

# Non-Conventional Mineral Binder-Bonded Lignocellulosic Composite Materials: A Review

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The construction industry suffers from unsustainability and contributes more than any other industrial sector to carbon emissions that lead to global warming. Increasing economic and environmental concerns related to conventional energy- and CO<sub>2</sub>-intensive building materials have propelled the rapid and sustained expansion of research in the area of plant-based inorganic mineral binder-bonded materials for the construction industry. The resulting composites can be qualified as eco-responsible, sustainable, and efficient multifunctional building materials. So far, most of these research efforts have not received as much attention as materials based on ordinary Portland cement (OPC). To address this gap, this review focuses on mineral binder-based lignocellulosic composites made from non-conventional inorganic mineral binders/cements with low embodied energy and low carbon footprint, namely hydrated lime-based binders, magnesium-based cement, alkali-activated cement, and geopolymers, as sustainable alternatives to OPC-bonded lignocellulosic composites (state-of-the-art). The emphasis here is on the application potentials, the influence of production parameters on the material properties/ performance, and recent advancement in this field. Finally, a prediction is provided of future trends for these non-conventional mineral binder-bonded lignocellulosic composites.

*Keywords:* Non-conventional mineral binders; Inorganic bonded composites; Sustainable construction; Natural fibers; Bio-based materials

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## INTRODUCTION

Lignocellulosic composites are engineered products manufactured from wood or other lignocellulosic raw materials, together with a suitable matrix/binder. Depending on the nature of the matrix, they are classified as organic matrix composites such as particleboards, fiberboards, and oriented strand boards (OSB) as well as inorganic mineral matrix composites such as those made from ordinary Portland cement (OPC), gypsum, and magnesia, which have been widely used in the manufacture of reinforced panels and structures in construction. Organic matrix-bonded composites are usually processed, depending on the raw materials, specific design, and end-use, by spray/hand lay-up under heat and pressure application, resin transfer molding, *etc.* They can also be processed by extrusion, compression molding, and injection molding as used for wood-plastic composites (WPC) (Nagavally 2016; Rajak *et al.* 2019). The focus of this study, however, is on inorganic mineral binder-bonded composites for construction applications; these are products containing lignocellulosic materials fully encapsulated in a continuous inorganic matrix. Inorganic-bonded composites are generally processed at ambient temperature by

casting, compact casting, and pressure (Irle *et al.* 2013; Stark *et al.* 2010), although, in some cases, additional heat may be applied to aid curing.

The most common inorganic-bonded composites incorporate gypsum, ordinary Portland cement (OPC), and magnesia cement as the matrix. Gypsum- and magnesia-bonded composites are mostly restricted to interior applications due to their moisture sensitivity, while the more durable OPC composites are used in both interior and exterior applications. The growing interest and adoption of lignocellulosic materials to manufacture mineral binder-bonded composites for construction applications are mainly triggered, among other advantageous properties, by their wide geographical distribution and ease of handling/processing (Mohammed *et al.* 2015). The matrix/binder system adopted to produce inorganic mineral binder-bonded composites, however, has far-reaching impacts on the properties, performance, and environmental profile (Shalbfafan *et al.* 2020).

Generally, the construction industry is not sustainable, owing to the high consumption of materials with high energy content and associated CO<sub>2</sub> emissions (Murphy *et al.* 2010; Ghavami *et al.* 2017). In a bid to reduce such negative impacts, scientists are challenged to develop more innovative customized, environmentally sound, non-conventional materials for indoor and outdoor use. Over the years, considerable efforts have been made in the development and application of novel inorganic mineral binder systems that show remarkable potential for producing alternative multifunctional engineered materials for construction applications (Walker and Pavía 2014; Sarmin *et al.* 2014; Miller *et al.* 2017; Soltan *et al.* 2017; Kang *et al.* 2019). These materials must feature sustainability characteristics that are superior to existing conventional materials, while retaining the main advantages of mineral binder-bonded materials in terms of higher mechanical properties, toughness, fire resistance, and durability against environmental influences and biodeterioration agents (Shalbfafan *et al.* 2016; Bahrami *et al.* 2019; Shalbfafan and Thoemen 2020). Non-conventional mineral binder-bonded lignocellulosic composites have increasingly emerged as an important class of alternative construction materials. They are obtained by combining the functional properties of the mineral binders with the advantages of lignocellulosic materials to give customized products of enhanced thermal stability, moisture preclusion, and improved mechanical performance. These products can meet the critical requirements of low carbon footprint and sustainability as well (Sarmin *et al.* 2014; Shalbfafan *et al.* 2016; Moslemi and Begum 2017; Jin *et al.* 2017; Shalbfafan *et al.* 2020). Additionally, non-conventional mineral binder-bonded lignocellulosic composites have the advantage of being able to utilize a wide range of organic and inorganic raw materials, including the valorization of agricultural residues and industrial wastes. The resulting products not only equal the conventional materials from OPC in terms of performance and cost-effectiveness, but they are also ecologically responsible.

Research related to the production of composites using inorganic mineral binders is in rapid flux. A majority of the reviews on this topic have focused primarily on Portland cement-bonded lignocellulosic composites, as these are the most widely used class of low-cost inorganic bonded composites (Frybort *et al.* 2008; Kang *et al.* 2019). Recently, however, other types of inorganic mineral binder materials have been increasingly studied. Some of these are of ecological importance and exhibit less embodied energy. Therefore, this study reviews the developments in the use of these rather lesser-described binders for mineral binder-bonded lignocellulosic composite technologies.

This study is structured in two major sections: the first section discusses mineral binder-bonded lignocellulosic composites based on hydrated lime binders, magnesium-

based cement, and alkali-activated and geopolymer binders, focusing on their application potentials, the influence of production parameters on their properties/performance, as well as recent advancement. Another section provides a conclusion and description of the future trends for non-conventional mineral binder-bonded lignocellulosic composites.

## LIME-BASED LIGNOCELLULOSIC COMPOSITES

Lime is a calcium-rich inorganic mineral composed primarily of oxides and hydroxide, with several applications in construction, environmental protection, industrial and agricultural processes. Generally, lime is obtained by burning limestone ( $\text{CaCO}_3$ ) at temperatures above  $900\text{ }^\circ\text{C}$  to produce highly caustic quicklime ( $\text{CaO}$ ), with a corresponding release of  $\text{CO}_2$  (Eq. 1). The quicklime ( $\text{CaO}$ ) is subsequently slaked with water to produce hydrated lime or air lime ( $\text{Ca(OH)}_2$ ) (Eq. 2) (Kang *et al.* 2019). Lime can also be recovered as a by-product from sugar refineries (lime sludge), welding (calcium carbide sludge), acetylene, and paper industries, where lime materials are used as chemical feedstocks. Lime-based materials can partially replace and/or substitute high embodied energy materials for certain applications in construction. This increases the overall ecologically friendly nature of the products by reducing or eliminating  $\text{CO}_2$  emissions generated during the production of buildings and building material, mostly associated with the consumption of ordinary Portland cement (OPC) (Walker *et al.* 2014).



In itself, pure hydrated lime ( $\text{Ca(OH)}_2$ ) or air lime is a common non-hydraulic binder whose hardening depends on a process called carbonation (*i.e.*, reaction with  $\text{CO}_2$  from the air) to produce calcium carbonate. In construction applications, however, limes are rarely used alone as the main cementitious binder but rather in combination with reactive additives (clay or pozzolana) to form a hydraulic cement. These exhibit better binding efficiency, shorter setting time, and enhanced strength properties, while retaining the sufficient durability characteristic of lime materials. This type of hydraulic cement can be used alone or as a partial substitute for OPC in non-performance secondary applications such as masonry, plastering, and production of composites for walling and insulation (Billong *et al.* 2011; Fic *et al.* 2013; Brzyski *et al.* 2017; Page *et al.* 2017).

### Lime–Lignocellulosic Composites, Performance

While lime has been a construction material since antiquity (Carran *et al.* 2012; Veiga 2017; Shao *et al.* 2018), its use in modern construction was curtailed with the introduction of OPC in the 19<sup>th</sup> century, which possesses better strength and other performance properties at relatively lower cost (Kang *et al.* 2019). However, increasing demand for low-energy, carbon-negative, and sustainable building materials has reignited interest in the development of lime-based construction materials of improved properties using bio-aggregates (Cizer *et al.* 2010; Billong *et al.* 2011; Walker *et al.* 2011; Lorca *et al.* 2014; Pavlík and Užáková 2016; Payá *et al.* 2017; Kang *et al.* 2019; Abdellatef *et al.* 2020).

*Acoustic, thermal, and hygrothermal properties*

Lime-lignocellulosic composites have been well studied in terms of their acoustic, thermal, and hygrothermal properties (*i.e.*, movement of heat and moisture). One of such materials is the lime-hemp concrete or “Hempcrete”, which was developed in the late 1980s/early 1990s in France as a sustainable alternative material for the restoration of historic buildings and to lessen the weight of ordinary Portland cement (OPC) concrete (Dai *et al.* 2010; Walker *et al.* 2014). It consists mainly of hemp shives (Kurd), the woody core of the hemp stalk after fibers have been extracted, and a lime-based binder. Other additives such as pozzolans, natural hydraulic lime, and a certain percentage of OPC may be included to enhance the binder properties such as reduced setting times, improved binder efficiency, and mechanical performance (Pochwała *et al.* 2020). Lime-hemp concrete exhibits excellent acoustic, thermal and hygrothermal properties as well as moisture transport qualities. These are mainly due to the highly porous structure (macropores) of the hemp shives, which allow it to absorb an enormous amount of water, and show good transpirability (Glé *et al.* 2011; Arizzi *et al.* 2015). Consequently, the highly porous lightweight composite displays higher thermal capacity, lower thermal conductivity, and higher water vapor permeability with the ability to buffer moisture and temperature changes better than OPC concrete (Evrard and De Herde 2010; Walker and Pavía 2014; Kinnane *et al.* 2015; Abdellatef *et al.* 2020). Accordingly, Hempcrete is increasingly being used in parts of Europe and Canada as a sustainable envelope material for wall cladding, exterior coatings, floor tiling, and insulation of ceiling and roof (Murphy *et al.* 2010; Page *et al.* 2017; Pochwała *et al.* 2020).

