and the data presented in the studies on the effect of biochar on cementitious composites containing fly ash, it could be suggested that mix containing 50% fly ash and 10% RGD is a nominated candidate for evaluating the effect of biochar on the properties of the sustainable concrete.

Accordingly, this study examines the effect of biochar and whole RGD on the properties of conventional concrete and sustainable HVFC. The results of this study address three research gaps, including i) the influence of biochar dosage on the compressive behavior (most importantly the ductility) and microstructure of the mentioned concrete, ii) the long-term effect of biochar on the compressive behavior of the concrete, i.e., examining the role of the retained water and its removal in load bearing capacity of concrete, and iii) the potential of using whole RGD in HVFC rather than the fine RGD powder. More importantly, this study explains the rule of biochar in concrete based on the available data. Accordingly, the effects of biochar dosage and using whole RGD on CS, porosity, stress-strain behavior, and microstructure of conventional concrete and HVFC at 28 and 90 days were examined by testing and analyzing the properties of 60 standard concrete cylinders. The effect of biochar on the carbon footprint of conventional and sustainable concrete was also evaluated based on the carbon dioxide equivalent of concrete components at the material production stage.

2. Methodology

2.1. Materials

In this study, general use (GU) ordinary Portland cement (Lafarge, ON, Canada), class F fly ash (Ocean Contractors, Halifax, NS, Canada), biochar (RDA Atlantic Inc., Halifax, NS, Canada), and RGD (USA Gypsum, Denver, PA, USA) were used as binders. Beside binders, river sand, crushed coarse aggregates (Casey Metro, Halifax, Canada), PLASTOL 341 superplasticizer (EUCLID Chemical, Cleveland, OH, USA), and tap water were utilized to manufacture specimens. The biochar was produced by the pyrolysis of waste wood chips at the temperature of 400℃ for 210 seconds. The water absorption of the biochar was 80.64% (wt%) based on the tea bag test. This water includes both retained water in the pores and the actual water absorbed by the biochar shell. Figure 1 shows the results of size distribution analysis by both sieve analysis and dry laser diffraction measurement techniques. Tables 1 and 2 depict the characteristics of aggregates and the chemical compositions of biochar, respectively. Table 3 also demonstrates the major oxides in the fly ash and GU cement measured by the lithium-tetraborate fusion technique.

Table 1. Characteristics of aggregates

Aggregate type	SSD water absorption (%)	Dry density (kg/m^3)
Coarse aggregates*	0.49	2646.76
Fine aggregates**	1.58	2524.42

[1] Saturated-surface-dry
* Measured based on ASTM C127-15 [56]
** Measured based on ASTM C128-22 [57]

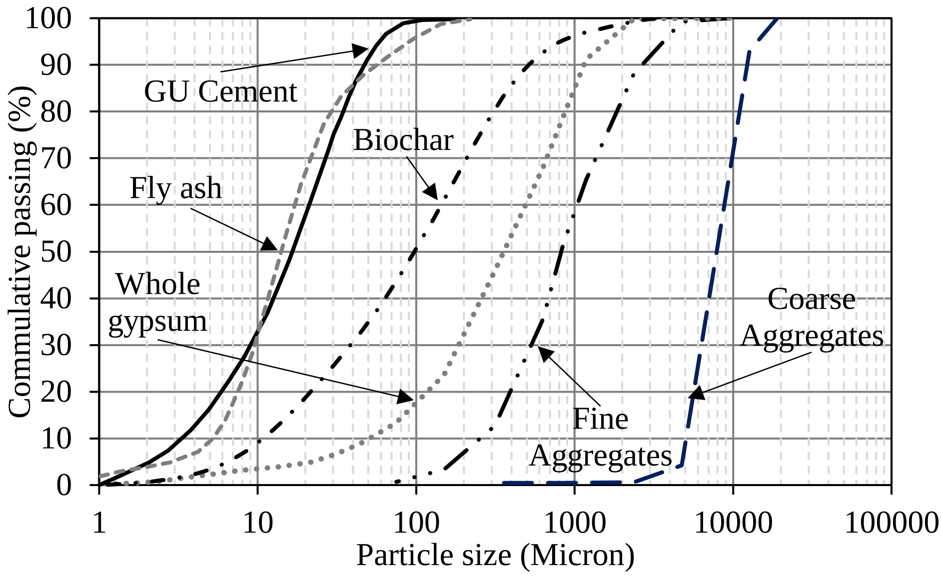

Figure 1. Particle size distributions of mix constituents (Note: the grading curves of fly ash and GU cement retrieved from Hansen and Sadeghian [7]**. Biochar, GU cement, and fly ash particle size were measured by volume).**

Table 2. Chemical elements 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

Table 3. Major oxides in fly ash and OPC

Oxides (wt%)	Al_2O_3	CaO	Fe_2O_3	MgO	Na_2O	SiO_2
Fly ash	20.87	1.44	6.55	1.84	1.06	59.39
GU cement	4.8	63.4	2.9	2.3	0.2	20

2.2. Test Matrix

Tables 4 and 5 depict the reference mix design and the OPC replacement proportions with each SCM in the mixtures, respectively. The amount of superplasticizer was adjusted based on the workability of the fresh concrete and the recommendations provided by the manufacturer company.

Table 4. Reference mixture per 1 m^3

Concrete component	Fine aggregate	Coarse aggregate	GU cement	Water
Weight (kg/m^3)	979.48	1197.45	490.5	236.4

Table 5. Test matrix

Mix Number	Mix Id	Binder replacement (wt%)			Number of specimens
		Biochar	Fly ash	RGD	
1	B0-F0-G0	0	0	0	6
2	B2.5-F0-G0	2.5	0	0	6
3	B5-F0-G0	5	0	0	6
4	B6.5-F0-G0	6.5	0	0	6
5	B0-F50-G0	0	50	0	6
6	B0-F50-G10	0	50	10	6
7	B2.5-F50-G10	2.5	50	10	6
8	B5-F50-G10	5	50	10	6
9	B6.5-F50-G10	6.5	50	10	6
10	B8-F50-G10	8	50	10	6
				Total	60

2.3. Specimen Preparation

This study followed the ASTM C192/C192M-18 [58] for specimen preparation. First, the aggregates were mixed in a mixer till the fine and coarse aggregates were uniformly combined. Next, binder(s) was(were) added to the mixer, and the ingredients were blended for 3 minutes. After that, the combination of water and superplasticizer was added to the mix in three steps. After achieving a homogenous mix, the concrete was poured into cylindrical molds with a diameter of 10 cm and a height of 20 cm according to the ASTM C192/C192M-18 [58]. The molded specimens were then sealed and held in the laboratory condition for 24 hours. Following the mold removal after 24 hours, the specimens were stored in the moist curing room till their testing ages. Figure 2 depicts the preparation process.

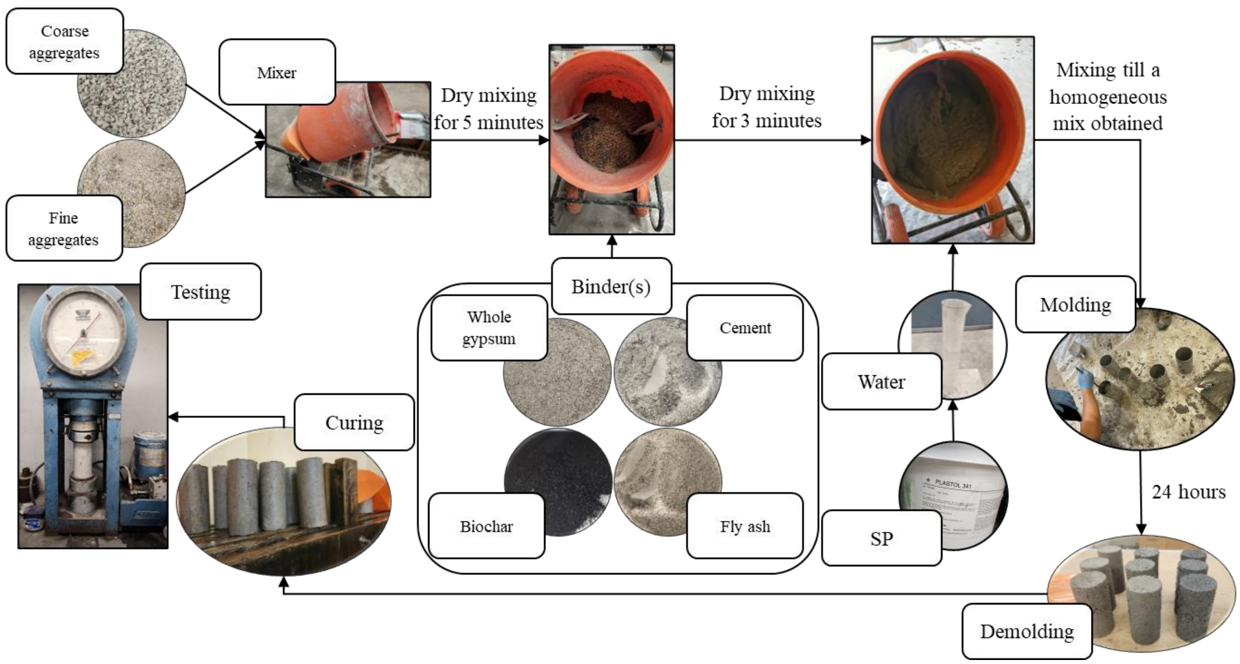

Figure 2. The preparation process

2.4. Testing

In this study, the CS, porosity, stress-strain behavior, and microstructure of the specimens were examined. The following subsections describe test setups and procedures.

2.4.1. Compressive Strength (CS)

This study measures the CS of the specimens after 28 and 90 days. The 28- and 90-day CSs of the specimens were measured by a Forney compression machine and a 2 MN Instron test frame with a loading rate of 0.5 mm/min in accordance with ASTM C39/39M-23 [59], respectively. To minimize the effect of retained water in the specimens containing biochar, the tests were conducted a day after taking specimens out of the curing room at 28-day measurements. However, since inside the tested specimens were still moist in the 28-day tests, the 90-day cured specimens were kept outside of the curing room, in the lab condition, for a longer period, until the weight of specimens stabilized in measurements of two consecutive days. This was to ensure that the internal moisture of the specimens was adjusted based on normal conditions, minimizing the potential effect of the biochar-retained water on artificially strengthening the specimens. Oven drying was avoided prior to testing since the rapid evaporation of the retained water could form microcracks and impact the strength of the specimens. The average of three compression tests was reported for each mix design at 28 and 90 days.

2.4.2. Stress-Strain Behavior

In this study, the stress-strain behavior of 90-day specimens was tested under compression load with a rate of 0.5 mm/min using a 2 MN Instron test frame in accordance with ASTM C469/C469M-22 [60]. In the test, a series of 4 linear potentiometers (LPs) was used to measure the axial and circumferential displacements at the middle height of specimens. The LPs' position was fixed to the specimens using a metal bracing system that connected to the specimens by six bolts, i.e., three bolts per ring. Figure 3 shows the test setup that was used to measure the displacements by LPs. The displacement of each LP and the corresponding load were recorded during the test and used to determine the stress-strain diagrams of the specimens. The modulus of elasticity (E) of the specimens was also calculated in accordance with ASTM C469/C469M-22 [60].

