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Green Concrete Development: Evidence from Waste Concrete and Hemicellulose Utilization

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Abstract

Natural resource scarcity, CO_2 emissions, and solid waste generated from the construction industry are major global environmental and developmental challenges, posing threats to the sustainability of terrestrial and aquatic ecosystems. In response to this multifaceted issue, recent studies focus on developing non-cement concrete, distinct from traditional cement-based compositions, by utilizing recycled concrete and wood waste molded at high temperature and pressure. Although wood hemicellulose shows adhesive properties and bonds particles at lower temperatures, it has not been studied in non-cement concrete. Hence, the present study focuses on developing green concrete using recycled concrete and hemicellulose, and further enhancing its strength with chitosan. The study used the press molding method with different pressing temperatures. The results, compared with conventional cement mortar and other wood components, revealed that hemicellulose-based green concrete exhibited superior bending strength compared to the other the components and even surpassed the strength of conventional cement mortar. Furthermore, an elevation in temperature to 60˚C resulted in enhanced strength, but a further increase to 160˚C led to delamination and thus a reduction in strength. Moreover, hemicellulose, when substituted by 50% of its weight with chitosan, further enhanced the strength of the concrete. The results also showed that hemicellulose has the potential to produce green concrete from abundant plants in a time interval of no more than ten minutes.

Keywords

Recycled Concrete Powder, Cellulose, Lignin, Hemicellulose, Chitosan, Oyster Shell

1. Introduction

Amidst natural resource scarcity, CO_2 emissions remain a major global environmental and developmental challenge, with the construction industry accounting for about 39% of total energy-related emissions. Such emissions are aggravated by the high demand for buildings, which consume significant amounts of resources for concrete, including 75% of sand deposits [1] and 4.1 billion tonnes of cement in 2019 [2], thus endangering the sustainability of life on land and underwater, and limiting resource availability for other competing uses such as agriculture and industry. Hence, it is vital to find an alternative for more environmentally friendly construction practices.

As a solution, construction and demolition (C&D) waste recycling have attracted significant attention because of its large amount generation [3]. However, most of the studies have focused on partial recycling, which requires incorporating new materials, and therefore, achieving complete recycling remains elusive.

To achieve the goal of a complete loop of waste recycling, the usage of plant-based waste and concrete waste show great potential to achieve promising results in both waste recycling and resource conservation. Therefore, in 2020, a concrete named Botanical Concrete was produced using only waste concrete powder and waste wood powder by the hot-pressing method, resulting in the production of a hardened body with high bending strength [4]. In this study, a high pressure of 50 MPa at a high pressing temperature ranging from 180˚C to 220˚C was applied, and it was observed that the Botanical Concrete bending strength was directly linked with the pressing temperature, pressing pressure, and mass ratio. The study concluded that such high pressure is difficult to apply while mass production and recommended that a lower pressing pressure is preferred. Therefore, the next study has investigated the effect of a lower pressing pressure, which showed that in an interval of 10 MPa to 50 MPa, the Botanical Concrete bending strength decreases as the pressure reduces [5]. Furthermore, the study demonstrated that obtaining a higher bending strength requires a higher content of wood powder, the same or higher amount than the concrete powder.

To improve the dimensional stability and water resistance of the Botanical Concrete as well as using lignin waste, which is abundantly produced around the world [6], incorporation of lignin with a pressing temperature of 180˚C to 240˚C and 30 MPa to 50 MPa pressing pressure was conducted [7]. The results showed that the addition of lignin up to 20% enhanced the Botanical Concrete bending strength and its dimensional stability. The study also discussed that the hardening and bonding properties are provided by lignin when heated over its glass transition, at which the hemicellulose decomposes.

The investigation of the bonding mechanism of the Botanical Concrete, which was molded out of normal concrete powder, carbonated concrete, sand, and wood powder, under 50 MPa pressing pressure and 220˚C temperature showed that there is no direct chemical interaction between ordinary concrete during the formation of Botanical Concrete; however, the alkalinity of ordinary concrete could promote chemical changes in wood, and its impact is greater than the pressing temperature [8]. However, all the studies have used a pressing temperature over 160˚C and neither of the studies have developed the Botanical Concrete at a lower or at room temperature pressing to further reduce environmental impacts and facilitate concrete production, as a high pressing temperature to 220˚C still requires a large quantity of energy and imposes environmental challenges.

