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REVIEW ARTICLE

State-of-the-art review on the use of lignocellulosic biomass in cementitious materials

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Abstract: The lignocellulosic biomass wastes cause some burden on the environment; meanwhile, the concrete industry is faced with large amounts of carbon dioxide emissions and raw mineral materials consumption. The use of lignocellulosic biomass wastes in cementitious materials not only provides an alternative to deal with the wastes but also favors the sustainable development of concrete industry. This review first introduces the characteristics of lignocellulosic biomass and then examines its effect on the mechanical properties, shrinkage, cracking, and some other properties of cement composites. Results show that lignocellulosic biomass can be directly used for three purposes: reinforcements, aggregates, and cement replacements. Although the lignocellulosic biomass cannot always enhance the mechanical properties of cementitious materials, it can improve toughness, shrinkage, cracking, heat insulation, etc. Additionally, some concerns with the use of lignocellulosic biomass are summarized, for which some physical and chemical modification methods (heating treatment, boiling treatment, torrefaction treatment, etc.) are identified to change the structure or remove amorphous components of lignocellulose biomass or prevent it from directly contacting cementitious materials. This review can provide some guidance for designing sustainable cementitious materials with lignocellulosic biomass.

Keywords: Lignocellulosic biomass; cementitious materials; cement; aggregate; fiber; treatment

1 Introduction

With the development of this society, more and more people are concerned about environmental issues such as energy consumption and global warming. The use of biomass materials provides a sustainable way to address these issues. Nowadays, lignocellulosic biomass has been paid much attention by researchers worldwide, because it is renewable, CO_2-neutral, and sustainable green material on earth [1-3]. For concrete industry, the lignocellulosic biomass is of great interest to be used to develop cement-based composites due to several benefits. Firstly, lignocellulosic biomass can be obtained from many sources, such as wood, crops, vegetables, etc., which makes it rather cheap and accessible. Secondly, the lignocellulosic biomass wastes are conventionally disposed of in landfills, which brings about high transport costs and incineration problems due to their high water content and low calorific value [4]. The recycling of these wastes is a sustainable way to solve this issue. Thirdly, large amounts of mineral materials (including limestone, clay, etc.) and energy are consumed in

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concrete industry [5, 6]. The use of lignocellulosic biomass as raw materials in concrete can save these mineral resources and eliminate the destruction of the natural environment. Meanwhile, the concrete with lignocellulosic biomass shows good thermal insulation and efficient energy saving [7]. Lastly, the annual yield of concrete is more than 14 billion tons, and the concrete industry accounts for 7% of global CO_2 emissions [8]. Similarly, Olivier reported that in the production of cement, both the carbonate oxidation in the cement clinker production process and the fuel combustion generated 4% of global CO_2 emissions, with a total amount of roughly 8% [9]. Thus, the use of lignocellulosic biomass in concrete industry is in accordance with the CO_2 emission reduction strategy.

Lignocellulosic biomass wastes can be used in the form of ash in cementitious materials. Burning biological products like wood, plants, energy crops, etc. can produce various lignocellulosic biomass ashes. Many types of lignocellulosic biomass ashes (such as bagasse ash, corn cob ash, etc.) have been commonly used in cement-based composites to partially replace cement [1, 10-12]. Due to the existence of large amounts of amorphous silica within these ashes, they are able to serve as pozzolanic materials, similar to fly ash and silica fume. The chemical characteristics of lignocellulosic biomass ashes are dependent on the combustion technology, combustion temperature, and biomass source [13], which may affect their reaction with cementitious materials. However, it needs to be noted that the non-coal derived biomass ashes have not been allowed to be used in concrete by the EN-450, EN197-1, American Concrete Institute (ACI), and American Society of Testing Material (ASTM) [14, 15]. In addition to lignocellulosic biomass ash, lignocellulosic biomass wastes can be directly used in cementitious materials, without being burned into ashes, in multiple forms including powders, chips, shavings, fibers, etc. Normally, based on the size, they can be used for different purposes, replacing aggregates or cement and serving as reinforcements in cement-based composites. In summary, the direct application of lignocellulosic biomass wastes to cement-based composites can bring in some benefits: reducing the self-weight, improving the thermal and acoustic insulation capacities, and increasing the toughness and even strength if used appropriately [16-20].

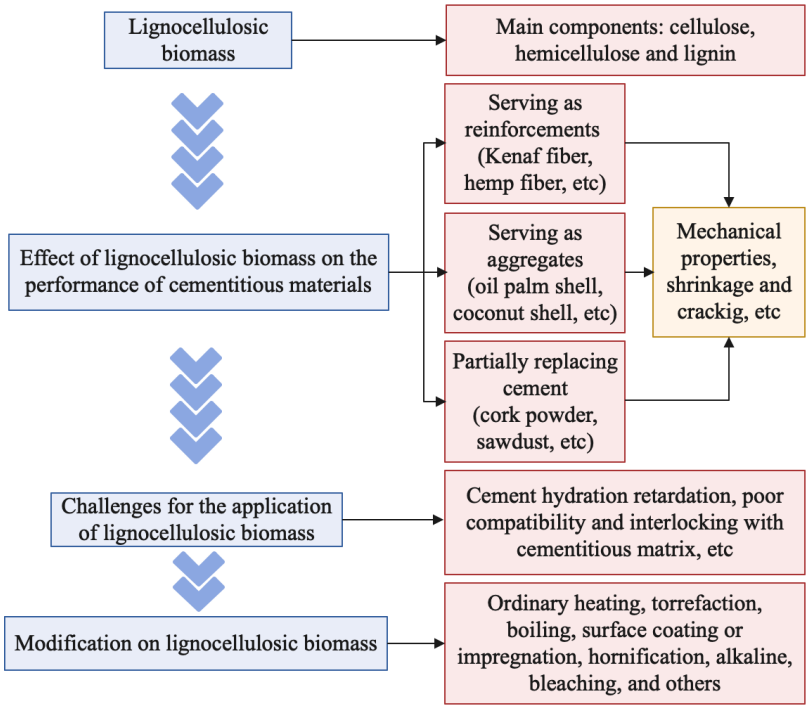


Fig. 1. Flowchart of this review

The application of lignocellulosic biomass ash in cementitious materials is not the focus of this review paper and will be reviewed in another paper. This review mainly focuses on the direct application of lignocellulosic biomass wastes without combustion. From the perspective of functionality, lignocellulosic biomass wastes of different sizes can be directly used as reinforcements, aggregates, and cement replacements in cementitious materials. So far, some reviews have been done on the application of lignocellulosic biomass in cementitious materials. Onuaguluchi and Banthia reviewed fiber type, fiber characteristics, and their effects on the fresh (consistency, setting time, and plastic shrinkage) and

hardened (drying shrinkage, mechanical strength, and durability) properties of plant fiber reinforced cement composites [21]. Similarly, Pacheco-Torgal reviewed vegetable fiber characteristics and properties and their effect on the properties (including durability) of cementitious materials; also, some matrix and fiber modification methods related to the durability issues were identified [22]. In addition, Hamada et al. reviewed the use of oil palm shell as an aggregate in cement concrete and summarized its effect on the fresh (density and workability), hardened (strength, elastic modulus, etc.), and durability (water absorption, drying shrinkage, resistance to sulfate attack, etc.) properties of cement concrete [23]. However, the existing reviews mainly focus on the use of lignocellulosic fibers, without comprehensively summarizing the use of lignocellulosic biomass as aggregates and even without involving the use of lignocellulosic biomass as cement replacements. Therefore, this review attempts to summarize the use of lignocellulosic biomass in cementitious materials from multiple aspects including serving as reinforcements, aggregates, and cement replacements and then points out some challenges and corresponding handling methods.