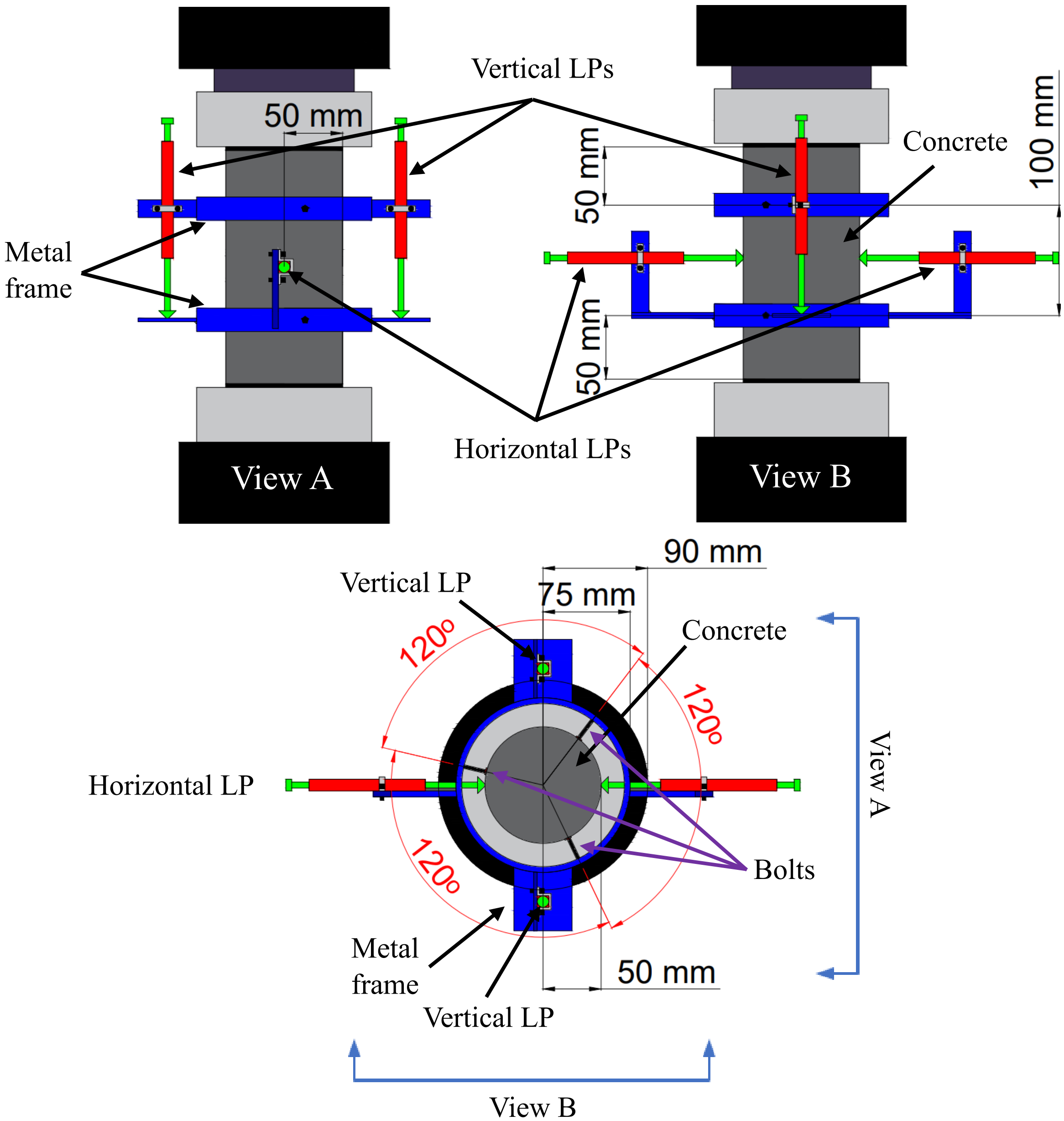

Figure 3. Test setup and instrumentation

2.4.3. Ultrasonic Pulse Velocity (UPV)

Ultrasonic pulse velocity (UPV) is a common non-destructive technique to evaluate the porosity of concrete specimens. Earlier studies also showed that the UPV could be used to estimate the mechanical strength and elastic modulus of concrete [10,61]. Accordingly, the test was used to determine the effect of biochar dosage on the porosity of specimens. The test was conducted based on ASTM C597-22 [62] by transmitting the ultrasonic wave through the specimens using 50kHz transducers. The transducers contacted on the opposite sides of the specimens, coated by a thin layer of honey as a couplant. The wave transmission length of the specimens was also accurately measured using a digital caliper. The UPV was then computed by dividing the traveling length by the traveling time.

2.4.4. Scan Electron Microscopy (SEM)

This technique was used to assess the influence of the replacement rate of OPC with biochar and whole RGD on the microstructure of the specimens. The test was conducted on the 90-day specimens following the completion of the compression test. After the compression test, the specimens were dried in an oven for 24 hours to halt the hydration. The SEM samples were then taken from the core of the dried specimens. Afterward, the samples were coated with a 20 nm layer of gold-palladium powder via the Leica EM ACE200 machine and scanned using the HITACHI S-4700 scanning electron microscope.

3. Results and Discussions

3.1. Unit Weight

3.1.1. Conventional Concrete

Figure 4 depicts the effect of OPC substitution with biochar on the unit weight of conventional and sustainable concrete. The data showed that increasing the biochar dosage to 2.5% reduced the 28-day unit weight of the specimens by 2.7%. However, the unit weight of the specimens stabilized after the 2.5% substitution dosage such that increasing the dosage to 6.5% had a tiny effect on the 28-day unit weight of the specimens. Nonetheless, the 90-day unit weight of the specimens was continuously reduced by up to 3.81% when the replacement dosage rose to 5%. Boosting the biochar dosage from 5 to 6.5%, however, had a negligible effect on the parameter at 90 days. The reduction in the unit weight of specimens is aligned with the poor structure and low density of the biochar and showed the potential for a rise in the porosity of the specimens by increasing the biochar percentage.

Figure 5 displayed that the slope of unit weight reduction of the 90-day specimens significantly changed after 5% cement replacement with biochar. The change in the unit weight reduction gradient, which resulted in stabilizing the unit weight, might be explained by the biochar-retained water. In this hypothesis, since biochar has the potential to retain a significant amount of water from the fresh mix, the increase in the biochar dosage decreased the amount of free water in the fresh mix and raised the total amount of water that was reserved by biochar. Although hydration reactions used a significant portion of this water at low biochar dosage. Exceeding the biochar dosage increased the reserved water, leading to an increase in the ratio of retained water to the required water for hydration reactions. This increase could ultimately raise the amount of retained water in biochar, neutralizing a portion of the unit weight reduction due to biochar addition at 28 days. In the case of 90-day specimens, however, the extra drying period reduced the amount of retained water. Accordingly, only a small portion of water remained in the pores due to the size of biochar pores and the capillary effect. Therefore, this extra water removal could provide a clearer perspective of the effect of biochar on the unit weight of concrete. This finding may encourage researchers to investigate the exact theory behind this change in future studies.

The results also demonstrated that the specimens had lower unit weight at 90 days compared to 28 days. This might be because of the additional period taken for water removal of the specimens prior to 90-day measurements. Besides neutralizing the possible effect of the biochar-retained water on the CS of the specimens, it could show the amount of retained water removed from the specimens, particularly biochar, in the additional period. The action reduced the unit weight of the specimens by 0.46% compared to 28-day measurements in the B0-F0-G0 specimens. Increasing the substitution dosage to 2.5% lessened the unit weight reduction, such that the 90-day unit weight of the specimens was boosted by 0.37% compared to 28-day specimens. However, a further increase in the biochar dosage promoted the 90-to-28-day unit weight reduction rate of specimens. This increase in the unit weight loss due to the rise in the biochar dosage could verify the presence of biochar-retained water in 90-day specimens. In the case of testing specimens based on the ASTM standard [59] applying compression load to the biochar in the presence of the retained water could increase the hydraulic cavity pressure inside the biochar pores because of their size, which is in nano- to micro-meters [23]. This could increase the load-bearing capacity of the biochar and ultimately enhance the CS of the specimens. This also highlighted the possible participation of biochar-retained water in the 28-day measurements. Besides, the data identified 2.5% replacement as an optimum dosage for the unit weight enhancement between 28 and 90 days.

3.1.2. Sustainable Concrete

The data revealed that replacing 50% of cement with fly ash reduced the unit weight of the specimens by 3.31 and 3.22% at 28- and 90-day measurements, respectively. This might be due to the lower density of fly ash compared to ordinary Portland cement. The results also presented that adding 10% RGD to the mixer further decreased the unit weight of the specimens by 1.45 and 1.12% relative to B0-F50-G0 at 28 and 90 days, respectively.

Figure 4 also illustrates that escalating the substitution percentage to 8% lowered the unit weight of the sustainable concrete by 4.8 and 3.14% in 28- and 90-day tests, respectively. However, a notable increase in the unit weight of 28-day specimens was observed at 6.5% substitution dosage compared to the B5-F50-G10, limiting the unit weight reduction to 1.76 and 2.33% at the B6.5-F50-G10 with respect to the B0-F50-G10 at the 28 and 90 days, respectively. The reduction in unit weight of specimens is consistent with the porosity of the biochar compared to other concrete ingredients. The difference in the gradient of unit weight reduction between the conventional and sustainable concrete could also emphasize the effect of binders' particle size distributions and their chemical reactivity on the performance of biochar in concrete.

The data also showed that replacing cement with 50% fly ash and 10% RGD lowered the unit weight reduction of the specimens between 28 and 90 days from 0.46% to 0.18%. This alteration might be attributed to two key factors: 1) reduced water removal from the specimens and 2) improved microstructural enhancement in the B0-F50-G10 specimens between 28 and 90 days. Less water removal could show potential for a lower void content and penetration, which are the well-known effects of adding fly ash to concrete because of its filler effect. The microstructural enhancement could also be attributed to the pozzolanic reactions of fly ash and RGD, which intensified after 28 days. This is because, in contrast to conventional concrete, in which the majority of the cementitious reactions took place in the first 28 days of curing, the pozzolanic reactions primarily initiated after 28 days. Increasing the pozzolanic reactions due to the rise in the fly ash dosage and the delayed RGD reactions boosted the water demand of the composite after 28 days. Accordingly, more water was used by the reactions between 28 and 90 days in the mix containing fly ash and RGD, which could potentially reduce the amount of free water in the pores, lowering the removed water from the 90-day specimens. The expanded reaction of RGD could also lower the size of pores between 28 and 90 days, lessening the porosity and thereby the water retention capacity of concrete.

Additionally, because of this higher water demand after 28 days, the biochar-retained water could enhance the delayed reactions, i.e., pozzolanic and RGD reactions, by serving as a water reservoir. Accordingly, increasing biochar dosage gained the microstructural improvement of the system, raising the 90-to-28-day unit weight ratio. Utilizing biochar-retained water in the delayed reactions also deducted the retained water, lessening the amount of removed water during the extra period applied after 90 days. Increasing the 90-to-28-day unit weight of specimens containing 2.5 and 5% biochar by 0.18 and 0.95%, respectively, was evidence of the effect of biochar on promoting the reactions. Increasing the biochar dosage to 8% further magnified the unit weight improvement by 1.56%. Theoretically, biochar could also offer spaces for the expanded reaction products of RGD, which not only reduced the impact of these reactions on concrete microcracking but also could improve the mechanical properties of biochar.

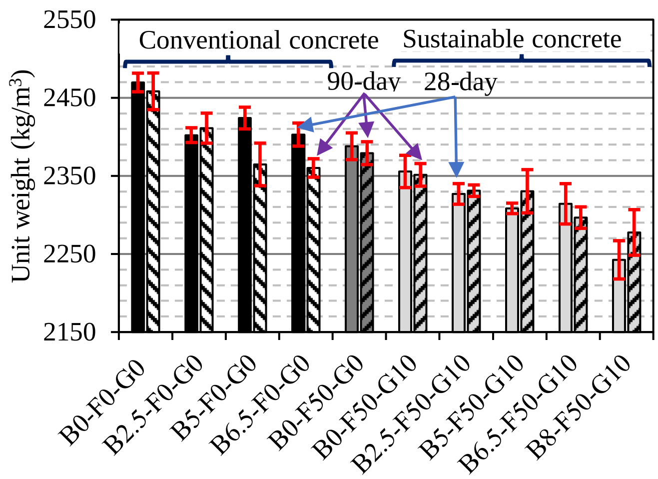

Figure 4. Unit weight of the specimens

Figure 5 illustrates the influence of biochar percentage on the unit weight of the specimens. According to the results, when a significant share of biochar-retained water was removed from the specimens, the reduction trend changed to an almost linear function. Findings also demonstrated that the difference between the 28- and 90-day unit weights of the specimens increased by boosting the biochar dosage in conventional concrete specimens. This might be because almost 80% of the hydration reactions in the conventional concrete occurred during the first 7 days of curing [63], reducing the demand for the biochar-retained water for the remaining reactions while the moist curing increased the available water for the biochar's water retention. Accordingly, the extra drying period of the specimens reduced a substantial portion of the additional retained water, shifting the pattern to almost a linear function. Equation 1 presents formulas that could be used to estimate the effect of biochar dosage variation on the unit weight of conventional and sustainable concrete by the correlation coefficient (R^2) of 96.62 and 87.66%, respectively.

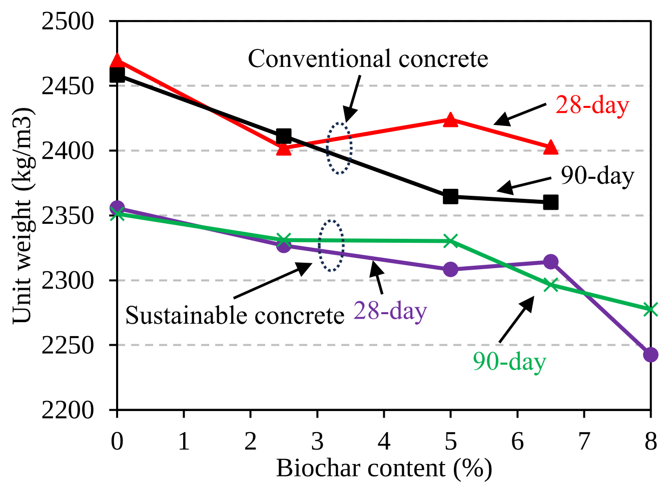

Figure 5. Influence of biochar content on the unit weight

$$UW(BC) = \begin{cases} -15.822BC + 2453.9 & Conventional\ concrete \quad 0 \le BC \le 6.5 \\ -8.712BC + 2355.7 & Sustainable\ concrete \quad 0 \le BC \le 8 \end{cases} \quad (1)$$

where UW and BC represent the unit weight (in kg/m^3) and biochar dosage (in percentage) of the 90-day specimens after the water removal period, respectively.