Therefore, developing a waste-based concrete with a lower pressing temperature and pressure is required to explore and open new insights within this novel field of study. In addition, hemicellulose, which is reported to yield higher strength than that of lignin and can be obtained in a shorter pressing time compared to lignin [9], has not been reported within the field of non-cement concrete. Hence, developing green concrete using hemicellulose at a lower pressing pressure and temperature, even at room temperature, is the central objective of this study.

Hemicellulose is one of the most abundant renewable resources on earth [10]. It can be found in every plant and constitutes a significant part of the pulping and paper industry byproduct, which is released together with lignin and used as a burning substance. It is also one of the main components of black liquor, comprising 30% - 50% of it [11]. Approximately 1.3 billion tons of weak Kraft black liquor are produced every year and usually concentrated by evaporation and burned to produce energy [12].

The global annual production of hemicellulose is approximately 60 billion tons [13]-[16]. Therefore, it is a great green and sustainable resource to be used to alleviate the use of unsustainable natural resources and reduce emitting CO_2.

In the adhesive industry, the substitution of expensive synthetic adhesive with green adhesives is of interest from an ecological standpoint. This substitution can not only alleviate the shortage of fossil resources and reduce CO_2 emissions but also prevent the emission of toxic substances such as formaldehyde generation when chemical binders are used [17].

The utilization of hemicellulose as a binder and in other applications is an ongoing area of research. However, it has not yet been used for producing concrete along with recycled concrete. In this study, hemicellulose is used as a binder for green concrete development.

Based on this background, the present study focuses on producing concrete using hemicellulose to achieve higher strength with lower environmental impacts. For a better understanding, cellulose and lignin were also used in the study to compare their bending strengths with that of hemicellulose. The study also uses marine biomass as a substitute for hemicellulose.

In this study, seventeen cases using six types of materials as binders with ordinary cement concrete powder were prepared. To the best of our knowledge, this is the first attempt to investigate and produce green concrete using hemicellulose by pressing. Hence, this study aims to advance this line of research by developing concrete that is producible at room temperature with lower molding pressure to generate stronger non-cement concrete.

2. Materials and Methods

2.1. Materials

The raw materials used for the experiment in this study include recycled concrete powder, cellulose, lignin, hemicellulose, chitosan, and oyster shell, gathered from multiple sources. **Figures 1(a)-(g)** shows photographs of the materials used in the experiment.

Figure 1. Raw materials: (a) Recycled concrete powder; (b) Waste wood powder; (c) Hemicellulose; (d) Cellulose; (e) Lignin; (f) Oyster shell; (g) Chitosan.

The mix proportion of the waste concrete used for production is presented in **Table 1**.

Table 1. Recycled concrete mix proportion.

w/c	Water (kg/m^3)	Portland cement (kg/m^3)	Sand (kg/m^3)	Gravel (kg/m^3)
0.42	173	412	777	1038

To obtain a dried mass of the concrete powder, the powder was dried in an oven at 120˚C for 24 hours. Then, the dried concrete was kept in a zip-locked plastic bag until usage for the green concrete production.

For wood powder, Japanese cedar sawdust was utilized, which was dried in an oven at 96˚C for 24 hours in the laboratory and then sieved to obtain a powder with particles less than 0.6 mm. To unveil the individual impact of wood major components, hemicellulose, cellulose, and lignin were used. Oyster shell, a by-product of oyster, and chitosan, a biopolymer derived from chitin found in the exoskeletons of crustaceans such as shrimp and crabs, were also used in the experiment.

All the materials used in the experiment were zip-locked and stored before the experiment.

The samples were prepared through press molding using a hot-pressing machine. The pressing apparatus could simultaneously heat and apply pressure to the specimens from both sides attached to pressing discs.