Overall, this review mainly consists of four parts: firstly, the characteristics of lignocellulosic biomass are presented in detail; secondly, the effects of various lignocellulosic biomass on the performance of cementitious materials are examined; thirdly, the challenges with the application of lignocellulosic biomass are identified; lastly, the modification methods on lignocellulosic biomass are reported. The flowchart is shown in **Fig. 1**. The purpose of this review is to provide some guidance for modifying lignocellulosic biomass and then designing more reliable and sustainable cementitious materials with modified lignocellulosic biomass.

2 Lignocellulosic biomass

Based on the definition by ASTM, biomass is taken as "a substance wholly comprised of living or recently living material" [24]. Generally, biomass can be categorized as lignocellulosic biomass and non-lignocellulosic biomass [25]. Although the composition of lignocellulosic biomass varies with the biomass source, its main components including cellulose, hemicellulose, and lignin remain unchanged, as introduced in **Table 1**.

Table 1. Characteristics of cellulose, hemicellulose, and lignin

	Cellulose	Hemicellulose	Lignin
Structure	A semicrystalline polysaccharide consisting of D-glucopyranose units	Polysaccharide mixtures in hemicellulose include pentose sugars hexose sugars, and sugar acids	Phenolic polymeric material with three primary precursors
Chemical stability	Easily hydrolyzed by acid but resistant to strong alkali and oxidizing agents	Easily hydrolyzed by acid and partly soluble in water and alkali	Not soluble in water and hydrolyzed by acid, but soluble in hot alkali, readily oxidized, and easily condensable with phenol
Thermal stability	Harder to be decomposed than lignin and hemicellulose	Easier to be decomposed than cellulose and lignin	Decompose over a wider temperature range than hemicellulose and cellulose
Water absorption	Quite hydrophilic	Higher hydrophilic capacity than cellulose and lignin	Hydrophobic

Cellulose is a kind of polysaccharide $(C_6H_{12}O_5)_n$ that is formed through the link of D-glucopyranose units with β-(1-4)-glycosidic linkages (see **Fig. 2**). It is normally polymerized with a degree of around 10000 [26-28]. Cellulose is interlinked through hydrogen and van der Wall bonds to form microfibrils that are closely held together by lignin and hemicellulose. Strictly, the cellulose can be divided into tightly packed crystalline cellulose and amorphous cellulose [27,28]. Compared to amorphous cellulose, crystalline cellulose is more stable and less prone to be degraded because it attached to each other by non-covalent hydrogen bonding [29]. Because the majority of cellulose is crystalline cellulose, it is harder for cellulose to be decomposed than lignin and hemicellulose [30]. Additionally, cellulose is easily hydrolyzed by acid but is more resistant to strong alkali and oxidizing agents [31]. The decomposition of cellulose produces glucose ($C_6H_{12}O_6$) with three hydroxyl groups that form intramolecular and intermolecular hydrogen bonds [32].

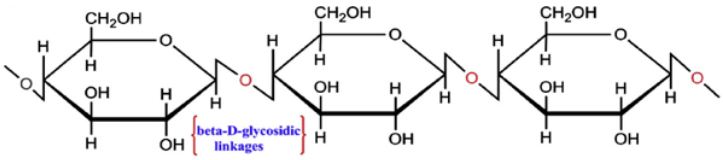


Fig. 2. Structure of cellulose [28]

Hemicellulose is another abundant biopolymer in lignocellulosic biomass, but with a more complex structure than cellulose [27]. Hemicellulose contains different sugar units than cellulose containing only 1,4–β-D-glucopyranose units. The commonly seen polysaccharide mixtures in hemicellulose are pentose sugars (e.g., xylose and arabinose), hexose sugars (e.g., glucose, mannose, and galactose), and sugar acids (e.g., glucuronic acid and galacturonic acid), with the chemical structure shown in **Fig. 3** [33]. Compared to the linear semi-crystalline structure of cellulose, hemicellulose lacks a crystalline structure because it has large amounts of side chains. The hemicellulose is polymerized with a degree of around 50-300 [29], much lower than that of cellulose. Thus, hemicellulose can be much easier to be degraded in the acidic or hot aqueous medium than cellulose [33]. It is also easily hydrolyzed by acid and soluble in alkali [31]. Compared to lignin and cellulose, the decomposition temperature of hemicellulose is lower [30]; also, the hemicellulose has higher water absorption capacity, contributing the most to the hydrophilic nature of lignocellulosic biomass [34].

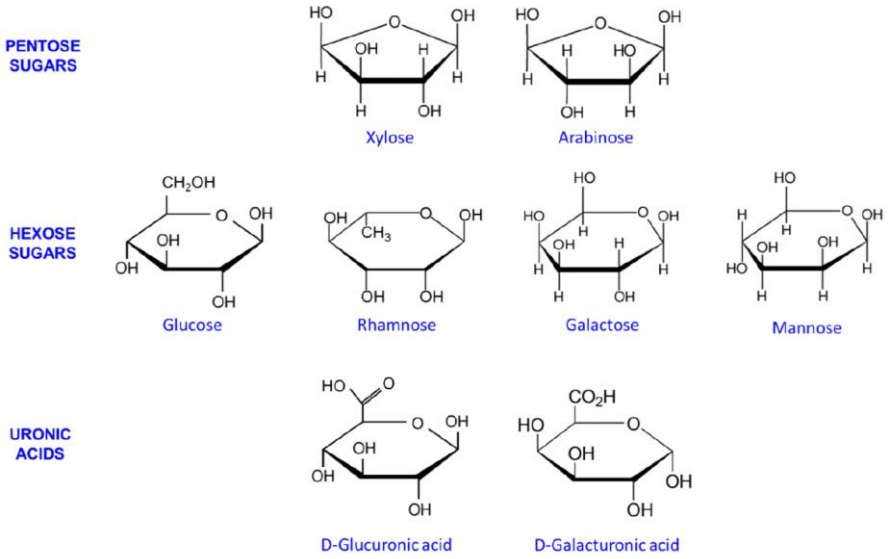


Fig. 3. Chemical structure of hemicellulose sugars and sugar acids [33]