3.2. Compressive Strength (CS)

3.2.1. Conventional Concrete

Figure 6 demonstrates that replacing OPC with biochar by 2.5% in conventional concrete enhanced the 28-day CS of the specimens by 2.3%. However, raising the substitution to 5% reduced the 28-day CS of the specimens by 11% relative to the B0-F0-G0 specimens. The existence of an optimum in the biochar dosage could be explained by four mechanisms. First, adding biochar to the fresh mix reduced the water-to-cement ratio of the mix by retaining free water in the biochar-effective zone, increasing the compactness of the fresh mix. This retained water was then gradually released over time and used for the hydration reactions. However, the over-increase in the biochar dosage could raise water retention such that the cement had insufficient water for initial reactions, lowering the early-age strengthening [33,64]. Second, the biochar potential to provide additional and available water as well as nucleation sites intensified the early-age and internal hydration, boosting the compressive strength of concrete [10,24]. Nonetheless, excessive biochar dosage could trigger the dilution effect, reducing the strengthening reactions. Third, the filler effect of biochar could enhance the mechanical strength of the concrete since the biochar could fill the voids and strengthen the cement paste and interfacial transition zone [65]. The specific shape of biochar particles with angular edges could improve the interlock in the cement paste [66]. However, adding an excess amount of biochar to the mix adversely impacted the mechanical strength of the composite due to the loosening effect. In this scenario, the increase in the biochar particles raised the distance between cement particles and boosted the porosity of the cement paste, thereby lowering the strength. Accordingly, the over-increase in biochar raised the number of pores and weakened the concrete [14]. Fourth, while the biochar addition to the composite increased the porosity of the system, it was able to modify the size of concrete voids from large pores to those of the nano-scale [23]. This change in the size of pores could strengthen the concrete because large pores had more impact on the mechanical strength of cementitious composites than small voids [67]. Over-escalating the substitution dosage, however, might result in the accumulation of biochar particles, forming large gaps in the cement paste and thereby reducing the strength of the composite. It is anticipated that the optimum dosage of the biochar is notably affected by pyrolysis temperature and duration. The increase in either of the two factors boosts the porosity of biochar and lessens the thickness of the biochar shells, lowering the strength and stiffness of the biochar particles. Accordingly, it could reduce the optimum substitution rate. This theory could also explain the discrepancies in the proposed optimum substitution dosages in the literature [17].

Lower OPC content in the mix with higher substitution dosage could also be an essential element for the strength reduction. OPC-water reactions were the primary source of hydration reactions in conventional concrete, and biochar was an inert material that catalyzed the reactions by supplying retained water and providing nucleation sites. Consequently, lower OPC content could limit biochar-induced hydration intensification, lessening the overall hydration reactions and thereby lowering the strength of concrete.

Increasing the biochar dosage to 6.5% raised the 28-day CS of the specimens by 9.3% compared to B5-F0-G0, such that the specimens containing 6.5% biochar had only a 2.87% lower strength than those of B0-F0-G0. This pattern was also repeated in the 90-day specimens. A similar behavior was also reported by Qin et al. [35] where the specimens containing 3.2% biochar obtained lower strength than those containing 0.65 and 6.5% at both 28 and 90 days. Given the specific edge shape of biochar particles, the aggregation and agglomeration of biochar particles could be a potential factor for the mechanical properties improvement of concrete in a 6.5% replacement dosage. The strength increase could also accentuate the importance of evaluating the effect of higher dosage on the mechanical properties of the concrete.

The data revealed a change in the strength variation at 90-day measurements. By increasing the substitution rate by up to 5%, the specimens' 90-day strength gradually decreased by 15.59%. However, the increase in the biochar dosage from 5 to 6.5% raised the 90-day CS of the specimens by 5.63%. The data also displayed that the increase in the biochar dosage boosted the gradient of strength reduction such that increasing the dosage from 2.5 to 5% had a greater impact on the CS of the specimens relative to that from 0 to 2.5% (see Figure 7). This pattern shift also highlighted the importance of examining the long-term performance of biochar on the properties of cementitious composites. Three primary factors could potentially contribute to this change in the influence of biochar dosage: i) a decrease in the OPC content, ii) biochar deterioration, and iii) a potential reduction in biochar-retained water.

It is well-known that biochar boosted the early-age hydration of cementitious composites [10]. Therefore, the specimens containing biochar as cement replacement in small dosages could achieve higher reactions and strength at an early age [22]. However, the OPC particles in the reference specimens gradually reacted over time in the presence of sufficient water, i.e., in the moist curing room. Consequently, when the OPC reactions were completed in the long run, the less OPC content in the concrete containing biochar lowered the strength of the concrete. Given this theory, it could be concluded that the interaction between the biochar properties and the total hydration reactions, which could be a function of the hydration acceleration rate as well as cement content and reactivity, could be used to determine the optimum biochar dosage at different ages. This theory primarily focuses on the biochar's potential to provide water and nucleation sites as well as its inertness in the composite. However, the filler effect of the biochar might result in a CS improvement in the low percentages (<2.5%). Using biochar containing a high dosage of silica and alumina could also change the performance of biochar in the mix due to the potential reactions between the biochar's elements and cement paste.

Biochar deterioration in the cement paste exposure might be another factor contributing to this pattern change. According to Xu et al. [23], exposure of biochar to the alkaline environment of cement paste could deteriorate the biochar by reducing its hardness and modulus of elasticity, as well as increasing the size of biochar pores. Their findings also showed that an increase in the exposure time could raise the adverse impacts of the alkaline environment on the biochar properties. Therefore, it is reasonable to assume that longer exposure to cement paste deteriorated the properties of biochar particles in the 90-day specimens, reducing the strength of biochar particles. Accordingly, although the cement reactions continued improving the microstructure of the composite, biochar deterioration negatively affected the CS of the composite over time. This factor could scale down the strength improvement of the concrete containing biochar compared to that of reference specimens, which resulted in a lower strength in the specimens containing biochar at 90 days. This reduction in the strength enhancement could also shift the optimum biochar dosage of 90-day specimens to a lower dosage than that of 28-day.

The partial removal of the biochar-retained water might also participate in this shift. The 28-day tests were conducted a day after taking specimens from the curing room according to the ASTM standard [59]. However, because of observing wet cement paste inside the broken specimens, additional time was considered for removing a portion of biochar-retained water before the 90-day tests. This additional drying time could lessen the retained water in biochar pores as the unit weight of specimens was decreased in the specimens containing biochar between 28- and 90-day measurements (see Figure 4). Lowering the retained water could shrink the load-bearing capability of the biochar-retained water and increased the share of biochar in the load resistance. This hypothesis was supported by the fact that the tested specimens had less internal moisture at 90-day measurements than at 28-day ones. This could efficiently mitigate the artificial strengthening effect of the hydraulic cavity pressure of biochar-retained water at 90-day measurements and strengthen the adverse effects of increasing biochar dosage on the porosity and CS of the specimens. It could also be suggested that the effect of the retained water on the strength reduction depends on the size, quantity, and connection of the biochar pores since these factors significantly influence the amount of biochar-retained water.

Figure 7 illustrates that increasing the biochar dosage from 0 to 6.5% reduced the strength improvement of the specimens between 28- and 90-day measurements from 35.75% to 24.59%, respectively. The three factors mentioned above could be responsible for lowering the strength improvement rate when the biochar percentage rose. Besides, the effect of biochar on the hydration acceleration might account for a portion of the decrease in the strength improvement. Biochar addition to the mix improved the early-age hydration and promoted the early-age strength of the specimens. The higher the biochar dosage, the greater the early-age hydration. Given the inertness of the biochar particles and their chemical stability, the ultimate level of hydration products was determined by the amount of cement in the mix. Therefore, replacing OPC with biochar led to a reduction in the ultimate level of hydration products. This would cause lower ultimate strength of the specimens when the biochar dosage increased. These two elements, namely the reduction in the ultimate amount of hydration products and the early-age hydration acceleration in the presence of biochar, could also potentially lessen the strength enhancement rate.

3.2.2. Sustainable Concrete

Figure 6 presents that replacing OPC with fly ash by 50% reduced the 28- and 90-day strength of the specimens by 56.87 and 50.70%, respectively. The data also showed that adding the whole RGD product by 10% to the concrete containing fly ash reduced the 28-day CS of the specimens by 18.66%. However, the data demonstrated that the addition resulted in 20.11% higher strength at 90-day measurements. Findings also revealed that adding RGD to the mix boosted the strength improvement in the 28-to-90-day interval from 51.63% to 123.9%. The delayed reactions of RGD and its potential to form an alkaline-ionic environment could be key factors that promote the strength improvement. Kocaba [68] reported that increasing the gypsum percentage in the cement paste postponed the peak in the hydration heat diagram. The study also identified a second peak in the hydration heat evaluation, corresponding to the reactions between alumina from the pozzolans and sulfur from gypsum. Zhang et al. [69] also revealed that gypsum reactions with water in the mix produced an SO_4^{2-} environment, which not only accelerated the hydration of C3S but also increased the formation of ettringite. Their findings also showed that the reaction of gypsum in the cement environment was mainly controlled by the Al^+ and SO_4^{2-}. The released Ca^+ ions could also boost the formation of C-S-H gels in the interaction with the silica elements of the pozzolans, enhancing the microstructure of the composite. RGD-water reactions could result in a similar pattern. However, because of the lower activity of the RGD compared to gypsum, the RGD reactions might be delayed relative to the natural gypsum in the lack of preheating. Its potential to activate fly ash by offering the ionic environment and to supply Al^+ ions from the fly ash to form an ettringite structure could explain the boost in the strength improvement. The second peak of the hydration heat could also promote the dissolution of fly ash, boosting its reactions with cement and RGD.

These results highlighted the potential of using whole RGD in the concrete. The approach reduced the usage of OPC in the concrete containing 50% fly ash by 20% while improving the 90-day CS. The use of whole RGD instead of fine RGD also prevents the return of almost 70% of RGD (RGD with a particle size greater than 0.3 mm) into landfills, eliminating the adverse environmental impacts of gypsum drywall and its leachate on the nearby ecosystem. Nonetheless, the data also accentuated a notable drawback of the concrete, low early-age strength, compared to conventional concrete. Solving this issue could establish an effective eco-friendly waste management technique for the hazardous waste and reduce the OPC demand and carbon footprint of the concrete.

The data showed that replacing cement with biochar generally reduced the 28-day CS of the specimens. Increasing the biochar dosage to 2.5 and 5% decreased the 28-day CS of the specimens by 3.97 and 22.62%. However, increasing the biochar dosage from 5% to 6.5% rebounded the CS of specimens by 16.92%. Further increase in the biochar dosage to 8%, however, scaled down the strength by 10.63%. The general trend could be explained by the effect of replacing reactive binders with an inert porous material, i.e., biochar, and the interaction between the fly ash, RGD, and biochar, which might adversely affect their filler effect. Increasing fillers in the composite might also boost their dilution effect, impacting the strength of the composite. The specimens containing 5% biochar demonstrated a minimum 28-day strength, suggesting that the significant drop in the CS upon increasing the biochar dosage from 2.5% to 5% resulted from the maximum interaction between the fillers, triggered by their particle size distributions. Increasing the dosage to 6.5%, however, reduced the impact of these interactions. These results highlighted the importance of further research on the performance of biochar in concrete at different dosages to determine the reasons behind this variation in the effect of biochar dosage on the CS of concrete.

Figure 6 also presents that replacing the cement with biochar by up to 2.5% in the sustainable concrete had a negligible impact on the 90-day CS of the concrete. However, boosting the substitution dosage to 5% could maximize the strength reduction rate, decreasing the strength of specimens by 22.79%. Expanding the biochar dosage from 5% had less impact on the strength of the specimens such that the specimens containing 6.5 and 8% biochar had approximately the same strength with the 26.89% strength reduction compared to the B0-F50-G10 (See Figure 7). Similar explanations could be applied to describe the effect of biochar on the 90-day CS of the composite. In addition to those factors, substituting OPC with biochar reduced the initially reactive binder in the sustainable concrete, responsible for starting hydration reactions and raising the temperature and alkalinity of fresh concrete. This reduction in the hydration heat and alkalinity of the cement paste might negatively affect the reactivity of fly ash and RGD, reducing or delaying the pozzolanic and RGD reactions. The repeated influence of 5% replacement in all the measurements also emphasized the importance of further study on this dosage. Accordingly, given the potential benefits of biochar in carbon sequestration and the low impact of biochar on the 90-day CS of the specimens at a 2.5% substitution rate, this study proposed replacing cement with biochar by 2.5% in the sustainable concrete.

