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Biochar addition for 3DCP: a preliminary study

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This contribution presents the first results of an ongoing research aimed at highlighting the possible reduction in the environmental impact of concrete through the synergy between two interconnected strategies: the exploitation of by-products, in this case biochar, for the realization of 3D printable cementitious conglomerates. Thanks to the use of biochar, the mixes presented are characterized by an excellent dimensional stability in the fresh state, evaluated through the extrusion test. Regarding the hardened state properties, the contribution highlights the effects of biochar-to-cement ratio, water-to-cement ratio (in combination with biochar content) and sand-to-cement ratio on the flexural and compressive strength of the mixes. The evaluation of CO₂ emissions shows that a proper mix design could result in a significant reduction in CO₂ emissions (up to 43%) while maintaining good mechanical performance (compressive strength of at least 60 MPa).

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Keywords: 3D concrete printing; biochar; compressive strength; CO₂ emissions.**1. Introduction**

In recent years, the building sector is characterized by a growing interest in the research for solutions that can help reduce environmental impact. This can be seen in the growing number of studies focused on reducing the environmental impact of the most widely used material in the construction field: concrete. There are several studies in the relevant literature that aim to use wastes and by-products in cementitious conglomerates as partial replacements for traditional aggregates or as filler additions: recycled PET (Beibei Xiong et al, 2021), (Mercante et al, 2018), nanoparticles obtained by pyrolysis of peanuts and hazelnut shells (Khushnood et al, 2016), (Restuccia et al, 2018), biochar (Restuccia et al, 2016a), demolition wastes (Restuccia et al, 2016b), (Rao et al, 2007). In any case,

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the effort to reduce environmental impact should be combined with satisfactory performance specifications for the proposed mixtures. In addition to this first strategy, another way to pursue the reduction of environmental impact could be to optimize raw materials by exploiting the use of advanced construction processes such as 3D printing. In fact, in this case, it is possible to realize particularly complex sections by depositing the material only where necessary, solving topological optimization problems (Martens et al, 2018). However, cementitious conglomerates for 3D printing applications must be characterized by dimensional stability (Falliano et al, 2020a), i.e., must be capable of maintaining their shape in the fresh state without the aid of formwork (Nerella et al, 2019). This characteristic is usually achieved through the use of appropriate chemical additives in the material design mix or by mixing accelerators at the print head (Muthukrishnan et al, 2021).

The present contribution is along this line of research. In fact, this contribution presents the first results of an ongoing research that aims to combine the two strategies of environmental impact reduction previously presented: the definition of 3D printable cementitious conglomerates obtained through the use of by-products, in particular biochar, the same used by the authors to improve the fracture energy of lightweight concretes (Falliano et al, 2020b). In fact, the incorporation of micro-sized particles gives rise to impediments along the development of cracks, thus improving the ductility and energy absorption capacity of cementitious composites (Ahmad et al, 2015). The effects of biochar-to-cement ratio, sand-to-cement ratio and water-to-cement ratio on the compressive and flexural strength are presented. In addition, the CO₂ emission of the presented mixtures is also evaluated and discussed.

2. Materials and testing conditions

Specimens are prepared using Portland cement CEM I 52.5R and tap water. The sand used was sieved to have a maximum diameter of 1 mm. As the goal is to make a cementitious conglomerate suitable for 3D printing applications, this choice allows to facilitate the extrusion of the material through the printing nozzles, usually characterized by dimensions between 15 and 50 mm. Three different sand-to-cement ratios, s/c in Table 1, are employed: 70%, 127% and 280%. The increase in the viscosity of the material was achieved through the addition of biochar. The biochar-to-cement ratios, b/c in Table 1, are: 5%, 9%, 11% and 23%. To limit the water-to-cement ratio as much as possible, a polycarboxylate ether superplasticizer was used. The superplasticizers-to-cement ratios, sp/c in Table 1, are: 0%, 5%, 6% and 10%. Lastly, the water-to-cement ratios, w/c in Table 1, are: 28%, 37%, 39% and 51%. The dimensional stability of the mixtures in the fresh state is evaluated through the extrusion test, discussed in (Falliano et al, 2020 c). Regarding the evaluation of properties in the hardened state, each of the series shown in Table 1 consists of 3 prismatic specimens characterized by dimensions equal to 40 mm x 40 mm x 160 mm, in accordance with UNI EN 196-1. Flexural and compressive strengths are evaluated using a CONTROLS test frame, model 65-L1301/FR, after 28 days of curing in air at 20±2 °C. In particular, the load rate for the flexural tests is equal to 50 N/s, while that of the compression tests is equal to 2500 N/s. For each test the peak load was recorded to evaluate the flexural strength and the compressive strength. Lastly, the CO₂ emissions of the presented mixes are also evaluated based on the amount of CO₂ emissions for each component, as explained in (Falliano et al, 2022).

