



RESEARCH ARTICLE

STRATEGIES FOR OPTIMIZING THE BIOCHAR CONTENT OF WOOD CHIPS, PEANUT SHELLS, AND RICE HUSKS IN CEMENTITIOUS MATERIALS

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Abstract

The characteristic GHG emissions from cement production, primarily due to decarbonation and fossil fuel consumption, are deeply concerning. Reducing the clinker content in cement is the fastest way to lower the carbon footprint, particularly through the addition of biochar. This promising approach is the subject of several studies. However, numerous factors come into play, ranging from the type of biomass used to the curing of the material. This is, in fact, the source of the diversity in results and the resulting contradictions. The common thread among these studies remains the negative impact of high biochar content. This review article aims to demonstrate the importance of certain pyrolysis parameters on the properties of biochars, but above all to highlight the levers that could enable an increase in biochar content in cementitious materials while preserving mechanical and durability properties. In the literature, the beneficial effect of biochars on mechanical properties, dry shrinkage, crack resistance, reduced water absorption, etc., is only significant when the biochar content is less than or equal to 1%. The porous nature of biochar, which is both beneficial and detrimental, limits the amount of biochar that can be added to or substituted for cement. However, alternatives such as pretreatment and accelerated carbonation hardening allow for additions of up to 5% or even 10%. This review summarizes the individual factors (biochar fineness, pretreatment, accelerated carbonation, etc.) that facilitate biochar incorporation. Unlike existing studies that treat these factors separately, we recommend a standardized protocol that combines them to achieve a high biochar content without compromising strength and durability.

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Introduction:-

By far the most widely used construction material in the world for the past century [1]. The amount of concrete poured annually worldwide is approximately 7.4 billion tons [2]. Although extremely durable, it has a very high carbon footprint due to the materials it contains [1]. Among these, one stands out due to its considerable impact compared to the others: cement, which accounts for 74–81% of concrete's total CO₂ emissions [3]. The cement industry is responsible for 5–7% of anthropogenic CO₂ emissions and ranks 4th in terms of CO₂ emissions into the atmosphere, behind oil, coal, and natural gas [3]. In 2023, annual cement and clinker production is estimated at 4.1 and 3.8 billion tons, respectively [4]. Given increasing urbanization, rising demand remains evident, and has been approximately 30% per decade since 2016 [3]. Clinker production is considered highly “energy-intensive,” also requiring a significant amount of nonrenewable natural resources, namely limestone and clay, which are dwindling over the years. The production of 1 ton of ordinary Portland clinker requires approximately 1.22 tons of limestone and 0.31 tons of clay [5]. The chemical reactions involved in the decarbonation of limestone generate 0.525 t of CO₂; when combined with thermal energy emissions, this results in 0.82 t of CO₂ per ton of clinker [6]. In cement production, numerous artificial (slag, fly ash) or natural (limestone, pozzolans, etc.) materials with a low carbon footprint are used as additives to clinker to address and improve the durability properties of the cements containing them and reduce their carbon footprint.

In this effort to maximize the replacement of clinker with low-carbon cementitious materials, one of the administrative and regulatory barriers lies in standardization, particularly at the European level. Indeed, the reference “product” standard EN 197-1 [7] is based on the composition of common cements and allows only a limited number of constituents in their composition [7]. Furthermore, the composition ranges are strictly defined according to the type of material and with a tolerance of +/-2.5%, which limits their use. These limitations hinder the optimization of cementitious materials and thus the reduction of cement's carbon footprint. This is why an approach based on demonstrating the performance of cements would help overcome these obstacles. This is the focus of the RILEM TC-312 PHC technical committee [8]. Excessive concentrations of CO₂ in the atmosphere contribute to global warming. If no action is taken, the risk of catastrophic climate change will be much higher and irreversible. As a result, strategies are being implemented by industry stakeholders, including reducing the clinker content in cement by using other additives that are much less carbon-neutral [9], or require less energy to produce. To this end, several conventional materials have been used for decades, but they remain insufficient given the urgent need to decarbonize the sector. Faced with this pressing need for a paradigm shift, a particular material has been attracting attention for several decades and is the subject of numerous studies as a construction material: biochar. Its use in construction addresses a dual concern: biomass waste management in West Africa and the reduction of GHGs. Biomass waste management remains a major challenge we face.

Beyond taking up space, it releases GHGs (CO₂ and CH₄) into the atmosphere when burned or left exposed to the open air [10]. Numerous studies have been conducted on the use of raw biomass waste, particularly palm kernel shells [11], wood waste [12], rice straw stalks [13], rice husks [14], and couch grass straw [15] in construction. However, the incorporation of biomass into cementitious materials disrupts hydration reactions, leading to delayed setting and consequently a loss of material strength [16]. Converting biomass into biochar is an effective way to chemically stabilize the material and contribute to the strength and durability of cementitious materials. According to Gupta et al., biochar's three properties: chemical stability, low thermal conductivity, and low flammability make it suitable for use as a construction material [17]. Thanks to its porous structure, biochar has water retention capacity; water is released over time, contributing to hydration and increased strength [18]. Daniel Suarez-Riera et al. [19] demonstrate a 13% and 30% increase, respectively, in compressive and flexural strength at 28 days for a 2% biochar addition compared to the control [19].

Most applications of biochar in cementitious materials use a low biochar content, rarely exceeding 5%. The overall objective of our study is to enable greater incorporation of rice husk, wood chip, and peanut shell biochars into cementitious materials. To this end, we will first review studies on biochars in general, and specifically on these three types of biochars. Next, we will identify the various strategies found in the literature that allow for optimizing the biochar content in cementitious materials without compromising their strength and durability. The purpose of my work is to demonstrate methods for increasing the typical biochar content incorporated into cementitious materials while maintaining the materials' properties intact. To visualize the structure of the research field on the incorporation of biochar from peanut shells, wood chips, and rice husks into cementitious materials, a bibliometric analysis based on keyword co-occurrence was performed using the VOSviewer software. Figure 1 shows the resulting map.