Figure 7 depicts that increasing the biochar dosage to 2.5% maximized the strength enhancement within the 28-to-90-day interval, leading to a 2.17% higher 90-to-28-day CS ratio. This improvement reduced the effect of biochar on the CS of the specimens such that the strength reduction of specimens containing 2.5% biochar decreased from 3.97% at 28 days to 1.98% at 90 days relative to B0-F50-G10. However, raising the dosage to 5% reduced the 90-to-28-day CS ratio such that the strength improvement was equal to that of the B0-F50-G10. The data also showed that exceeding the dosage from 5% to 6.5% amplified the adverse impact of biochar dosage on the strength improvement of the specimens by decreasing the strength improvement percentage from 123.41 to 80.93%. Nonetheless, increasing the dosage to 8% boosted the strength percentage to 102.94%. Biochar's potential to offer accessible water for the fly ash and RGD reactions could boost the delayed reactions, enhancing the strength improvement of the system. Nevertheless, since the OPC was replaced with biochar, the escalation in the biochar dosage reduced the OPC content and lessened the hydration heat and alkalinity of the system. Although the lower alkalinity of the environment reduced the biochar’s deterioration, it decreased the reactivity of RGD and fly ash as their performance and reactivity were enhanced by an increase in the hydration heat and alkalinity of the system. The increase in the biochar dosage also boosted the potential of the dilution effect, which could adversely affect the strength of the concrete. Based on the aforementioned, it could be assumed that the balance points between these effects resulted in the behavior. At first, the potential of biochar to boost reactions could overcome other factors while the biochar dosage rose to 2.5%. Between 2.5 and 6.5%, the influence of the dilution effect and less OPC content on the system outweighed the beneficial impact of the biochar. In the higher dosages, the strong effect of lowering cement in reducing the alkalinity of the system and thereby the biochar deterioration might dominate over the impacts dilution effect and less OPC content, promoting the strength improvement rate.

The magnified influence of biochar dosage in lessening the CS of the sustainable concrete at 90 days compared to 28 days could also raise concerns about the significant impact of the applied drying time at 90 days. This is because fly ash and RGD needed less water at early ages compared to OPC. This lower water demand increased the potential free water in the fresh mix, which could be retained by biochar particles. The lower cement percentage also reduced the water demand during the curing time. These factors could increase the amount of retained water in biochar pores. Without additional drying time, this retained water could strengthen the biochar particles by resisting the applied load through hydraulic cavity pressure, leading to a lower strength reduction rate in the specimens containing a higher biochar dosage. However, employing additional drying time could reduce a portion of retained water and thereby the hydraulic cavity pressure, which might reduce the system's strength. Accordingly, further studies are required to examine this hypothesis as it could shed light on the effect of biochar on the long-term properties of cementitious composites in practical applications.

Results also indicated that increasing the biochar dosage from 2.5% had a stronger impact on the CS of the sustainable concrete compared to that of the conventional concrete. For example, adding 6.5% biochar to sustainable concrete reduced its strength by 9.52 and 26.89% at 28 and 90 days, respectively. However, the conventional concrete containing the same dosage had 2.85 and 10.83% lower strength compared to the reference specimens at the testing ages, respectively. This significant impact might result from the dilution effect led by the overuse of fillers and their interactions. This intensification also highlighted the importance of the particle size distributions of the binders and biochar in biochar's effect on concrete properties. The variation in the effect of biochar on the strength improvement of specimens between 28 and 90 days could also attract attention. Using biochar in conventional concrete reduced the strength improvement of the specimens by 5 to 9%. However, increasing the biochar dosage to 2.5% enhanced the strength improvement rate of the sustainable concrete by 2.17%. This difference could be caused by the delayed reaction of fly ash and RGD in the composites and their demand for biochar-retained water. Accordingly, it could emphasize the importance of cementitious materials, particularly their level of reactivity and reaction time, in evaluating the performance of biochar in cementitious composites.

Equation 2 illustrates models for estimating the 90-day CS of conventional and sustainable concrete. The model proposed that the optimum biochar dosage for maximizing the strength in conventional concrete would be 0.705%.

$$CS(B) = \begin{cases} 0.1668B^3 - 1.4836B^2 + 1.8439B + 45 & 0 \leq B \leq 6.5 \ \textit{Conventional concrete} \\ 0.0766B^3 - 0.9104B^2 + 1.5062B + 26.683 & 0 \leq B \leq 8 \ \textit{Sustainable concrete} \end{cases} \quad (2)$$

where CS and B are the compressive strength (in MPa) and biochar dosage (in percentage with respect to the weight of binders).

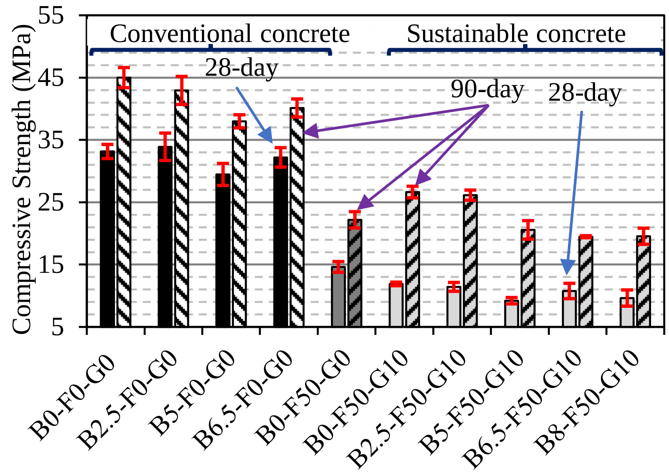

Figure 6. CS of the specimens

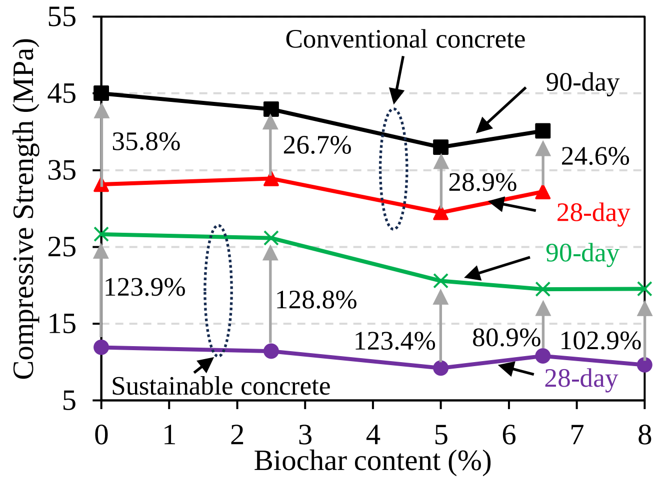

Figure 7. Effect of biochar content on the CS

3.3. Ultrasonic Pulse Velocity (UPV)

3.3.1. Conventional Concrete

Figure 8 presents that raising the biochar dosage to 6.5% gradually reduced the 28-day UPV of the conventional concrete by 7.79%. This reduction might arise from the low density and high porosity of the biochar particles compared to other concrete ingredients. Figure 9 depicts that the reduction rate of UPV was almost constant in the range of 0 to 6.5% biochar dosage. These results conflicted with the unit weight variations shown in Figure 5 and the CS pattern observed in Figure 6. The inconsistency between the UPV and unit weight results was vivid when the biochar dosage increased from 2.5%. Despite the reduction trend in UPV, escalating biochar dosage from 2.5% to 5% increased the unit weight of the specimens at 28 days. It could be assumed that a trade-off between biochar porosity, biochar-retained water, biochar agglomeration, and the effect of biochar on the composite compactness could explain this behavior.

Biochar is a porous material that can act as a filler and water reservoir in the cement paste. It could improve the compactness of the system by absorbing a significant amount of free water in the fresh concrete. This improvement in the compactness increased by raising the biochar dosage. Typically, this retained water was used by cementitious reactions, boosting the early-age hydration. However, curing the specimens in a highly humid environment, particularly in moist- and water-curing conditions, could result in a new round of water retention by biochar particles. This retained water could stored in the biochar because of the low water demand of the specimens at the stage and the size of the biochar pores, leading to the capillary effect. Accordingly, it could raise the weight of specimens, specifically in high dosages. Besides, the potential of biochar agglomeration in the specimens containing a high biochar dosage could boost the impact of biochar dosage on the unit weight of the specimens by forming large pores.

Therefore, it could be suggested that the unit weight reduction between 0 and 2.5% was an obvious effect of biochar porosity. The increase in compaction and the retained water due to raising the biochar dosage could cause the unit weight enhancement between 2.5 and 5% replacement. However, since the wave transition velocity is lower in the water than solid, the retained water could not compensate for the effect of the increased porosity on UPV results. Increasing the biochar dosage from 5 to 6.5% in the concrete, however, could result in agglomeration of biochar particles, which could lead to the alignment of the effect of biochar addition on the unit weight and UPV.

The inconsistency between the UPV and CS trends might arise from the different factors that affect UPV and CS. The UPV was primarily affected by the porosity of the system. However, the CS could be affected by two substantial factors: porosity and bonding. Typically, OPC alternatives improve the mechanical properties of the cementitious composites through pozzolanic reactions or reducing voids in the composite. However, the biochar had different mechanisms for improving the mechanical strength of the composite through bond enhancement. First, biochar reduces the large voids by converting them to small ones. Second, biochar improves the reactions by providing water for internal hydration. Third, biochar's special shape could improve the interlock in the cement paste and enhance the interfacial transition zone. Except for reaction enhancement, which could reduce the system's porosity, all the mechanisms were independent of the porosity. Besides, the retained water might also account for a portion of the conflict at 28 days.

The results also depicted that increasing the biochar dosage by up to 6.5% could generally reduce the 90-day UPV of the specimens by up to 4.64%. An abnormal increase in the average UPV of the specimens was also observed when the biochar dosage was raised from 2.5 to 5%. Nonetheless, examining the standard deviations, shown in Figure 8, revealed that the maximum UPV of the specimens containing 2.5% biochar was higher than that of 5%. Therefore, it could be suggested that either material uncertainties or measurement errors might cause the abnormal increase in the 90-day UPV. Taking this assumption, it could be stated that the UPV pattern matched the unit weight trend at 90-day measurements. However, it was incompatible with the CS trend, in which the strength increased by raising the biochar dosage from 5 to 6.5%. The previously mentioned factors could explain the conflict between the effect of biochar dosage on UPV and CS in the range of 5 to 6.5% substitution. Besides, the alignment between UPV and unit weight results as well as the shift in the 28-day CS trend when the biochar dosage changed between 0 to 5%

might emphasize the importance of biochar-retained water. This is because the parameter could be responsible for the changes based on the extra drying time applied to the 90-day specimens.

Figure 9 also shows that increasing the biochar dosage could boost the UPV improvement between 28 and 90 days. However, the analysis of the average values revealed an exception to the positive effect at 2.5% replacement; the high standard deviation suggests that material and measurement uncertainties could cause this lower enhancement. These data also revealed that the porosity of the specimens decreased during the time interval when the biochar dosage rose, which was inconsistent with the decreasing rate of the CS and unit weight improvement. Given the mentioned mechanisms, the dilution effect and biochar deterioration could be responsible for the negative gradient of the CS improvement trend. However, the conflict between the improvement rate of UPV and unit weight could highlight the rule of biochar-retained water, since it was the primary factor that changed during the 28 and 90-day measurements, considering the additional drying time. This is because the release of fulvic acid organics during biochar deterioration could form soluble calcium salts stored in the specimens [23]. The coincident reduction in the CS improvement rate with the decrease in the porosity also emphasized the potential effect of biochar-retained water removal on the CS improvement rate.

3.3.2. Sustainable Concrete

Figure 8 depicts that replacing OPC with fly ash by 50% reduced the UPV of the specimens by 9.91 and 3.70% at 28 and 90 days, respectively. The lower reactivity of fly ash compared to OPC and the dilution effect from overusing fly ash could cause this reduction. The data also showed that the mix containing 50% fly ash had a 6.89% higher UPV improvement between 28 and 90 days than the B0-F0-G0, potentially due to fly ash's pozzolanic reactions.

The results also showed that adding 10% RGD to the mix reduced the UPV of the specimens by 1.8 and 4.48% at 28 and 90 days, respectively. This could be attributed to the presence of paper fibers in the whole RGD, as well as the dissimilarity in the microstructure between the RGD-water reaction products and the OPC-fly ash reactions. These factors could also address the conflict over the effect of RGD on the UPV and CS. Given the wooden microstructure of the paper fibers, these fibers could increase the porosity of concrete by halting the hydration reactions in their ITZ and their shrinkage. However, the fibers might reinforce the cement matrix under loading, delaying the axial splitting failure mode and thereby increasing the CS of the specimens. RGD could also promote the formation of dense ettringite microstructures in the system, which were more porous than the uniform microstructures formed between fly ash and OPC. Despite their porosity, these dense ettringites could increase the specimen's load-bearing capacity of the concrete. The microstructural analysis section provided a detailed discussion of how RGD affected the specimens' microstructure.