The bending strength of the samples were measured through three-point loading.

2.2. Experimental Procedure

The production of the green concrete was conducted using the hot-pressing technique of the Botanical Concrete [4]. All the experimental parameters and ranges are shown in **Table 2**. As shown in the table, the experiment cases are divided into three groups, each with several cases. Each case used a different material as a binder and was pressed at three different temperature degrees. The first group used binders such as wood, cellulose, hemicellulose, lignin, chitosan, and oyster shell, individually with the recycled concrete powder.

Table 2. Materials and Experimental condition.

Cases	Materials (gr)								Pressing condition		
	RCP	W	L	C	H	Ch	O	Water	H (˚C)	P (MPa)	t (min)
RCP-W	12	6	-	-	-	-	-	1.5	24˚C, 60˚C, and 160˚C	25	10
RCP-L	12	-	6	-	-	-	-	1.5			
RCP-C	12	-	-	6	-	-	-	1.5			
RCP-H	12	-	-	-	6	-	-	1.5			
RCP-L-C-H	12	-	2	2	2	-	-	1.5			
RCP-Ch	12	-	-	-	-	6	-	1.5			
RCP-W-Ch	12	3	-	-	-	3	-	1.5			
RCP-L-Ch	12	-	3	-	-	3	-	1.5			
RCP-C-Ch	12	-	-	3	-	3	-	1.5			
RCP-H-Ch	12	-	-	-	3	3	-	1.5			
RCP-L-C-H-Ch	12	-	1	1	1	3	-	1.5			
RCP-O	12	-	-	-	-	-	6	1.5			
RCP-W-O	12	3	-	-	-	-	3	1.5			
RCP-L-O	12	-	3	-	-	-	3	1.5			
RCP-C-O	12	-	-	3	-	-	3	1.5			
RCP-H-O	12	-	-	-	3	-	3	1.5			
RCP-L-C-H-O	12	-	1	1	1	-	3	1.5			

RCP: recycled concrete powder, W: waste wood powder, L: lignin, C: cellulose, H: hemicellulose, Ch: chitosan, O, oyster shell, H: pressing heat, P: pressing pressure, t: pressing time.

The moisture content of the recycled concrete powder was controlled by adding water based on the dry weight of the powder, which was acquired by drying in an oven at 120˚C for 24 hours. Furthermore, the proportion of the concrete powder and the binder was based on the dried weight of the concrete and the dry weight of the targeted binder. The weights were measured using an electrical scale with 0.1g precision. The mix proportion was maintained at 2:1 (concrete powder:binder) along all the cases. For mixture, the concrete and water, with a quantity of 12.5% of the dried mass of the concrete, were mixed for 1 minute to obtain concrete with 12.5% moisture content. Then, the moist concrete powder and individual binder were mixed for another 1 minute. The mix proportion and mixing method was determined as the most effective after several trials. After that, the mix was placed in a mold and pressed at 25 MPa for 10 minutes. The experiment was conducted at three different temperatures: cold press (24˚C), warm press (60˚C), and hot press (160˚C) individually, to unveil the effect of temperature.

In the second and third groups, all the weighing and mixing procedures of the first group were followed. However, it is worth mentioning that, since these groups used two types of binders, first, the dry mass of both binders was mixed and then added to the moist concrete powder at once and mixed for 1 minute manually. The rest of the test followed the first group experimental procedure. Figure 2 illustrates the concrete molding procedure.

