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Suitability of biochar as supplementary cementitious material (SCM) or filler: waste revalorization, a critical review

Viabilidad del biochar como material cementante de reemplazo (MCR) o filler: revalorización de residuos, review crítica

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Abstract

For decades, researchers on materials science have highlighted the potential of biochar as a CO_2 adsorption medium and the possibility of its incorporation into other materials to reduce the overall carbon footprint. This present study is a critical review of a selection of articles about biochar potential as a material on the construction industry. Biochar is a promising material in order to mitigate GHG emissions when added to cementitious materials, reducing its carbon footprint through a dual effect: CO_2 sorption and replacement of cement or aggregates. Literature evidenced that replacement ratios of around 2-8 of cement wt% improved or leveled with conventional cementitious composites. However, some recent studies have shown that the incorporation of biochar up to >10% replacement ratios have the potential to improve the composites. Based on this premise, the present review emphasizes on the durability and long-term properties of biochar cementitious composites by providing up-to-date discussions of the studies on the matter and the future perspectives of the research in order to develop more eco-efficient concretes or mortars.

Biochar, Eco-efficient cement, Cementitious composites

Resumen

Durante décadas, investigadores en ciencia de materiales han destacado el potencial del biochar como un medio de absorción de CO_2 y la posibilidad de incorporación en otros materiales compuestos con el fin de reducir su huella de carbono. El presente trabajo es una review crítica compuesta de una selección de artículos enfocados en el potencial del biochar como un material en la industria de la construcción. El biochar es un material prometedor para reducir los gases de efecto invernadero cuando es incorporado a materiales cementantes, al reducir su huella de carbono a través de un efecto dual: absorción de CO_2 y el reemplazo de cemento o agregados. La literatura indica que tasas de reemplazo entre 2 a 8% en peso de cemento desarrollan cementantes con mejores o iguales propiedades a las de un cementante convencional. No obstante, estudios recientes destacan la posibilidad de reemplazar >10% de cemento y obtener compósitos con mejores propiedades. Con base en esta premisa, la presente investigación enfatiza las propiedades relacionadas a la durabilidad y largo plazo de compósitos base cemento con biochar, proporcionando discusiones actualizadas y perspectivas futuras de investigación con el objetivo de desarrollar concretos y morteros con mayor eco-eficiencia.

Biochar, Cementantes eco-eficientes, Compósitos cementantes

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1. Introduction

Concrete is the most abundant composite material available nowadays. Its constituents are mainly cement, water and aggregates -as fillers-, the former has the most important role overall: binding the mixture and providing the characteristic properties of hardened concrete -versatility, strength and durability-. Due to its crucial role in concrete mixtures, the carbon footprint associated with cement production has been extensively analyzed in several studies (Andrew, 2019; Busch *et al.*, 2022; IPCC, 2018; Liao *et al.*, 2022; Mahasenan *et al.*, 2003; Nidheesh & Kumar, 2019; Scrivener *et al.*, 2016), ranging from around 0.81kgCO_2-eq to 0.90kgCO_2-eq per kg of cement produced (Bellona Foundation, 2018; Plaza *et al.*, 2020; Samad & Shah, 2017; Shen *et al.*, 2016). Concerning anthropogenic greenhouse gas (GHG) emissions, cement production contributes to around 5-8% of global emissions owing to its energy intensive production process, mostly related to acquisition of raw materials, calcium carbonate calcination, fuel burning and logistics (Barcelo *et al.*, 2014; Miller *et al.*, 2021).

One of the foremost solutions that have been proposed in literature to mitigate the above-mentioned environmental impact is the use of supplementary cementitious materials (SCMs) (Siddique & Khan, 2011; Snellings, 2016). Conventional SCMs include blast furnace slag(Amran *et al.*, 2021; Lee *et al.*, 2019), fly ash (Y. Han *et al.*, 2022; Park *et al.*, 2021) and silica fume (Ibrahim, 2021; Tavares *et al.*, 2020), however, in recent years other candidates have been explored, such as: agro-industrial waste (Aprianti *et al.*, 2015; Chand, 2021; Manan *et al.*, 2021; Ramos *et al.*, 2022; Siddika *et al.*, 2021), livestock manure (Leng *et al.*, 2019; Rehman *et al.*, 2020), municipal solid waste (Caprai *et al.*, 2019; L. Chen, Wang, *et al.*, 2022; Ryłko-Polak *et al.*, 2022; Tome *et al.*, 2020), glass waste (Bueno *et al.*, 2020; Ibrahim, 2021), forest and vegetable crops and residue (Martirena & Monzó, 2018; Puga *et al.*, 2022; Restuccia *et al.*, 2020; Sirico *et al.*, 2020) among other organic wastes also known as biomass. The disposing of these wastes traditionally includes their revalorization as fertilizers, open-air landfilling or open-air incineration.

The aforementioned methods of disposal have revealed to significantly disrupt the subsoil and groundwater integrity through phenomena such as eutrophication and liberation of GHG emissions into the atmosphere (Abiriga et al., 2020; Speight, 2020). Parallel to this, biomass is a suitable material for thermal conversion processes such as pyrolysis for energy production, fuel production or element recovery (Fahimi *et al.*, 2020; Fiameni *et al.*, 2021; Leng *et al.*, 2019; Pandey *et al.*, 2022; Shashvatt *et al.*, 2018).