Table 1. Mix proportions.

Series	b/c [%]	w/c [%]	sp/c [%]	s/c [%]
Mix 1	11	37	10	280
Mix 2	5	39	6	280
Mix 3	23	51	0	280
Mix 4	9	28	5	70
Mix 5	9	28	5	127

3. Results and discussion

All the series shown in Table 1 exhibited excellent dimensional stability in the fresh state. In fact, the samples exhibited neither initial slump values (instantaneous settlement), nor slump values accumulated subsequently (settlement after the end of the extrusion test). With respect to the hardened state properties, it is interesting to highlight the effects of biochar content, water content, and sand content on the flexural and compressive strengths. The effect of the biochar content is highlighted in the comparative histogram between the mixes with the highest aggregate-to-cement ratio, equal to 280%, shown in Fig. 1. A decrease in biochar percentage (from 11% to 5% of cement weight) results in an increase of approximately 17% and 15% in compressive and flexural strength values, respectively. Minor discrepancies in water-cement ratios (37% and 39%, respectively) are counterbalanced by a change in the superplasticizer content (10% and 6%, respectively). Therefore, doubling the amount of biochar leads to a small decrease in compressive strength.

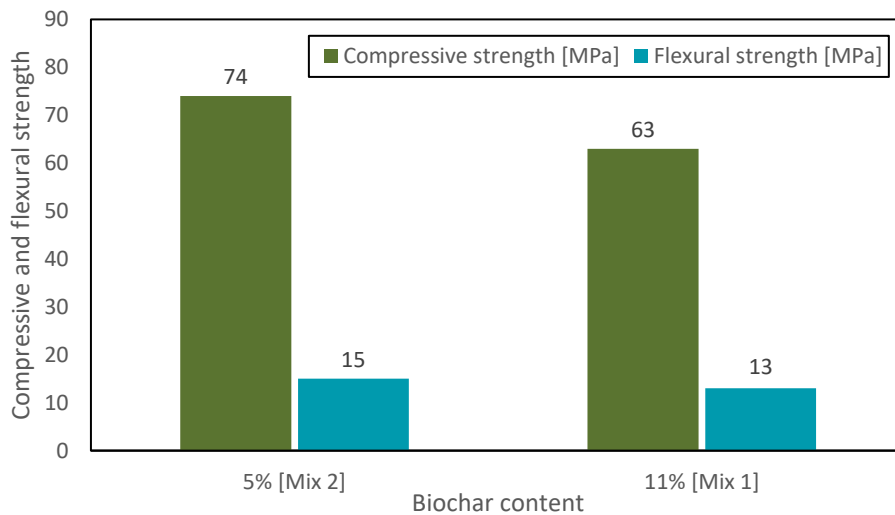


Fig. 1. Effect of biochar content on compressive and flexural strength.

Interestingly, a further increase of the biochar content from 11% to 23% with respect to the cement weight does not lead to a significant reduction of the strengths, which are found to be similar in the two different cases, as shown in Fig. 2. Looking at the other differences in terms of mix design between the two mixes compared in Fig. 2, it is possible to search for a justification for this experimental evidence.

First of all, it must be emphasized that, despite the high water-to-cement ratio, mix 3 also gave excellent results on dimensional stability in the fresh state. This is due to the water absorption property of biochar, the amount of which is more than doubled compared to mix 1. The high strength achieved, comparable to that obtained with a much lower water-to-cement ratio (37% in the first case, with a necessary addition of superplasticizer, and 51% in the second case, without superplasticizer) and, as mentioned, with a lower amount of biochar, could be due to the internal curing phenomenon promoted by biochar, thus improving cement hydration. This very interesting result, especially when applied to cementitious conglomerates to be processed through 3D printing, will be investigated more deeply, and presented in a forthcoming study.