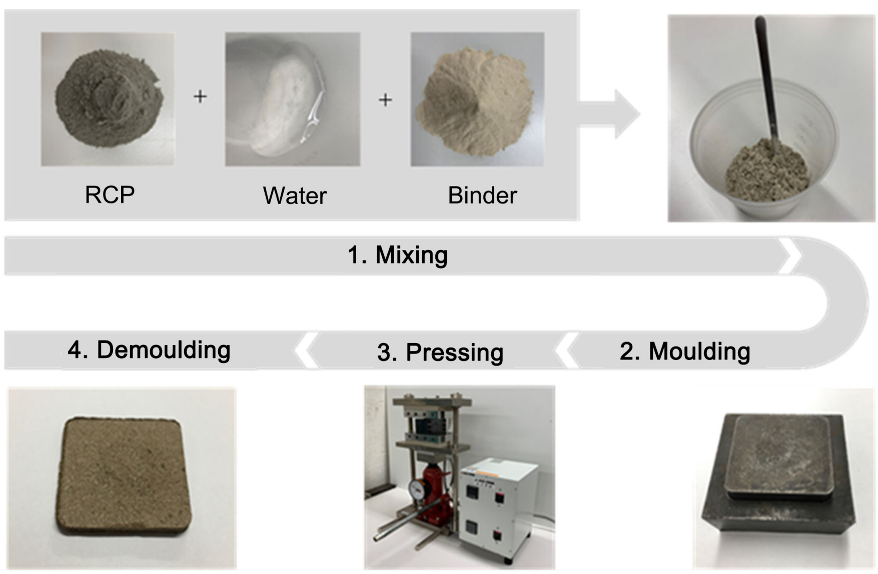

Figure 2. Concrete molding method.

Upon the end of pressing, the specimens were demolded, and the curing period started. Although the specimens were strong enough right after demolding, to obtain further strength, all the specimens were kept at room temperature (24˚C) for a 72-hour curing period prior to the bending strength test.

After 72 hours of curing, all the specimens' dimensions were measured and prepared for the bending test. The bending strength of the hardened body was evaluated by a three-point bending test with a 40 mm support distance. Additionally, a cement mortar specimen was made and cured for 72 hours to compare its bending strength with that of the green concrete. The dimensions of the green concrete specimens are 50 mm × 50 mm × 5 mm, and as a sample, the photographs of the specimens pressed at 60˚C are shown in **Figures 3(a)-(e)**.

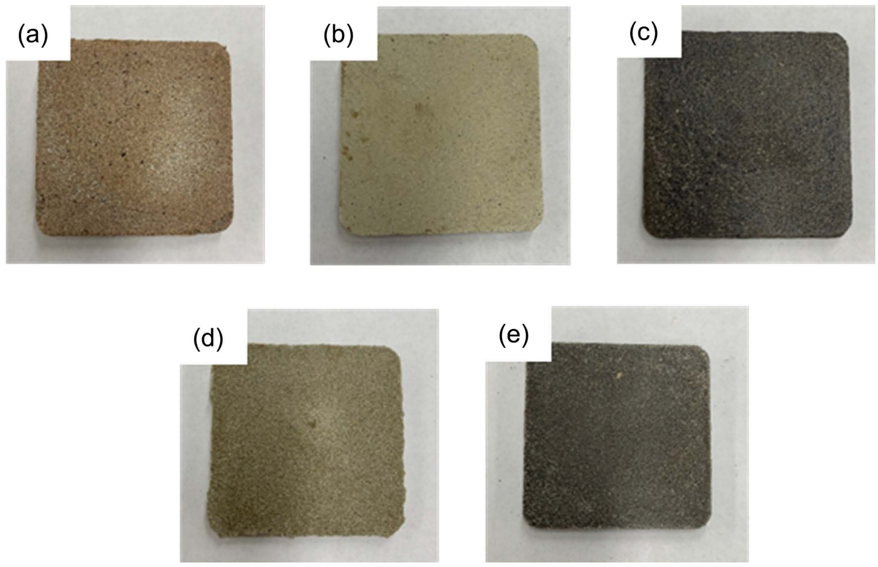

Figure 3. Green concrete using different binders pressed at 60˚C: (a) RCP-W; (b) RCP-L; (c) RCP-C; (d) RCP-H; (e) RCP-L-C-H.