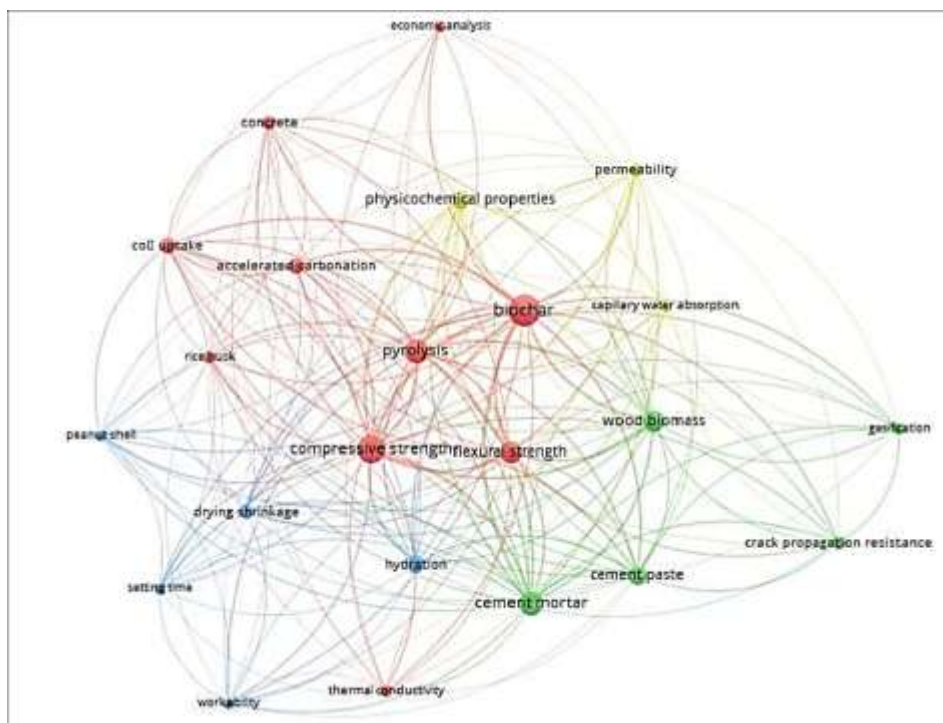


Fig.1. VO Sviewer mapping of keyword co-occurrences

Biochar:-

Known for thousands of years[20] for its use as a soil amendment, charcoal is now referred to as biochar. Some of these applications were primarily aimed at conserving or restoring degraded ecosystems, with the added benefit of carbon sequestration [21] [22]. In fact, it is considered by the IPCC to be a carbon storage solution [10]. Biochar is a porous, carbon-rich material containing oxygen-containing functional groups and aromatic surfaces [23]. It is produced through the thermochemical conversion of biodegradable materials under specific conditions. Several types of waste materials, such as algal and food waste, agricultural residues, biodegradable forestry and municipal waste, animal waste, and urban activated sludge, are considered primary resources for this purpose[22][24]. Their thermal conversion generally yields three byproducts: biochar, synthesis gas, and bio-oil in varying proportions.

Biochar Production Methods:-

Biochar production involves various methods, each with its own advantages and limitations [25]. The choice of production method is primarily guided by several material-related factors, such as the type of biomass[26], moisture content, desired biochar properties, and available resources, which influence the choice of method[25]. Among these methods, we will focus primarily on three.

Pyrolysis:-

Pyrolysis is the most commonly used process [27] [28]. It is the best method for producing biochar with significant yields [29]. When lignocellulosic biomass is used, hemicelluloses decompose first at temperatures between 197 and 257°C, followed by cellulose between 237 and 347°C, and finally lignin, which is pyrolyzed between 277 and 497°C[30]. Pyrolysis parameters differ depending on whether or not one wishes to maximize biochar yields. In fact, this is what has allowed pyrolysis to be classified into two main categories: slow pyrolysis and fast pyrolysis. Pyrolysis can be considered slow or fast depending on temperature, heating rate, residence time, and pressure[31].

Slow pyrolysis:-

Slow pyrolysis has been used for thousands of years, primarily for the production of charcoal [20]. Characterized by a low heating rate and a long residence time [32], it promotes the production of biochar. During biomass pyrolysis, the pyrolysis temperature range is typically 400-500°C, with a heating rate of 0.1 to 1°C/s over 5 to 30 minutes[32]. The values of these parameters are not fixed; they may vary from one study to another. In the literature, biochars obtained by slow pyrolysis have a cation exchange capacity (CEC) and ash content that are much higher than those

obtained by fast pyrolysis [33]. The increase in CEC and ash content are related because acidic functional groups are reduced[34].

Rapid pyrolysis:-

Unlike slow pyrolysis, fast pyrolysis maximizes bio-oil production. Typically, fast pyrolysis processes yield 60–65% bio-oil, 15–25% (10–30% [35] charcoal, and 10–20% non-condensable gases, depending on the feedstock used [20]. It is carried out at temperatures ranging from 600–1000°C, at a heating rate of 10–200°C/s [20]. The increase in temperature can significantly increase the pore volume and specific surface area of biochars [36]. The essential characteristics of fast pyrolysis for obtaining bio-oil are [37]: High heating and heat rates at the reaction interface; Very short residence time < 2 s.

Gasification:-

Gasification is one of the methods for converting biomass that prioritizes the production of synthesis gases (H₂, CO₂, CH₄, CO, N₂) over biochar[24] [38]. It is a complex process, limited by the choice of feedstock [39]. It involves the following four steps: drying, pyrolysis, combustion/oxidation, and reduction [40]. The composition of synthesis gases produced by biomass gasification depends primarily on the biomass, the type of reactor, and the gasification agent [41]. Gasification occurs at high temperatures (>700°C) in the presence of gasification agents (air, oxygen-enriched air, water vapor, carbon dioxide) [42]. When air is used as the gasification agent, N₂ is the primary product (41.6–61.6%) (vol%), while steam and oxygen combined produce a large proportion of CO (approximately 42.5–52%) (vol%) [43]. The biochar produced has a low yield of 5–10% with increased porosity, due to the release of volatile matter influenced by the rise in temperature [39]. Daniel Suarez-Riera et al.[44] studied the effect of two types of biochar on the mechanical properties of cement pastes and mortar. The results showed that gasified biochar has a higher water retention capacity than pyrolyzed biochar, due to its high internal porosity [44].

Hydrothermal carbonization (HTC):-

HTC, also known as wet pyrolysis [29], is a process for converting wet biomass (sewage sludge, algae, kitchen waste, etc.) into a material called “hydrochar” [24]. In this process, biomass with a high water content is introduced into a closed reactor containing liquid, at a temperature range (180–250°C) and under a water vapor pressure determined by the temperature [45]. Depending on whether the processing temperature is < 250°C, [250-400°C], or > 400°C, the resulting products are, respectively, biochar (hydrothermal carbonization), bio-oil (hydrothermal liquefaction), and gaseous products (thermal gasification) [31]. HTC involves four steps: (i) hydrolysis of biomass into monomers, (ii) pH reduction, (iii) polymerization through dehydration of the monomers, and (iv) aromatization [39].