The residual ashes obtained after thermal conversion of biomasses are commonly outlined under the term biochar. Biochar is the pyrogenic residual waste obtained after the thermochemical conversion of a given biomass (Arif *et al.*, 2020; Bergman *et al.*, 2022; Vieira *et al.*, 2022). Biochar has four main production processes: pyrolysis, hydrothermal carbonization, gasification and torrefaction (Bartoli *et al.*, 2020; Roychand *et al.*, 2021). Moreover, literature indicates the characteristic calcination temperatures at which biochar is conventionally obtained ranging from 300° to 1000°C (Yaashikaa *et al.*, 2020). These temperatures and methods of thermal conversion heavily influence the properties and reactivity of the resultant biochar (Cosentino *et al.*, 2018; Tan *et al.*, 2020). Generally, these ashes are known to be carbon neutral or have negative impacts through energy production and, in some cases, even function as CO_2 sequestrating agents which result in further GHG emission abatements (Gupta *et al.*, 2018; Ibarrola *et al.*, 2012).

In view of the favorable environmental performance of biochar it has been proposed in a wide number of studies as an SCM (Jittin *et al.*, 2020), as an admixture (Akhtar & Sarmah, 2018; Gupta *et al.*, 2018), or as filler *(Cuthbertson et al.*, 2019) in cementitious mixtures in order to produce carbon-neutral composites by reducing their carbon footprint. Considered as an SCM, its abatement potential positions biochar at an advantage point amidst the rest of SCMs which, although possessing a smaller carbon footprint than cement, rarely exhibit abatements.

Furthermore, the advent of clean energies (Buonocore *et al.*, 2021) has shifted attention away from coal-burning production industry into more eco-efficient ways of obtaining energy and materials.

In this context, given its high availability, its prevalent chemical composition and environmentally troublesome disposal, pyrogenic residual ash -biochar- becomes a suitable construction material. Notwithstanding the prospective benefits of biochar as a replacement material in the construction industry, the major challenge for its adoption as a suitable replacement material for cementitious composites lies in its vast mutability and variability. Not only the way various biochars interact and react within a cementitious matrix is widely variable, but also the chemical composition and intrinsic properties differ greatly from biochar to biochar (Suarez-Riera *et al*., 2020). In consequence, in-depth analysis of the reactions and interactions that ensue from the addition or the replacement of cement with biochar is needed, in order to diminish the negative influence on performance and durability of cementitious binders.

Literature indicates that low percentages of substitution have been predominantly explored, whereas relatively high percentages of substitution are generally dismissed due to poor strength-related performance and durability concerns (Danish *et al*., 2021). However, in accordance with standards such as ASTM C618 and ASTM C311, the test procedures to determine pozzolanic activity require a 20% substitution (ASTM C311-18, 2018; ASTM C618-19, 2019). In addition, there is a paucity of data concerning the properties influencing long-term interactions between biochar properties and cementitious materials, indicating an important research gap.

In the present review paper, a critical review of the biochar merits and demerits on cementitious matrixes is outlined, state-of-the-art research from January 2017 to June 2022 concerning properties of biochar cementitious composites is examined from environmental, durability, and long-term performance viewpoints, key indicators of suitability of biochar as SCM are reviewed and future opportunities of research to develop appropriate biochar cementitious composites are discussed throughout the review; authors find it timely to conduct a systematic evaluation of recent advances, challenges, future outlooks and applications regarding biochar as a potential SCM or filler.

Thence, this is a unique review, providing up-to-date discussions on biochar as a functional material in the construction industry, its properties, characteristics, environmental performance, durability perception and long-term performance.

1.1 Selection criteria

There are plenty research works regarding biochar cementitious blends, however, not all of them provide the same level of in-depth discussion and their contributions on long-term performance, environmental performance and durability are limited. In addition, not all of them provide the same level of detail discussing residence time, temperature and thermochemical conversion method. The purpose of this review is to serve as a guide for researchers to assess the feasibility and potential applications of different biochar as supplementary cementitious material by exploring some challenges such as (1) which properties are affected by the replacement of cement with biochar; (2) the potential of biochar cementitious composites to improve ecoefficiency in contrast with regular blends; and (3) the long-term expected performance of composites containing biochar.

The systematic literature search was conducted in two electronic databases: ScienceDirect and Dimensions.ai. The search was limited to publications from January 2017 to June 2022. A combination of the following keywords in conjunction with the Boolean operators AND and OR were used to narrow the scope of the search: biochar, cementitious, carbon footprint, abatement, construction, material, building, SCM, replacement. A supplementary search was conducted by revising the references lists on the articles obtained.

The eligibility criteria was determined by: (i) published in full-text and in English, (ii) high priority was given to articles that discuss the properties of biochar blends in conjunction with their expected long-term and environmental performance, and (iii) articles that discuss the hydration kinetics of biochar blends; meanwhile, the exclusion criteria was determined as follows: (i) review or conference articles, (ii) articles merely focusing on biochar as is and not in cementitious composites, (iii) articles that provide no details on the thermochemical conversion process and the biomass feedstock.