Figure 8 also demonstrates that raising the biochar dosage in the sustainable concrete by up to 6.5% could gradually reduce the UPV of the specimens by up to 5.71% at 28 days. However, the 28-day UPV of the specimens was approximately constant when the biochar dosage changed from 6.5 to 8%. The data showed that this pattern shift might be caused by material uncertainty or measurement error. This is because the maximum value observed in the B6.5-F50-G10 was higher than that of the B8-F50-G10. The added porosity by the biochar pores could be attributed to the UPV decrease. The stabilization in the UPV could also encourage researchers to investigate the effect of biochar dosage on the porosity of the specimens at high dosages. Given the high standard deviation of the specimens containing 6.5% biochar, it could be suggested that the 28-day UPV pattern aligned with the 28-day unit weight trend shown in Figure 4. However, it had inconsistency with the 28-day CS pattern, particularly in the range of 5 to 6.5% substitution dosage. The biochar's strengthening mechanisms and factors explained in Section 3.3.1 might cause this conflict, given that the added porosity had little to no effect on the mechanisms.

The findings also revealed that raising the biochar dosage to 2.5% reduced the 90-day UPV of the specimens by 0.96%. Expanding the biochar dosage beyond 2.5% to 6.5% boosted the UPV reduction rate, decreasing UPV from 4.21 km/s to 4.07 km/s (see Figure 9). However, increasing the dosage to 8% raised the specimens' UPV by 1.96% compared to the B6.5-F50-G10. The porous structure of biochar could account for the UPV reduction. However, its filler effect and potential to extend the RGD and fly ash reactions by providing water might lessen its adverse impacts on the porosity of the specimens. Nonetheless, increasing the biochar dosage from 2.5% to 6.5% could cause a dilution effect, increasing the reduction rate of the UPV. The interruption between the performance of fillers, i.e., fly ash, RGD, and biochar, could also contribute to the boost in the reduction rate. The strange increase in the UPV of the specimens due to the rise in the biochar dosage to 8% also accentuated the importance of examining higher dosages on the properties of the concrete in future studies. Theoretically, the lower OPC content in the mix could collaborate on increasing the UPV at 8% by reducing the alkalinity of the environment, lessening the biochar deterioration, and thereby reducing its effect on the concrete’s porosity. Except for the improvement in the UPV due to raising the biochar dosage to 8%, the UPV trend at 90 days was aligned with the unit weight and CS patterns shown in Figures 4 and 6.

Figure 9 depicts that increasing the biochar dosage to 2.5% boosted the 90-to-28-day UPV ratio by 0.08%. This might be because of the simultaneous effect of biochar as filler and water provider for the RGD and fly ash reactions. However, raising the dosage from 2.5 to 6.5% gradually reduced the ratio by 3.08%, likely due to the amplified dilution effect and magnified influence of biochar deterioration at high dosages. Escalating the biochar dosage to 8% enhanced the UPV improvement rate by 1.51% compared to B6.5-F50-G10. Accordingly, the rise in the UPV improvement ratio was maximized at 2.5% replacement and minimized at 6.5%. This pattern was similar to that of the CS. Nonetheless, the agglomeration of biochar and their interlock could not explain the increase in the UPV improvement rate at 8% substitution. Therefore, further research is required to determine the reason for this improvement. The potential effect of retained water in the biochar pores on the 28-day unit weight of the specimens could also explain the difference between the UPV improvement pattern and that of unit weight.

The data also revealed that increasing the biochar dosage had a stronger impact on the UPV of conventional concrete than sustainable concrete at 28 days. The higher impact in the conventional concrete might be due to the high alkalinity of the mix since OPC reactions could boost the alkalinity of the paste, thereby raising the biochar deterioration. This deterioration increase could boost the porosity of the biochar and consequently concrete.

The 90-day measurements showed a reverse trend, in which substituting OPC with biochar had more of an impact on the sustainable concrete, except for 2.5% replacement. The fulvic-acid organics produced by the biochar deterioration might lessen the alkalinity of the cement paste over time. This could significantly affect the reactivity of fly ash and RGD since their reactivity depends on the alkalinity of the system. This hypothesis could also support the negative effect of the biochar increase on the UPV of the specimens, as increasing the biochar could promote acid production.

The data also revealed that, except for B2.5-F0-G0, increasing the biochar dosage by up to 6.5% increased the 90-to-28-day UPV ratio of conventional concrete. However, it reduced the UPV improvement ratio in sustainable concrete. This reduction in the enhancement rate could result from two factors: 1) the potential interruption in the filler effect of biochar in the presence of RGD and fly ash, and 2) the effect of fulvic-acid organics on reducing the alkalinity of the environment. This underscored the importance of optimizing the biochar dosage and particle size distribution based on the binders used in a cementitious composite.

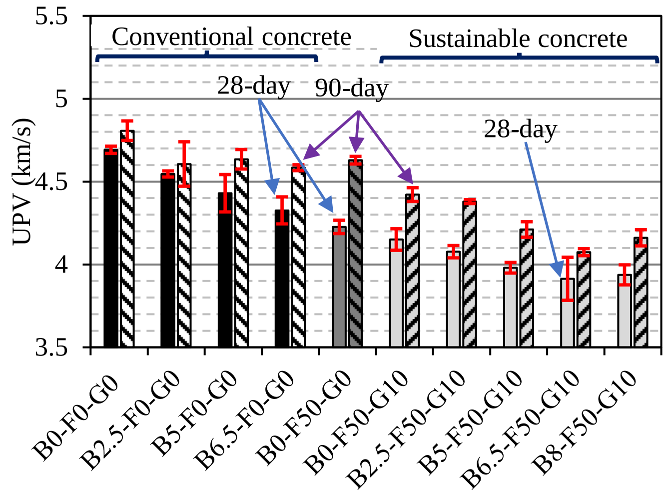

Figure 8. UPV of the specimens

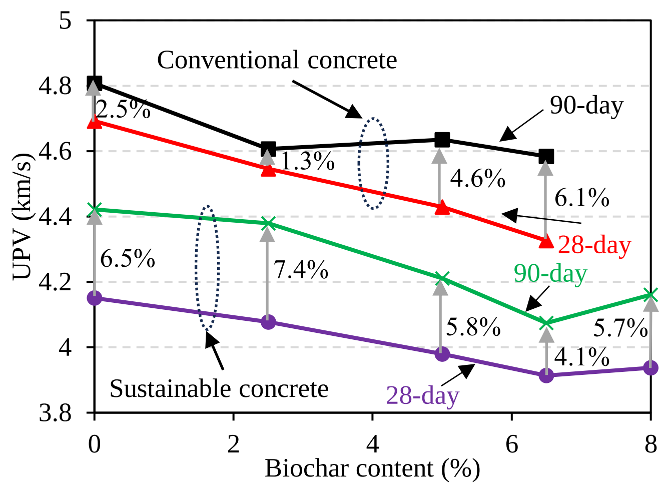

Figure 9. Effect of biochar content on the UPV

3.4. Stress-Strain Behavior

3.4.1. Elastic Modulus

Figure 10 shows the stress-strain curves of the specimens under the compression load at 90 days. Besides the effect of biochar on the CS of specimens discussed in Section 3.2, Figure 11 depicts that increasing the biochar dosage to 5% reduced the elastic modulus (E) of conventional concrete by 18.85%. However, expanding the dosage to 6.5% rebounded the E of conventional concrete by 15.49%. In sustainable concrete, however, boosting the biochar dosage to 8% gradually lowered the E of the concrete by 41.79%. The outcome also showed that the gradient of the reduction trend in the sustainable concrete dropped from 1872.1 to 739.67 after 5% replacement. These data could highlight the importance of a 5% replacement, as the effect of biochar dosage on E of concrete was changed at the percentage. The reduction in the E of the specimens by increasing the biochar dosage could be attributed to its structure, stiffness, and deterioration.

Given the similarities between biochar and lightweight aggregates, the porous structure of biochar could enable biochar to deform more than the solid cement paste. According to Li et. al. [70], the specific structure of biochar could also change the failure mechanism of concrete. In normal concrete, the cracks were initiated from the aggregates' interfacial transition zone (ITZ) and propagated toward the cement paste. However, given the low strength of biochar particles, the cracks simultaneously formed in the biochar particles and their ITZ. Despite this coincidence failure, the correlation between the biochar orientation and its load-bearing capacity in the composite, which resulted from its anisotropic structure, prevented a sudden failure of all biochar particles concurrently. Accordingly, the biochar particles were continuously and sequentially fractured during the test, gradually increasing the porosity of concrete and thereby reducing its E. The porous structure of the biochar could also increase the stress concentration in the biochar pores under loading [70], facilitating the formation of initial cracks in the biochar particles. This hypothesis also aligned with the effect of lightweight aggregates on the elastic properties of concrete [71,72].

A literature [73] suggested that the parallel model can be used to calculate the E of concrete based on the properties of its ingredients and their volume fracture. Given the model, increasing the biochar dosage could increase its volume fraction in the mix. This increase in the fraction could ultimately reduce the E of the system since the biochar had a lower E than the concrete [23,70]. This was also aligned with the model developed for simulating biochar-modified concrete by Li et. al. [70].

The biochar deterioration could also affect the E of the system by decreasing the E of biochar and producing fulvic-acid organics. According to Xu et al. [23], exposing the biochar to a cement environment could reduce its young modulus by 59.32% by degrading the biochar cross-linking network structure. Moreover, the deterioration could release the fulvic acid organics [23]. The produced fulvic acid could affect the size of pores in the concrete and weaken the cementitious bonds between the particles, increasing the deformation of specimens under a specific load [74]. This could ultimately reduce the range of the elastic behavior of cement paste and promote its plastic behavior.

The water retention of the biochar may have contributed to the increase in the modulus of elasticity of the conventional concrete containing 6.5% biochar. The biochar's water retention feature could condense the concrete by absorbing free water in the fresh mix. Accordingly, increasing the biochar dosage could reduce the free water in the fresh mix, lowering voids and boosting the E of the system. However, further research is required to determine influential factors that affect the increase in the E and CS of conventional concrete containing 6.5%.

Figure 11 also displays that the proposed formula for normal-weight concrete by ACI 318-19 (22) [75] could estimate the E of the reference specimens, i.e., the B0-F0-G0, with high accuracy. However, it could not precisely predict the E of specimens containing biochar. The reduction in E due to the usage of biochar was more than the decrease predicted by the formula based on the strength loss. Differences in the strengthening mechanisms of biochar compared to those of other SCMs, the specific failure mode of biochar-modified concrete, and the biochar deterioration could contribute to this decrease. Besides, the biochar porous structure could be a substantial factor that promoted this change. This is because it enabled biochar to deform more than the solid elements. The difference might also be attributed to the significant impact of biochar on the porosity and unit weight of the concrete since the advanced formula for the E of concrete highlighted the critical rule of the unit weight [75] in estimating the E of cementitious composites. Therefore, the results emphasized the importance of further research to determine the primary factors that lead to this reduction. This difference might also encourage researchers to develop a prediction model for the elastic modulus of the concrete containing biochar.