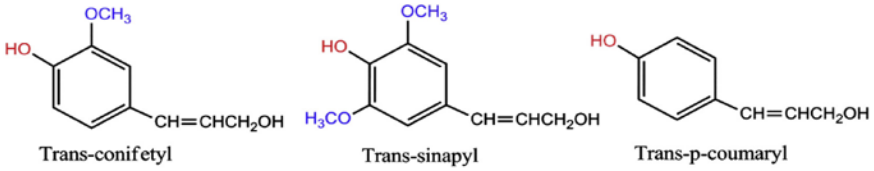


Fig. 4. Precursors of lignin [28]

Lignin has a complex three-dimensional structure that is formed through the copolymerization of aliphatic and aromatic constituents, with a very high molecular weight [31]. Unlike cellulose and hemicellulose, the exact chemical structure of lignin remains unclear because the original structure of lignin is often modified during the isolation process. Moreover, the isolation method affects the chemical structure of lignin to a large extent. For example, the lignins obtained from Kraft process and lignosulfonate process have different sulfonate groups [33]. So far, some functional groups (such as hydroxyl, methoxyl, and carbonyl groups) within lignin have been identified [27]. The three primary precursors in lignin are trans-coniferyl, trans-sinapyl, and trans-p-coumaryl (see **Fig. 4**) [28,35]. Lignin has lower water absorption capacity than hemicellulose and cellulose, showing hydrophobic behavior, which makes it insoluble in water under ambient conditions [26]. It is even not hydrolyzed by acids but soluble in hot alkali, readily oxidized, and easily condensable with phenol [27]. Besides, the

decomposition temperatures of different oxygen functional groups associated with lignin vary to a large extent, so the decomposition temperature range of lignin is wider than hemicellulose and cellulose [30,34].

3 Effect of lignocellulosic biomass on the performance of cementitious materials

The lignocellulosic biomass can be used in different forms based on their sizes, and they can function for different purposes: serving as reinforcements, serving as aggregates, and partially replacing cement. The mechanical properties, shrinkage and cracking, and other properties (thermal insulation, permeability, etc.) are summarized in this review paper. Moreover, more attention is given to compressive strength, flexural strength, tensile strength, elastic modulus, autogenous shrinkage, drying shrinkage, etc. that can be experimentally measured following ASTM C39 (or C109), ASTM C78 (or C293), ASTM C496, ASTM C469, ASTM C1698, ASTM C157, etc.

3.1 Serving as reinforcements

The first application of lignocellulosic biomass is to use lignocellulosic fibers as reinforcements in cementitious materials. Several typical lignocellulosic fibers including kenaf fiber, hemp fiber, coconut fiber, etc. that are used to reinforce cementitious materials are shown in **Fig. 5**. The effects of lignocellulosic fibers on the mechanical properties, shrinkage, cracking, and other properties of cementitious materials are introduced as follows.

(a) (b) (c)

Fig. 5. Appearance of (a) kenaf fiber [36]; (b) hemp fiber [37]; and coconut fiber [38]

3.1.1 Mechanical properties

In terms of mechanical properties, large amounts of research have been conducted, as summarized in **Table 2**. There are some variations in the effect of lignocellulosic fibers on the strength of cementitious materials, depending on fiber type, fiber length, and fiber content. The compressive strength, flexural strength, and tensile strength can be improved by fibers if a proper fiber dosage and fiber length is used, but they are not increased in the same proportion. On one hand, some research indicates that the compressive strength is less increased than the flexural strength or tensile strength. Chakraborty et al. reported that when the optimal quantity (around 1 wt.%) of jute fibers is used, the flexural strength and compressive strength increased by 16% and ~9%, respectively, compared to the plain mortar [39]. Li et al. reported that under the optimum conditions (0.36% fiber dosage, 20 mm fiber length, 20 mm aggregate size, and wet mix method), the compressive strength and flexural strength of hemp fiber reinforced concrete increased by 4% and 9%, respectively [40]. On the other hand, some research indicates that the increase of compressive strength is more significant than that of the flexural or tensile strength. Ahmad et al. indicated that 50 mm-long and 1.5% coconut fiber increased the splitting tensile strength, compressive strength, and flexural strength of high strength concrete up to 20.4%, 25%, and 3%, respectively, compared to plain high strength concrete [41]. Ramli et al. reported that the compressive and flexural strengths of concrete improved up to 13% and 9%, respectively, with the incorporation of coconut fibers [42]. Zakaria et al. indicated that 0.1%-0.25% fiber volume and 10 mm-15 mm fiber length could improve the compressive, flexural, and tensile strengths significantly, resulting in the maximum increase percents of 33%, 23%, and 38%, respectively [43]. Therefore, there is not a consistent conclusion about which one (compressive strength, flexural strength or tensile

strength) can be improved more by lignocellulosic fibers. The strength improvement can be attributed to the bond between fibers and cementitious matrix, reducing the stress that causes ruptures [38].

Although the above-mentioned research indicates that the strength of cementitious materials can be improved by lignocellulosic fibers, the opposite effect also can be seen in some research. Hwang et al. reported that the compressive strength of cementitious composites could be reduced by coconut fibers [38]. Zhou et al. reported that kenaf fibers could decrease compressive strength by 12.2-46.2% although they increased the flexural strength by 30.7-66.9% [44]. Benaniba et al. indicated that the flexural strength of date palm fiber reinforced concrete was increased at low fiber dosages, but the compressive strength was decreased at all considered dosages (0-30%) [45]. Overall, the compressive strength may be negatively affected by lignocellulosic fibers while the flexural strength can be still improved. More interestingly, the effect of fibers on compressive strength depended on the strength grade of the matrix in addition to fiber characteristics; for the matrix with high strength grade, the compressive strength was decreased more by fibers compared to low strength grade [44]. The reduction of compressive strength can be related to the clustering of fibers, introducing some voids into the specimens, and the weak bond between fibers and the matrix [38,44].