Increasing the flax fiber content and the addition of a foaming agent lowered the thermal conductivity ( $\lambda$ ) of flax-lime materials to the range of 0.13 to 0.14 W/m K and 0.20 W/m K, respectively, thereby resulting in a low-density material (Fic *et al.* 2013). Rahim *et al.* (2015) investigated the moisture-related properties (sorption isotherm and water vapor permeability) and moisture buffer values (MBV) of flax-lime concrete, in comparison to hemp-lime concrete. They found that the flax-lime concrete exhibit high hygric performance as well as a moisture buffer capacity equal to that of the hemp-lime material, with MBV values of 2.02 g/ (m<sup>2</sup> %RH) and 2.27 g/ (m<sup>2</sup> %RH) for hemp and flax concretes, respectively. These findings demonstrate that flax materials, when combined with a lime-based binder, can be upcycled into a sustainable green “flax-lime concrete” with potential functional applications in buildings. The capillary water absorption of hybrid hemp-flax lime-based concrete with a 9:1 hemp shive to flax fiber ratio was determined more by the overall aggregates/binder ratio, which is influenced by the complex porosity and increased tortuosity of the capillary networks, than by the water absorption capacity of the individual fibers. At the same time, the thermal insulation properties remained within acceptable limits (Page *et al.* 2017).

Belakroum *et al.* (2018) studied the thermal and acoustic properties of lime-date palm fiber composites. At a fiber volume range of 20 to 50%, the thermal conductivity ranged from 0.21 to 0.091 W/m K and sound absorption capacities in medium and high frequencies were enhanced. Moisture buffering was also higher in these variants than in those with fewer fibers. The use of various particle dimensions in lime-OPC-wood particle composites resulted in different densities and thermal conductivity values of the composites (Park *et al.* 2019). Thermal conductivity ranged from 0.0742 to 0.1078 W/m K for big and small particles, respectively. Furthermore, wood-lime boards with larger particles had lower moisture content at 60% relative humidity (RH) but higher moisture

content at 70% RH, implying that for relatively low RH, wood-lime boards with larger particles are more suitable as building materials.

### *Mechanical properties*

Table 1 shows the production parameters and strength properties of various lime-based lignocellulosic composites. Hempcrete is typically applied as a “light work” non-structural building skin, cast around load-bearing structural frames in the construction and thermal upgrading of buildings (Walker and Pavía 2014). Substantial initial compressive strength development of the Hempcrete at the time of application is important to support its wet weight, which is more than twice its dry weight (Hirst *et al.* 2010; Murphy *et al.* 2010; Walker and Pavía 2010).

Owing to the rough surface, flax fibers exhibited very good adhesion with the lime matrices, resulting in a ductile composite material (Fic *et al.* 2013). Compressive strength ranged from 0.45 to 0.65 MPa, as a function of the material bulk density. The lowest strength and bulk density were observed in materials in which a foaming agent was applied due to high corresponding porosity. Hydrophobization of flax straw by impregnation with flaxseed oil resulted in an accelerated binding process of the lime matrix with improved mechanical properties. The exact mechanisms, however, by which this effect of the accelerated binding process occurs in the presence of flax oil need to be further elucidated. The incorporation of flax fibers to a hybrid hemp-flax lime-based composite with a hemp-shive-to-flax-fiber ratio of 9:1 significantly enhanced the compressive strength, compactness, and ductility of the material with a corresponding reduction in shrinkage (Page *et al.* 2017).

Sunflower piths, due to their morphological similarities with hemp shives, were investigated in terms of their geometrical features related to the casting technique (Nozahic *et al.* (2012). The hemp and sunflower materials showed large similarities in terms of the mechanical properties of the resulting concrete. As the stalk is a sunflower by-product, which is available in large quantities, it is therefore a possible alternative to hemp shives for use in plant concrete. Unlike most lignocellulosic fiber composites, where the interface between the aggregates and the binder is the weakest point of the system, in lime-sunflower piths composites it is mainly the binder that provides the mechanical performance.

Three batches of relatively large fibers of giant reed (*Arundo donax*) with a length of 4, 8, and 12 cm were incorporated into a hydraulic lime mortar matrix to produce composites with a target application as ductile prefabricated bricks (Badagliacco *et al.* 2020). The addition of fibers led to a significant increase of the post-fracture flexural toughness compared to unreinforced mortars, but slightly reduced compressive strength. Of the three fiber batches, the medium variant (8 cm) had the greatest effect on strength. The extraordinarily high growth rate (similar to bamboo) and capability to produce huge amounts of biomass make giant reed (*Arundo donax*) an interesting candidate for application in composite production.

Di Bella *et al.* (2014) compared the effect of two types of natural fibers, namely sisal (*Agave sisalana*) and kenaf (*Hibiscus cannabinus*), and synthetic polypropylene fibers on the mechanical and durability properties (freeze/thaw and salt spray test) of the resulting lime plasters. The lime plasters reinforced with natural fibers resisted cracking and exhibited mechanical properties as well as freeze/thaw resistance comparable to that of plasters with polypropylene fibers. They exhibited higher resistance to degradation after aging time in the salt spray environment, with lower weight loss (14%) after exposure compared to those with polypropylene (18%). The resistance was attributed to the

hydrophilic nature of the natural fibers, which enables them to partially absorb water from the salt solution and thus preserving the plaster (Di Bella *et al.* 2014). Thus, sisal and kenaf fibers can serve as suitable alternatives to polypropylene fibers in lime plaster applications.

**Table 1.** Overview of Lime-based Lignocellulosic Composites

Type of fibers	Amount of fibers (%)	Length of fibers (mm)	Type of casting	Density (kg/m <sup>3</sup> )	Fillers	Maximal strength of composites reinforced with fibers (MPa) *	Reference
Hemp shives	n/a	n/a	Casting	308.3 to 357.8	n/a	n/a	Pochwała <i>et al.</i> (2020)
	10 - 75 (wt%)	n/a	Casting	n/a	n/a	<b>FI:</b> 4 MPa for 50% fiber, <b>CM:</b> ~17 MPa	Murphy <i>et al.</i> (2010)
	n/a	n/a	Casting	265.8 to 461.2	Metakaolin and OPC	n/a	Barnat-Hunek <i>et al.</i> (2015)
	20 – 22 (wt%)	n/a	Casting d-100mm, h-200 mm	290 to 350	Metakaolin and crushed bricks	<b>CM:</b> 0.47 MPa for 22% fiber	Abdellatef <i>et al.</i> (2020)
Flax fibers	n/a	n/a	Casting, 50 x 50 x 50 mm <sup>3</sup> and 100 x 100 x 100 mm <sup>3</sup>	250 to 500	Hemp fibers/shives	<b>CM:</b> 0.25 MPa	Page <i>et al.</i> (2017)
Flax fibers	n/a	10 to 20 mm	Casting, 10x10x10 mm <sup>3</sup>	440 to 1290	Microsilica, OPC	<b>CM:</b> 0.65 MPa	Fic <i>et al.</i> (2013)
Sisal fibers	0.2 (wt%)	5 mm	Casting	n/a	Polypropylene fibres	<b>FI:</b> 0.39 MPa <b>CM:</b> 0.39 MPa	Di Bella <i>et al.</i> (2014)
Kenaf fibers	0.2 (wt%)	5 mm	Casting	n/a	Polypropylene fibres	<b>FI:</b> 0.30 MPa <b>CM:</b> 0.33 MPa	Di Bella <i>et al.</i> (2014)
Palm fibers	20 to 50 (wt%)	0.063 to 5	Casting, d-80 mm, h-160 mm	680 to 950	n/a	<b>CM:</b> 0.6 MPa for 20% fiber	Belakroum <i>et al.</i> (2018)
Giant reed fibers	0.5 to 2 (wt%)	40 to 120	Casting, 40 x 40 x 160 mm <sup>3</sup>	1788 to 1875	n/a	<b>CM:</b> 7.51 MPa for 1% fiber	Badagliacco <i>et al.</i> (2020)
Wood particles	16.7 (wt%)	0 to 1 and 2.36 to 4.75	Casting, 100 x 100 x 20 mm <sup>3</sup>	875 to 984	OPC	n/a	Park <i>et al.</i> (2019)
Sunflower pith particles	5.3 (wt%)	10 to 30	Compression casting, 40 x 40 x 40 mm <sup>3</sup>	1084 to 1310	Pumice sand	<b>CM:</b> 4.52 MPa for 5.3% fiber	Nozahic <i>et al.</i> (2012)

\* FI: Flexural Strength, CM: Compression Strength, n/a: information not available

## Influence of Production Parameters on the Strength Properties of Lime Composites

A well-known property of hydrated-lime is that it can be easily combined with a wide range of natural and synthetic additives to yield materials of improved properties (Walker and Pavía 2010; Payá *et al.* 2017; Martínez-García *et al.* 2019). Various studies have investigated the influence of binder composition and hydraulicity on the mechanical strength of lime-hemp composites and stiffness (Murphy *et al.* 2010). Hirst *et al.* (2010) reported that strength properties of Hempcrete are not prominently dependent on the individual strength of the binder, but are rather a function of a complex interaction between the hemp and lime, influenced by mixing, density, and age.