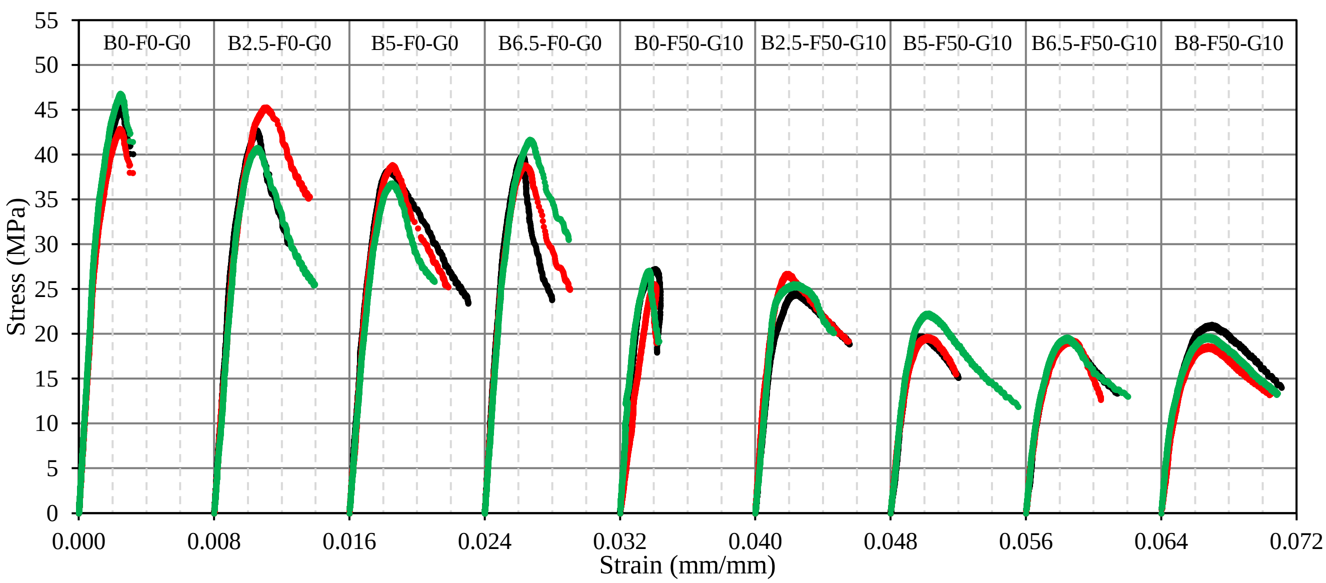

Figure 10. Stress-strain curves of the specimens at the 90-day test

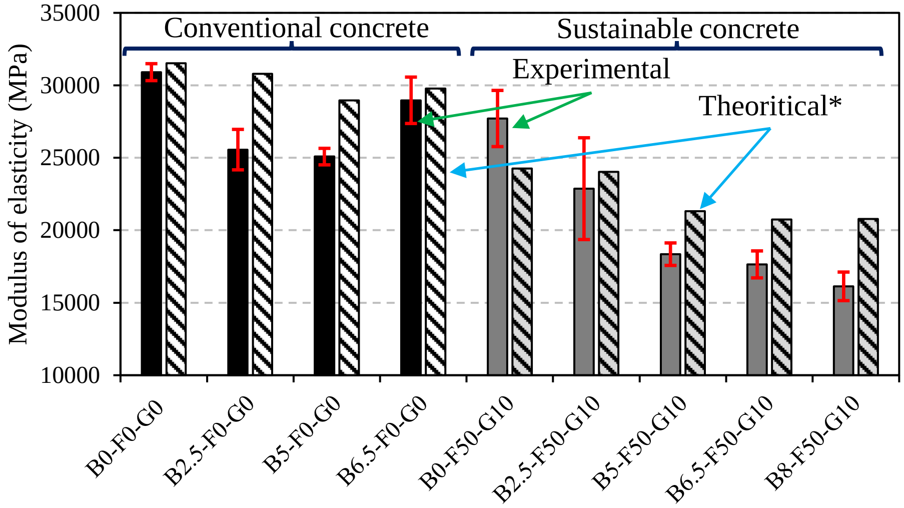

Figure 11. Effect of biochar content on the elastic modulus of the 90-day specimens

Results also demonstrated that using RGD significantly enhanced the E of B0-F50-G10 specimens compared to the ACI 318-19(22) [75] prediction. This surge in E could be caused by the effect of RGD on forming dense ettringite structures, reducing the porosity of concrete. This was also consistent with earlier findings on the elasticity development acceleration of cementitious composites in the presence of gypsum [76]. It also underscored the potential of using original RGD products in concrete since earlier findings [76] were based on the use of natural gypsum in the cement clinker.

3.4.2. Peak Broadness

Figure 10 also illustrates that adding biochar to the concrete could broaden the peak at the stress-strain diagrams. Table 6 shows the full width between the strains at 90% and 80% of the peak load. The limitation, 90% of the peak load, came from a sudden failure of the reference specimens, B0-F0-G0. The data showed that raising the biochar dosage to 2.5% boosted the peak broadness by 53.5% at 90% peak load in conventional concrete. However, a further increase in the biochar dosage reduced the peak broadness at the level such that the peak broadness at the B6.5-F0-G0 was approximately similar to that of the B0-F0-G0. The peak broadness at the 80% strength level was increased when the biochar was raised to 5% and then reduced by 26.29% at 6.5% substitution.

The data also showed that the combination of fly ash and RGD narrowed the peak at the 90% peak load level by 43.9%. This significant drop could be due to the added brittleness by RGD reactions. The RGD reactions could fill the pores with ettringite products [77]. These expansive products could cause microcracks within the concrete, which could link to form major cracks under loading, resulting in a narrow peak and significant strength drop after the peak load. Figure 12 shows the expansive ettringite structures and the linked microcracks.

The results also displayed that adding biochar to the sustainable concrete could compensate for the effect of RGD on the peak broadness. The broadness at 90 and 80% peak load levels increased by 207.15 and 247.22% at a 2.5% replacement, respectively. Nevertheless, increasing the dosage in the range of 2.5 to 6.5% could narrow the peak by 16.26 and 23.46% at 90 and 80% peak load levels, respectively. However, boosting the biochar dosage to 8% changed the effect, as the peak broadened by 37.27 and 48.51%, respectively.

Table 6. Strains of the mixtures at the 80% peak load and the peak broadness

Mix ID	Full Width (mm/mm)	
	At 90% peak load	At 80% peak load
B0-F0-G0	0.00129	-
B2.5-F0-G0	0.00198	0.00263
B5-F0-G0	0.00173	0.00289
B6.5-F0-G0	0.00129	0.00213
B0-F50-G10	0.00072	0.00104
B2.5-F50-G10	0.00222	0.00362
B5-F50-G10	0.00192	0.00293
B6.5-F50-G10	0.00186	0.00277
B8-F50-G10	0.00255	0.00412

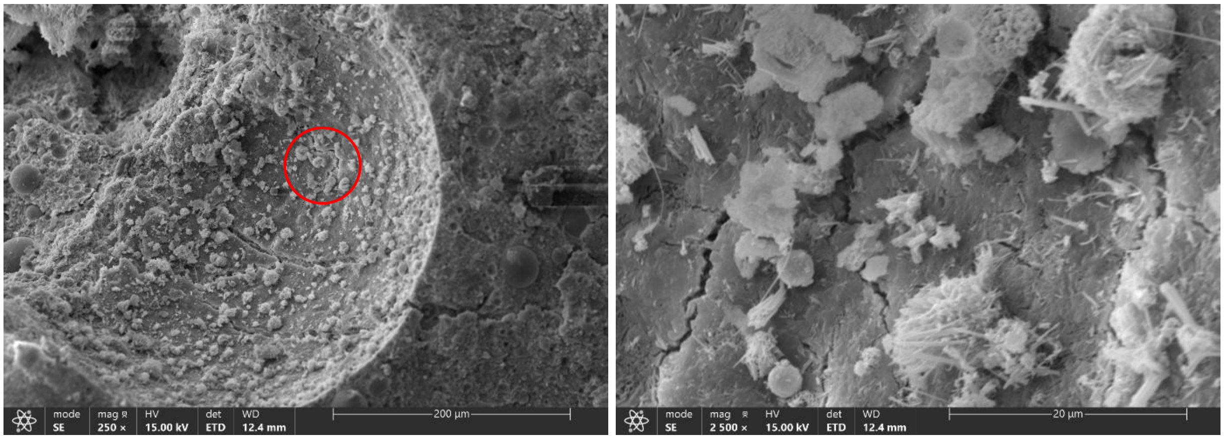

Figure 12. Expansive ettringites and linked microcracks

Four features of biochar could contribute to improving the peak broadness of the curve by increasing the biochar dosage. First, the porous structure of biochar could allow more deformability compared to the solid structure of other concrete ingredients. This increase in deformability could boost the deformation of the system as its stiffness was reduced. Therefore, it could increase the width between the strains at the load levels and that of the peak load at the left side of the peak. Second, the biochar's specific microstructure could allow a gradual failure, similar to the behavior observed in the lightweight aggregates [71,72]. Moreover, the anisotropic structure of biochar and the dependency between its orientation and load-bearing capacity in the concrete could cause a gradual failure, dissipating energy and increasing the ductility of the system [70]. This gradual failure could increase the distance between the strains at the peak load and those at the 80 and 90% peak loads at the right side of the peak. Third, the specific shape of the biochar could theoretically delay the microcrack propagation, which could increase the strain at the peak load and enhance the strain of the system after the peak. This could be caused by the difference in the crack initiation mechanisms in aggregates' and biochar's ITZs, explained in Section 3.4.1. Fourth, the viscoelastic behavior of biochar [70] and the release of the fulvic acid during its deterioration [23] could boost the plasticity of the concrete microstructure, increasing its plastic behavior domain and thereby the strain widths. Despite these features, excessive biochar dosage could leverage the concrete's porosity. This could shorten the crack propagation path and facilitate the formation of critical linked cracks. Accordingly, it could significantly reduce the width between the strain at peak load and the strains at the 80 and 90% peak load levels.

3.4.3. Peak and Ultimate Strains and Post-peak Behavior

Table 7 displays the average strains of the specimens at the peak and failure loads. In this study, 20% strength loss was considered the failure criterion if the specimens did not suddenly break after the peak, i.e., the B0-F0-G0 and B0-F50-G10. In these two cases, the maximum strain was determined by the maximum strain experienced by specimens, not the failure point. This limitation was applied since the specimens containing biochar could carry up to 38% strength loss before the crush.

The results also displayed that adding biochar to conventional and sustainable concrete could raise the strains at the peak load and failure points. The strains in conventional concrete increased by 11.07 and 35.63% at 2.5% replacement, respectively. Nonetheless, boosting the biochar percentage from 2.5 to 6.5% lessened the strains so that the B6.5-F0-G0 had approximately the same strain at the peak load and 9.38% higher strain at the failure point compared to B0-F0-G0. In the sustainable concrete, adding 2.5% biochar raised the strains at peak load and failure points by 13.92 and 112.5%, respectively. Further increase in the biochar dosage to 8% promoted the strains at peak load and failure points by up to 44.85 and 147.2% compared to those of the B0-F50-G10, respectively. However, peak load strain was slightly decreased in the B5-F50-G10 compared to that of B2.5-F50-G10. The ultimate strain was also reduced by 13.94% and 12.64% compared to that of B2.5-F50-G10 at 5 and 6.5% replacements, respectively. The increase in the deformability of the specimens aligned with the higher ductility and fracture energy of the concrete containing biochar in three-point bending tests in earlier studies [24,78] and the delayed peak load strain found by Li et. al. [70].

Five factors could explain the improvement in the strain at peak load: 1) The specimens containing biochar had lower E; 2) the acid released during the biochar deterioration could boost the plastic behavior of the cement paste; 3) biochar's potential for higher deformability (based on its viscoelastic behavior) than the solid concrete ingredients; 4) the gradual and sequential failure of biochar particles based on their orientations and anisotropic behavior; and 5) the specific cracking mechanism of biochar particles and their ITZ, which could delay the formation of crucial cracks. The post-peak ductility of the specimens containing biochar could also be due to 1) the plastic behavior added to the cement paste by the released acid during biochar deterioration; 2) the gradual failure of biochar particles, which could delay the formation of the critical cracks; and 3) the edge shape of the biochar, which could increase the required crack path for the failure and reinforce the cement paste.

However, an excessive increase in the biochar dosage could lower the crack path in the system. This is because it could increase the porosity of the concrete, reduce the distance between the biochar particles in the cement paste, and potentially lead to biochar agglomeration. Given the biochar's low strength and failure mechanism, the lower distance and agglomeration could boost the strength loss at post-peak due to a significant load applied to the biochar particles. The difference between the observed patterns in conventional concrete and sustainable concrete could also emphasize the effect of the binders used in the concrete on the performance of biochar, as this factor could affect the alkalinity of the concrete and thereby the biochar deterioration. The particle size distributions of the binders might also affect the peak and ultimate strain since this factor could influence the porosity of the concrete.

Table 7 also showed that adding biochar to the system could result in a smoother post-peak behavior than the reference specimens. The data demonstrated that the width between the strains at peak and failure loads could be maximized by up to 139.47 and 1050% in conventional and sustainable concrete at 5 and 2.5% substitution rates, respectively. Biochar's gradual failure and its effect on increasing the plastic behavior of the cement paste could contribute to this significant rise in smoothness. However, an over-increase in the biochar could reduce the post-peak width by increasing the porosity of the system and shortening the crack propagation path.

Table 7. The strain of specimens at peak and failure loads

Mix ID	Average strain at peak load (mm/mm)	Average ultimate strain (mm/mm)	Width between strains at peak and failure loads (mm/mm)
B0-F0-G0	0.00244	0.00320	0.00076
B2.5-F0-G0	0.00271	0.00434	0.00163
B5-F0-G0	0.00250	0.00432	0.00182
B6.5-F0-G0	0.00244	0.00350	0.00106
B0-F50-G10	0.00194	0.00216	0.00022
B2.5-F50-G10	0.00221	0.00459	0.00238
B5-F50-G10	0.00206	0.00395	0.00189
B6.5-F50-G10	0.00258	0.00401	0.00143
B8-F50-G10	0.00281	0.00534	0.00253

Findings also displayed that using a combination of fly ash and RGD lessened the strains at peak and failure loads by 20.49 and 32.5%, respectively. This reduction could be attributed to the effect of RGD on the formation of dense ettringite structures [76] and their potential to increase the microcracks [77]. The first factor could boost the E of the system, reducing the deformation and ultimately the strain of the specimens at the peak load. The latter could affect the strains at peak and ultimate points, as it could facilitate crack propagation and expansion. Based on the aforementioned, although biochar could lower the strength of concrete, its usage in sustainable concrete containing fly ash and RGD was necessary to avoid a brittle failure.