(iv) articles which include properties or applications of biochar blends which are not suitable for the construction industry -like heavy metal removal or waste water treatment-. A total of 379 items were retrieved from the electronic databases ScienceDirect and Dimensions.ai, 329 and 50, respectively. Thereupon, 41 items were excluded during a preliminary screening due to duplicity, leaving 338 items for the next stage of screening which is based on the eligibility criteria described above, from which 293 items were excluded due to the insufficient information provided regarding thermochemical conversion process or properties covered and shallow discussion about environmental and durability relation or the lack thereof; a total of 45 items were selected for in-depth full-text revision. Full-text revision refined the total of items that adhered to the criteria, resulting in a total of 20 items fit for the present review. The selected items are presented in Table 1.

Article	Proposed use of biochar	Biomass feedstock
(Gupta & Kua, 2018)	Admixture	Mixed wood sawdust
(Dixit *et al*., 2019)	Cement replacement	Mixed wood sawdust
(Gupta, Krishnan, *et al*., 2020)	Cement replacement	Wood wastes and coconut shells
(Gupta, Palansooriya, *et al*., 2020)	Admixture	Sorghum, dairy manure, cotton stalk, mixed wood waste, Vermont biochar, Wakefield biochar, Hoffman biochar
(Tang *et al*., 2020)	Cement replacement	Municipal solid waste
(X. Chen *et al*., 2020)	Cement replacement	Municipal sludge
(Gupta, Muthukrishnan, *et al*., 2021)	Cement replacement	Rice husk and mixed wood sawdust
(Gupta, Kashani, *et al*., 2021)	Cement replacement	Peanut shell
(X. Yang & Wang, 2021a)	Cement replacement	Rice husk
(X. Yang & Wang, 2021b)	Cement replacement	Commercial biochar (Korean)
(Praneeth *et al*., 2021)	Sand replacement	Enhanced poultry litter
(Maljaee *et al*., 2021)	Cement replacement	Olive stone, rice husk and forest residues
(Dixit *et al*., 2021)	e (UHPC)	Marine clay and wood waste
(Sikora *et al*., 2022)	Cement replacement	Wood chip (commercial/Polish)
(X. Han *et al*., 2022)	Binder (AAS)	Residue of pine and cedar
(Castillo *et al*., 2022)	Cement replacement	Poultry litter
(F. Wu *et al*., 2022)	Admixture	Miscanthus (commercial/Dutch)
(Kanwal, Khushnood, Khaliq, *et al*., 2022)	Admixture	Bagasse sugarcane
(Kanwal, Khushnood, Shahid, *et al*., 2022)	Admixture	Bagasse sugarcane
(L. Chen, Zhang, *et al*., 2022)	Admixture	Waste wood

Table 1 Proposed use of biochar in cementitious composites, and biomass feedstock

2. Biochar as a functional material

Biochar is the pyrogenic residue of biomass thermochemical conversion processes where oxygen conditions are limited, during which water evaporation occurs first, followed by the release of volatiles and finally the setting of porous-carbonaceous ashes remains. In the broadest sense, biochar is a charcoal-like solid material typically composed of carbon (C), of porous-carbonaceous nature and low bulk density. From a structural point of view, biochar and activated carbon are similar materials given their porous nature and amorphous carbon content, however, their surface functional groups differ. There are several key factors of the thermochemical process governing biochar properties: temperature range, heat rate, biomass-feedstock, pretreatment or refinement, residence time and pressure (W. J. Liu *et al.*, 2015).

Considering its origin, biochar is a highly available material worldwide thereby reducing stress on the supply chain and exploitation of raw materials. Once obtained biochar needs no special processing and can be utilized directly, nonetheless, pretreatments and refinements might prove useful for targeted applications (Shanmugam *et al.*, 2022). Even when considering a pretreatment or refinement process biochar is still considered a low-cost byproduct.

Biochar has long been considered a suitable material for carbon sequestration aimed at reducing GHG emissions, as a matter of fact, the Intergovernmental Panel on Climate Change (IPCC) considers biochar as one of the six methods proposed to permanently mitigate carbon emissions on account of its carbon fixing capabilities -acting as a carbon sink- and negative carbon footprint (IPCC, 2018; Neogi *et al.*, 2022).

A noteworthy benefit of replacing cement with biochar in cementitious composites could be defined as ecoefficiency-strengthening; namely, a dual-purpose effect: on one side, the potential to strengthen the composite while decreasing its carbon footprint, on the other, fixing carbon on its microstructure for decades-long storage (Yin *et al.*, 2021).

On the same note, certain biochar properties are of interest for alternative cementitious composites development, such as density, surface area, electrical resistivity, porosity, particle shape, hydrophilicity, and surface functional groups, among others. These properties have a direct influence on mechanical behavior, workability, durability and adsorptive efficiency of composites containing biochar.

In regards to characterization techniques for biochar composites, the present paper focuses mainly on those relevant to cementitious composites development and application; several comprehensive reviews and research titles on biochar extensive characterization -which falls beyond the aims of the present review- have been already published. For the full-frame characterization -including mechanical behavior- of biochar as is, the reader is referred to the comprehensive research papers of Yaashikaa *et al.* (Yaashikaa *et al.*, 2020), Shanmugam *et al.*(Shanmugam *et al.*, 2022), and the references contained therein.