Table 2. Mechanical properties of cementitious materials with lignocellulosic fibers

Type	Content	Length	Results	Ref.
Coconut fiber	0.5%, 1%, 1.5%, and 2% by cement mass	25 mm, 50 mm, and 75 mm	Improve compressive, splitting tensile, and flexural strengths, and energy absorption and toughness, with 50 mm long +1.5% dosage being the best.	[41]
Jute yarn	0.1%, 0.25%, 0.5%, and 0.75% by concrete volume	10 mm, 15 mm, 20 mm, and 25 mm	Improve compressive, flexural, and tensile strengths significantly with 0.1%-0.25% fiber volume and 10-15 mm fiber length.	[43]
Kenaf fiber	1%, 1.5%, and 2% by mass	5-10 mm (40%) and 10-15 mm (60%)	Decrease compressive strength but increase flexural strength; reduce compressive toughness but increase flexural toughness.	[44]
Coconut fiber	2% by cement mass	5 cm	Improve elastic modulus, strength, toughness index, and absorbed energy; also, the addition of superplasticizer results in more improvement.	[48]
Jute fiber	0.25%, 0.5%,0.75% and 1% by volume	10-20 mm	Compressive strength, flexural strength, and splitting tensile strength are improved the most by 0.5% fiber; the modulus of elasticity is improved the most by 1% fiber.	[46]
Kenaf fiber	1%, 2%, and 3% by volume of the mixture	6, 12, and 18 mm	2%-3% amount and 12 mm length give the optimum results.	[51]
Jute fiber	0.5% by volume for concrete; 1% by volume for mortar	20 mm	JFRCC with GGBS/PC matrix achieves higher strength than that with PFA/PC matrix, but the latter possesses higher impact resistance and absorbs more impact energy than the former at the ages of 14 and 28 days.	[47]
Coconut fiber	1%, 2%, 3% and 5% by cement mass	2.5, 5, and 7.5 cm	5 cm length+5% amount results in the best overall mechanical and dynamic properties.	[49]
Bagasse and Hemp fibers	0.8 %	3-12 mm (bagasse fiber) 6 mm (hemp fiber)	Bagasse fiber increases flexural strength while hemp fiber increases toughness.	[50]
Coconut fiber	0.6% , 1.2%, 1.8%, and 2.4% by binder volume	20-30 mm	Improve compressive and flexural strengths up to 13% and 9%, respectively.	[42]
Date palm fiber	0-30%	7 mm	Increase flexural strength at low fiber dosages but decrease compressive strength.	[45]

It needs to be noted that the optimum fiber content may vary for different mechanical properties.

For example, Bheel et al. indicated that the compressive strength, splitting tensile strength, and flexural strength were improved the most by 0.5% jute fiber, which were 10.14%, 12.50%, and 11.11%, respectively, higher than those of plain concrete; however, the modulus of elasticity increased from 2.45% to 14.91% as the jute fiber content increased from 0.25% to 1% [46]. Besides, the matrix type and the use of superplasticizer affect the strength and elastic modulus of cementitious materials. Zhou et al. reported that jute fiber reinforced cementitious composites (JFRCC) with ground granulated blast furnace slag (GGBS)/portland cement (PC) matrix achieved higher compressive strength, splitting tensile strength, and flexural strength than those with pulverized fly ash (PFA)/PC matrix [47]. Khan and Ali reported that 1% superplasticizer could improve the modulus of elasticity, compressive strength, flexural strength, and splitting tensile strength by 93%, 92%, 36%, and 39%, respectively, compared to coconut fiber reinforced concrete without superplasticizer [48].

Regarding the toughness of cementitious materials, it is widely recognized to be improved by lignocellulosic fibers [38-40,49], indicating a higher energy absorption capacity. For example, Ahmad et al. reported that the toughness indices in compression and flexure for concrete reinforced with 50 mm and 1.5% coconut fiber increased by 23.4% and 94%, respectively; and the corresponding total energy absorptions in the compression and flexure were increased by 72.5% and 162%, respectively [41]. The improved toughness can be because fibers transfer the stress from the matrix and restrain the propagation of cracks [38]. In addition to fiber length and fiber content, the matrix strength and the use of admixture also affect the toughness of fiber-reinforced cementitious materials. Zhou et al. reported that, with the increase of the matrix strength grade, the compressive toughness decreased, and the flexural toughness increased firstly and then decreased; however, the toughness index was barely changed [44]. Khan et al. indicated that coconut fiber reinforced concrete (CFRC) had improved absorbed energy and toughness indices than plain concrete; and compared to the CFRC without superplasticizer, 1% superplasticizer could improve the flexural total absorbed energy, flexural toughness index, compressive total absorbed energy, compressive toughness index, splitting-tensile total absorbed energy, and splitting-tensile toughness index by 17%, 4%, 92%, 51%, 87%, and 7%, respectively [48]. Because of the improved toughness, the fiber-reinforced cementitious materials may be suitable for machinery foundation floor in factories and shatter and earthquake resistant construction [39].

In addition, the dynamic tests indicated that as the fiber content increased, the damping ratio increased but the fundamental frequency and dynamic modulus of elasticity reduced; also, 5 cm long fibers resulted in higher damping than other fiber lengths [49]. The pull out test indicated that all hemp fibers were pulled out from the matrix, while roughly 1/3 of bagasse fibers were broken with very small displacements, so hemp fibers and bagasse fibers contributed more to the composite toughness and flexural strength, respectively [50].

3.1.2 Shrinkage and cracking

Cementitious materials normally have low tensile strength and can easily crack due to shrinkage, bending, freeze-thawing, or other reasons, which not only reduces the load-carrying capacity of cementitious materials but also allows the ingress of aggressive agents to harm the durability of cementitious materials, eventually shortening the service life of concrete structures [52,53]. The incorporation of lignocellulosic fibers into cementitious materials provides a possible way to improve the cracking resistance. Lignocellulosic fibers positively influence the plastic cracking of cementitious composites. With the increase of coconut fiber volume from 0 to 4%, the number of cracks decreased from 11 to 0, and the cracking index decreased from 2.95 to 0 mm [38]. The possible reason can be the internal curing effect of fibers, the homogeneous distribution of linear stresses responsible for cracking facilitated by fibers, higher elastic modulus of fibers than the matrix, and bridging effect of fibers [38,54]. Besides, Guo et al. suggested that 0.25% and 0.5% kenaf fibers by weight of cement could reduce autogenous shrinkage and drying shrinkage cracking of cement paste significantly [36]. The reduced autogenous shrinkage can be related to the internal curing capacity of lignocellulosic fibers that is more related to their physical morphology than chemical composition [55]. Even, in a hot-dry environment, the addition of date palm fibers (DPF) to self-compacting concrete (SCC) could reduce the early drying shrinkage and cracking risks although the compressive strength was slightly reduced, and 0.1% volume fraction and 2 cm length of DPF reduced early age drying shrinkage by 50% compared

to plain SCC specimens [56].