3. Results and Discussions

Some of the green concrete specimens hot-pressed at 160˚C were delaminated when they were removed at the end of the hot pressing, with the manufacturing conditions shown in **Table 2**. At 160˚C, case RCP-W, which contained concrete and wood powder, was not delaminated, but those containing other binders, such as cellulose, hemicellulose, lignin, oyster shell, and chitosan, were delaminated. The reason may be as follows: these binders' glass transition in wet condition is considerably lower than 160˚C, and thus, at this temperature, they readily flow and do not create a strong bond unless they cool down lower to the glass transition temperature [18]. In addition, as the concrete was pressed at 25 MPa, making it difficult for the water to evaporate and escape during the press, high steam pressure is created inside the concrete, and when the pressure is higher than the internal bonding at removal, delamination occurs [18]. Thus, before opening the hot press, a cooling period is recommended to lower the temperature and allow the binder to create a strong bond [18]-[20].

The bending strength results of the delaminated specimens are reported here to highlight the effect of hot-pressing conditions on the concrete strength and properties and to consider a suitable pressing procedure.

3.1. Wood and Its Individual Components, Chitosan and Oyster Shell Effect on the Bending Strength

Figure 4 shows the bending strength of the green concrete made of recycled concrete powder and different bio-based additions, such as wood, hemicellulose, lignin, cellulose, chitosan, and oyster shell, which were measured after 72 hours of curing. As can be seen in the figure, the green concrete bending strength varies based on different binder materials. Additionally, the strength of each concrete with the same mixture and materials differs when exposed to different pressing temperatures. Since this study's experimental method and the green concrete hardening method follow the binderless particle boards production and solidification mechanism, in which pressing temperature is one of the most important manufacturing parameters to create internal bonds and increase bending strength [21], the concrete in case RCP-W pressed at room temperature did not harden, and therefore its strength was not measurable. A higher pressing temperature of 60˚C formed a hardened body with a 0.5 MPa bending strength, indicating the bonds created by the partial activation of lignin and hemicellulose because wood lignin and hemicellulose soften in a wide range of temperatures from 20˚C to 180˚C based on wet and dry conditions and different native wood [22]-[25]. At 160˚C press, a stronger hardened body with 2.7 MPa was obtained because at such a high temperature both the lignin and hemicellulose soften even in dry conditions [25], thus increasing the strength.

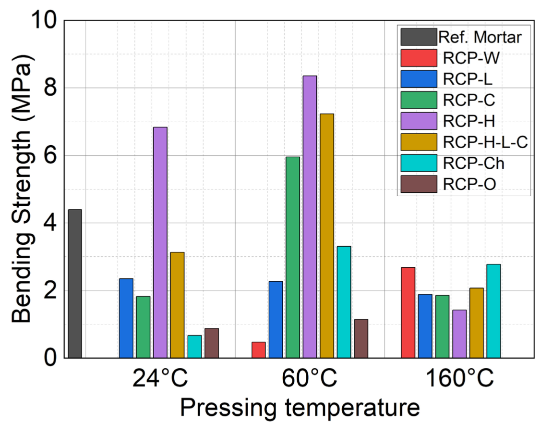

Figure 4. Bending strenght of green concrete made of different bio-based binders at three pressing temperatures.

As shown in **Figure 4**, the RCP-L solidified at all the pressing temperatures. At room temperature and 60˚C, it showed a strength higher than that of the RCP-W, but at 160˚C, its bending strength decreased due to a slight delamination phenomenon. However, the RCP-L bending strength is similar at all the pressing temperatures, 2.4 MPa, 2.3 MPa, and 1.9 MPa, respectively. The lower bending strength and delamination at a high temperature of 160˚C could be linked to the evaporation that may have occurred so quickly that the lignin dried before flowing and developing the strength [9]. It may indicate that the most strength of the RCP-W enhanced at high temperature is not due to lignin. Furthermore, lignin needs more time to soften than hemicellulose [9], and therefore at 10 minutes pressing, it could be inferred that mostly it was hemicellulose which softened and thus provided strength to the concrete in case RCP-W.