### *Hydraulicity of lime*

A more hydraulic lime binder resulted in a higher rate of mechanical strength gain and ultimate compressive strengths than pure hydrated lime relying solely on carbonation for strength development. The hydraulic additions in the commercial binder induce a partial hydraulic set, which is a faster process (Murphy *et al.* 2010). However, the influence of the lime binder hydraulicity on strength development, both in flexion and compression, decreased with increasing hemp content, particularly in compressive strength. On the other hand, the flexural strength of the materials was more influenced by the density of the composites compared to compression strength. Increasing binder hydraulicity induced by hydraulic lime and Portland cement improved the initial (24 h to 5 days) strength development and durability of the Hempcrete materials but did not significantly affect their ultimate strength after one year (Walker *et al.* 2014). Other authors have also reported advancement in the use of various agricultural wastes/residues as pozzolanic additives to improve the hydraulic properties, binding efficiency, and ecological profile of hydrated lime-based binders for construction purposes (Billong *et al.* 2011; Payá *et al.* 2017; Nayaka *et al.* 2018; Martínez-García *et al.* 2019).

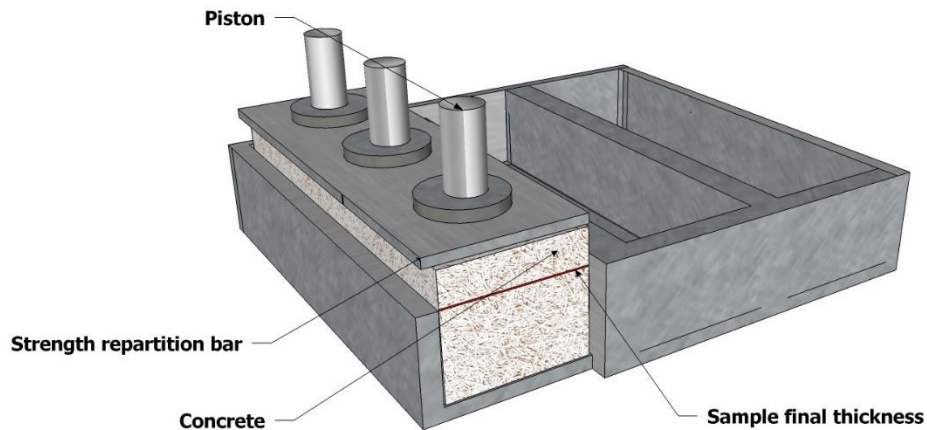
### *Casting technique*

In addition to the use of pozzolanic and hydraulic binder additive blends in formulated lime binders for lime-lignocellulosic composite production, the casting technique is another production parameter with a critical influence on the performance properties of the materials. Various molding techniques have been employed depending on the geometry, dimensions, and volume of fibers added in the lime paste.

Casting with a subsequent vibration process to eliminate the volume of pores, and hence, the porosity of the final product is a conventional casting technique to produce mineral binder-bonded lignocellulosic composites with a low volume of incorporated fibers mixed in a highly workable paste. The major objective is to achieve homogenous dispersion, adequate bonding, and sufficient interconnection between the aggregate and the matrix and, as a consequence, better mechanical performance of the composite. Casting by compaction (Fig. 1) is usually utilized for higher ratios of lignocellulosic aggregate/binder. Metal molds equipped with a movable bar are compressed by an external force compacting the binder-aggregate mixture until a specific thickness is achieved.

Nozahic *et al.* (2012) concluded that casting by compaction shows the high capability of the plant aggregate to be reorganized in layers and then to form concrete with orthotropic mechanical behavior. This was found to be an interesting way to greatly increase the compressive strength without increasing the density of the material in the same proportion, as it results in orthotropic properties. Indeed, the effectiveness of this technique

is determined by the shape of the plant particle, because long and flat objects have the ability to be reorganized into layers during compaction.



**Fig. 1.** Schematic diagram illustrating the compaction casting technique

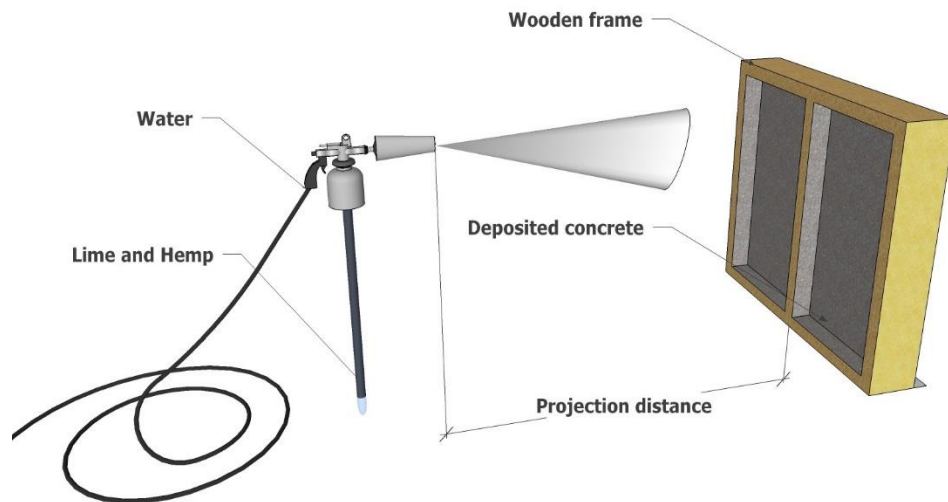
The influence of the casting technique has also been investigated in terms of reducing the long setting and drying times of lime-based composites. This in turn shortens the labor time on site, reduces costs, and improves the initial hydraulic set of the concrete. A study investigated the influence of a projection casting process (Fig. 2) varied at distances from 0.5 m to 3.0 m on the mechanical (flexural strength, compression strength, and hardness) and thermal properties of Hempcrete.

Projection casting is a recently developed process in which a mixture of lime, shives, and water is projected either directly onto a wall formwork or into a block mold (Elfordy *et al.* 2008). Initially, a dry premix of lime and shives is conducted through a hose by air, and pressurized water is added just before the hose outlet. The projection process was adopted to minimize the time for the hemp component to absorb significant amounts of water, on the one hand, and to limit the total setting time to that required for lime carbonation, on the other, while achieving better compaction and higher density. A significant increase in the mechanical properties of the Hempcrete material was observed with increasing mortar density as a function of the projection distance. However, highest strength was achieved at a 1-m-projection distance, and a moderate density variation occurred within a given projected block, both along the length and across the width. This observation is comparable to the classical spray coating processes, where too short distances result in “splashing” of the projected material on the substrate. This leads to a poor stacking of the deposited particles. On the other hand, when the projection distance increases and becomes too long, the kinetic energy of the particles decreases and the compaction of the material on the substrate becomes less efficient (Elfordy *et al.* 2008). The authors attributed the improved mechanical properties to the higher compaction, and hence, higher density achieved with the projection casting method. In contrast, almost no compaction occurs with the conventional mixing and casting technique, resulting in highly porous materials with very low bulk density and poor mechanical strength after curing.



Other studies have also confirmed that density, which is a function of compaction, as a crucial production factor affecting the performance of the lime-hemp concrete (Barna-Hunek *et al.* 2015; Abdellatef *et al.* 2020).

Nonetheless, the higher compaction and density values achieved with the projection casting process (Elfordy *et al.* 2008) meant that its usefulness as a viable thermal insulator in temperate climates was compromised due to the increased thermal conductivity of Hempcrete. However, it opens up new possibilities for the elaboration of lime-based load-bearing composites with bio-aggregates in applications where a favorable compromise between thermal insulation and mechanical strength properties can be achieved.



**Fig. 2.** Schematic diagram illustrating projection casting of lime-based composites

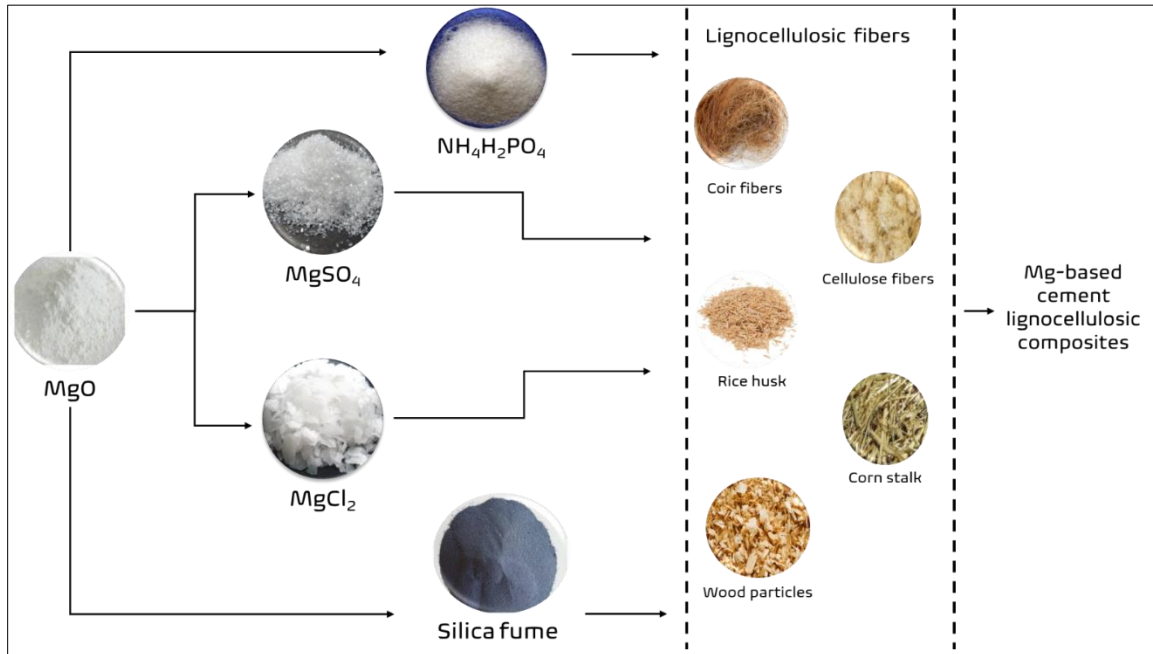
## LIGNOCELLULOSIC COMPOSITES FROM MAGNESIUM-BASED CEMENT

The major motivation for the development of MgO-based cement has been driven by environmental aspects such as lower calcination temperatures compared to those of Portland cement. Equally, the ability of MgO to absorb CO<sub>2</sub> from the atmosphere (as much CO<sub>2</sub> as was emitted during manufacture) makes it a carbon-neutral cement (Walling and Provis 2016). The following presents recent research on various Mg-based composites containing a wide range of fibers and their properties (Fig. 3.).