3.5. Microstructural Analysis

Figure 13 illustrates the interfacial transition zone (ITZ) between biochar-cement paste and aggregate-cement paste in conventional and sustainable concrete specimens. The aggregate-cement ITZ predominantly consisted of calcium hydroxide, ettringite, and calcium silicate hydrates (CSH). However, the distribution of hydration products in the biochar ITZ relied on the location of the products relative to biochar particles. If hydration products were formed on the side shells of the biochar, the ITZ would resemble that of the aggregates. However, ettringite and CSH were the predominant hydration products formed on top of the biochar pores, i.e., where the water escaped. The variation in the hydration products distribution may be ascribed to the wall effect of the biochar side shells and the potential water release from the biochar pores. The released water could enhance the water-to-cement ratio in the cement paste adjacent to the top of the pores, amplifying the hydration. Calcium hydroxide is a soluble phase of hydration products. It could be dissolved in biochar-provided water over time and produced Ca^{2+} ions. These ions enhanced the alkalinity of the mix and formed lime water, which could generate calcium carbonate and CSH phases, enhancing the microstructure of concrete. On the other hand, releasing the functional groups during the biochar deterioration could promote a Ca-rich layer on the surface of the biochar's side shell. The dependency between the biochar orientation and the hydration product distribution was consistent with Zhu et. al. [79].

The SEM analysis of the biochar-cement paste ITZ in sustainable concrete showed that the effective zone of the biochar-retained water on dissolving the calcium hydroxide was more confined to the zone near the biochar pores than that of conventional concrete. This may be because of the delayed reactions of RGD and fly ash, which used a portion of released water from biochar. The lower the free water, the lower the dissolution of calcium hydroxide. Consequently, a larger volume of calcium hydroxide could be observed in SEM images in the biochar-cement paste ITZ in sustainable concrete.

The analysis also showed that while the biochar pores connected to the ITZ were filled with hydration products (Figure 13-f and h), the majority of the biochar pores were empty. This could allude to the low level of interconnectivity between the biochar pores and their size. Moreover, the cracks in the biochar structure and the hydration product-free surfaces of the biochar particles could display the biochar fracture under compression. This could demonstrate the potential contribution of biochar's low strength to reduce the strength of concrete specimens containing biochar. According to Li et. al. [70], the biochar's low strength could result in a simultaneous fracture in the biochar and its ITZ under loading, which might cause the hydration-free surfaces in the SEM images.

The data also confirmed that the biochar had good compatibility with cement paste in both conventional and sustainable concrete. Results also showed the irregular shape of biochar could result in a better interlock in biochar-cement paste ITZ than that of the aggregate-cement paste. However, the clear boundary between the biochar and cement paste emphasized the lack of noticeable reaction between the two in the specimens. This highlighted that although the biochar could intensify the hydration reactions by providing additional nucleation sites and water, it did not react with the cement paste. Accordingly, using it as an additive could be more efficient in cementitious composites.

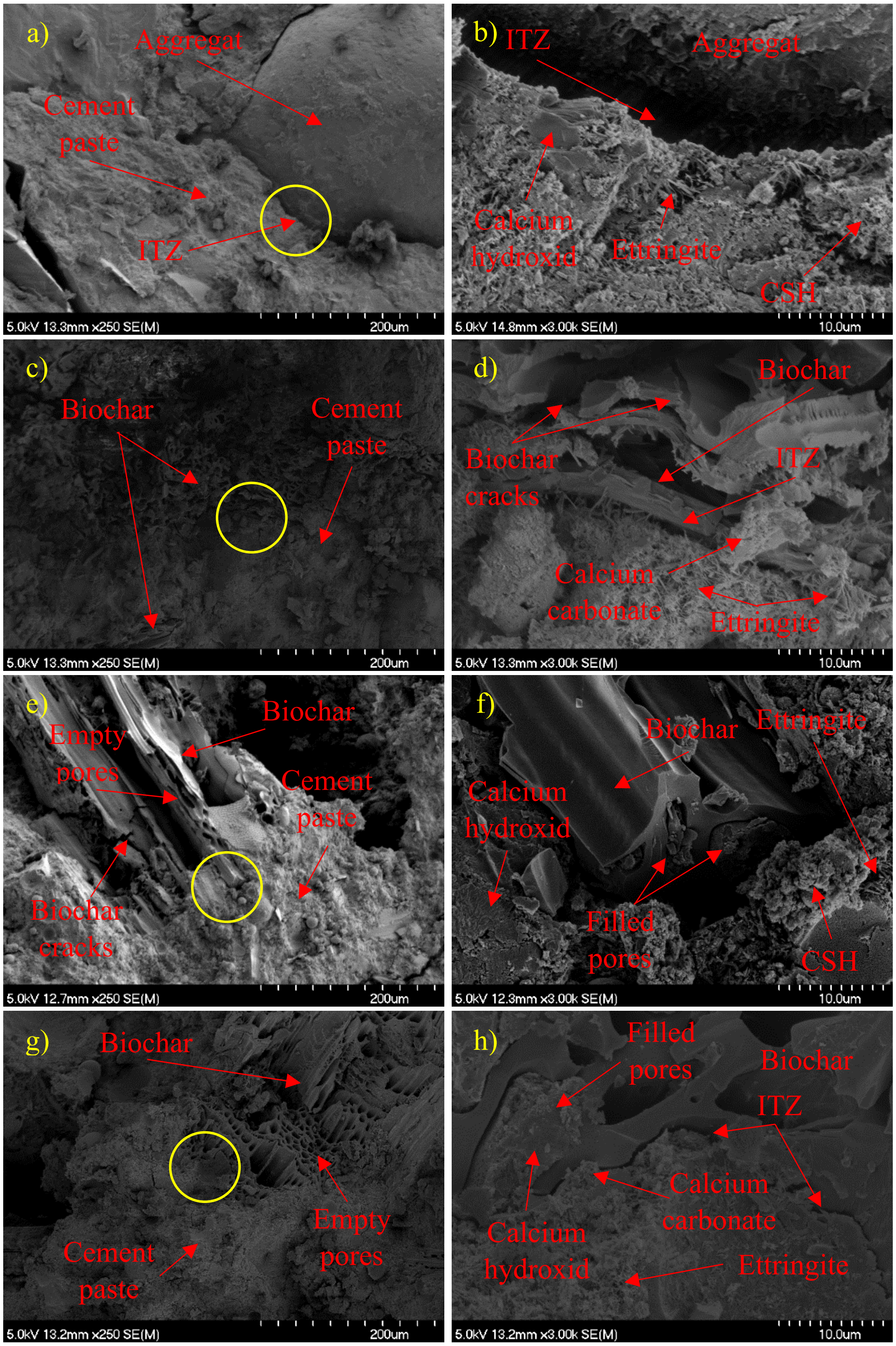

Figure 13. SEM of a & b) the aggregate-cement paste, and biochar-cement paste in c & d) conventional concrete, and e to h) the sustainable concrete

Figure 14 shows the effect of RGD on the microstructure of concrete. Since the RGD included fibers, it could be easily identified by the RGD-fiber structures. According to the SEM result, RGD promoted the formation of a dense ettringite in the concrete at the microstructural level. The formed ettringites gathered close to the fly ash particles and attempted to interact with them. Furthermore, since no calcium hydroxide was detected in the zone, it could be concluded that using RGD reduced the calcium hydroxide crystals next to the RGD particles. The significance of removing the fibers from RGD is further illustrated in Figure 14, where it is evident that the fibers accumulated at certain points in the mixture forming voids in the concrete. Figure 15 demonstrates a fly ash particle close to a biochar in the sustainable concrete, partially covered in CSH and ettringite crystals. Since more reactions were visible on the surface of this fly ash particle than on that shown in Figure 14, it could be stated that the biochar-retained water boosted the fly ash-RGD reactions by providing additional water.

Figure 16 and Table 8 display the position and chemical composition of the points analyzed by the SEM-Energy Dispersive X-ray Spectroscopy (EDS), respectively. Although calcium and oxygen were the dominant chemical elements in the aggregate-cement paste ITZ, the results showed a significant reduction in calcium dosage adjacent to the biochar particles. The data also revealed that the dosage of alumina was raised in the biochar's effective zone. This increase could show the potential of biochar to offer nucleation sites in the concrete. Although the reduction in the Ca to Si ratio and increasing the aluminum content could highlight the potential promotion in the formation of CASH in concrete, further research is required since, according to Zhu et. al. [80], the change in the chemical elements of the system could be caused by the changes in the chemical elements in other hydration phases.

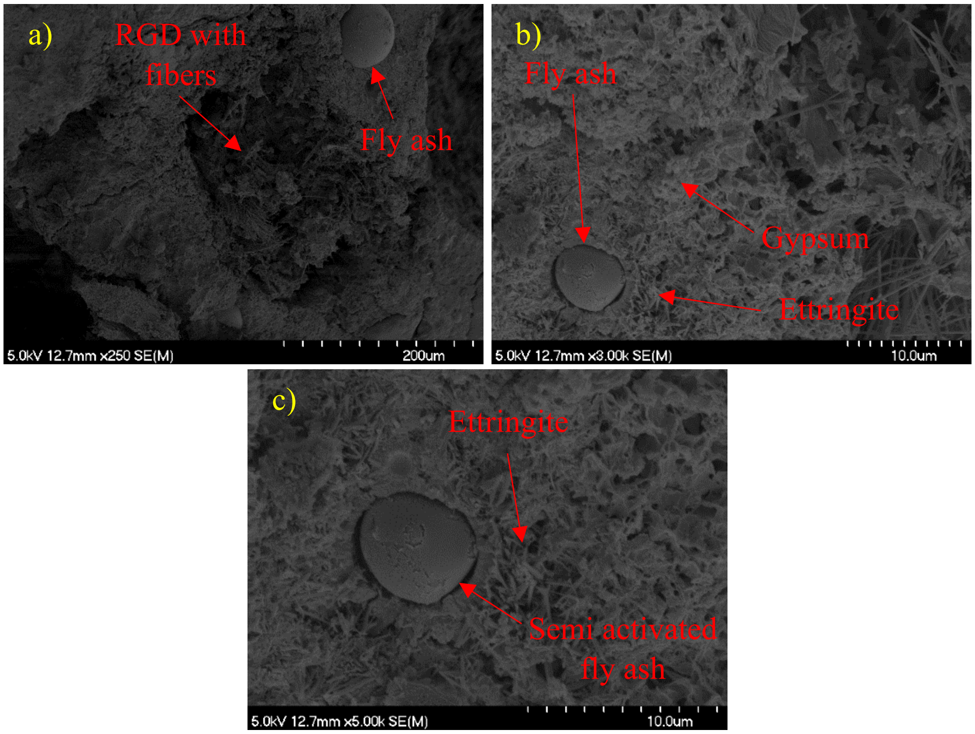

Figure 14. Effect of RGD on the microstructure of the sustainable concrete

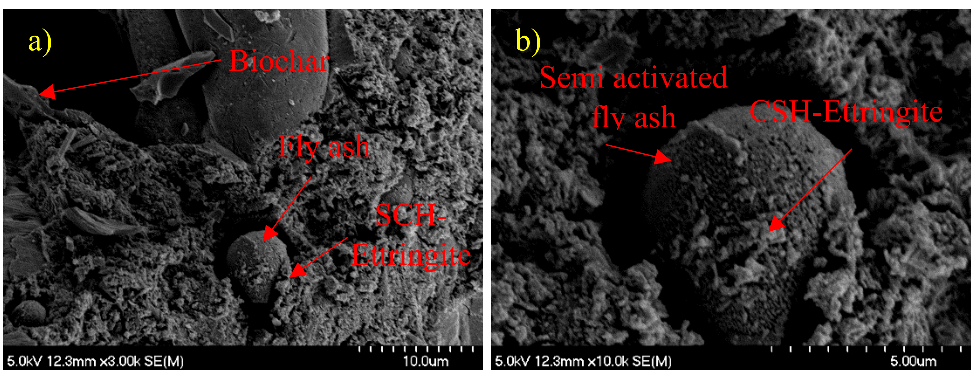

Figure 15. Fly ash activation by RGD

Table 8 also represents the higher carbon dosage at point 6, indicating the potential boost in carbonization of cement paste on the biochar's boundary. This aligned with Xu et al. [23], which showed that biochar deterioration released functional groups, including COOC, into the cement paste. Points 9 to 11 also presented that the products on the fly ash particles contained sulfur elements, which fit with the potential fly ash activation by RGD. The data also showed that adding biochar to the mix containing fly ash and RGD slightly affected the alumina dosage; however, it significantly boosted the oxygen and sulfur dosages near biochar in the sustainable concrete.