Table1: Summary of the 3 biochar production processes

Criteria	Pyrolysis		Gazification	Carbonization hydrothermal
	slow	rapid		
Biomass	dry		dry	wet
Température (°C)	300-500	600-1000	>700	<250
Atmosphère	Inert		Air, O ₂ , vapor, CO ₂	Liquid
Main produit	biochar	oil	syngas	hydrochar
Application of biochar in cement	Common	Not recommended	rare	rare

Effect of pyrolysis source and parameters on biochar properties:-

Effect of raw material composition:-

The waste materials used for biochar production are diverse but can generally be classified into two subgroups based on their composition. These are: lignocellulosic biomass and non-lignocellulosic biomass. In this section, we will specifically discuss the impact of the composition of lignocellulosic biomass on the properties of biochar. Lignocellulosic biomass consists primarily of three polymers (lignin, cellulose, and hemicellulose) in varying proportions depending on the biomass source. Cellulose and hemicellulose are responsible for volatile products, while lignin primarily produces biochar and a small amount of water during pyrolysis [46]. The proportion of

hemicellulose, cellulose, and lignin in the biomass determines the amount of carbon stabilized in the biochar [23]. Indeed, each component has a decomposition temperature that allows for the formation of stable aromatic carbon. However, the high lignin content of the raw materials is proportional to the formation of aromatic carbon, which is positively correlated with the stability of the biochar [39]. When lignocellulosic biomass is used for biochar production, a high carbon content is observed, unlike with non-lignocellulosic biomass [47]. The study by Xin Chen et al. shows a carbon content of 8.9% in biochar derived from municipal sludge pyrolyzed at 500°C for 1 hour [48]. In contrast, a study conducted at the same pyrolysis temperature and residence time on lignocellulosic biomass shows a significantly higher carbon content, reaching 87.13% [49]. According to the study by Souradeep Gupta et al. (2020) on the incorporation of lignocellulosic and non-lignocellulosic biochars into cement mortar, the type of biomass also affects the mechanical strength of the added cementitious materials [50]. Wood biochar prepared at 500 °C resulted in a slight 7% increase in 28-day strength compared to the control, while non-lignocellulosic algae biochar reduced the mortar's strength by 15% [50]. For a comprehensive analysis of the effect of biomass composition on biochar, IR and NMR spectroscopy analyses are necessary and help highlight the thermal decomposition of cellulose and lignin, as well as the formation of aromatic structures [51].

Effect of pyrolysis parameters:-

The temperature:-

The pyrolysis temperature plays a key role in determining the properties of biochar. As the temperature rises, volatile organic compounds are released, leaving voids in the material that form an open-pore network with a high specific surface area. As the temperature continues to rise, the specific surface area also increases until it reaches an optimum, after which it decreases. The surface area can improve water retention capacity and, to some extent, the potential for biochar to interact with its environment [25]. The pores in biochar serve as water storage sites and, at times, as nucleation sites for hydrated products. When the temperature is [500-700°C], volatile matter decreases drastically[52] and the carbon skeleton becomes more robust.

The degree of decomposition of cellulose, hemicellulose, and lignin influences the extent to which the physical structure of biomass changes during pyrolysis [17]. Cellulose, hemicellulose, and lignin decompose at different temperatures [17]. This is why an ATG/DTG thermal analysis of raw materials is essential to determine the appropriate pyrolysis temperature and associated mass losses. Leandro Rodriguez Ortiz et al. studied the influence of temperature on the volatiles in walnut shells and kernels [53]. Respective reductions from 37.35±5.30% to 31.94±1.27% and from 59.19±0.96% to 50.03±1.22% for kernel and shell biochars are observed when the temperature increases from 400 to 600°C[53]. The modification of biochar's structural properties under the influence of temperature affects the fresh and hardened properties of the cementitious materials to which they are added. While the nature of the biochar influences mechanical strength, temperature also plays a significant role. In their study on the addition of rice straw biochar to cement paste, Xiaoyu Jiang et al. show that compressive strength at 28 days' increases with temperature and that a 32.5% increase in mechanical strength is achieved with a 1% addition at 500°C[54]. Above 500°C, strength tends to decrease, although it remains higher than the control [54]. Xin Chen et al. confirm in their study that temperature affects mechanical strength at 28 days and go further by stating that increasing the temperature accelerates setting time and improves the degree of hydration[48]. These results show a 5.8% increase in 28-day strength compared to the control when municipal sludge biochar is produced at 500°C and then substituted for 2% of the cement[48]. Previous studies are insufficient to definitively determine the impact of temperature on the properties of cementitious materials, but they unanimously show that a pyrolysis temperature of 500°C yields the best results regardless of the biomass used.

Heating rate:-

The heating rate is a parameter that influences the nature and composition of the charcoal to some extent [32]. Depending on whether the heating rate is low or high, the biochar yield changes. A low heating rate ensures that no thermal cracking of the biomass occurs, and consequently the biochar yield is high [32]. Whereas a high heating rate leads to the depolymerization of the biomass into volatile compounds[55] resulting in increased porosity. The work of Sevgi Sensöz and Dilek Angin on the pyrolysis of safflower seed cake shows biochar yields of 34.2%, 30%, and 29.7% when the heating rate is 10°C/min, 30°C/min, and 50°C/min, respectively [55]. There is no significant effect of heating rate, despite the decrease in yield. However, a significant difference is observed when the temperature increases from 400°C to 600°C, with yields ranging from 34.2% to 26.3% at a heating rate of 10% [55]. The authors conclude that, in reality, the pyrolysis temperature is the primary factor influencing yield, and that the combination of low temperature and a slow heating rate maximizes biochar yield.

Residence time:-

Increasing residence time promotes the repolymerization of biomass components by giving them sufficient time to react [32], thereby increasing the degree of carbonization of the biochar. Longer residence times reduce the vapors produced during secondary reactions, resulting in a higher proportion of charcoal [17]. Mouhamed et al and Tsai et al observed that residence time does not significantly affect biochar yield, but rather the composition of bio-oil and gaseous products [56] [57]. Studies on the effect of residence time on yield generally show that residence time alone is insignificant and that other associated parameters such as temperature and heating rate have a much greater influence on biochar yield and certain biochar characteristics [58].

Use of biochars in materials:-**Biochar-cementitious composite:-**

The use of biochar in construction materials dates back decades. Several studies have been conducted, and the results are promising in many cases. In the literature, biochar is incorporated into cement not only to reduce its carbon footprint but also to improve the mechanical, thermal, and durability properties of cementitious materials. While there are many studies, they remain limited in scope and rarely lead to practical implementation. In 2013, the Ithaca Institute took action and decided to construct its first building using a biochar-based plaster composed of 50% biochar, 30% clay, and 20% sand [59]. Biochar is also used in other building materials, notably lightweight biochar-concrete panels, biochar-lime bricks, and insulating plasters for interiors (biochar-clay) and exteriors (biochar-cement or lime) [60]. Their properties are enhanced and are undergoing extensive performance testing. While biochar incorporated into cement reduces its carbon footprint, it also possesses essential properties that make it unique. Preliminary results from the institute confirm this, demonstrating good insulation and excellent humidity regulation in the building. A study by Gupta et al. reveals another aspect of biochar, as a coating for polypropylene (PP) fibers reinforced in concrete [61]. The results were satisfactory, with a 28-day flexural strength of 14 MPa for the biochar-coated polypropylene (PP) fiber-reinforced concrete, compared to 11 MPa for the uncoated PP fiber-reinforced concrete [61].