### Magnesium Phosphate Cement (MPC)

Magnesium phosphate cement (MPC) is a new type of fast-setting inorganic mineral binder. Curing is based on an acid-base neutralization reaction of metal cations from calcined magnesia (MgO) with phosphate anions from ammonium dihydrogen phosphate (NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub>) or potassium dihydrogen phosphate (KH<sub>2</sub>PO<sub>4</sub>). This leads to the formation the hydration products struvite (MgNH<sub>4</sub>PO<sub>4</sub>·6H<sub>2</sub>O) or K-struvite (MgKPO<sub>4</sub>·6H<sub>2</sub>O), respectively (You *et al.* 2015; Wagh 2016; Qin *et al.* 2018a; Wang *et al.* 2018). The initial development of MPCs was motivated by the need for an entirely inorganic and nontoxic chemically-bonded ceramic (CBC) with fast-setting properties for dental application (Wagh 2016). Due to their favorable properties, they have now found a wide range of applications, including materials for nuclear shielding and immobilization/

encapsulation of highly hazardous industrial wastes (Wagh *et al.* 2001; Covill *et al.* 2011; Wagh 2016), coatings and adhesives (Laufenberg and Aro 2004; Ahmad *et al.* 2018; Qin *et al.* 2018a; Ahmad *et al.* 2020) as well as for rapid repair of concrete structures and high-performance civil engineering applications (Jeong and Wagh 2003; Laufenberg and Aro 2004; Amiandamhen *et al.* 2016; Wagh 2016; Walling and Provis 2016; Ahmad and Chen 2020).



**Fig. 3.** Schematic diagram illustrating the production of Mg-based composites

### *Magnesium phosphate cement (MPCs) – lignocellulosic composites*

Previous studies have confirmed the unique advantages of MPCs over other inorganic mineral binders, making them suitable and highly adaptable for versatile applications in the production of mineral binder-bonded composite. These include ready availability of the raw materials (natural minerals) and compatibility with existing equipment and processes for cementitious products (Jeong and Wagh 2003; Laufenberg and Aro 2004), low water requirements, low dry shrinkage (Li and Chen 2013; Wagh 2016; Ahmad *et al.* 2018), and fast setting at ambient temperature with high early and long-term strength (You *et al.* 2015; Lu and Chen 2016; Ahmad *et al.* 2018, 2019).

Similar to other rapid-curing cementitious materials, MPCs are also very brittle (low fracture toughness) with low strain capacity and crack resistance, limiting their utility in applications with high impact or flexural loads (Li *et al.* 2015; Qin *et al.* 2018a; Wang *et al.* 2018; Zhang *et al.* 2020). A wide range of natural and synthetic fibers, however, can be used with the MPC matrix to produce materials with interesting properties (Péra and Ambroise 1998; Amiandamhen *et al.* 2016; Qin *et al.* 2018a; Wei *et al.* 2019; Zhang *et al.* 2020). Natural plant fibers are more competitive in improving the poor fracture toughness and crack resistance of MPC than their synthetic counterparts as well as in terms of reducing the carbon footprint, embodied energy and costs while improving the overall ecological profile of the composite (Amiandamhen *et al.* 2016; Zhang *et al.* 2020). Moreover, curing of MPC is not affected by plant extractives, as fast hydration and setting behavior of the MPC take precedence over the interaction of binder cations with the soluble

extractives such as simple sugars and hemicelluloses in natural fibers (Wang *et al.* 2017; Wang *et al.* 2018). This renders the MPCs suitable for the production of high value-added composites from a wide variety of lignocellulosic wastes/agricultural residues compared to other cementitious binders (Laufenberg and Aro 2004; Amiandamhen *et al.* 2019). Unlike OPC and lime-based binders, which form entirely alkaline hydrate products, the MPC slurry has a near-neutral pH. This is due to the acid-base reaction, which makes it highly compatible with lignocellulosic materials without additional costs of energy-intensive processes such as pre-treatment, drying, and precuring (Laufenberg and Aro 2004; Wagh 2016; Ahmad *et al.* 2018).

Most importantly, MPCs can incorporate a high proportion of lignocellulosic fibers and allow shorter curing time at near room temperatures to yield composites that are stable over a wider range of pH. Still, they possess the superior technical properties of ceramics such as hardness, high compression, and resistance to chemical attack (Jeong and Wagh 2003; Laufenberg and Aro 2004; Wagh 2016; Qin *et al.* 2018a; Wang *et al.* 2018).

Despite the suitability of MPC for the development of plant-based composite products, there have been fewer studies on plant-based MPC concrete compared to OPC and lime-based concretes (Amiandamhen *et al.* 2019). Various studies report on concretes based on lignocellulosic wastes/agricultural residues and MPC binder formulations modified by various mineral admixtures (Table 2). Amiandamhen *et al.* (2019) recently reviewed these plants-based MPC materials.

### **Magnesium Oxychloride Cement (MOC)**

Magnesium oxychloride cement (MOC), also known as sorel cement, is a non-hydraulic (air-hardening) cementitious material of low alkalinity, which is formed by mixing calcined magnesium oxide (MgO) powder with aqueous magnesium chloride (MgCl<sub>2</sub>) solution. MOCs, in general, exhibit several properties superior to OPC including higher setting speed, lower thermal conductivity and higher temperature resistance, better mechanical resilience (shock-resistance), and lower shrinkage and creep. In addition, they possess high resistance to chemicals and abrasion, high fire resistance, low carbon emission, less embodied energy, and high durability (Montle and Mayhan 1974; Zhou and Li 2012; Karthikeyan *et al.* 2014; Li *et al.* 2020). Its production process is relatively low cost (lower calcination temperatures) and makes use of a wide array of raw material sources, including industrial waste streams, further lowering the overall environmental profile of its products (Li *et al.* 2003; Walling and Provis 2016; Li *et al.* 2020). The primary application of MOC is as flooring material and concrete repair works, owing to its fast curing, resistance to accumulation of static charge, very hard, tough, and durable nature as well as its attractive, marble-like appearance (Sorell and Armstrong 1976; Walling and Provis 2016). Tables 2 and 3 present the production parameters and mechanical properties of lignocellulosic composites based on magnesium cement.

#### *Magnesium oxychloride cement (MOC) lignocellulosic composites*

In addition to its traditional use in flooring applications, certain unique properties of MOC cement have promoted its adoption in the manufacture of composites with functional properties. These include its capacity to bond with large quantities and a wide variety of inert fillers ranging from mineral to lignocellulosic materials with high strength properties at the same time (Karthikeyan *et al.* 2014; Walling and Provis 2016). MOC cement, just like the OPC and MPC, is a typical brittle material (Chen *et al.* 2019a). The incorporation of fibers is an effective approach to increase its toughness.

**Table 2.** Overview of Mg-based Lignocellulosic Composites

Type	Type of Fiber	Amount of Fibers (%)	Length of Fibers (mm)	Formation Technique	Density (kg/m <sup>3</sup> )	Fillers	Strength (MPa)*	Ref.
MPC	Wood waste	20 wt%	n/a	Casting, 160 × 160 × 15 mm	1650	Alumina, Red mud	CM: ~56 MPa for 20% fiber	Wang <i>et al.</i> (2017)
	Waste-paper	n/a	n/a	Casting, 165 × 165 mm	1112 to 1177	FA	CM: 3.36 MPa	Donahue and Aro (2010)
	Sugar maple	n/a	n/a	Casting, d- 6.35 mm L- 25.4 mm	n/a	PC, VCAS, WO	n/a	Chi and Englund (2014)
	Acacia species	n/a	1mm	Casting, 218×77×40 mm <sup>3</sup>	630 to 950	FA	CM: 4.51 MPa	Amianda mhen <i>et al.</i> (2017)
	Rape stalk (RS)	5 to 30 wt%	3 to 15 mm	Casting	575 to 1650	FA	CM: 25 MPa for 5% fiber	Liu and Chen (2015)
	Corn-stalk (CS)	5 to 30 (Vol. %)	9 and 14 mm	casting	557 - 2253	FA	CM: 9.44 MPa for 30% fiber	Ahmad <i>et al.</i> (2018)
	Corn-stalk (CS)	n/a	n/a	Casting 70 x 70 x 70 mm <sup>3</sup>	n/a	n/a	CM: 3.83 MPa	Wei <i>et al.</i> (2019)
	Coir fiber (CF)	3 (Vol. %)	5 to 30 mm	Casting 40 x 40 x 40 mm <sup>3</sup>	n/a	FA	FI: 13.48 MPa for 3% fiber	Zhang <i>et al.</i> (2020a)
	Coir fiber (CF)	1 to 4 (Vol. %)	20 mm	Casting 40 x 40 x 40 mm <sup>3</sup>	n/a	FA	FI: 7.31 MPa for 3% fiber	Zhang <i>et al.</i> (2020b)
Coir fiber (CF)	3 wt%	5 to 30 mm	Casting 40 x 70 x 40 mm <sup>3</sup>	n/a	FA	CM: 56.5 MPa for 3% fiber	Jiang <i>et al.</i> (2020a)	
MOC	Sawdust	n/a	n/a	extrusion	996 - 1210	PAM, CMC, and Perlite	FI: ~3.7 MPa	Zhou and Li (2012)
	Corn-stalk (CS)	**20 to 120 wt%	1.25 to 4.75 mm	Casting 40 x 40 x 160 mm <sup>3</sup>	816	EVA Emulsion	FI: 3.65 MPa CM: 4.69 MPa	Li <i>et al.</i> (2017)
	Corn-stalk (CS) and sawdust	5, 10, and 15 wt%	n/a	Casting 40 x 40 x 160 mm <sup>3</sup>	n/a	CBP and TP	CM: ~ 73 MPa	Chen <i>et al.</i> (2019)