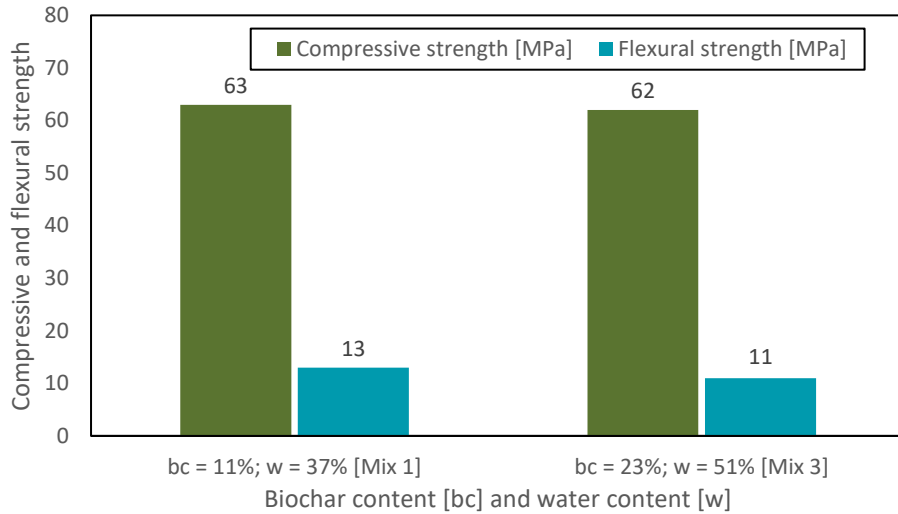


Fig. 2. Effect of biochar content and water content on compressive and flexural strength.

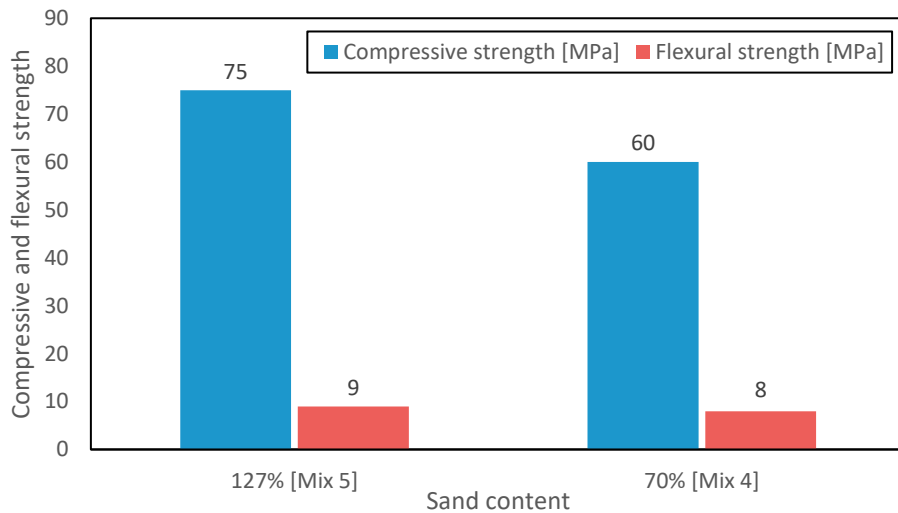


Fig. 3. Effect of sand content on compressive and flexural strength.

Coming to the effect of the sand-to-cement ratio, increasing the sand content from 70% to 127% of cement weight results in an increase of the mechanical strengths equal to 25% and of 12% for the compressive and flexural strength, respectively, Fig. 3. This evidence could be due to the higher aggregate interlock effect.

Lastly, representative mixes (mix 2, mix 3, and mix 5) are selected to highlight the influence of different biochar-to-cement ratio and aggregate-to-cement ratio on the carbon footprint of the presented cementitious conglomerates. The choice of these representative specimens is aimed to include different biochar contents (from low to high contents: 5%, 9%, and 23% with respect to cement weight) and different aggregate content (127% and 280% with respect to cement weight). Using the methodology illustrated in (Falliano et al, 2022), the following values of CO₂ emissions in kg per cubic meter of cementitious conglomerates are obtained: 433 [kg CO₂/m³] for mix 3; 498 [kg CO₂/m³] for mix 2; 750 [kg CO₂/m³] for mix 5. The largest contributor to CO₂ emissions is cement, which makes these results quite clear. Therefore, the optimization of the mix design achieves a reduction in CO₂ emission of about 43%, while maintaining compressive strengths of at least 60 MPa.

4. Conclusions

This contribution highlighted the first results of an ongoing research activity focused on 3D printable mortars characterized by the addition of biochar. In particular, the use of these special mortars makes it possible to reduce the environmental impact with a twofold strategy: the use of waste materials, biochar, and the use of cutting-edge technologies, 3DCP, allowed by the excellent dimensional stability at the green state of the mixes.

The results are summarized as follows:

- Biochar acts as a VEA.
- Compressive strengths are higher than 60 MPa, with a peak value of 75 MPa.
- Flexural strengths are higher than 8 MPa, with a peak value of 15 MPa.
- Due to the significant contribution of the cement in the assessment of CO₂ emissions, the mixes characterized by the highest biochar-to-cement ratio and sand-to-cement ratio turned out to be those associated with the lower CO₂ emissions.
- It is possible to reduce the CO₂ emission by up to 43% while maintaining very high mechanical strength.

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