The green concrete containing cellulose (RCP-C) hardened at all the pressing temperatures. As demonstrated in **Figure 4**, at room temperature (24˚C), the bending strength is 1.8 MPa and further increased to 6 MPa when molded at 60˚C. However, further temperature elevation to 160˚C led to delamination and a reduction in strength to 1.9 MPa, comparable to the strength achieved at the room temperature pressing. In the same way, the concrete made of hemicellulose and the waste concrete (RCP-H) successfully hardened at all the pressing temperatures. As shown in **Figure 4**, the concrete pressed at room temperature exhibits 6.8 MPa, the highest bending strength among others, even higher than that of conventional cement mortar. It is approximately three times more than the bending strength of the RCP-L, because the bond strength provided by hemicellulose is stronger than that of lignin [9]. The bonding strength of the RCP-H increased as the temperature increased, and therefore the specimen molded at 60˚C showed 8.4 MPa bending strength, indicating a sudden increase in bending strength at this temperature. However, at 160˚C, the mix readily flowed during the hot press so that it was difficult to maintain it under 25 MPa pressure. At the end, the sudden downloading, neglecting a cooling period, caused severe delamination and as a result, the strength dropped to 1.4 MPa. It is very interesting to obtain high strength at lower temperatures to not only decrease environmental impacts but also relieve economic and manufacturing challenges.

To unveil the synergistic effect of the binders from wood components, in the case of RCP-L-C-H, an equal combination of hemicellulose, lignin, cellulose nanofiber, and waste concrete was used. As shown in **Figure 4**, the concrete hardened at all the pressing temperatures, but the highest strength was achieved at room temperature and 60˚C, respectively. Similar to the RCP-H and RCP-C cases, its bending strength increased from 3.1 MPa at room temperature to 7.2 MPa at 60˚C, but at 160˚C, delamination occurred, and the strength dropped to 2.1 MPa. It follows the bending strength pattern observed in the RCP-H case, which may imply the integration of hemicellulose with the other binders and its significant role in enhancing the concrete strength.

The green concrete containing chitosan and waste concrete (RCP-Ch) had a bending strength of 0.7 MPa at room temperature pressing, but it increased to 3.3 MPa at 60˚C, representing a significant enhancement. However, pressing at 160˚C led to concrete delamination, thus reducing the bending strength to 2.8 MPa.

The oyster shell and waste concrete (RCP-O) hardened at room temperature and 60˚C, but at 160˚C, severe delamination occurred, rendering its bending strength unmeasurable. As shown in **Figure 4**, at room temperature pressing, its bending strength was 0.9 MPa, slightly increasing to 1.1 MPa when the pressing temperature was raised to 60˚C.

3.2. Effect of Partial Substitution of Wood and Its Components with Chitosan, and Oyster Shell

Figures 5(a)-(e) depict the bending strength of the green concrete, incorporating chitosan and oyster shells as substitutes. As demonstrated in Figure 5(a), when 50% of the wood was replaced with chitosan in case RCP-W-Ch, the bending strength enhanced with a similar trend at all the pressing temperatures, indicating the creation of more and stronger cellulose-chitosan and hemicellulose-chitosan bonds. This enhancement aligns with previous reports indicating that chitosan improves the strength of wood-pulp webs [26] [27].