3. Physical and chemical properties related to durability and long-term performance of biochar composites

3.1 Physical and morphological properties

The replacement of Portland cement with biochar is expected to yield composites with lower densities and diminished compressive strength, mainly due to the porous nature of biochar. These reductions in density and strength impact heavily on the expected durability of the composites altogether. Due to the high temperatures set during thermochemical conversion process of biomass into biochar many physical and morphological properties are defined during this stage of production.

Physical and chemical properties tend to be proxies of durability of the overall composite and provide information about its expected long-term performance. The present section reviews some relevant physical and morphological properties related to cementitious materials and their relation with durability and long-term performance of cement-based composites.

3.1.1 Porosity, particle shape, surface area and density

Porosity, surface area and density are some of the most important physical properties of any material with cementitious applications -SCMs, admixtures or fillers-, as these define their contribution to the potential features and performance of cementitious composites.

Given its porous nature, biochar usually presents a low bulk density and high absorption capabilities, these properties carry over to cementitious composites containing biochar resulting in blends with higher porosity, higher absorption rates and a complex pore network. This transfer of properties has been the major concern for researchers seeking to develop more durable cementitious composites containing biochar, as these properties affect durability and resistance directly; workability and setting times are also affected.

Some authors (Gupta & Kua, 2018; Mrad & Chehab, 2019) consider the high porosity of biochar as beneficial and conceive it as an internal curing agent, consistently supplying water to the composite through further hydration. It differs from conventional water curing methods where water is spread through the surfaces, limiting its effectiveness due to its shallow penetration, whereas biochar acts as a water retention agent inside the matrix. In biochar, porosity owes its variability to the escaping of volatile matter during thermochemical conversion and, like with other properties, the rising of temperature promotes an increase in porosity and surface area.

Dixit *et al.* (Dixit *et al.*, 2019) found that the increase in size of biochar -fine, medium and coarse- also influences the size and overall availability of pores. Greater pore sizes serve as bridging points between micropores and mesopores, increasing interconnectivity and allowing water absorption and retention to increase as well.

Other carbon-based materials like graphene and nano-sheets have shown shape-dependent properties and features (Tatrari *et al.*, 2021; Yoo *et al.*, 2019).

Notwithstanding, in the case of biochar, a study by Suárez-Riera *et al*. (Suarez-Riera *et al*., 2022) investigated the influence of biochar shape on the mechanical performance of cement-based composites and based on their results concluded that sphere-shaped, rod-shaped or sheet-shaped biochar have no significant influence on the mechanical behavior or even workability, unlike graphene, nanotubes or nanosheets. Nonetheless, the addition of biochar -regardless of the particle shape- improved compressive strength, flexural strength and fracture energy more than 20% as compared to reference.

Density is interrelated with porosity, as porosity values increase density values decrease; likewise, when particle size decrease surface area experiences an increase. Yang & Wang (X. Yang & Wang, 2021a) and Praneeth *et al*. (Praneeth *et al*., 2021) reported that an increase in biochar proportion is strongly correlated with a decrease in the density of the resultant composite. By itself, a reduction in bulk or skeletal density is not a deleterious effect, however, it suggests the potential applications of biochar composites as lightweight energy-efficient composites.

Achieving denser biochar composites is possible up to a certain proportion of biochar addition/replacement -which is highly dependent on biomass feedstock and the calcination process-, according to Akhtar & Sarmah (Akhtar & Sarmah, 2018), if this proportion is exceeded then the hydration products are insufficient to fill the pores of the biochar and eventually lead to a more porous and brittle composite instead. Low density composites tend to exhibit low strength, affecting its durability and long-term performance.

Surface area is an important property to evaluate as it influences hydration products growth, workability, and is positively correlated with density and porosity. Surface area has direct incidence on adsorptive efficiency -carbon fixing-. High calcination temperatures and biomass feedstock influence porosity, surface area and contaminant fixing capacity.

The results obtained by the research of Castillo *et al*. (Castillo *et al*., 2022) indicates that higher biochar substitution proportions can yield positive results in mechanical performance: at 90 days testing every proportion of substitution -10, 15 and 20 wt%- achieved higher strength, from 10% to over 32% increases; in contrast with what several other investigations have reported suggesting lower substitution proportions (Qin *et al*., 2021; Rodier *et al*., 2017); this is mainly attributed to the high calcination temperatures -600°C to 800°C- related to other investigations, since this increase in temperature is positively correlated to an increase in porosity and surface area, thus improving properties like water absorption, reactivity and bonding/bridging in the cementitious matrix.

3.1.2 Transport properties

In cementitious composites several transport properties are important, namely: water absorption, permeability, thermal conductivity, electrical resistivity, sulfate resistance and chloride diffusion.

Biochar presents a complicated pore network -regardless of biomass feedstock-, which contributes to diffusion effects and entrapment of contaminants. Its low thermal conductivity and high porosity allow it to be considered as an insulating agent. Greater porosity leads to a decrease in thermal conductivity -biochar as is possesses a low thermal conductivity-, turning it into a better thermal insulation agent (Tan *et al*., 2020). This is an important property when developing lightweight composites or concretes.

In the investigation of Cuthbertson *et al*. (Cuthbertson *et al*., 2019), thermal conductivity and acoustic properties were evaluated in concretes with biochar addition. Since thermal conductivity and acoustic properties -sound absorption- are associated with porosity it is expected that biochars have the potential to act as thermal insulators and sound dissipators. The authors reported a linear decrease in density of concretes in relation with higher biochar addition, however, substitutions up to 12% were identified to be optimal for maintaining integrity and exhibit good sound and thermal insulation. These results are directly translated into energy savings due to thermal-efficiency of buildings employing these biochar-amended concretes.