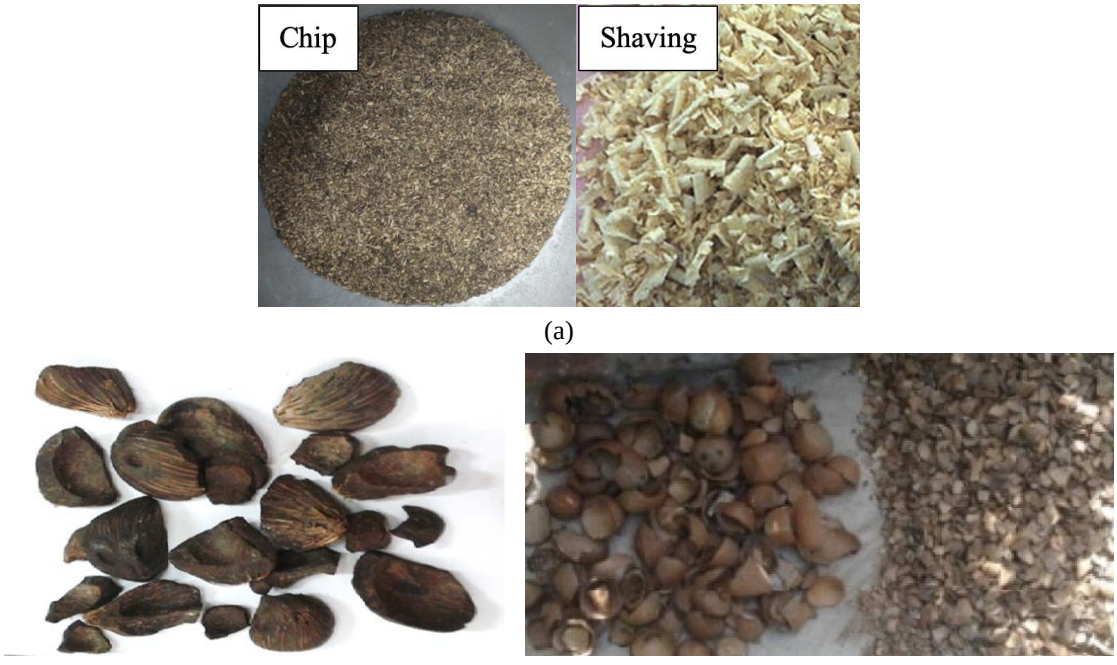
3.1.3 Other properties

Lignocellulosic fibers could reduce the thermal conductivity of cementitious materials and thus could be used as thermal insulation materials in buildings [19]. It was reported that the date palm fiber reinforced concrete could be classified as hygroscopic and breathable material with excellent moisture buffering capacity and was highly recommended to be used for construction applications [57]. The hygrothermal behavior of concrete with 15 wt.% date palm fibers was studied at the wall scale, which showed that this material had very good hygrothermal performances and consequently could be used for thermal insulation and hygric regulation inside the buildings [58]. Also, increasing the date palm fiber content could increase the insulating capacity of mortar by reducing its thermal conductivity [45]. In addition, the treatment method of fibers affects the effectiveness of thermal insulation. The cementitious materials with pyrolyzed bagasse fibers were weaker heat conductor materials than those with alkaline treated bagasse fibers, which showed that pyrolyzed bagasse fibers were better to be used to prevent heat transfer into buildings and consequently save energy [59].

The permeability of jute fiber reinforced concrete is 9%, 18.18%, 31.82%, and 36.40% lower than that of plain concrete with 0.25%, 0.5%, 0.75%, and 1% of jute fiber, respectively [46]. It can be seen that a higher fiber dosage results in a lower permeability, which is helpful to the durability of concrete. However, the fiber dosage needs to be limited to a certain amount. Ramli et al. reported that the coconut fiber reinforced concrete could suppress the deleterious effect caused by aggressive environments, in which the coconut fiber dosage should not be more than 1.2% of the binder volume due to its natural degradation [42].

3.2 Serving as aggregates

Several commonly used lignocellulosic biomass wastes including wood chip and shaving (**Fig. 6**(a)), oil palm shell (**Fig. 6**(b)), coconut shell (**Fig. 6**(c)), etc. have been reported to serve as aggregates in cementitious materials. This type of lignocellulosic biomass wastes can be called lignocellulosic aggregates from the perspective of cementitious materials. The effects of lignocellulosic aggregates on mechanical properties, shrinkage, cracking, and other properties of cementitious materials are introduced as follows.



(a)

(b) (c)

Fig. 6. Appearance of (a) wood chip and shaving [60, 61]; (b) oil palm shell [23]; and (c) coconut shell [62]

3.2.1 Mechanical properties

The mechanical properties of cementitious materials with lignocellulosic aggregates are shown in **Table 3**. Many researchers suggested that the addition of lignocellulosic aggregates as lightweight aggregates normally reduced the mechanical properties of cementitious materials [18,60,62,63]. Guo et al. incorporated wood chips into mortars to partially replace 5% and 10% sand and then suggested that wood chips could reduce the flexural and compressive strengths of mortars significantly while increasing their toughness [60]. Mohammed et al. indicated that the replacement of fine aggregate with wood chipping in concrete resulted in the reduction of compressive strength, splitting tensile strength, and flexural strength [18]. Kanojia and Jain indicated that as the amount of coconut shells increased, the compressive strength reduced; moreover, 40% replacement of conventional coarse aggregate by coconut shell decreased the 7-day and 28-day compressive strengths by 62.6% and 21.5%, respectively [62]. Mannan and Ganapathy found that the use of oil palm shell (OPS) could reduce flexural strength, splitting tensile strength, compressive strength, and elastic modulus of concrete [63]. Although the strength is reduced, the use of lignocellulosic biomass as lightweight aggregates can develop lightweight concrete. Gunasekaran et al. indicated that the flexural strengths of coconut shell concrete (CSC) with 0.42 and 0.44 water/cement (w/c) ratios were 17.53% and 16.42% of the compressive strengths, respectively; the splitting tensile strengths of CSC with 0.42 and 0.44 w/c ratios were around 10.11% and 9.17% of the compressive strengths, respectively; the impact resistance of CSC was higher than that of conventional concrete; and the bond strength was comparable to those of normal concrete and other concretes with lightweight aggregates [64]. Therefore, coconut shells were proved to fulfill the requirements for use as lightweight aggregates [64]. A similar conclusion was reported by Mannan and Ganapathy who found that the 28-day compressive strength of oil palm shell concrete was 20-24 MPa, meeting the requirement for structural lightweight concrete [63]. In addition, the toughness of cementitious composites can be improved by lignocellulosic biomass. Guo et al. found that the incorporation of wood chips into mortar could increase its flexural toughness and toughness index, which might be because wood chips arrest cracks of mortar and consequently results in a more ductile failure mode; also, the untreated wood chips lead to higher toughness than the thermally treated wood chips [60].

Table 3. Mechanical properties of cementitious materials with lignocellulosic aggregates

Type	Content	Results	Ref.
Wood chip	5% and 10% in replacement of sand by weight	Reduce compressive and flexural strengths; increase toughness; reduce weight.	[60]
Wood chipping	10%-30% in replacement of fine aggregates by weight	Reduce weight and strength.	[18]
Coconut shell	Varying mix proportions	Can be used as aggregates to develop structural lightweight concrete.	[64]
Coconut shell	10%-40% in replacement of coarse aggregates by volume	Reduce compressive strength; 40% replacement reduces 7-day and 28-day compressive strengths by 62.6% and 21.5%, respectively.	[62]
Oil palm shell	Varying mix proportions	Reduce flexural strength, splitting tensile strength, compressive strength, and modulus of elasticity, but can be accepted to develop structural lightweight concrete.	[63]

3.2.2 Shrinkage and cracking

Mannan and Ganapathy showed that the drying shrinkage of oil palm shell concrete was 14% higher than that of control concrete at 90 days [63]. This seems to be a concerning issue. The possible reason can be that the open textured and irregular surface of oil shell concrete increases the loss of free water.