\* FI: Flexural Strength, CM: Compression Strength, \*\* value is relative content of corn stalk, n/a: information not available, FA: Fly Ash, PAM: Polymer Polyacrylamide, CMC: Carboxymethyl Cellulose, EVA Emulsion: Ethylene-Vinyl Acetate Emulsion, CBP: recycled Clay Brick Powder, TP: Tourmaline Powder

**Table 3.** Overview of Mg-based Lignocellulosic Composites (Contd.)

Type	Type of Fiber	Amount of Fibers (%)	Length of Fibers (mm)	Formation Technique	Density (kg/m <sup>3</sup> )	Fillers	Strength (MPa)*	Ref.
MOSC	Cellulose fibers	3 wt%	1.15	Vacuum and compression 30 × 100 × 5 mm <sup>3</sup>	n/a	CaCO <sub>3</sub> 85 wt%, glass fiber 1.8 wt%	FI: 9.65 MPa for 3% fiber	Gomes & Camarini 2014
	Rice husk	10 to 30 (vol. %)	n/a	Casting 70.7×70.7×70.7 mm <sup>3</sup>	800 to 2000	Citric acid (wt 5%), boric acid (wt 5%)	CM: 124.8 MPa for 10% fiber	Wang <i>et al.</i> 2020
	Rise husk	10 to 32 (vol. %)	n/a	Casting 70.7×70.7×70.7 mm <sup>3</sup>	451 to 1722	Citric acid (wt 5%)	CM: 70.4 MPa for 10% fiber	Qin <i>et al.</i> 2018b
MS	Pine and eucalyptus cellulosic fibers	8 wt%	0.2 to 2.2	Vacuum and compression 200 × 200 × 6 mm <sup>3</sup>	1390 to 1520	limestone 23 wt%	FI: 9.23 MPa for 8% fiber	Marmol & Savastano 2015

\* FI: Flexural Strength, CM: Compression Strength, n/a: information not available

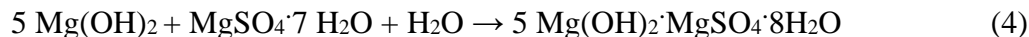
Adding lignocellulosic waste materials/agricultural residues has been considered as an economic and environmentally friendly method that enhances the technical properties of MOC concrete (Zhou and Li 2012; Li *et al.* 2017; Chen *et al.* 2019a). Additionally, MOC cement exhibits a characteristic yellowish color, which is close to the color of natural wood, as well as low alkalinity, and resistance to the inhibitory effect of extractives. These properties have made it desirable for the production of mineral binder-bonded composites from wood particles as well as a wide range of biomass wastes/agricultural residues, without the aging and setting problems commonly experienced with (highly alkaline) OPC binder (Zhou and Li 2012). Nevertheless, MOC's poor moisture resistance in exterior applications has slowed down its wide application as composites for construction engineering using hydrophilic lignocellulosic fibers.

Hence, the improvement of the poor water resistance and other technical properties of MOC has attracted much attention from researchers, because it has a crucial influence on the use of MOC-based materials under exterior conditions (Guo *et al.* 2018).

### Magnesium Oxysulfate Cement (MOSC)

Magnesium Oxysulfate Cement (MOSC), a non-conventional material belongs to the class of Mg-based cement. Compared to OPC, MOSC features various advantages such as fire resistance, low thermal conductivity, low density, and especially adequate compatibility with lignocellulosic materials (Gomes and Camarini 2014). Along with other Mg-based cements, MOSC cements are considered environmentally friendly due to their lower carbon emissions released predominantly in the calcination process of the raw material (Mo *et al.* 2016). MOSC is formed by the reaction of hydrated magnesium oxide and magnesium sulfate at ambient temperature:





#### *Magnesium oxysulfate (MOSC) cement– lignocellulosic composites*

A limited number of studies have been conducted in the field of MOSC cement-bonded lignocellulosic boards. Cellulosic fibers-MOSC composites were prepared using calcium carbonate as an inert filler (Gomes and Camarini 2014). Flexural strength displayed a marginal decrease with the increase of aging cycles; however, the durability of the fiberboards after aging was still very promising. Recently, Wang *et al.* (2020) prepared lightweight building materials from rice husk bonded with MOSC and a foaming agent. For the preparation process, a small fraction of citric acid was added to the reagent mixture. The weak citric acid enhanced the compressive strength, as it catalyzed the formation of finer crystals. Rice husk-MOSC composites, however, were quite sensitive to various wetting-drying cycles. After numerous aging cycles, the compression strength decreased considerably, particularly when the porosity of the sample was high. When the rice husk content was 10 wt%, compression strength was not affected, but, when the rice husk content was increased to 70 wt%, it declined significantly. In an analogous study (Qin *et al.* 2018b), the compression strength of rice husk–MOSC composites increased with increasing density while insulation properties worsened. A higher amount of rice husks in the composite eventually leads to higher drying shrinkage. A direct comparison with other studies is presented in Tables 2 and 3.

The application of MOSC cement is often limited due to the lack of mechanical strength compared to OPC. Lightweight MOS cement-lignocellulosic composites could be used for interior non-structural applications as they have exceptional sound-, fire-, and water-resistance as well as thermal insulation properties (Gomes and Camarini 2014). The less alkaline environment arising from the hydration process has a positive effect on the curing behavior compared to OPC, as lower amounts of alkaline soluble polysaccharides are dissolved in the cement paste, which affects crystallization.

#### **Magnesium Silica Cement (MSC)**

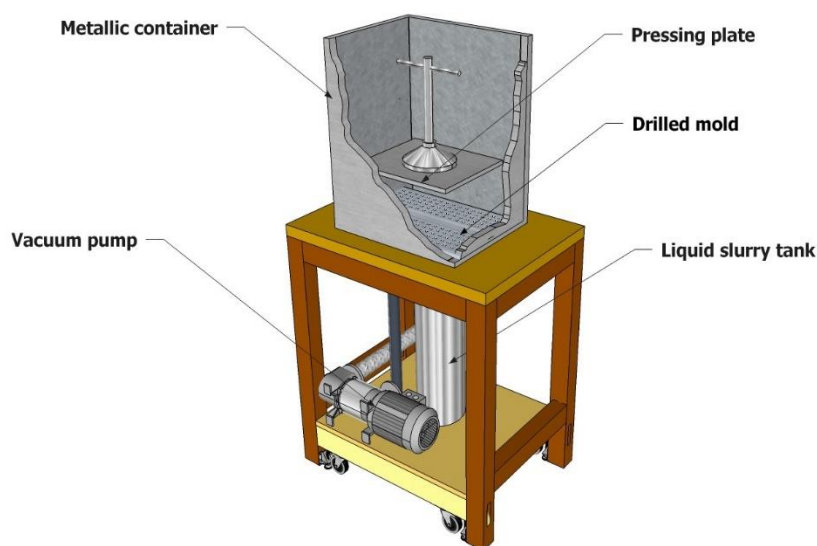
Magnesium silicate hydrate (MSH) is a matrix formed from the reaction of MgO and silica (silicon dioxide). The factors controlling the formation of MSH include the ratio and characteristics of the reagents, alkalinity of the mixture, and temperature (Jin and Al-Tabbaa 2014). Comparing OPC and Magnesium Silica Cement (MSC), the two main hydrated compounds from the former one are calcium silicate hydrate and portlandite ( $\text{Ca(OH)}_2$ ), whereas, in the case of the latter one, MSH and brucite ( $\text{Mg(OH)}_2$ ) are formed (Jin and Al-Tabbaa 2014; Zhang *et al.* 2014; Lothenbach *et al.* 2015). Even though there are many similarities in the crystal structure, it is assumed that MSC required more water than OPC to form completely hydrated gels (Chen *et al.* 2013). An excess of water at the initial stages can be problematic because it can increase the number of capillary pores and, as a result, decrease the mechanical performance (Marmol and Savastano 2015).