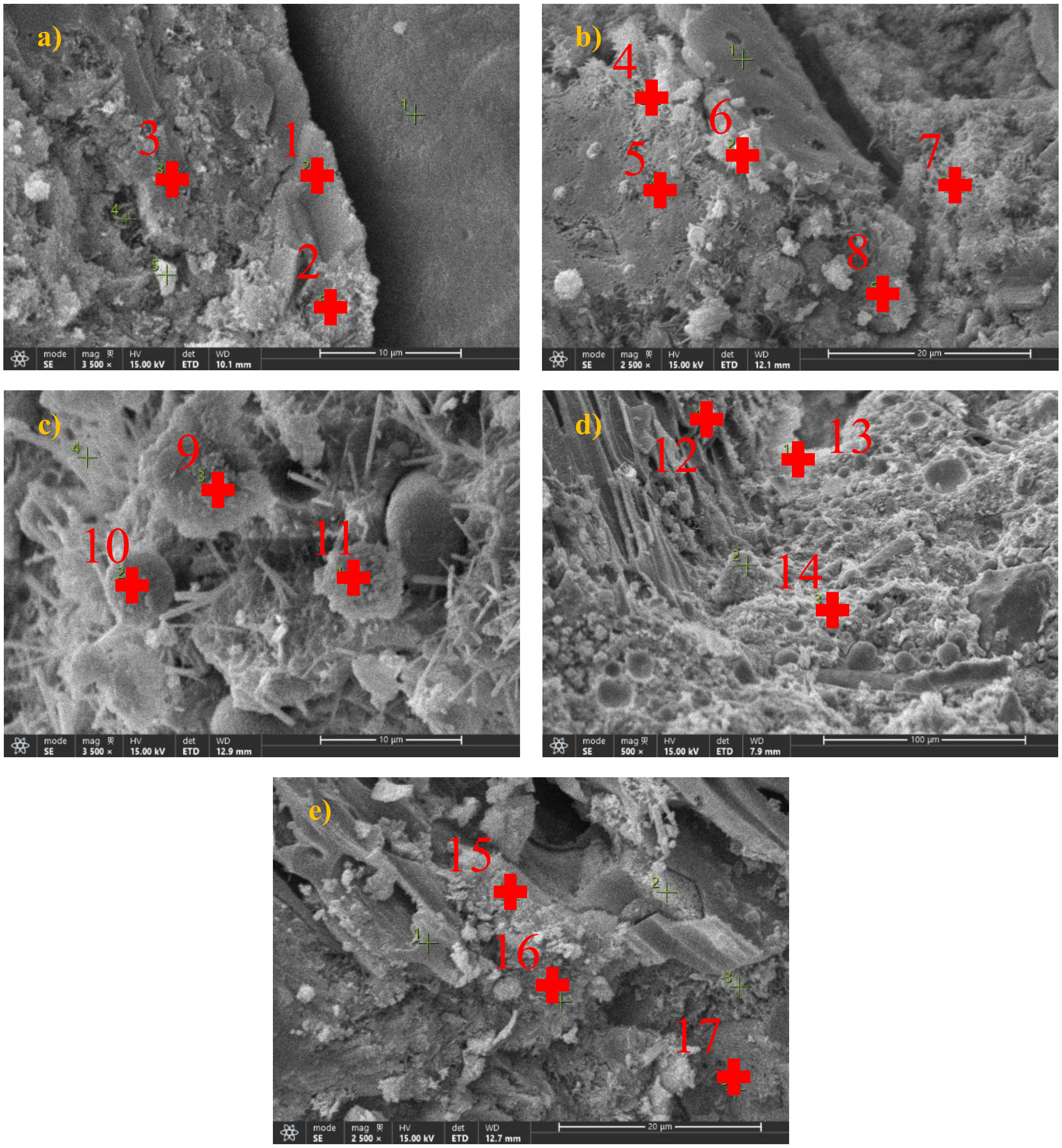

Figure 16. The points considered for EDS a) Aggregate-cement paste ITZ and b) Biochar-cement paste ITZ in conventional concrete, c) RGD-fly ash reactions and d) Biochar-cement paste ITZ in sustainable concrete, and e) partial filling of biochar pores by cement paste

Table 8. Chemical elements at the EDS points

Point number	Chemical element (wt%)					
	C	O	Al	Si	Ca	S
1	13.0	39.8	0.1	7.1	40.0	-
2	18.2	39.3	1.2	14.6	25.0	-
3	14.0	32.8	1.3	8.6	42.2	-
4	15.9	40.1	3.3	10.6	24.5	-
5	16.6	32.1	2.0	13.3	28.6	-
6	33.3	40.9	0.6	4.0	10.0	-
7	16.6	48.1	3.2	8.2	20.6	-
8	17.5	37.9	1.7	14.4	24.2	-
9	16.1	41.8	3.5	9.3	18.3	0.3
10	17.3	24.3	2.7	7.7	15.1	0.0
11	16.6	41.4	3.8	9.2	17.8	0.4
12	19.4	45.9	3.6	5.9	16.6	2.3
13	15.5	51.1	2.2	12.2	16.3	0.0
14	15.0	53.2	4.3	5.5	17.6	3.1
15	19.9	43.6	2.8	10.7	19.6	-
16	5.9	15.3	6.4	7.7	61.9	-
17	5.8	17.1	1.8	9.9	58.0	-

4. Environmental Impact

This section evaluates the carbon footprint of conventional and sustainable concrete containing 0 and 2.5% biochar. Although the results demonstrated that increasing the biochar dosage could significantly impact the 90-day strength of specimens, the biochar potential for reducing the carbon footprint of concrete could compensate for the strength loss. Therefore, the carbon dioxide emissions of concrete containing biochar could be assessed to determine its potential.

The current analysis estimated the carbon dioxide emissions of concrete by focusing on the carbon dioxide equivalent of its ingredients at their production stage. This was because the carbon dioxide emissions of other parts such as the transportation phase strongly depended on the type of vehicle and the destination distance. Accordingly, the weight of concrete ingredients in each mix and their carbon dioxide equivalent (CO_2-eq), listed in Tables 9 and 10, were used to calculate the

CO_2-eq of the mixtures. It is worth noting that the biochar carbon footprint varied between -2.0 to -3.3 kg CO_2-eq/kg based on the biomass source and the pyrolysis procedure [81]. However, based on the required preparations, including biomass preparation and heating as well as biochar grinding, the total carbon footprint of the biochar could shift to -1.3 kg CO_2-eq/kg [12].

Table 9. Evaluated mix designs

Concrete component	Mix design			
	B0-F0-G0	B2.5-F0G0	B0-F50-G10	B2.5-F50-G10
Coarse Aggregates (kg)	1197.45	1197.45	1197.45	1197.45
Fine aggregates (kg)	979.48	979.48	979.48	979.48
OPC (kg)	490.5	478.24	196.2	183.94
Water (kg)	236.4	236.4	236.4	236.4
Biochar (kg)	0	12.26	0	12.26
Fly ash (kg)	0	0	245.25	245.25
RGD (kg)	0	0	49.05	49.05

Table 10. Carbon dioxide equivalent concrete ingredient

Concrete component	Carbon dioxide equivalent (kg CO_2-eq/kg)
Coarse aggregates*	0.0459
Fine aggregates*	0.0139
OPC**	0.82
Water***	0
Biochar***	-1.3
Fly ash**	0.027
RGD****	0.034

*: Source [3]
**: Source [82]
***: Source [12]
****: Source [83]

Table 11 illustrates that replacing 2.5% of the binder(s) with biochar in conventional and sustainable concrete could lessen the CO_2-eq of concrete by 5.52 and 10.28%, respectively.

Therefore, given the global climate change crisis and the urgent need to mitigate carbon dioxide emissions, using a low dosage of biochar in concrete might be worthwhile since the substitution could merge the carbon dioxide emissions of concrete and provide a sustainable waste management technique for biochar. Besides the environmental benefits of biochar usage in concrete, reusing biochar in concrete could financially profit the industry by lessening the carbon tax and the price of concrete production.

Table 11. The carbon footprint of the mixes

Concrete component	Mix design			
	B0-F0-G0	B2.5-F0-G0	B0-F50-G10	B2.5-F50-G10
Coarse aggregates (kg CO_2-eq)	54.96	54.96	54.96	54.96
Fine aggregates (kg CO_2-eq)	13.61	13.61	13.61	13.61
OPC (kg CO_2-eq)	402.21	392.16	160.88	150.83
Water (kg CO_2-eq)	0	0	0	0
Biochar (kg CO_2-eq)	0	-15.93	0	-15.93
Fly ash (kg CO_2-eq)	0	0	6.62	6.62
RGD (kg CO_2-eq)	0	0	16.68	16.68
Total	470.78	444.8	252.75	226.77

5. Future Studies

The data suggested that the biochar agglomeration could improve the strength of concrete at higher dosages. Accordingly, further research on the optimum dosage of the biochar is proposed. It also demonstrated that the poor structure of biochar and its deterioration significantly affected the strength of concrete containing biochar. Therefore, developing a technique for assessing the strength of biochar particles and fortifying them, as well as reducing the effect of their deterioration on the properties of biochar and biochar-modified concrete would be a major step toward using biochar in concrete. Moreover, assessing the biochar's long-term performance in various environments is also required to ascertain the service life of the concrete structures made of biochar-modified concrete. The potential effect of retained water on the properties of the concrete also accentuates the necessity of precisely examining the effect of additional drying time on the properties of the concrete containing biochar.

The data also displayed the potential of employing the whole RGD as SCM in the concrete. However, it could be argued that the brittleness of the concrete containing RGD, its low early age strength, and the presence of fibers throughout the RGD could restrict the applications of RGD-modified concrete. Therefore, future studies can focus on these subjects as well as the durability of RGD-modified concrete to examine the feasibility of its usage in the industry.

6. Summary and Conclusions

This study aims to examine the effect of biochar dosage and its retained water removal as well as using whole RGD on the properties of conventional and high-volume fly ash concrete. Accordingly, 60 concrete cylinders were manufactured and tested at 28 and 90 days to evaluate the effect of the materials on the mechanical strength, stress-strain behavior, porosity, and microstructure of the composites. The main outcomes of the study are listed below:

- Adding biochar to concrete by up to 6.5% increased the porosity of conventional and sustainable concrete due to the porous structure of the biochar, resulting in a reduction in the unit weight (by up to 3.99 and 2.33%, respectively) and UPV (by up to 4.64 and 7.86%, respectively).
- Increasing the biochar dosage by up to 6.5% in the conventional and sustainable concrete lessened the compressive strength (by up to 15.58 and 26.88%, respectively) and elastic modulus (by up to 6.29 and 41.79%, respectively) at 90 days. However, the 28-day strength of the specimens was maximized at a 2.5% replacement rate. This difference emphasized

the importance of measuring the long-term effect of biochar in cementitious composites, as the biochar deterioration, lower cement content, and biochar retained water could significantly affect the long-term properties of the system.

- Using 2.5% biochar could significantly enhance the ductile behavior of conventional and sustainable concrete, by increasing the peak broadness (by 53.49 and 208.33% at the 90% peak load level, respectively) and post-peak smoothness (by 114.47 and 981.82%, respectively).
- Biochar-retained water could promote hydration and carbonization by boosting the formation of calcium carbonate, CSH, and CASH in the biochar ITZ, particularly adjacent to the open pores, in conventional concrete. In the sustainable concrete, the water also boosted fly ash-RGD reactivity and the sulfur dosage.
- Replacing 2.5% of OPC with biochar in conventional and sustainable concrete could reduce the carbon footprint of the concrete by 5.52 and 10.28% compared to the reference mixtures (B0-F0-G0 and B0-F50-G10), respectively.
- RGD has the potential to be used as a cement replacement in high-volume fly ash concrete to increase both the strength of the concrete and the reactivity of fly ash. However, the low early-age strength and brittleness of the concrete may limit its industrial applications.

Based on the results, this study recommends a 2.5% replacement of cement with biochar in conventional and sustainable concrete, as this dosage has minimal impact on compressive strength while significantly enhancing its ductility and reducing its carbon footprint.

Acknowledgment

The authors would like to acknowledge Alireza Sadat Hosseini and Ali Alinejad for their help in the specimen preparation and testing phases. The authors also thank the concrete and structural lab personnel at Dalhousie University for their cooperation in the experimental phase of this study. Dr. James Forren's cooperation and support would also be appreciated. We also would like to express our sincere gratitude to Dr. Eric Moreau at the Mechanical Engineering Department's FIB-SEM facility, Dr. Ping Li at the Department of Biology, and the staff at the Mineral Lab. The authors also acknowledge USA Gypsum and RDA Atlantic Inc. for providing recycled gypsum drywall and biochar. We also thank Dalhousie University's VPRI Next Wave Competition and the Nova Scotia Graduate Scholarship (NSGS) program for their financial support.

Data Availability

Data will be available upon request.

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