3.2.3 Other properties

Bederina et al. found that the addition of wood shavings reduced the thermal conductivity and thus increased the insulating capacity of concrete; also, when the content of wood shavings was low, the thermal conductivity of river sand concrete was slightly higher than that of dune sand concrete, but this

difference tended to disappear if the wood shavings content was high [16]. The pretreatment method and binder type also affect the thermal conductivity of concrete. It was reported that the pretreated corn stalks increased the thermal conductivity of concrete compared to untreated ones; for the same type of corn stalk, the thermal conductivity of magnesium phosphate cement based concrete was higher than that of ordinary Portland cement or geopolymer based concrete [65]. The thermal conductivity of concrete is positively related to its density. In addition, Gunasekaran et al. examined the durability of coconut shell aggregate concrete (CSAC), with coconut shells as recycled lightweight aggregates, and found that the durability of CSAC was comparable to that of other conventional lightweight concretes [66]. Therefore, it could be seen that the use of lignocellulosic biomass as lightweight aggregates does not harm the durability of concrete and even is favorable to its heat insulation.

3.3 Partially replacing cement

As mentioned in the Introduction part, the replacement of cement with lignocellulose biomass ash (that is, burning lignocellulosic biomass wastes into ashes) is not the focus of this study. This section mainly reviews the direct use of lignocellulosic biomass wastes without combustion to partially replace cement. The appearance of several kinds of lignocellulosic biomass wastes that are used to replace cement is shown in **Fig. 7**. Because the existing studies about this topic are limited, this part only reviews the mechanical properties, shrinkage, and cracking of cementitious materials, as introduced below.

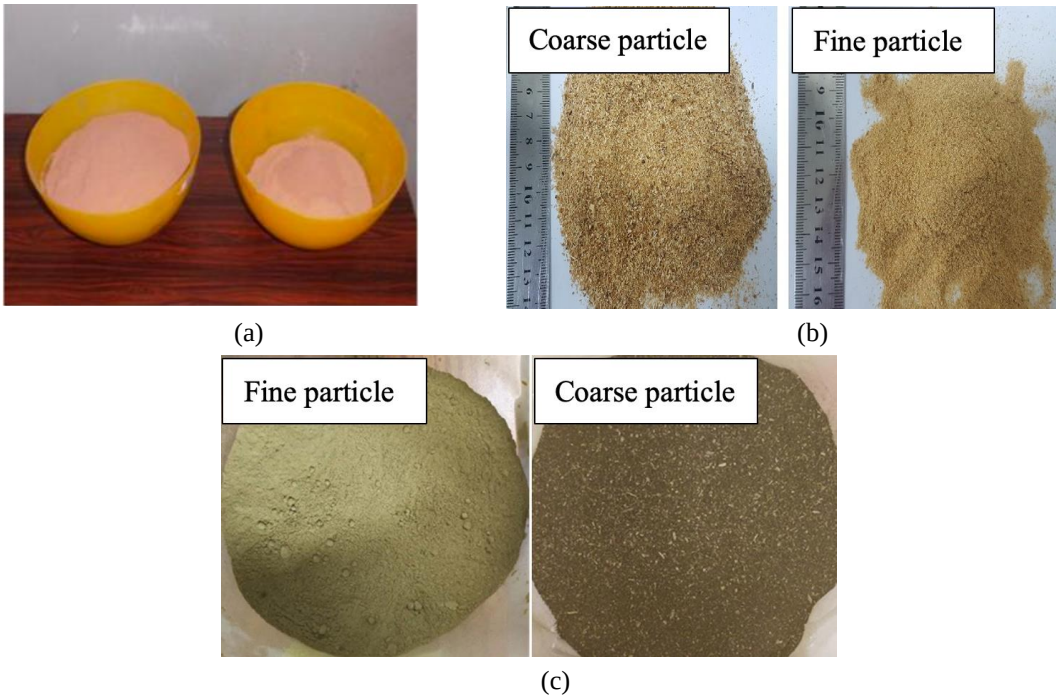


Fig. 7. Appearance of (a) cork powder [67]; (b) sawdust [68]; and (c) hemp powder [69]

3.3.1 Mechanical properties

Usman et al. reported that the compressive strengths of self-compacting concrete with cement partially replaced with sawdust were reduced by 11%, 27%, and 34% at the replacement contents of 2%, 5%, and 7%, respectively, which was due to the increased air content that increased the overall porosity and weakened the internal structure [68]. Also, the water/cement ratio increases when cement is replaced with lignocellulosic biomass waste, which may further increase the porosity of concrete. In addition to porosity, the strength of cementitious materials is closely related to cement hydration. Guo et al. examined the hydration of cement paste with hemp powders that are obtained by grinding the flowering materials of hemp products after extracting the cannabidiol [69]. It was reported that hemp powders could delay cement hydration due to some hemicellulose, lignin, and impurities within hemp powders. Therefore, it can be concluded that the strength reduction caused by lignocellulosic biomass is not only due to the increased porosity but also attributed to the retarding effect on cement hydration.

The strength of concrete can be controlled by adjusting the dosage of lignocellulosic biomass or the chemical composition of cement. Matos et al. showed that although a reasonable partial replacement of cement with cork powders in self-compacting concrete reduced the strength of concrete, a good strength level (C30/37) and suitable durability still can be obtained [67]. Usman et al. showed that the strength reduction could be compensated by using cement with higher amounts of dicalcium silicate and tricalcium silicate [68].

3.3.2 Shrinkage and cracking

Although the strength was reduced, the replacement of cement with coarse and fine sawdust could reduce the drying shrinkage due to the internal curing of sawdust [68]. The authors also suggested that the reduction in drying shrinkage could reduce the micro-cracking and thus improve the durability. The use of sawdust in cementitious materials could promote both sustainable development and durable infrastructures.