#### *Magnesium silica (MS) cement – lignocellulosic composites*

Few studies have reported on magnesium silica (MS) cement-lignocellulosic fiber composites. Marmol and Savastano (2015) investigated the degradation of MSC reinforced with pine and eucalyptus wood fibers. In order to cast the slurry, a dewatering process using a vacuum pump connected with the drilled mold was utilized, followed by a pressing step (Fig. 4.). This technique, analogously with other casting methods, has been widely

used to produce OPC-fiber composites and corresponds to the Hatschek process (Ardanuy *et al.* 2015). The MS cement binder was able to maintain the strength properties of the composite and to preserve the wood fibers' integrity after several aging cycles, while OPC suffered from strength losses. After 28 days of curing, OPC-wood fiber composites exhibited higher flexural strength than MS cement composites. The authors attributed the lower strength to the higher water-cement ratio required for the hydration of MgO-SiO<sub>2</sub>. Aging over 200 cycles had no negative effect on flexural strength of MSC composites. In contrast, OPC composites suffered a significant loss of Modulus of Rupture after ageing. Also, in the case of MSC composites, the interfacial transition zone (ITZ) was densified, thus improving the adherence between the fiber and the matrix. In OPC composites, however, the presence of portlandite on the fiber surface can degrade the fiber and reduce the mechanical properties, while in MSC, such mineralization did not occur.

The authors suggested that during aging, unreacted magnesium hydroxide reacts with SiO<sub>2</sub>, forming a resistant M-H-S matrix that protects the fibers (Marmol and Savastano 2015). Durability (maintaining strength) during aging and resistance to weathering appear to be the distinctive property of MSC – lignocellulosic composites.



**Fig. 4.** Schematic diagram illustrating the vacuum dewatering process followed by pressing. A laboratory-scale Hatschek process

### Factors Affecting the Performance of Mg-based Cement

Similar to other inorganic mineral binder-bonded composites, the performance of Mg-cement-bonded composites as green building materials is critically dependent on the production parameters.

#### *Effect of modifiers/fillers, MPC*

The hydration reaction of MPC binder in the presence of water proceeds very rapidly, accompanied by an increase in hydration temperature, owing to the exothermic setting (curing) reactions. This results in difficulties with mixing and casting, with attendant effects on the mechanical properties of the resulting material (Yang *et al.* 2014). In addition, due to the instability of MPC hydration products to water, MPC materials may exhibit compromised performance when exposed to long-term outdoor conditions or humid

environments, further reducing the strength properties of the material (Lu and Chen 2016; Wang *et al.* 2017). Thus, modifiers are needed to control the reactions and setting rate as well as to improve the resistance of MPC products to water. Borax and boric acid are the most common effective additives for reducing the hydration rate and curing temperature of MPC, to allow enough working time (Hall *et al.* 2001; Yang *et al.* 2014; You *et al.* 2015; Wang *et al.* 2017). However, issues persist in terms of stability and mechanical performance in a highly humid environment. To circumvent this, incorporation of mineral admixtures or industrial by-products as modifiers into MPC imparted significant improvements in moisture resistance, mechanical and functional properties of the resulting MPC concrete while lowering the overall product costs (Chi and Englund 2014; Liang *et al.* 2019a). These modifiers include fly ash and silica fume from utility plants (Wagh *et al.* 1997; Li and Chen 2013; Liu and Chen 2015; Li *et al.* 2016; Zheng *et al.* 2016; Liao *et al.* 2017; Ahmad *et al.* 2018), mineral wastes (Fan and Chen 2015; Wang *et al.* 2017), metallic slag (Tan *et al.* 2016; Chen *et al.* 2017) as well as natural (mineral) pozzolanic materials such as metakaolin (Lu and Chen 2016; Li *et al.* 2016; Liu and Chen 2016; Shi *et al.* 2019).

The principal mineral modifier used for lignocellulosic-MPC composites is fly ash (FA), an industrial by-product (Table 2). FA can significantly improve the technical properties of the MPC system, even at higher mass contents (up to 40 wt%) (Ding and Li 2005a). Additionally, FA inclusion at certain mass content can reduce the total heat evolution during hydration, increase setting time and flowability of fresh MPC paste as well as improve the mechanical strength and moisture-related properties of MPC mortar (Donahue and Aro 2010; Li and Feng 2010; Li and Chen 2013). The addition of FA improves the microstructural compactness of MPC paste, and thus its mechanical performance, through the dual mechanisms of physically filling the pores of MPC paste and the participation of its alumina ( $\text{Al}_2\text{O}_3$ ) and amorphous silica ( $\text{SiO}_2$ ) components in the hydration reaction to form a new cementitious gel. This new product (gel) makes the MPC structure denser while generating more binder in the mixture (Laufenberg and Aro 2004; Ding and Li 2005a,b; Donahue and Aro 2010; Li and Feng 2010).

The effect of admixing OPC, wollastonite (WO), and vitrified calcium aluminosilicates (VCAS) on the bonding interface of sugar maple (*Acer saccharum*) wood/MPC composites was evaluated using a mixture design (Chi and Englund 2014). A combination of WO and VCAS in an MPC mix mutually promoted the interfacial shear strength ( $\tau_i$ ) of the composites. The materials modified with VCAS exhibited higher water sorption than those modified with WO and OPC with a corresponding rapid loss of bond integrity after one day of immersion in water. The MPC binder content (level), however, is the primary factor affecting the interfacial bond properties of the wood/MPC materials, irrespective of the type of modifier used (Chi and Englund 2014).

Alumina ( $\text{Al}_2\text{O}_3$ ) and red mud ( $\text{Fe}_2\text{O}_3$ ) were used as sustainable additives to improve the water-resistance and strength properties of MPC particleboards made from wood waste (Wang *et al.* 2017). The partial replacement of MgO with alumina or red mud at an Mg/Al or Mg/Fe optimal molar ratio of 10:1 facilitated the formation of amorphous Mg-Al or Mg-Al-Fe phosphate gel, respectively, which enhanced compressive strength. While alumina-MPC particleboards exhibited improved short-term (24-h) water resistance and strength retention, red-mud-modified boards exhibited higher long-term water resistance and enhanced strength retention with reduced water absorption after 72-h water immersion. This is attributed to the stable scaly-like Mg-Al-Fe phosphate gel. Additionally, red mud imparted excellent compatibility of MPC binder with wood waste and facilitated the extension of both the initial and final setting time of the MPC mix.



*Effect of modifiers/fillers, MOC*

Various fillers/modifiers have been utilized to improve MOC composites. In a recent study, recycled clay brick powder (CBP) was investigated as an alternative low-cost waste pozzolanic material for MOC mortar reinforced with agricultural residues, namely corn stalk and wood sawdust (Chen *et al.* 2019). Due to the higher water absorption capacity of the CBP, the fluidity and workability of freshly blended agro-waste MOC mortars consistently decreased with increasing CBP content up to a maximum CBP content of 15%. Other properties such as specific gravity increased, while the flexural strength (more than 25% decline) and water absorption capacity of the MOC mortars significantly decreased with increasing CBP content up to 35%. Due to the pore filling and pozzolanic potential of the CBP, the MOC composites became denser and more brittle with increasing CBP content, thereby limiting the ingress of moisture into the structure, while adversely impacting the flexural strength.

Another study assessed the effects of adding ethylene-vinyl acetate (EVA) emulsion as a modifier on the water resistance, density, thermal conductivity, and mechanical properties of corn-stalk-MOC (C-MOC) composites (Li *et al.* 2017). Microscopic analysis revealed that EVA emulsion exhibited good compatibility with the magnesium oxychloride cement by interacting synergistically with the fiber and matrix components. It thus enhances their interfacial bonding performance by forming a thin film on the outer surface of the corn stalk, increases the roughness of the straw surface, and reduces the gap between the stalk. Increasing EVA emulsion content resulted in a corresponding reduction of the water absorption capacity of C-MOC composites, while it increased the density and coefficient of thermal conductivity, respectively (Li *et al.* 2017).

*Effect of modifiers/fillers, MOSC*

Wang *et al.* (2020) showed that the addition of the weak acids citric acid and boric acid can significantly increase the compressive strength of MOSC composites at different curing ages. Citric acid was more effective and greatly enhanced the compressive strength after 28 days of curing. This can be attributed to citric acid's ability to catalyze the production of a greater number of fine needle-like crystals. Adding tartaric acid gave analogous results regarding the enhancement of flexural and compression strength (Wu *et al.* 2017). Microscopic observations indicated that tartaric acid promotes the formation of the needle-like high-strength phase of  $5\text{Mg}(\text{OH})_2 \cdot \text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  and inhibits the formation and growth of the  $\text{Mg}(\text{OH})_2$  crystal phase.

*Effect of plant aggregate type, size, and volume content*

The type and volume content of plant-derived aggregates are important parameters with critical influence on the density and performance of mineral binder-bonded wood and plant fiber-based composites. Liu and Chen (2015) investigated rape stalk-MPC materials obtained by incorporating different sizes and volume contents of rape stalk (RS) in an MPC binder formulation containing 20% fly ash. RS-MPC concretes were realized, which satisfied the requirements for structural and insulating purposes. At RS contents < 20%, the thermal conductivity decreased linearly with increasing RS size, owing to the morphological (honeycomb) structure and lower thermal conductivity of the rape stalk compared to the MPC matrix. However, improved early strength gain values of the concrete were observed with increasing RS content, while the concrete mixtures with the smallest RS size exhibited the highest early strength gain in the first hour. The compression strength of RS-MPC concrete decreased with increasing rape stalk content and size, due to

the difficulty in packing large-sized RS aggregates at higher contents, thereby resulting in the development of voids in the concrete. This indicates that, aside from the poor RS/matrix interfacial adhesion and low mechanical strength of the rape stalk, increasing porosity of the composite material as a function of rape stalk size and content is a major factor contributing to the reduction of compressive strength of RS-MPC concrete (Liu and Chen 2015).