Hydrophilicity -or water absorption and retention- is another important property worth evaluating in biochar cementitious composites. The findings of Praneeth *et al.* (Praneeth *et al.*, 2021) indicate a correlation between biochar content and water absorption and void content on composites. However, some other authors (Maljaee *et al.*, 2021) reported a decrease in capillary absorption, this effect can be attributed to the filler effect -which is linked to particle size- filling the pores of composites with hydrated products blocking to some degree the superficial penetration of water.

The work of Gupta & Kua (Gupta & Kua, 2018) evaluated the effect of pre-soaked biochar acting as water reservoirs -internal curing agent- in cement mortar It was found that pre-soaked biochar improved the degree of hydration, proving the hypothesized gradual release of trapped water for internal curing even when the external conditions stopped supplying water.

Free chloride ions are a major threat to reinforced concrete due to the de-passivation effect, leading to corrosion and the formation of solids on the surface of the reinforcement. This volume increase derives in internal stresses and the eventual cracking of the concrete layer. Phases of cement like C_3A and C_4AF are known to interact chemically with free chloride ions.

In the light of it, one of the most concerning deleterious phenomena associated with chloride ingress is the conversion of AFm phases to Friedel’s salt and the C-S-H dissolution due to this ion exchange in a NaCl rich environment eventually leading to diminished mechanical and durability performances (Glasser *et al.*, 2005). Gupta *et al.* (Gupta, Muthukrishnan, *et al.*, 2021) evaluated sulfate resistance and chloride diffusion in biochar cementitious composites, crucial transport properties for durability.

The findings of the study suggest that biochar addition helps to preserve strength in an 8-11% proportion even after 120 days of exposure to a NaCl-medium. In the case of sulfates expansion experiences a reduction in the order of 60-68% in contrast with the references. Both cases correspond to a 1-2 wt% mixed wood and rice husk biochar substitution.

As specimens age the free water content decreases and the hydration products fill some of the available pores leading to an increase on the electrical resistivity property, as evidenced by the study of Yang & Wang (X. Yang & Wang, 2021a) in which the electrical resistivities of all specimens increased with time and the more replacement proportion an even greater electrical resistivity was achieved. In contrast with the reference specimen, 2 and 5 wt% replacement derived in a 4.4% and 13.8% increase at 28 days. In the same note, a study carried out by Ram *et al.* (Ram *et al.*, 2022) identified the effect of interconnectivity of capillary pores on chloride transportation and identified that due to the formation of secondary hydration products the pores are clogged and the interconnectivity reduced, thus a decrease in chloride diffusion is expected.

3.1.3 Workability

The works of Cuthbertson *et al.* (Cuthbertson *et al.*, 2019) and Maljaee *et al.* (Maljaee *et al.*, 2021) indicate that manual mixing could derive in poor homogenization due to the agglomeration tendency present in biochars; coupled with its hydrophilicity, the setting times and flow test are affected in a deleterious manner since biochar tends to have a high absorption capacity depriving the paste of water during mixing. The studies carried out by Tan *et al.* (Tan *et al.*, 2020) and Chen *et al.* (X. Chen *et al.*, 2020) also reported a reduction in fluidity of around 3% for 1% addition to 36% for 10% addition and an inverse relation between water absorption and fluidity of fresh paste, respectively.

In both studies a common trait is identified: the finer biochar particles present a greater reduction in workability as compared to coarse biochar particles. In all the previously mentioned studies biochar addition or substitution has demonstrated to have a greater influence on workability, setting times and flow of fresh pastes which ultimately leads to a higher w/b ratio, pre-soaking of biochar or the use of superplasticizer to compensate this loss of fluidity.

3.1.4 Filler effect

While pozzolanic activity refers to chemical reactivity through $Ca(OH)_2$ consumption (M. Wu *et al.*, 2021), latent hydraulic activity refers to materials with high contents of CaO and possibly SiO_2 self-reacting with water to form C-S-H gel or C-A-S-H compounds *(Sivakumar et al.*, 2021). Both types of reaction develop more hydration products -even at older ages- which is beneficial to the cement paste.

On its part, filler effect is characterized by being inert and non-reactive on its own; nevertheless, filler effect favors the development of a more densified microstructure through two mechanisms: firstly, the filling of interstitial spaces between cement grains, thus promoting densification; and secondly, an increase in the surface area available for the nucleation of hydration products and C-S-H gel. Normally, filler effect is more evident during the early stages of hydration, but its effects have been confirmed in extended periods of study as long as 120 days (Gupta, Palansooriya, *et al.*, 2020). Filler effect can coexist with either pozzolanic or latent hydraulic behavior, in fact, most SCMs present filler effect at some point regardless of the nature of the material reaction -either pozzolanic or latent hydraulic-.