4 Challenges with the application of lignocellulosic biomass

Although some efforts have been made to expand the application of lignocellulosic biomass in cement-based composites, there remains some concerns. Firstly, lignocellulosic biomass is reported to retard cement hydration and prolong its setting time. On one hand, the degradation of lignin, hemicellulose, and impurities in lignocellulosic biomass produces some saccharides that disturb the formation of calcium silicate hydrate and affect cement hydration [27,31,70-73]. On the other hand, some researchers argue that a protective layer around the hydrated cement grains or a chelate complex with the cations present in the hydrated cement can be formed, which prevents the inside zone of cement grains from hydrating [74]. The delaying of cement hydration can slow down the strength development of cement-based composites. For engineering projects in a normal construction environment, this phenomenon may not be desired because it may expand the construction period. However, it should be pointed out that for those projects in hot environment, it may be favorable because fast cement hydration may bring in some problems, such as cracking and poor strength development. Secondly, lignocellulosic biomass possesses high water absorption/desorption capacity, generating volumetric changes in the cement-based composites. Thus, a complex and poor interfacial transition zone can be formed around lignocellulosic biomass, showing bad compatibility [75], which may harm the mechanical properties of cement-based composites. Finally, the impurities (wax, pectin, etc.) on the surface of lignocellulosic biomass provide some barriers for the interlocking with the cementitious matrix [76]. In light of the above-mentioned concerns, various strategies are proposed to make modifications on lignocellulosic biomass, including removing amorphous components (lignin, hemicellulose, and impurities), changing the structure, and preventing direct contact with cement-based composites, as introduced below.

5 Modification on lignocellulosic biomass

5.1 Ordinary Heating Treatment

Ordinary heating treatment refers to heating the lignocellulosic biomass at a temperature of less than 200 ℃ in an air-circulating environment. The crystallinity of cellulose in lignocellulosic biomass can be improved after heating treatment. Wei et al. found that the treatment could increase the crystallinity of sisal fibers from 20.27% to 22.67%, 26.42%, and 23.86% when heating for 4h, 8h, and 16h, respectively [77]. The increased crystallinity of cellulose is favorable to the tensile properties of lignocellulosic biomass. This is confirmed by Rong et al. who suggested that the heating treated sisal fibers at 150 ℃ for 4 h showed superior tensile properties [78]. Moreover, the heating treated lignocellulosic biomass could improve the strength and durability of cement-based composites compared to the untreated one. Yew et al. indicated that concrete with heat-treated oil palm shell (OPS) aggregates has higher compressive strength than that with untreated OPS aggregates; the rapid chloride penetration test showed that the former has lower passed charge than the latter, indicating improved resistance to ion penetration, which may be due to the reduction in inner conductivity of pores and less capillary porosity [79]. However, the heated OPS could increase the drying shrinkage of concrete compared to untreated OPS, which may be because the heat treatment on OPS changes the physical properties of aggregates, resulting in better adhesion between cement paste and OPS [79].

5.2 Torrefaction Treatment

Torrefaction (mild pyrolysis) is a thermal treatment process that is conducted in the inert gas atmosphere at a low temperature between 200 and 300 ℃ [80]. Because of the distinct chemical and thermal reactivity of cellulose, hemicellulose, and lignin, they undergo different chemical transformations during torrefaction [81]. At low torrefaction temperatures, most of the hemicelluloses degrade into volatile components; however, cellulose and lignin are more thermostable and less devolatilized [81]. When the torrefaction temperature is higher than 250 ℃, cellulose (mainly amorphous part) takes greater participation in the devolatilization, resulting in considerable mass losses [82]. During the torrefaction process, the lignin structure is thermally modified by way of cleavage of aryl ether linkages (e.g., β-O-4 linkages) and condensation-like reactions. In general, after the torrefaction treatment, most hemicellulose can be removed, while there remains much cellulose and lignin. Because the degradation of hemicellulose produces some saccharides that delay cement hydration, the torrefied lignocellulosic biomass can delay cement hydration less through removing hemicellulose and some impurities. This is verified by Govin et al. who reported that torrefied wood shavings at 240 ℃ and 260 ℃ could promote gypsum dissolution or consumption, ettringite formation, and hydration of silica phases compared to natural wood shavings [83]. Similarly, the torrefaction treatment on bagasse at 200 ℃ and 250 ℃ was reported to increase the setting time of cement-based materials compared to untreated bagasse [84]. Not only cement hydration, the interfacial transition zone between wood chips and the cement matrix also benefits from torrefaction treatment, which can reduce the side effect of wood chips on the flexural strength and compressive strength of cement mortar [60].

5.3 Boiling Treatment

Boiling treatment refers to immersing lignocellulosic biomass into boiling water for a certain duration and then rinsing it with water. The boiling treatment can remove some water-soluble substances, accountable for setting delay and incompatibility problems with the matrix, and clean the surface of lignocellulosic biomass [85]. Ali-Boucetta et al. reported that the setting time for the non-fiber mixture increased from 4 h 10 min to 5 h 30 min if raw date palm fibers were involved; however, compared to untreated fibers, the water boiling treated fibers reduced the delaying time by 15%-33%, proportional to the boiling time [86]. The main reason may be attributed to the reduction of water-soluble sugars, which is confirmed by Sellami et al. who found that the amounts of water-soluble sugars for untreated Diss, boiled-but-not-washed Diss, and boiled-and-washed Diss were 30.78%, 1.95%, and 0.72%, respectively [87]. In addition, the boiling treatment could improve the fiber toughness, fiber tensile strength, and fiber/concrete bonding strength [88, 89]. All improvements caused by boiling treatment lead to the increase of flexural strength and compressive strength of cement-based composites [85-87].

5.4 Surface Coating or Impregnation Treatment

Surface coating or impregnation treatment refers to using coverings on lignocellulosic biomass to prevent direct contact with the cement-based composites. The commonly seen coverings include polymers, oil, and even cement paste. Ahmad and Fan adopted polyester, vinylester, polyurethane, and epoxy to coat sisal fibers, respectively, and concluded that the mechanical properties of cement-based composites could be improved by coated fibers, with polyurethane coating being the best, which was possibly attributed to the significant improvement in interfacial bonding [90]. Similar conclusions were also reported by other researchers who showed that the styrene-butadiene polymer coated lignocellulosic biomass could increase the strength and stiffness of cement-based composites as compared to the untreated one by improving the bonding with the matrix [91,92]. Regarding oil coating, it was reported that the linseed-oil coating on boiling treated Diss fibers increased the resistance to considerable tensile stresses compared to natural Diss fibers; in compression, the brittle failure did not happen, and the samples almost remained intact [87]. In addition, cement impregnated miscanthus could result in higher compressive and flexural strengths than untreated miscanthus, which was not only due to the rougher surface and higher density associated with treated miscanthus but also due to the disappearance of the delaying effect of sugars after impregnation [20]. However, the samples with cement impregnated miscanthus still had much lower compressive and flexural strengths than those

without miscanthus, which was mainly attributed to the defect bond between miscanthus aggregates and cement paste.