MPC composites containing large corn stalk (LCS) and small corn stalk (SCS) were investigated in terms of mechanical and physical properties. Increasing corn stalk content resulted in a corresponding decrease in the compressive strength and rate of strength gain during curing for CS-MPC concrete and increased the thermal conductivity and water absorption coefficient of the concrete accordingly. Corn stalk size significantly influenced the compressive strength of CS-MPC concretes. The SCS-MPC concrete exhibited higher thermal conductivity and lower water absorption coefficient compared to the LCS-MPC concrete (Ahmad *et al.* 2018).

The effects of coconut coir fibers (CF) with different lengths and curing times were studied in terms of flexural strength, load-displacement behavior, and flexural toughness of MPC with CF lengths varying from 0 to 30 mm at various curing ages (Zhang *et al.* 2020). At both curing times, flexural strength and toughness of MPC initially increased with increasing CF length, but then they decreased when CF length exceeded 20 mm. Ductility and elastic modulus displayed continuous increasing and decreasing trends, respectively with increasing CF length. After a curing period of 28 days, compressive strength, elastic modulus, and secant modulus decreased with increasing CF length. The CF length of 20 mm was the threshold for the maximum energy absorption, beyond which the value declined following a convex curve (Zhang *et al.* 2020b).

Jiang *et al.* (2020a) studied the effects of CF volume concentrations (0% to 4%) at two curing ages (7 and 28 days) on MPC flexural properties. CF showed similar effects on the flexural performances for MPC at both curing times, with specimens at 28 days exhibiting greater flexural strength, higher stiffness, and flexural toughness than those at 7 days. In addition, increasing CF volume concentration up to 3%, sequentially enhanced the flexural toughness and compression strength (Jiang *et al.* 2020).

### Hybridization

A hybrid usage of polyacrylamide (PAM) and carboxymethyl cellulose (CMC) was adopted as a cost-effective and efficient rheology-enhancing additive for wood-MOC cement composites (Zhuo and Li 2012). The authors were able to produce extruded light-weight wood-MOC cement composites to be used as full-scale door frame and door panels, with specific dry densities close to 1.0, and nailability similar to natural hardwood. Incorporation of perlite, by replacing 50 wt% sawdust in the composite, resulted in materials with less die swell and better performance in resisting high temperatures. Aside from factors influencing the MOC binder properties and fiber-matrix interfacial interaction, the fiber content and distribution is a critical factor that greatly affects the performance properties of plant fiber-MOC composites (Li *et al.* 2017). The flexural strength of agro-waste MOC composites was enhanced by the addition of corn stalk (CS) and sawdust (SD), while the water sorption of mortars increased with increasing fiber content (Chen *et al.* 2019a). The induced increase in the flexural strength with the incorporation of agro-waste aggregates (*i.e.*, CS and SD) was mainly attributed to the arbitrary distribution of the fibers in the mixture, thereby improving the continuity of the composite material and reducing internal defects when loaded. Contrarily, the

incorporation of CS and SD fibers in the MOC reduced the compressive strength. This can be explained based on two factors: First, the incorporation of the inert fibers results in less compact materials, due to the poor fiber-matrix interfacial bonding in the MOC structure. In addition, it decreases the proportion of cementitious materials and thereby further reduces the hydration products responsible for bonding.

## LIGNOCELLULOSIC COMPOSITES FROM GEOPOLYMERS AND ALKALI ACTIVATED CEMENT

### Geopolymers: Properties and Applications

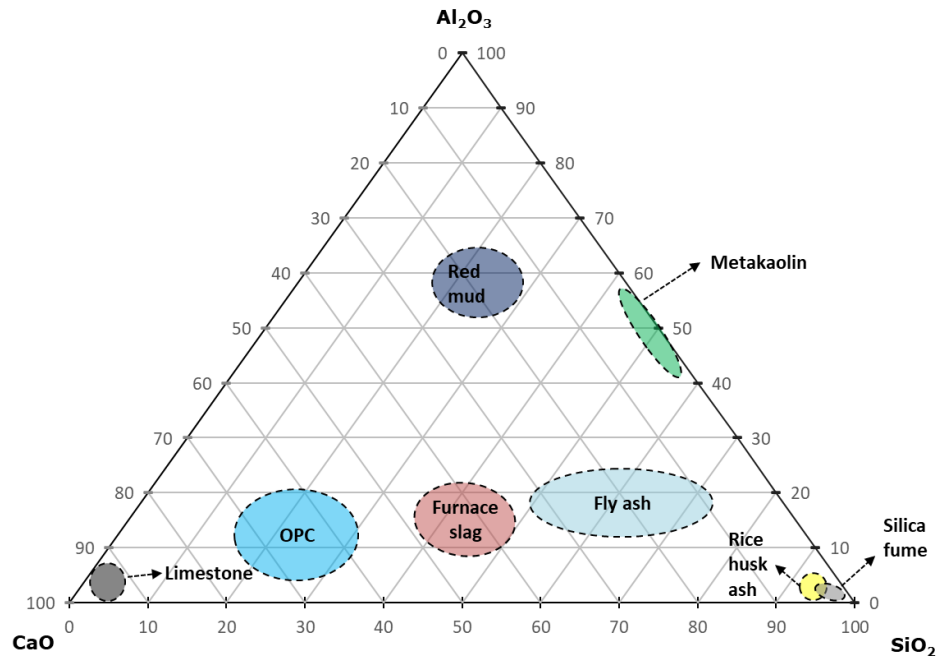
Geopolymers (mineral polymers resulting from geochemistry or geosynthesis) are inorganic polymer materials formed by polycondensation of mineral substances rich in silicon (Si) and aluminum (Al) in a strongly alkaline or acidic environment. When such (geopolymer) materials harden at ambient/room temperature like regular cement, they are referred to as “geopolymer cement”. If they only harden under the influence of heat treatment, they are called “geopolymer binders” (Davidovits 2013). Geopolymer materials have emerged as a low embodied energy and eco-responsible alternative to conventional construction materials. They can be produced from a wide range of natural minerals or industrial waste streams such as metakaolin, fly ash, slag, silica fume, natural pozzolanic (ashes) materials as well as calcium or magnesium-containing minerals (Shalbfan *et al.* 2016; Bazan *et al.* 2020; Korniejenko *et al.* 2020a; Lemouagna *et al.* 2020).

There is disagreement in the scientific community as to whether geopolymers should be classified as a subset of alkali-activated materials (AAM). According to a state-of-the-art report by RILEM (RILEM TC 224-AAM 2014), Alkali-Activated Materials (AAM) are defined as “the broadest classification, encompassing essentially any binder system derived by the reaction of an alkali metal source (solid or dissolved) with a solid silicate powder”. These can be derived from a myriad of natural and industrial waste materials as precursors and regard geopolymers as a subset of this classification. However, for this study, geopolymers are treated as a separate group.

Detailed information on the various industrial by-products and waste materials used for geopolymer production including the reported compositions, mixtures, compressive strengths, and curing conditions for their pastes and mortars can be found in the review of Silva *et al.* (2019). The major oxide concentrations of various precursors used for the production of geopolymers and alkali-activated cements are depicted in a ternary phase diagram (Fig. 5.).

Owing to their high compressive strength, low shrinkage, resistance to corrosive environments, and high thermal stability as well as a high capacity to encapsulate foreign materials within their structure (Silva *et al.* 2019; Liang *et al.* 2019b; Korniejenko *et al.* 2020a; Lahoti *et al.* 2020; Lemouagna *et al.* 2020), geopolymers have found useful applications not only in the construction industry but also for the immobilization of toxic waste and heavy metals, as well as coatings in various industries (Bazan *et al.* 2020; Jiang *et al.* 2020b). Currently, geopolymer composites are the most promising environmentally friendly alternative to traditional cementitious materials including OPC, because their use could significantly reduce CO<sub>2</sub> emissions as a result of the low carbon footprint of the various raw materials from which they can be prepared (Sarmin *et al.* 2014; Shalbfan *et al.* 2016; Silva *et al.* 2020). These distinctive properties have allowed the development of eco-responsible construction materials from both natural minerals and industrial wastes/

residues, *viz.* geopolymer cement, mortars, and concrete with similar or even better performance properties than traditional materials. Still, despite their high compressive strength, geopolymer materials possess major weaknesses of low ductility and brittle cracking behavior. This limits their use in possible structural functions. These weaknesses are quite similar to those of other hardened inorganic mineral binders/cement such as OPC and magnesium cement (Silva *et al.* 2019; Bazan *et al.* 2020; Korniejenko *et al.* 2020a; Silva *et al.* 2020).



**Fig. 5.** Ternary phase diagram (CaO-SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>) indicating major compositions (%) of various precursors used for the production of geopolymers and alkali-activated cement compared to OPC

#### *Geopolymer-based lignocellulosic composites*

A lot of studies have been conducted to develop geopolymer composites reinforced with both natural and synthetic fibers to improve their ductility, flexural strength, and fracture toughness (Korniejenko *et al.* 2020a; Silva *et al.* 2020). Detailed reviews have been published regarding the recent advances in the production of natural fiber-reinforced materials with potential construction applications and their performance (Sarmin *et al.* 2014; Korniejenko *et al.* 2020b; Silva *et al.* 2019, 2020). Therefore, this review focuses on the influence of the curing conditions and activator composition, as well fiber surface treatment on the performance of lignocellulosic fiber-reinforced geopolymer composites. Tables 4, 5, 6, and 7 summarize the production parameters and mechanical properties of lignocellulosic fiber-reinforced geopolymer composites in the literature.