Several studies have considered biochar as inert fillers or in conjunction with other SCM's, as well as evaluated its pozzolanic potential by $Ca(OH)_2$ consumption (Asadi Zeidabadi *et al.*, 2018; L. Chen, Wang, *et al.*, 2022; L. Chen, Zhang, *et al.*, 2022; Gupta, Kashani, *et al.*, 2021; Gupta, Muthukrishnan, *et al.*, 2021). Biochar is able to present pozzolanic potential or latent hydraulic activity depending upon its original biomass feedstock, chemical composition, amorphous phases and surface functional groups, whereas, its filler potential is determined merely by its fineness and the agglomeration of the biochar particles in the fresh paste.

3.2 Chemical properties

Evaluation of chemical properties of biochar are of vital importance to determine the overall performance of composites, due to these properties directly influencing the reactivity, functionality, and ultimately, potential application of biochar in cementitious composites.

3.2.1 Elemental and chemical composition

The main determinant factors of biochar chemical composition are the biomass feedstock composition itself and the pyrolysis conditions (Vassilev *et al.*, 2013). The basis of chemical properties are stability and reactivity. Furthermore, the pozzolanic reaction or hydraulic reaction are dominated by the content of $SiO_2+Al_2O_3+Fe_2O_3$ and CaO oxides, respectively. In materials, stability is conceived as the condition of materials to not react with the environment during conventional use and preserve its properties during its expected service time. Spokas (Spokas, 2010) identified a relation of O/C molar ratio to reliably predict stability in biochar, based on their results a ratio of < 0.2 are the most stable, with an estimated service time of 1000 years; whereas, a ratio between 0.2 and 0.6 offers more variability, oscillating between 100 and 1000 years of service life; lastly, ratios > 0.6 can preserve properties for over 100 years. O/C ratio also denotes the polarity and hydrophilicity of biochar (Jiang *et al.*, 2019). H/C ratio has been linked with aromaticity of carbon-based materials, is subject to variability due to thermochemical temperature and is inversely related with carbon content -to a low H/C ratio corresponds a high carbon content- (Sumaraj *et al.*, 2020). These two ratios area closely related.

Thereby, the determination of biochar elemental composition is a top priority when considering its application on the construction industry, given the importance of stability of any component of cementitious matrixes in order to prevent deleterious reactions like alkali-silica reaction (ASRs) or alkali-carbonate reactions (ACRs), which can lead to cracking and fracture over the span of several years of service (Adams & Ideker, 2020; Leemann *et al.*, 2022). For elemental composition characterization the most used techniques are ICP-OES, ICP-MS and CHNS analysis, extremely useful to determine the ratio of heavy metals in biochar composition. On the other hand, XRF is the most used characterization technique to determine the oxides in a material as a ratio of mass.

In the selected studies several biomass parent feedstocks were identified: Mixed wood wastes (MWW), coconut shells (CS), dairy manure (DM), sorghum (SOR), cotton stalk (CST), algae (AL), municipal solid waste (MWS), municipal sludge (MS), rice husk (RH), peanut shell (PS), commercial biochar (B), poultry litter (PL), olive stone (OS), marine clay (MC) and bagasse sugarcane (BS), among others; certainly, the variability of chemical compositions is enormous.

Biochar	O/C	H/C
MWW300 (Gupta & Kua, 2018)	0.41	--
MWW500 (Gupta & Kua, 2018)	0.08	--
MWW500 (Dixit *et al*., 2019)	--	0.03
CS500 (Gupta, Krishnan, *et al*., 2020)	0.27	0.05
MWW500 (Gupta, Krishnan, *et al*., 2020)	1.27	0.05
SOR500 (Gupta, Palansooriya, *et al*., 2020)	0.28	0.06
SOR600 (Gupta, Palansooriya, *et al*., 2020)	0.21	0.05
AL500 (Gupta, Palansooriya, *et al*., 2020)	0.46	0.05
CST210 (Gupta, Palansooriya, *et al*., 2020)	1.23	0.12
CST250 (Gupta, Palansooriya, *et al*., 2020)	1.04	0.11
CST290 (Gupta, Palansooriya, *et al*., 2020)	1.00	0.12
DM600 (Gupta, Palansooriya, *et al*., 2020)	0.88	0.08
MS300 (X. Chen *et al*., 2020)	1.22	0.13
MS500 (X. Chen *et al*., 2020)	1.96	0.05
MS600 (X. Chen *et al*., 2020)	1.92	0.01
RH (Gupta, Muthukrishnan, *et al*., 2021)	1.37	0.05
MWW (Gupta, Muthukrishnan, *et al*., 2021)	0.21	0.03
PS (Gupta, Kashani, *et al*., 2021)	0.64	0.05
PL (Praneeth *et al*., 2021)	1.70	0.05
OS (Maljaee *et al*., 2021)	0.27	0.03
MWW (Maljaee *et al*., 2021)	0.25	0.04
RH (Maljaee *et al*., 2021)	0.82	0.05
MWW (Dixit *et al*., 2021)	--	0.04

Table 2 O/C and H/C ratios for different biochar in selected studies

Evidently, as can be seen on Table 2, even sharing the same parent biomass feedstock no two biochars are the same, this is attributed to differences in pyrolysis process, pretreatments or even geographic availability, Certain biochars like RH and PL share the same trend -regardless of parent biomass-, in relation with SiO_2 and CaO ratios, respectively.

Literature has evidenced the higher the thermochemical conversion temperature the more carbon content, and thus a lower O/C ratio, indicating a more reliable biochar. Moreover, the highest H/C ratios correspond to the lowest temperatures, indicating a less aromatic biochar, in other words, less stable carbon in biochar.