The importance of biochar is recognized by the cement manufacturer Vicat, which has taken the initiative to innovate by adding biochar to clinker to produce a new carbon-negative binder, named “Liant Carat” in July 2022. The biochar from the company Soler used in this process is capable of sequestering 2.9 tons of CO₂ per ton of biochar [62]. The Carat binder underwent several performance tests and was used for the first time in the construction of the structural walls for a two-story office building project by the Corbioli company in Ambérieu-en-Bugey [62]. Large-scale testing of the binder’s performance was conducted as part of a project in which 850 m³ of Carat-based concrete was used to construct the building’s floors [63]. In the absence of established standards, the use of biochar as a cement additive continues, and progress is being made. Its impact on cementitious materials varies depending on several parameters, which must be understood to ensure its widespread use and, ultimately, standardization. The influence of biochar on cement, followed by an analysis of the intrinsic properties of cement-biochar composites, is detailed in Part 3.

Biochar-asphalt composite:-

The incorporation of biochar into asphalt concrete stems from its chemical composition, which is equivalent to that of carbon-based materials historically used as asphalt additives [64]. Indeed, these carbon-based materials, due to their high tensile strength, can increase the crack resistance of asphalt pavements [64]. Carbon fiber and carbon black are commonly used due to their availability [65]. Because of its high carbon content, biochar has the advantage of being included in this range of carbon-based materials used in pavements, with the aim of improving material performance. In their study, Zhao et al. [65] reported that biochar showed significant results in terms of the rheological properties and crack resistance of asphalt mixtures, unlike carbon fiber and carbon black [65]. An addition of up to 10% biochar exhibited resistance similar to that of the control [65]. Chebil, S. et al [41] also used biochar as a bitumen modifier for pavement applications. Among these results, we observe an increase in bitumen viscosity with the addition of 15% biochar, and higher rutting resistance after adding 5% biochar under multiple loading cycles (>100) [17] [41]. In summary, biochar is an excellent modifier for bituminous binders; not only does it improve performance under certain conditions, but it also sequesters carbon in pavements.

Biochar-polymer composite:-

Carbon-based materials (carbon black, graphene, biochar, etc.) used as fillers in polymer composites, in addition to having good interfacial adhesion [38], have the ability to improve the mechanical, electrical, and thermal properties of these composites [66]. Consequently, numerous studies on the incorporation of biochar into resin composites, rubber composites, and even plastic composites have been conducted [38].

Biochar is an eco-friendly and sustainable alternative to carbon black, the primary and standard reinforcing filler in rubber composites. Synthesized from non-renewable resources at temperatures $> 1600^{\circ}\text{C}$ in an oxidizing atmosphere, carbon black is known for its high carbon footprint [67]. It possesses a specific surface area, aggregate structure, and surface chemistry that enhance its capabilities as a filler in elastomers [68]. Given its key characteristics, biochar is attracting attention due to its carbon content and low carbon footprint. In the literature, biochar can be found in various rubber composites: natural rubber composites, nitrile rubber composites, styrene-butadiene rubber composites, and mixtures of natural rubber and butadiene [67]. To improve its effectiveness in rubber composites, treatments are sometimes necessary to modify its surface chemistry [69]. Steven C. Peterson and A. J. Thomas modified biochar to make it a viable co-filler; treatments suitable for the current rubber industry are being carried out [69]. A respective increase of 19% and 40% in tensile strength and toughness is observed with biochar first treated with CO_2 and then coated with lauric acid [69]. The study by Mauro Giorelli et al. demonstrates that the electrical performance of epoxy/biochar composites increases with the conductivity of Miscanthus biochar [66]. Nevertheless, the type of biochar and pyrolysis conditions can influence the electrical properties of the final composite [66]. However, it is necessary to pre-select the biochars used in order to achieve the desired results [66].

Incorporation of rice husk, peanut shell, and wood chip biochars into cementitious materials:-

The addition of biochar to cementitious materials was driven by its ability to sequester carbon over its lifespan. Biochar in a structure was previously considered a carbon sink. Over the years, various studies began to explore in depth the role, limitations, and both positive and negative effects of biochar in cementitious materials. This revealed its remarkable properties, which we will explore in this section, and attracted the attention of researchers in the field.

Impact of Adding Biochar on the Fresh Properties of Cementitious Materials:-

Workability:-

The incorporation of biochar into cementitious materials negatively affects workability. Indeed, part of the mixing water is absorbed by the pores of the biochar, thereby reducing the workability of the material. This limitation is a major reason for the restriction on the biochar incorporation rate, which rarely exceeds 5% [70]. The pore distribution of biochar is critical, as it significantly influences the mixture's water absorption capacity. It has been reported that biochar pores in the range of 5 to 30 μm are most effective at absorbing and retaining water [71]. Furthermore, fine, angular biochar particles also contribute to reduced workability. The angular shape of biochar tends to restrict the movement of the cement paste and particles [72]. Gupta et al. confirm these observations in their study, noting that angular wood sawdust biochar, which has a high water absorption capacity, exhibits the greatest reduction in flow compared to other biochars [47]. To overcome this undesirable effect and allow for a higher incorporation rate of biochar, a high-water-reducing superplasticizer is added in a dosage proportional to the biochar content. In the literature, the W/C ratio remains constant for all mixtures, and the dosage of the superplasticizer is varied until the desired workability is achieved. However, the use of the superplasticizer no longer has the desired effect at certain biochar dosages, and furthermore, it could delay the hydration of the material, thereby reducing early-age strength [73]. In some studies, biochar is pre-saturated before mixing, which ensures moisture transfer into the cementitious matrix while maintaining workability at a reduced W/C ratio. The study by Gupta et al. is a specific example of this, except that the workability of samples incorporating biochar pre-soaked for 6 hours prior to mixing is reduced, despite the addition of superplasticizers [74].

Hydration kinetics:-

Hydration is an essential process that leads to the formation of hydrates, which are responsible for the strength of cementitious materials. It is accompanied by the release of varying amounts of heat and is generally described in four stages: the initial stage, the induction stage, the acceleration stage, and the deceleration stage. Biochar, like any other additive, can accelerate or delay hydration, based on previous research. In a study by Gupta et al., the thermal evolution rate of cement pastes containing 1% and 2% mixed wood waste biochar (MWBC) showed a shortening of the induction period by approximately 30 minutes, followed by a slight acceleration of hydration [75]. Contradictory results were reported by Carolina et al. [76], who observed a prolongation of the induction period by 73.1%, 48.9%, and 77.9%, respectively, for additions of 1%, 2%, and 3% biochar compared to the control [76]. Cement pastes containing 1% fine and coarse wood sawdust biochar show, respectively, an increase in the amplitude of the first peak [77], corresponding to the hydration of tricalcium silicates (C3S), followed by dicalcium silicates (C2S) [78]. In some cases, the addition of biochar not only accelerates hydration but also amplifies the hydration peak. It has an accelerating effect. Biochar has the ability to provide additional nucleation sites where the formed hydrates precipitate. Indeed, the addition of biochar fills the voids between fine aggregates and cement particles, and their negative surface charges (due to the presence of OH^- groups) [75] attract the positively charged hydrated cement

particles around them [12]. This attraction has enabled the formation of nucleation sites, promoting an enhanced hydration reaction through the attraction of neighboring particles, leading to the precipitation of hydrates on their surfaces [12]. Consequently, biochar acts as a filler and contributes to the densification of the cementitious material [52], more so than its pozzolanic effect in some cases [79].