5.5 Hornification Treatment

Hornification treatment refers to the application of drying/wetting cycles on lignocellulosic biomass. After treatment, the polysaccharide chains of cellulose can be closely grouped, which provokes lower water absorption capacity and higher dimensional stability of lignocellulosic biomass by reducing the lumen [70,93]. This is favorable to the fiber-matrix interface performance. In addition, the cellulose content may also increase because of the removal of remains components during hornification [94]. It is widely known that cellulose contributes the most to the mechanical properties of fibers, so the increase of cellulose content may improve the mechanical properties of hornificated fiber reinforced cement-based composites. This is confirmed by some researchers who showed that the hornification treatment not only increased the stiffness and tensile strength of lignocellulosic fibers, without deteriorating their crystallinity index, but also improved the specific energy and modulus of rupture of cement-based composites [70,92].

5.6 Alkaline Treatment

Alkaline treatment (also known as mercerization) is a commonly seen chemical treatment method on lignocellulosic biomass. The hydroxyl groups of lignocellulosic biomass react with alkaline substances to reduce hydrogen bonding. The alkaline treatment can remove the lignin, hemicellulose, pectin, and wax, and even increase the crystallinity index of cellulose [95-97]. Of all alkaline substances, sodium hydroxide is the most commonly used. Sodium hydroxide can not only clean the surface of lignocellulosic biomass but also change the native cellulose I to cellulose II [98]. By chemical reaction, the OH-groups can be converted into ONa-groups. But the linked Na-ions can be removed while rinsing with water, which will result in the formation of a new crystalline structure, namely cellulose-II, that is thermodynamically more stable than cellulose-I. The alkaline treatment not only roughens the surface of lignocellulosic biomass, improving the interfacial adhesion with the matrix, but also reduces the retarding effect on cement hydration, which can increase the mechanical properties (including stiffness, toughness, and strength) of cement-based composites [71,99-102]. In addition, the alkaline treated lignocellulosic biomass could reduce the crack width and improve the multi-cracking property of cement-based composites, which endowed the composites with autogenous healing capacity when exposed to wet/dry cycles [103]. The non-cellulosic substances of lignocellulosic biomass can be reduced further by increasing the severity of alkaline treatment (concentration, temperature, and duration) [104]. However, the treated lignocellulosic biomass under too severe conditions may harm the performance of cement-based composites. de Klerk et al. reported that the post-peak strength and fiber/matrix bonding performance of cement-based composites could be enhanced at low concentrations of NaOH (2%, 6%, and 10%); however, the opposite result was observed at high concentrations of NaOH (20% and 30%) [105].

5.7 Bleaching Treatment

Bleaching treatment can be conducted to remove lignin from lignocellulosic biomass, with sodium chlorite ($NaClO_2$) being the most commonly used. $NaClO_2$ is highly soluble and stable in water. It is also widely known that the $NaClO_2$ is quite stable in alkaline condition [106]. To be an active bleaching agent, $NaClO_2$ should be used in acid condition. The decomposition of $NaClO_2$ produces chlorine dioxide (ClO_2) that reacts with lignin to get it removed [32,107]. Abdel-Halim even introduced a new way to delignify sugarcane bagasse by using both $NaClO_2$ and hexamethylene tetramine ($(CH_2)_6N_4$) [107]. The bleaching efficiency is determined by the concentration of hexamethylene tetramine, bleaching time, and PH. A higher content of $NaClO_2$ will be decomposed at a given bleaching time if higher concentration of hexamethylene tetramine is used; prolonging the bleaching time at a given hexamethylene tetramine concentration will increase the decomposition of hexamethylene tetramine; the gradual liberation of ammonia adjusts the PH through buffering effect, which can slow down the decomposition rate of sodium chlorite, resulting in increased efficiency in removing lignin. In addition to $NaClO_2$, hydrogen peroxide (H_2O_2) is another bleaching agent to remove lignin and is gradually replacing $NaClO_2$ because of its environmental friendliness [108,109]. Not only removing lignin, the

bleaching treatment also increases the exposure of cellulose and improves its crystallinity index, which can reduce the negative effect of lignocellulosic biomass on cement hydration [110,111].

5.8 Other Treatments

The modification methods on lignocellulosic biomass are various, and some other treatment methods are also adopted by researchers besides the above-mentioned treatments. For example, Amiandamhen et al. compared three treatment methods ($CaCl_2$, hot water, and a combination of hot water and $CaCl_2$) on kenaf fibers and found that cement boards achieved the best physical and mechanical properties through the combination of hot water and $CaCl_2$ [112]. However, not all treatments can improve the performance of cement-based composites. Ali et al. indicated that the fiber-concrete bonding strength could be reduced when the fibers were treated with 1% calcium chloride and 0.25% sodium alginate solution, which might be due to the reduction in the tensile strength and strain of fibers [88].

6 Conclusions

Nowadays, people are becoming more and more aware of the importance of sustainability of building materials. The involvement of lignocellulosic biomass in cementitious materials not only provides an alternative to dispose of lignocellulosic biomass wastes to a certain extent but also promotes the development of green building materials. By reviewing the existing research, some important conclusions are summarized below.

The lignocellulosic fibers are usually used as reinforcements in cementitious materials. They are reported to improve the shrinkage, cracking, toughness, and heat insulation properties of cementitious materials. In addition, the lignocellulosic fibers normally improve the flexural strength or tensile strength of cementitious materials; however, regarding compressive strength, there is not a consistent conclusion about whether it is increased or reduced.

The use of lignocellulose biomass as lightweight aggregates is reported to reduce the mechanical properties (including compressive strength, splitting tensile strength, flexural strength, etc.) of cementitious materials but is proven to be able to develop lightweight concrete. Moreover, it can improve the toughness and heat insulation capacity without significantly harming the durability of cementitious materials. However, it seems to increase the drying shrinkage of cementitious materials, which is a big challenge.

The study on the use of lignocellulosic biomass wastes to partially replace cement is limited so far. The existing studies indicate that the replacement of cement with lignocellulosic biomass can reduce the mechanical properties of cementitious materials due to the increased porosity and the retarding effect on cement hydration but, on the bright side, reduce drying shrinkage due to internal curing.

There are some concerns with the involvement of lignocellulosic biomass in cementitious materials, including delaying cement hydration, high water absorption capacity, hindering interlocking with the cementitious matrix, etc. To solve these problems, several treatment methods are proposed to change the structure or remove amorphous components of lignocellulose biomass or prevent it from directly contacting cementitious materials. It is shown that the properly modified lignocellulosic biomass can further improve the properties (strength, cracking resistance, etc.) of cementitious materials compared to raw lignocellulosic biomass. Therefore, sustainable cementitious materials with desired performance can be developed using modified lignocellulosic biomass to meet the needs of engineering projects.

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CRediT authorship contribution statement

Aofei Guo: Investigation, Funding acquisition, Writing – original draft, Formal analysis. **Zhihui Sun**: Conceptualization, Supervision. **Hu Feng**: Investigation, Supervision, Writing – review & editing. **Hong Shang**: Writing – review & editing. **Noppadon Sathitsuksanoh**: Writing – review & editing.

Conflicts of Interest

The authors declare that they have no conflicts of interest to report regarding the present study.

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