Increasing thermochemical conversion temperature has been proven to decrease O/C and this is also evidenced by an adsorption mechanism shift from cation exchange (CEC) to physisorption, which affects its adsorption capabilities; however, this effect is moderately ameliorated due to the increase in surface area associated with elevated temperatures (Enders *et al*., 2012; Rafiq *et al*., 2016). Biochar yield is also associated with O/C and H/C ratios; the higher the temperature of thermochemical conversion the less biochar yield is obtained, thence, while O/C and H/C ratios decrease so does yield proportion.

3.2.2 Pozzolanic reaction

Pozzolanic reactivity is evaluated by $Ca(OH)_2$ -calcium hydroxide- consumption, the most widespread tests to measure this reactivity are the Frattiini test (BS EN 196(5)), the modified Chapelle test and the pozzolanic activity index (ASTM C311); one noteworthy addition is the R3 method to determine reactivity of SCMs (Al-Shmaisani *et al*., 2022; Avet *et al*., 2016, 2022; Blotevogel *et al*., 2020). When the hydration product $Ca(OH)_2$ reacts with the amorphous silica of the SCM in the matrix of cementitious materials to produce C-S-H or C-A-S-H, it can be established that a pozzolanic reaction is taking place. The amorphous phase of silica is of utmost importance since non-soluble silica would not react with calcium hydroxide, which is the case of quartz -high content of crystallized SiO_2-, considered an inert filler.

Tang *et al*. (Tang *et al*., 2020) carried out a study in which the pozzolanic potential of municipal solid waste biochar was evaluated through the calcium hydroxide consumption test -the modified Chapelle test-, in comparison with other commonly used SCMs -Fly ash and GGBFS-. Their results suggest that MSWI biochar is less reactive than FA and GGBFS and confirmed the late age filler effect of biochar due to a poor mechanical performance during the early ages but a comparable strength after 90 days.

On a similar note, Liu *et al.* (W. Liu *et al.*, 2022) proposed bamboo biochar as SCM, having an SiO_2 of >44%, an Al_2O_3 of 15% and Fe_2O_3 of >9%, which equal a sum of >68%, shying away from the 70% requirement established by the ASTM C618 standard for a Class N pozzolan. Moreover, semi-quantitative analysis was performed on the XRD spectrum obtaining the ratio of crystalline phases vs. non-crystalline phases, 59.87% and 44.10% respectively, which directly influences the reactivity of bamboo biochar. In this study, replacement ratios of 0.2, 0.4, 1, 2, 3 and 4 wt% were evaluated and provided excellent performance in terms of strength with an increase for all replacement ratios in comparison with reference.

3.2.3 pH

Serviceability of cementitious composites is affected by numerous parameters such as strength, density, corrosion, carbonation, environment, humidity, chemical exposure, among others. When SCMs, fillers and admixtures are considered the hydration process and the internal reactions tend to become very complicated, due to the modified reactions between C-S-H gels, available $Ca(OH)_2$ and the properties of the material added to the mixture. pH is a useful parameter to determine the durability of cementitious composites through its key role in deleterious phenomena such as carbonation and steel corrosion; an acidic environment facilitates ion exchange and decalcification of hydration gels, thus cementitious composites are prone to corrosion, alkali-silica reaction (ASR) and carbonation (H. J. Yang *et al.*, 2021; Zhang *et al.*, 2021). Therefore, pH of biochar is an important property to evaluate biochar-cementitious matrix compatibility and long-term performance prediction agent. Low pH values tend to develop retarding effects and decalcification -detrimental effects for cement hydration-, on the other hand, high pH values are associated with a proper strength development and reinforcement steel protection in composites (Kochova *et al.*, 2017). Table 3 presents a recollection of the biochar pH values from the selected studies and their respective calcination temperature.

Reference	Biomass feedstock	Temperature (°C)	pH
(Gupta, Palansooriya, *et al.*, 2020)	Sorghum	500	7.43
	Sorghum	600	9.62
	Dairy manure	600	9.84
	Cotton stalk	210	5.82
	Cotton stalk	250	5.87
	Cotton stalk	290	6.33
	Mixed wood waste	500	10.14
	Algae	500	10.24
	Vermont biochar	--	9.61
	Agricultural biochar	--	9.36
	Horticultural biochar	--	9.61
(X. Chen *et al.*, 2020)	Municipal sludge	300, 400, 500, 600	7.38
(Gupta, Kashani, *et al.*, 2021)	Peanut shell	500	8.17
(X. Han *et al.*, 2022)	Pine and cedar	760	10.3
(F. Wu *et al.*, 2022)	Miscanthus	250	5.2

Table 3 Biochar feedstock, thermochemical conversion temperature and pH in selected studies

As is the case with other properties, temperature is a defining factor for pH values in biochar, Table 3 corroborates that higher temperatures of thermochemical conversion are positively correlated with higher pH values; however, most of pH values of biochars in the selected studies are above 7, indicating a mildly alkaline addition or substitution.