Several factors can contribute to improved hydration, such as the specific surface area of the biochar [50], its chemical composition, pre-soaking the biochar during air-drying [74], etc. The pyrolysis temperature plays an important role in hydration. Indeed, a low pyrolysis temperature results in incomplete decomposition of cellulose and hemicellulose, which are sources of sucrose and glucose formation [80], responsible for delaying hydration. A study by K. Kochova et al. found that the higher the sugar concentration, the greater the delay in the peak heat flow of primary hydration [81]. If the sugars tested were ranked by their impact on cement hydration from strongest to weakest, the order would be: sucrose, then glucose, and finally fructose [81]. The delay in cement setting is explained by two main mechanisms: the adsorption of sugars by cement grains and the chelation of Ca^{2+} ions. Sugar adsorbed by hydrated cement grains eventually forms a semipermeable layer, leading to delayed hydration [82]. In an acidic environment, certain functional groups of the dissolved sugar are capable of capturing Ca^{2+} ions present in the interstitial solution, which are intended for the formation of portlandite and C-S-H [81]. In addition to the presence of saccharides, the ash content of biochars and impurities can also hinder the hydration of cementitious materials [50].

Setting time:-

Setting time is the interval between the start and end of setting; it is the period during which the material hardens. The start of setting is often correlated with the start of the acceleration phase of the hydration kinetics. Both the start and end of setting can be shortened or prolonged following the incorporation of biochar, and there are numerous reasons for this. The incorporation of peanut shell biochar (500°C) at 3% by weight shortened the initial and final setting times by 40 and 88 minutes, respectively, compared to the control, unlike lower dosages, which showed no significant influence [83]. A similar effect was observed by Gupta et al (2018) using CO_2 -saturated and unsaturated sawdust biochars (300°C) [84]. Municipal sludge biochars pyrolyzed at 500°C and 600°C accelerated setting time, whereas those pyrolyzed at 300°C delayed it, for the same dosage of 2% by weight [48]. The setting time of cement pastes proved less sensitive to the presence of sugarcane and orange bagasse biochar; the results in Figures 5a and 5b showed that the end of setting occurs before the acceleration peak [76]. The reduction in setting time achieved by biochars could be an asset in terms of the speed of construction and project delivery.

Impact of adding biochars on the hardened properties of cementitious materials:-

The dosage that yields the best mechanical performance can vary from one study to another. In most cases, a low biochar dosage results in improved mechanical strength. In two different studies, the best compressive strength was achieved with a 1% dosage of olive pit biochar [85] and peanut shell biochar, although in one of them, a 3% dosage showed superior performance compared to the control [83]. In their study, Gupta et al. recorded the highest compressive and flexural strength at the same dosage of mixed wood sawdust biochar, resulting in improvements of 8.38% and 8%, respectively, compared to the control [77]. The 10% increase in strength following the addition of olive pit biochar is attributed to the high potassium content in the biochar, which is capable of reacting with cement particles during hydration [85]. The authors cite several reasons to explain this improvement in strength at low biochar dosages, including the fineness of the biochar [83], the biochar production conditions [86], and even the morphology of the biochar, which enhances the interlocking between the aggregate and the cement matrix [85]. The study by Restuccia et al. showed that dosages of 0.5% and 0.8% of hazelnut shell biochar and coffee grounds were more than sufficient to achieve the best mechanical strength [87].

A common finding across all studies is a reduction in mechanical performance when biochar is applied at high rates. This is not the case, however, when the biochar has pozzolanic properties, as adding up to 4% rice husk biochar slightly increased compressive strength compared to the control [85]. These results contrast with those of Xu Yang et al., who, despite adding only 2% rice husk biochar, observed reductions of 10.76% and 4.9% at 7 days and 28 days, respectively [88]. The reduction in compressive strength at later ages is less evident [88]. The addition of 2% wood chip biochar reduced the flexural strength of the mortar by 2% [89]. High biochar dosages should be avoided because the particles can agglomerate and create areas of weakness, increase porosity, and cause the water needed for hydration to be absorbed by the pores, resulting in incomplete hydration and a decrease in hydrated products [44]. Regarding flexural strength, it is more sensitive to pores in the tensile plane because these defects tend to open under tension [77]. Analysis of experimental data using machine learning identified initial saturation, the oxygen-to-carbon ratio, and soluble silicon content as the biochar characteristics that control mortar strength [90].

Table2: Variation in mechanical properties at 7 and 28 days for several studies incorporating biochar into cementitious materials

Biomass	T (°C)	Biochar content %	Appl.	Biochar as	Optimal content (%)	Change in mechanical properties at 28 and 7 days (%) / Control		Réf
						Rm	Rf	
Rice husks	450 - 550	2 ; 5 ; 10	Mortar	Sbst cement	2	↘ 4,9	-	[88]
Wood shavings	500	0,5 ; 1 ; 2 ; 4	Mortar	Sbst cement	1	↗ 5,76	↗ 3,07	[85]
Pine cones	500	3 ; 6	Mortar	Sbst cement	3	↗ 22,94	↘ 10	[91]
Dried distillery grains	500	1 ; 2 ; 3	Concrete	Sbst sand	3	↗ 24,56	-	[92]
				Sbst aggregate	3	↗ 28,07	-	
Sawdust	300	1 ; 2 ; 5	Mortar	Additive	1	↗ 18,97	↗ 4,66	[47]
Wood waste	-	1 ; 2 ; 5	Mortar	Additive	1	↘ 1,63	↗ 7,69	[44]
			Cement paste		2	↗ 13,11	↗ 23,52	
Mixed wood shavings	500	0,25 ; 0,5 ; 1 ; 2	Mortar	Additive	1	↗ 8,38	↗ 8	[77]
Rice husks	500	0,1 ; 0,25 ; 0,5 ; 0,75	Concrete	Sbst cement	0,1	↘ 7,79	-	[79]
Peanut shells	500	1 ; 3	Mortar	Sbst cement	1	↗ 20,51	-	[83]
Wood shavings	700	2	Mortar	Sbst cement	2	-	↘ 2	[89]
				Additive			↘ 2	
			Cement paste	Sbst cement			↘ 30	
				Additive			↗ 15	