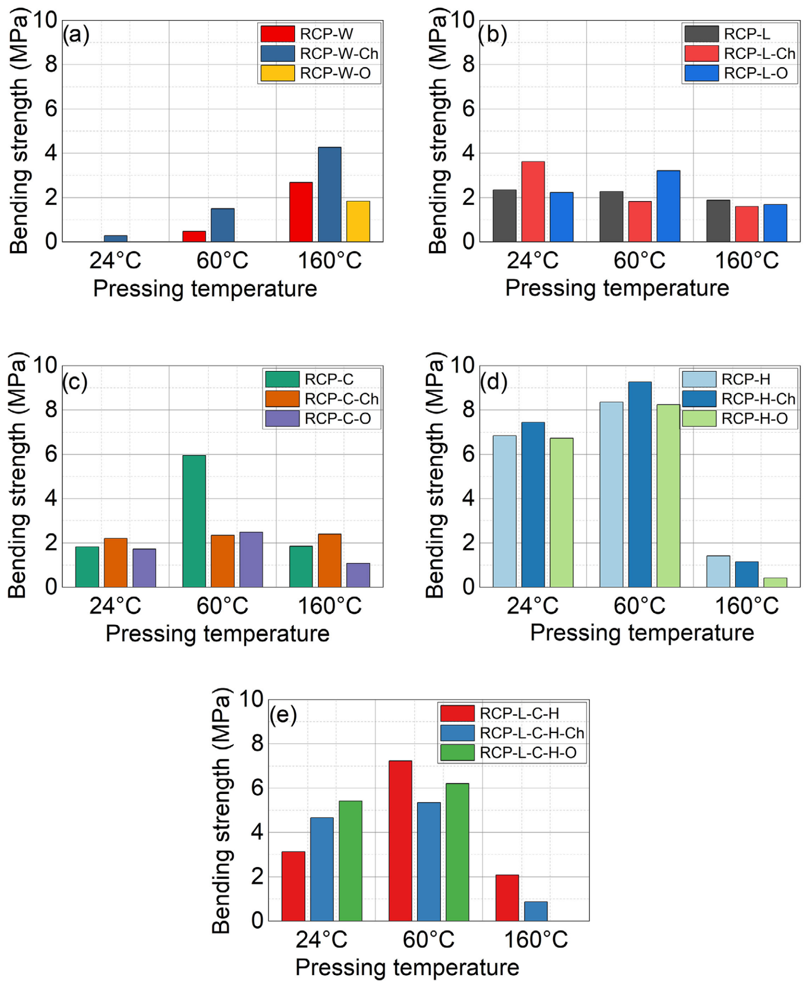

Figure 5. Bending strenght of the green concrete enhanced with chitosan and oyster shell: (a) RCP-W; (b) RCP-L; (c) RCP-C; (d) RCP-H; (e) RCP-L-C-H.

Oyster shells mainly consist of $CaCO_3$ [28], with a small amount of chitin [29]. Therefore, the effect of oyster shell powder addition could be expected to differ from that of sole chitosan. As shown in Figure 5(a), in case RCP-W-O, when oyster shell was substituted, a decline in strength was observed against all pressing temperatures with a similar trend, which may indicate the formation of fewer bonds due to the limited amount of chitin and chitosan present in the oyster shell.

Figure 5(b) demonstrates that the substitution of 50% lignin with chitosan in case RCP-L-Ch considerably improved the bending strength only at room temperature, indicating interactions due to hydrogen bonds between chitosan and lignin [30]. However, the strength was negatively affected at 60˚C and 160˚C. The highest strength was achieved at room temperature, 3.6 MPa. However, at 60˚C the strength reduced to 1.8 MPa, and at 160˚C the specimen slightly delaminated and thus the strength further reduced to 1.6 MPa. However, oyster shell replacement with 50% of the lignin mass in case RCP-L-O improved the strength of the concrete only at 60˚C press. At room temperature, it was 2.2 MPa and increased to 3.2 MPa at 60˚C, but at 160˚C due to delamination, the strength decreased to 1.7 MPa.

Substitution of cellulose with chitosan in case RCP-C-Ch slightly improved the concrete strength at room temperature and 160˚C but adversely affected at 60˚C pressing temperature, as illustrated in **Figure 5(c)**. This improvement indicates cellulose-chitosan interactions, which enhance mechanical strength [31]. In addition, the strength development could be related to the particle size of the cellulose, as it has been reported to increase specific surface area, ester and hydrogen bonds, and increase cross-linking degree between lignocellulose and chitosan [32]. However, replacement of cellulose with oyster shell in case RCP-C-O reduced the concrete strength across all pressing temperatures, same as the results of wood and lignin substitution with the oyster shell.