Some authors evaluated the pH values of composites instead of biochars. Dixit *et al.* (Dixit *et al.*, 2019) evaluated the pH of cement pastes with biochar replacement at 2-8 cement wt% proportions and their results suggest negative significance between biochar substitution and alkalinity of the overall cementitious matrix; reference paste exhibited a pH value of 12.80 while 2, 5 and 8% substitution showed values of 12.75, 12.60 and 12.10, respectively. Moreover, Chen *et al.* (X. Chen *et al.*, 2020) also evaluated the correlation between biochar pH and the biochar cementitious blend pH, based on their results no significant correlation was identified, biochar pH (7.38) showed negligible effect on modified cementitious pastes, exhibiting pH values in the order of 8.03-10.47 with replacement ratios of 2-4 cement wt%.

The study of Gupta *et al.* (Gupta, Kashani, *et al.*, 2021) corroborated the negligible effect of biochar addition or replacement in cementitious composites when the replacement ratios are low (< 3 wt%), regardless of the pH value of biochar as is. Wu *et a*l. (F. Wu *et al*., 2022) obtained pH values for Miscanthus biochar which exhibited marginally acid values: 5.1-6.3, whereas the pH values of biochar cement pastes were alkaline: 11.4-12.5 with addition of biochar at ratios of 1, 1.5 and 2.0 vt%. Meng *et al*. (Meng *et al*., 2021) carried out a study to determine the effects of different pretreatments for raw biomass feedstock on the properties of yielded biochar.

3.3 Mechanical properties

Replacement ratio played a determinant role in the development of mechanical properties of biochar cementitious composites. In a study using sludge biochar, low proportions (< 8%) of replacement or addition volumes biochar presented an internal curing effect and improved the compressive strength in magnitudes of 5% up to 29% (X. Chen *et al*., 2020). Some other findings follow a different trend: regardless of the type of parent biomass compressive strength decreased with a corresponding increase in biochar replacement (Maljaee *et al*., 2021). Nevertheless, recent findings with PL used as parent biomass feedstock, indicate a higher substitution potential -as high as 20%- and still develop a composite with equal or improved properties in comparison with reference cementitious blends (Castillo *et al*., 2022). In the aforementioned study, three ages were evaluated for compressive strength: 7, 28 and 90 days; in the first 7 days all substitution ratios showed a higher strength than reference, at 28 days reference blend surpassed the 20% replacement blend by a hefty 40%, while marginally surpassing the 10% replacement blend; lastly, at 90 days every replacement ratio surpassed reference and the 10% replacement blend obtained the best results overall.

4. Discussion and opportunities for future research

Biochar has proven to possess a great number of properties such as low skeletal and bulk density, contaminant adsorption, hydrophilicity, among others; these properties make it an attractive SCM or filler, for different applications in the construction industry.

Although several studies highlight the deleterious effects on strength development and densification of cementitious composites containing biochar, its carbon-sink properties and low density could be approached from a different viewpoint. The findings of Cuthbertson *et al.* (2019) are promising for biochar composites as heat and sound insulators. X. Han *et al.* (2022) proposed biochar as amendment agent for alkali-activated slags to further improve coral sands in soils. Some other findings support high substitution levels with an equal or higher strength development capability (Castillo *et al*., 2022; Ofori-Boadu *et al*., 2021), positioning biochar as a potential SCM or filler, with either pozzolanic or hydraulic potential. Biochar has also been proposed as a key component of pervious concrete (Qin *et al*., 2021; Tan *et al*., 2022; Xie *et al*., 2021), while some authors have proposed pyrolyzed char as bacteria carriers in autogenous-healing concretes or cement-based materials (Kanwal, Khushnood, Khaliq, *et al*., 2022; Kanwal, Khushnood, Shahid, *et al*., 2022).

Another positive outlook of biochar incorporation to cementitious composites is the reduction of the overall carbon footprint, as several studies have shown (Guo *et al*., 2022; Igalavithana *et al*., 2020; Tan *et al*., 2022).

The main findings of the present review were as follows:

- Biochar dosage and parent biomass feedstock played a critical role in the overall properties and strength development of the composite.
- Dosages of over 10-20% demonstrated to increase compressive and flexural strengths.
- Incorporation of biochar reduced the composite density; however, it acted as an internal curing agent.
- Dosage 2-8% cement replacement enhanced water absorption, increased capillary porosity and decreased water penetration due to saturation.
- Biochar incorporation to cementitious blends resulted in an abatement of GHG emisiions for 59 to 65 kg CO_2-eq for each tonne of produced composite.

The abatement potential of biochar containing composites -carbon footprint reduction- needs to be examined at length in future studies, due to the enormous variability existent between biochar from different parent biomass.

Greater focus should be placed on developing studies based on the durability issues related to biochar-mended composites mentioned in the selected studies in the present review.

To the best of our knowledge there are no studies which statistically correlate the thermochemical conversion parameters such as temperature, heating rate and residence time with certain properties of biochar related to durability in cementitious composites such as elemental/chemical composition, O/C and H/C ratios, pH, density, porosity, surface area and mechanical properties to establish the statistical significance of these properties in relation to long-term expectation of biochar cementitious composites in terms of durability and as carbon sinks.

Notwithstanding the lack of research about this particular subject, it can be inferred that biochar-amended composites pose as promising materials for the development of novel cement-based materials, such as pervious concrete, water purification, heavy metal removal, soil stabilization, bacteria carriers, heat and sound insulator, among others; while simultaneously acting as carbon footprint reduction agents. Assuredly, further investigations are needed to provide more precision and certainty over biochar-containing composites performance.

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