MS : Mechanical strength; FS : Flexural strength; Sbst : Substitute

Study of the durability of biochar-cement composites:-

Cementitious materials mixed with biochar are subjected to durability tests to assess their long-term resistance while maintaining their integrity. In this section, we will review the tests conducted and discuss the impact of biochar. The dry shrinkage of a material is the phenomenon by which it undergoes a reduction in volume due to the loss of moisture to the outside through evaporation. A low biochar content resulted in a significant reduction in dry shrinkage (36.2%), a reduction that was much more pronounced with the incorporation of sludge ash, bringing the

reduction rate to 55.17% compared to the control [91]. The addition of biochar to a cement paste incorporating the expansive additive MgO known for its ability to mitigate autogenous shrinkage significantly influenced this shrinkage to the point of achieving an expansion of 80 microstrains at 180 hours [93]. In their study, Gupta et al. succeeded in reducing dry and autogenous shrinkage through the use of biochar and rice husk ash; according to the authors, this reduction is due to the release of alkaline cations by the inorganic components of the biochar, which affects the structure and quantity of hydrated products [39].

A reduction in the water absorption of mortars is observed following the incorporation of a small amount of biochar; the opposite effect occurs as the biochar content increases [94]. Even when combined with silica fume, the results remain the same. Wood biochar combined with 2% and 4% silica fume shows a 74% and 75% reduction in water absorption compared to the control; whereas at a 6% addition, a 50% increase is observed [95]. As the amount of biochar increases, the composite's pore network expands, which can lead to high water absorption through the biochar's pores. Resistance to erosion is also determined, even though it is not at all common. We distinguish between resistance to acid attack and erosion by sulfate, chloride, and other ions. Resistance to erosion by sulfate ions after 120 days of exposure is much higher for silica-poor biochars (MWBC) than for silica-rich biochars (RHB) [75]. The latter are less resistant due to the dissolution of amorphous silica by sulfate ions, which weakens the particles to the point of cracking [75]. Studies demonstrate the importance of biochar on its own but also when combined with other amendments. The synergistic effect of biochar with other amendments improves certain properties and must be taken into account to maximize their potential.

Strategies for Optimizing the Biochar Content in Cementitious Materials:-

The diversity of biochars produced and their specific properties, combined with the various protocols used in the production of cement paste, mortar, and even concrete, make comparison and even reproducibility quite difficult. The results are comparable, and each procedure is unique. However, certain key factors that allow for an increase in the biochar content defy standard protocols and stand out. It is these factors, identified in the literature, that we will discuss in this section. The improvement of cementitious material properties through additives occurs in two ways: either physically or chemically. In terms of clinker substitution clinker being the main active component of cement the ideal approach would be to use an additive with cementitious properties, i.e., one containing oxides that can chemically react with the hydrates present during hydration. These additives, better known as pozzolanic materials, have high silica and alumina contents. Unlike ash, which is commonly used for its pozzolanic properties, biochar is generally chemically inert due to its production method and the type of biomass used. However, alternatives are employed, notably the pretreatment of biomass with acid or water. Pre-treatment of rice husk and sugarcane bagasse biomass with hydrochloric acid (0.1N) at 23°C modified the chemical and physical structure and improved the reactivity of the corresponding biochars [96]. The pretreated rice husk and sugarcane bagasse biochars, when substituted at a 5% replacement rate, showed a 36% and 54.8% increase in compressive strength at 28 days, respectively [96]. In fact, the amorphous silica contained in these biochars reacted with portlandite to form other hydrated calcium silicates responsible for mechanical strength [97]. Of all the biochars studied, rice husk biochar is the most likely to exhibit pozzolanic properties, due to the high silica content in its biomass.

The order in which the components are introduced may seem unimportant, but it contributes to whether or not the resulting mechanical properties of the composites are improved. Daniel Sueraz et al. (2020) demonstrate in their study a significant variation in the flexural strength of cement pastes incorporating 2% biochar as an additive [89]. Depending on whether the biochar is first mixed with water and the superplasticizer (pre-wetting) or with the cement (dry pre-mixing), the results differ; pre-wetting shows a 15% increase in strength compared to the control, whereas the latter shows an 8% reduction [89]. In another study by Daniel Sueraz et al., only pre-wetting was performed, and a significant improvement in the compressive and flexural strength of cement pastes containing 2% NB biochar was observed [44]. While the order of ingredient addition is important, the production method and particle size of the biochar are also critical, as among the two types of biochar used, the pyrolyzed biochar with a finer particle size (F) demonstrated the best performance [44].

The particle size distribution of biochar in the mixture is critical because it affects the intrinsic properties of the cementitious material. Fine particles tend to densify the matrix by filling existing voids or cavities. The substitution of cement with biochar in ultra-high-performance concrete is studied by Anjaneya et al., and according to them, the effect of biochar on the compressive strength of concrete depends on the size and quantity used to replace the cement [98]. Among the three particle sizes used, the fine particle size (<125 µm) added at 5% generally exhibited better strength, comparable to the control; the fine particles accelerated the hydration kinetics of the cement-biochar paste mixture [98]. It should be noted that the biochars were pre-soaked before mixing, which could explain this

increase in the degree of hydration, highlighting another important aspect, namely the condition of the biochars prior to incorporation into the mixture. Fine biochar particles (0.10-2 μm) offer greater early-age strength and water impermeability than coarse particles (2–100 μm) [77].

The natural state of biochar after pyrolysis is modified to achieve a specific goal. For example, saturating the biochar with CO_2 (or physically activating the biochar) before incorporating it into the mixture not only reduces CO_2 emissions but also improves mechanical strength through the mineralization of calcium carbonate. In a study by Gupta, the construction of a 0.03 m^3 roof slab incorporating 2.5% saturated biochar sequestered 1.47 kg CO_2 -eq [99]. Furthermore, the 2.5% biochar-saturated paste mixture can improve early compressive strength (7 days) by 25–30% and by 12-15% compared to the non-carbonated control and the mixture with 8% silica fume at 28 days, this same mixture offers similar strength to the previous mixtures [99].

Biochar that has been pre-soaked in water until saturated and then incorporated into the mixture acts as an internal hardening agent. This is because the water contained in the biochar's pores desorbs, improving internal hydration. The internal hardening provided by the saturated biochar particles improved mechanical strength by 40% and 30% compared to the control mortar and the mortar incorporating dry biochar, respectively specifically, flexural strength and splitting tensile strength and reduced porosity by 20% compared to the control [49].