Figure 5(d) shows that in both room temperature and 60˚C pressing, in case RCP-H-Ch, the bending strength of the concrete increased when 50% hemicellulose was replaced with chitosan, indicating the formation of covalent imine bonds [26], and thus yielding a higher strength compared to sole hemicellulose [33]. However, neither the strength nor the delamination changed considerably at 160˚C. At room temperature, the concrete strength reached 7.5 MPa while it was 6.8 MPa in case RCP-H, when pure hemicellulose and the concrete powder were used. In the same way, at 60˚C, the bending strength reached 9.3 MPa while it was 8.4 MPa in case RCP-H, indicating a significant strength enhancement at 60˚C.

As with the other materials, in case of RCP-H-O, the substitution of oyster shell with hemicellulose slightly decreased the concrete bending strength across all pressing temperatures, indicating a negative effect of oyster shell replacement in terms of bending strength. However, further studies are needed to understand the wider impact of oyster shell on green concrete properties.

Figure 5(e) shows the bending strength of the green concretes in cases RCP-L-C-H-Ch and RCP-L-C-H-O, which used waste concrete and a combination of hemicellulose, cellulose, and lignin as 50% of the binder mixture, and 50% chitosan in one group and in another group 50% replacement with oyster shell, respectively. The figure shows that both chitosan and oyster shell substitutions enhanced the bending strength at room temperature molding but had a negative impact at 60˚C and 160˚C. Interestingly, oyster shell addition yielded a higher concrete strength than chitosan in this case, indicating an inverse result in terms of combined binder partial substitution compared to single binder partial substitution. However, delamination still occurred at 160˚C.

Among all the cases, those containing hemicellulose showed the highest bending strength, even higher than conventional cement mortar and the RCP-W case. This is because in case of raw wood approximately 60% of wood is lignin and hemicellulose [34] [35], which may not be evenly heated during hot pressing due to wood's low thermal conductivity, potentially limiting their softening and bonding potential. Therefore, the strength of the RCP-W is lower than that of the hemicellulose-based concrete. Additionally, the bonding strength of hemicellulose is more than two times higher than that of lignin, contributing to its superior bending strength [9], and therefore it yielded the highest bending strength among the other components of wood such as lignin and cellulose.

Producing hemicellulose-based concrete with high bending strength does not require heat, high pressure, or extended time, and it is producible at room temperature, showing sufficient strength right after demolding, though further strength increase requires curing.

Moreover, hemicellulose, abundantly available around the world, is currently underutilized for construction material production. If utilized, it can contribute to reducing a significant amount of biomass waste, including lignin and hemicellulose in black liquor from the pulp industry, currently incinerated for energy production, thus further emitting CO_2.

Furthermore, for marine biomass utilization, the study showed promise in using chitosan and oyster shells in green concrete. These environmentally friendly materials can be used without adverse effects. When used alone, considerable strength was not attained, but when combined with botanical components, comparable or even superior results were achieved.

Hemicellulose-based concrete shows promise for developing green concrete, not only reusing demolished concrete and its fine powder generated during concrete recycling but also effectively utilizing abundant biomass waste to contribute to resource conservation, waste management and recycling, and climate change mitigation.

It is worth mentioning that a comprehensive study of the concrete mechanical properties, durability, chemical interaction, microstructural changes, and its scalability and economic viability are still elusive and need further investigations.

4. Conclusions

In this study, an innovative approach was undertaken to reduce CO_2 emissions associated with cement production and utilize waste from concrete debris recycling, waste wood, and other biomasses by grinding and hot-pressing these materials to produce green concrete. This method enabled the preparation of hardened bodies with considerable strength. The study yielded the following conclusions:

- Hemicellulose has significant potential for producing green concrete and other construction materials. Its effectiveness in producing concrete with enhanced bending strength surpassed that of raw wood, lignin, cellulose, and even conventional cement mortar.
- The hemicellulose-based concrete pressed at room temperature exhibited high

bending strength, which further increased when pressed at 60˚C. However, increasing the pressing temperature to 160˚C led to delamination and a reduction in strength. Therefore, elevated temperatures during production were unnecessary, allowing for the fabrication of construction material under ambient conditions.

- The incorporation of marine biomass such as chitosan and oyster shell further increased the strength of the green concrete. Thus, their positive synergistic effect can contribute to the properties of green concrete and recycling efforts.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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