Just like internal curing, external curing plays a role and significantly influences the final result. Given the current context and existing curing methods, carbonation curing could meet our expectations in two ways. Accelerated carbonation curing (ACC), which is less costly, is one of the carbon capture and storage processes and the most widely used method for storing carbon in cementitious materials [100]. Unlike natural carbonation, it is carried out under strict conditions, taking into account a high CO_2 concentration and a controlled environment (temperature and relative humidity). Furthermore, the carbonation reaction rate of cement paste reaches its maximum when relative humidity is (50-80%) [101]. Cementitious materials incorporating biochar are subjected to several curing tests: wet [47][49] [74], sealed [99] [102], air [74][103], underwater [80][103], and even carbon dioxide [80][99] [102] [104]. The finding is that the development of mechanical strength in the presence of biochar is more pronounced during air curing than in water; nevertheless, underwater curing still yields the material's maximum performance, especially in the long term [103].

Compared to sealed and underwater curing, carbonation curing generally exhibits better mechanical strength at the 7-day mark; beyond that, specifically at 28 days, CO_2 curing no longer affects strength and even tends to decrease compared to the curing methods mentioned above [99] [102] [105]. CO_2 curing made it possible to increase the biochar content of rice husks and wood flour substituted for cement up to 10%, and to improve mechanical strength at 28 days by 24.2%, even though porosity and water absorption increased [106]. Furthermore, the CO_2 -equivalent emissions of the mortar were reduced by 7.7% compared to the control [106]. Admittedly, CO_2 curing delivers compelling results in both mechanical and environmental terms—which sets it apart from other curing methods—but it has its own requirements and limitations. The duration of carbonation, the type of preconditioning used before carbonation, etc., make all the difference. A new technique is used to address the decrease in strength caused by hardening before curing: spraying water after carbonation until all the water is absorbed [107]. All the factors mentioned above are summarized in Figure 2 below.

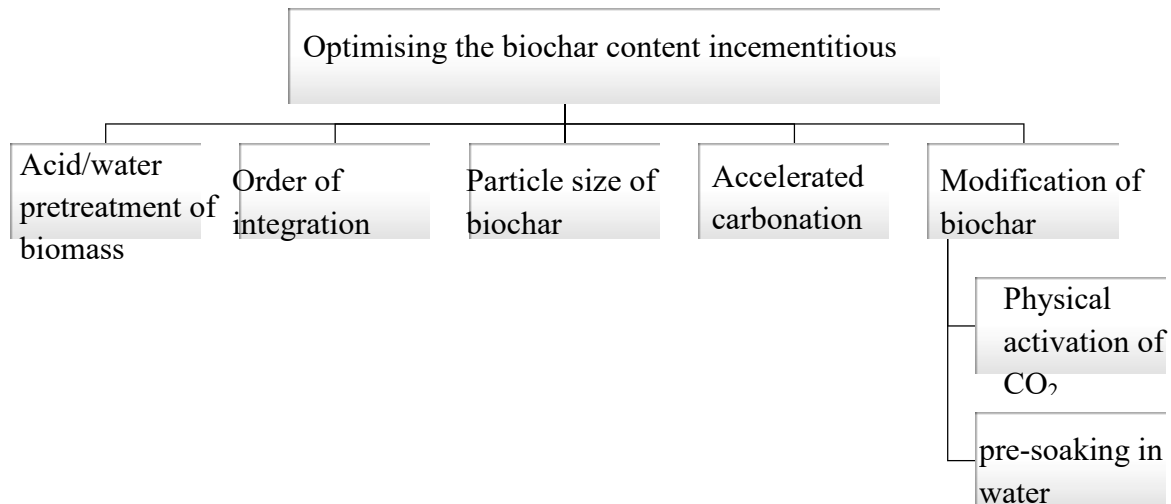


Fig.2.: Some strategies for optimizing the biochar content in cementitious materials

Benefits of adding biochar to cementitious materials:-

Reduced carbon footprint:-

The value of biochar in cementitious materials is not limited to improving mechanical properties and durability. The environmental aspect is significantly influenced by the reduction of the carbon footprint at various levels. First, in its current production method known as pyrolysis, which releases part of the carbon from the biomass in the form of CO and CO₂ and stores the remainder as a solid structure within the biochar [84]. The partial substitution of fossil fuels with renewable energy sources such as synthetic oil and gas derived from pyrolysis would be a major advantage and would further reduce GHG emissions [108]. The life cycle assessment (LCA) of the pyrolysis-biochar system reveals a reduction in net greenhouse gas emissions of -864 and -885 kg CO₂ eq per ton of dry matter from corn stalks and green waste, respectively, of which 62 to 66% stems from biomass carbon sequestration [109]. Next, its substitution for clinker, which aims to reduce emissions from decarbonation. An assessment of biochar's performance in reducing the global warming potential (GWP) of blended CEM II/A shows a reduction in net carbon emissions of 8%, 17%, and 26% for additions of 2%, 4%, and 6% biochar, respectively [70].

Finally, its ability to adsorb atmospheric CO₂ in structures and buildings could contribute to an additional 25% reduction in GHG emissions [17]. However, this ability to adsorb CO₂ is only possible due to the presence of basic functional groups [110], a specific surface area [111], and microporosity, which are responsible for the adsorption of gases and solvents [112]. CO₂ curing is performed on biochars prior to their incorporation into cementitious materials, as well as in mortars and concretes containing biochar. According to Maljaee et al., the saturation of biochars with CO₂ via adsorption does not affect their chemical or physical properties [73].

Improved crack propagation resistance:-

Toughness is a material's ability to absorb energy without breaking once a crack has formed. Numerous studies have shown that biochar can improve this toughness. The impact of biochar on toughness is much more pronounced in cement pastes than in mortars; for additions of 2% and 5%, an improvement of 175% and 240% is achieved for pastes, compared to an improvement of 17% and 30% and a reduction of 11% and 10% for mortars incorporating pyrolyzed and gasified biochar, respectively [44]. The rate of increase in fracture toughness is higher for smaller epoxy resin particles [113]; drawing an analogy with the biochar used, one could infer that the submicrometer size of pyrolyzed biochar is responsible for the increase in toughness [44]. According to Restuccia et al., the high carbon content of biochars alters the crack path by increasing its tortuosity; indeed, the carbon content confers increased strength and stiffness, inducing reinforcement mechanisms [114]. SEM analysis of cement pastes containing 0% and 0.8% hazelnut shell biochar shows a linear crack path for the control sample, whereas in the presence of biochar, the crack becomes much more tortuous, thus confirming the analyses by Restuccia et al [87]. Bamboo biochar incorporated at 1% by weight into the mortar increased both crack initiation and crack propagation resistance, although it reduced the stable crack propagation length [115]. The reasons cited are the filling and bridging effects of bamboo biochar in the cementitious matrix [115].

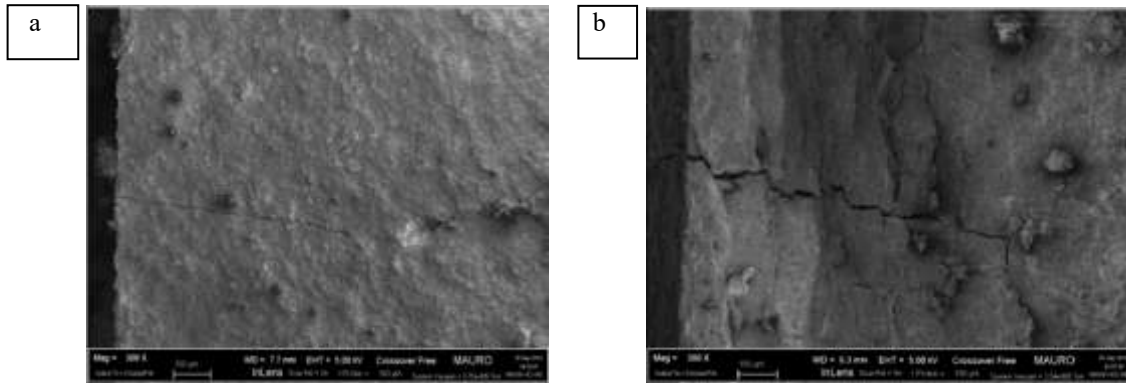


Fig.3. : Crack path on the surface of the cement paste: a) without biochar; b) with 0.8% hazelnut shell biochar [87]

Improved thermal insulation:-

Thermal conductivity is a material's ability to conduct heat due to a temperature gradient [116]. The low thermal conductivity of concrete[52] is further reduced by the incorporation of biochar, according to certain studies. A heat transfer analysis of biochar shows low thermal reactivity [117]. The work by Tan et al. identifies the porosity of biochar as the main reason for the reduction in thermal conductivity of mortar mixed with biochar derived from wood waste pyrolyzed at 400°C [118]. A decrease of 16%, 22%, 30%, and 39% compared to the control is observed for respective additions of 1%, 3%, 5%, and 10% biochar[118]. Rodier et al.[116] also observed a 45% and 30% reduction in the thermal conductivity of cement pastes to which 6% sugarcane bagasse biochar was added, at 28 days and 90 days, respectively [116]. Rice husk biochar mixed with clay exhibits the lowest thermal conductivity, ranging from 0.123 W/mK to 0.184 W/mK depending on the dosage [119]. Researchers agree that the porous nature of biochar largely due to the raw material used and the pyrolysis temperature [120] is the cause of this phenomenon. The porosity of biochars helps break thermal bridges in building materials and consequently improves thermal insulation in buildings, which will reduce energy consumption for heating or cooling buildings [17].

Absorption and shielding of electromagnetic waves:-

According to electromagnetic theories, the primary shielding mechanism involves reflecting radiation at the material's interface and dissipating the wave's energy within the material [121]. The combined effect of graphene and carbon nanotubes improves shielding performance against electromagnetic interference [121]. However, their high cost and poor distribution in cement under certain conditions limit their use, paving the way for research into other materials such as biochar [122]. Coals derived from woody biomass, particularly cellulose, lignin, and wood monoliths, exhibit good electrical conductivity, low weight, increased porosity, and a stable structure [123]. This makes them and their composites powerful contenders in the fields of electromagnetic wave absorption (EMWA)[124] and electromagnetic interference (EMI)[125]. A study conducted by Filipe et al (2020) presents a gypsum-biochar composite with increasing shielding efficiency, proportional to the biochar content, primarily in the microwave range [126]. The work of Khushnood et al (2016), incorporating 0.5% by weight of hazelnut and peanut shell biochar into cement, also demonstrates an improvement in total shielding effectiveness [122]. Compared to the control, the peanut biochar-cement composite yielded the best results, with an additional increase in total shielding efficiency of 353%, 223%, 126%, and 83% at frequencies of 0.94 GHz, 1.56 GHz, 2.46 GHz, and 10.0 GHz, respectively [122].

In the literature, the improved performance of carbon-based materials derived from lignin, cellulose, and wood monoliths in terms of electromagnetic wave adsorption and shielding is attributed to the water dispersibility of the precursors (lignin and cellulose) and their porous structure [123]. Indeed, these properties promote the uniform loading of functional groups, the dissipation of electromagnetic waves, and the enhancement of multiple reflections of EMWs in the case of wood monoliths [123]. The highly porous structure of biochars plays a decisive role in improving microwave absorption capabilities[127].

Conclusion:-

This review article discusses the use of biochar a bio-based material and carbon sink as an additive in cementitious materials to reduce the carbon footprint of cement. Consequently, the article addresses biochar production methods, its impact on the fresh and hardened properties of materials, durability, optimization of biochar content, and the benefits of its use. Slow pyrolysis of lignocellulosic biomass is more commonly reported in the literature, as it yields a high biochar yield and results in a relatively high carbon content; consequently, this leads to a reduction in CO₂ emissions and improved mechanical strength and crack propagation resistance. Due primarily to the porous nature of biochar, the workability, mechanical strength, and even the durability of cementitious materials tends to decrease, hence the need to limit the biochar content to reduce porosity and prevent potential particle agglomeration. To overcome this barrier posed by the incorporated biochar content and improve mechanical and durability properties, acid/water pretreatment of biomass, pre-soaking of biochar, physical activation with CO₂ of biochar, the use of fine biochar particles, and accelerated carbonation of the composites have been reported in the literature. Biochars have the ability to reduce the carbon footprint, improve the thermal conductivity of composites, and absorb electromagnetic waves. The discrepancies noted in the studies are partly due to the variability in biochar properties, caused by the biomass used and the pyrolysis parameters.

Thus, for the same biomass, two given studies may present different optimal rates. Among the studies reviewed, none considered the cement-biochar mixture as a new binder requiring comprehensive characterization according to standards applicable to hydraulic binders. To date, they have been limited to the addition of biochar and its effect on the mechanical properties and durability of cementitious materials. Beyond the lack of standardized tests, there is a near-total absence of full-scale applications. Regarding durability, erosion resistance has been little studied. Since this material is not currently recognized or authorized under the European cement standard EN 197-1, adopting a performance-based approach to cement would facilitate the development of manufacturing, standardization, and applications for this type of innovative cement composition. Another alternative would be to consider biochar not merely as an additive to cement, but rather as a component of a new type of binder, and thus develop a specific “product” standard for it, similar to alumina cements (EN 14647)[128] or supersulfated cements (EN 15743+A1)[129] . Such an approach will allow for a comparison of the performance of cement-biochar binders with that of conventional binders. This should also include the establishment of a standardized protocol for the cement-biochar mixture.

Abbreviations:-

- GHG: Greenhouse gases
- EN : European standard
- PHC: Performance testing of hydraulic cements
- IPCC : Intergovernmental Panel on Climate Change
- HTC : Hydrothermal carbonization
- IR : Infrared
- NMR : Nuclear Magnetic Resonance
- ATG : Thermogravimetric Analysis
- DTG : Thermogravimetric Derivative
- PP : Polypropylene
- MPa : MegaPascal
- W/C : Water-cement ratio

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