#### **Alkali-Activated Cement**

Alkali-activated (AA) cements are cementitious binders formed by activating aluminosilicates with highly alkaline solutions. Due to their less negative environmental impact, AA cement was adopted to substitute OPC. Besides, it is now widely accepted that AA cement has advantages over Portland cement in terms of energy cost, strength, and high-temperature resistance (Puertas and Fernández-Jiménez 2003).

**Table 4.** Production Parameters and Mechanical Strength Properties of Lignocellulosic Fiber Reinforced Geopolymer Composites\*

Precursor	Activator	Type of fiber	Fiber content	Length of fibers (mm)	Initial curing details	Type of casting details	Density of the composite (kg/m <sup>3</sup> )	Curing time (Days)	Mechanical Strength (MPa) **	Reference
Dehydroxylated halloysite	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Flax fiber	10 wt%	0.01-0.08 mm	40 °C for 2 days	bar-shaped, 120 × 10 × 6 mm <sup>3</sup>	n/a	28	FI: 70.2	Alzeer and Mackenzie (2013)
Metakaolin	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cornhusk Bundles	5 wt%	n/a	50 °C for 1 day	rectangular, 152.4 mm × 203.2 mm	n/a	1	FI: 8.8	Kriven <i>et al.</i> (2013)
Metakaolin	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Luffa fiber	10 % (vol.)	n/a	40 °C for 1 day	prism, 160 × 100 × 20 mm <sup>3</sup>	n/a	n/a	CM: 31.0 FI: 14.2	Alshaaer <i>et al.</i> (2017)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cotton fabric	2.1 wt%	n/a	105 °C for 3 h	n/a	1590 to 2020	28	FI: ~12.5	Alomayri <i>et al.</i> (2013a)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cotton fiber	0.5 wt%	10 mm	105 °C for 3 h	rectangular, 80 × 20 × 10 mm <sup>3</sup>	1800 to 2000	28	FI: 11.7	Alomayri <i>et al.</i> (2013b)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cotton fiber	0.5 wt%	10 mm	105 °C for 3 h	rectangular, 80 × 20 × 10 mm <sup>3</sup>	n/a	28	CM: 48	Alomayri <i>et al.</i> (2013c)

\* Adapted from Korniejenko *et al.* (2020b) and Silva *et al.* (2019), \*\* FI: Flexural Strength, CM: Compression Strength, n/a: information not available

**Table 5.** Production Parameters and Mechanical Strength Properties of Lignocellulosic Fiber Reinforced Geopolymer Composites\* (Contd.)

Precursor	Activator	Type of fiber	Fiber content	Length of fibers (mm)	Initial curing details	Type of casting details	Density of the composite (kg/m <sup>3</sup> )	Curing time (Days)	Mechanical Strength (MPa)**	Reference
Metakaolin	Na <sub>2</sub> SiO <sub>3</sub> (SiO <sub>2</sub> /Na <sub>2</sub> O) = 2.17	Sisal fiber	3 wt%	25 mm and 50 mm	55 °C for 1 day	<b>CM:</b> cylindrical, d-50 mm, h-100 mm <b>FI:</b> bar-shaped, 160 × 40 × 40 mm <sup>3</sup>	n/a	n/a	<b>CM:</b> 6 <b>FI:</b> 2.7	Correia <i>et al.</i> (2013)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cotton fabric	8.3 wt%	n/a	80 °C for 1 day	n/a	1900	28	<b>CM:</b> ~90 <b>FI:</b> 31.7	Alomayri <i>et al.</i> (2014b)
Metakaolin	Na <sub>2</sub> SiO <sub>3</sub> (SiO <sub>2</sub> /Na <sub>2</sub> O) = 2.17	Pine-apple leaf fiber	1.2 wt%	25 and 50	55 °C for 1 day	<b>CM:</b> cylindrical, d-50 mm, h-100 mm <b>FI:</b> bar-shaped, 160 × 40 × 40 mm <sup>3</sup>	n/a	n/a	<b>CM:</b> 3.3 <b>FI:</b> 2	Correia <i>et al.</i> (2013)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cotton fabric, (parallel to fabric)	8.3 wt%	n/a	80 °C for 1 day	n/a	n/a	28	<b>CM:</b> ~60 <b>FI:</b> 26	Alomayri <i>et al.</i> (2014a)

\* Adapted from Korniejenko *et al.* (2020b) and Silva *et al.* (2019), \*\* FI: Flexural Strength, CM: Compression Strength, n/a: information not available

**Table 6.** Production Parameters and Mechanical Strength Properties of Lignocellulosic Fiber Reinforced Geopolymer Composites\* (Contd.)

Precursor	Activator	Type of fiber	Fiber content	Length of fibers (mm)	Initial curing (details)	Type of casting (details)	Density of the composite (kg/m <sup>3</sup> )	Curing time (Days)	Mechanical Strength (MPa)**	Reference
Fly ash (Class F)	NaOH solution	Sorghum	2.0 wt%	< 10	60 °C for 3 h	CM: cylindrical, d-35mm, h-70 mm FI: rectangular, 360 × 60 × 25 mm <sup>3</sup>	~1500	7	FI: 5.4 CM: 22.9	Chen et al. (2014)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Flax fabric	4.1 wt%	n/a	60 °C for 1 day	n/a	n/a	28	FI: 91 CM: 23	Assaedi et al. (2015)
Fly ash and metakaolin	10 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Wood	10 wt%	0.7	40 °C for 1 day	CM: cubic, 40 × 40 × 40 mm <sup>3</sup> FI: prism, 40 × 40 × 160 mm <sup>3</sup>	1600 to 2100	28	CM: ~65	Duan et al. (2016)
Fly ash (Class F)	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Sisal fiber	3 wt%	ca. 10	Heat cured	n/a	n/a	14	FI: 4.5 CM: 41	Patel and Joshi (2016)
Fly ash, silica	16 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Coir fiber	0.75 wt%	25	Heat cured	cubic, 70 × 70 × 70 mm <sup>3</sup>	n/a	28	CM: 51.2	Siddharth et al. (2016)
76% metakaolin, 24% silica	14 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Bamboo	5 wt%	12.5-40	20 °C for 28 days	Cubic, 50 × 50 × 50 mm <sup>3</sup>	1550 to 1705	7	FI: 27.6 CM: 33.1	Sarmin (2016)

\* Adapted from Korniejenko *et al.* (2020b) and Silva *et al.* (2019), \*\* FI: Flexural Strength, CM: Compression Strength, n/a: information not available

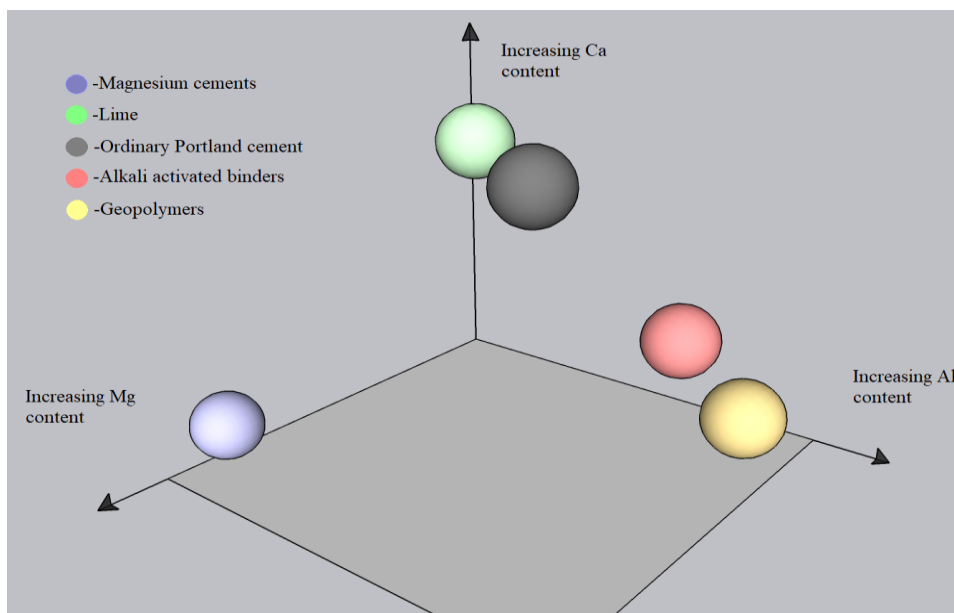
**Table 7.** Production Parameters and Mechanical Strength Properties of Lignocellulosic Fiber Reinforced Geopolymer Composites\* (Contd.)

Precursor	Activator	Type of fiber	Fiber content	Length of fibers (mm)	Initial curing (details)	Type of casting (details)	Density of the composite (kg/m <sup>3</sup> )	Curing time (Days)	Mechanical Strength (MPa)**	Reference
Fly ash (Class F)	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Pineapple leaf fiber	1.2 wt%	ca. 50	60 °C for 1 hour	n/a	1990 to 2020	28	FI: 7.1 CM: 58.2	Amalia <i>et al.</i> (2017a)
Fly ash (Class F)	8 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Cotton fiber	1 wt%	30	75 °C for 1 day	n/a	n/a	28	FI: 5.8 CM: 28.4	Korniejenko <i>et al.</i> (2016)
Fly ash (Class F)		Sisal fiber,	1 wt%	3					FI: 5.9 CM: 25.2	
Fly ash (Class F)		Coir fiber,	1 wt%	3					FI: 5.2 CM: 31.4	
Fly ash (Class F)		Raffia fiber	1 wt%	3					FI: 3.0 CM: 13.7	
Fly ash (Class F)	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Coconut trunk fiber	0.5 wt%	30 to 50	70 °C for 1 hour	n/a	n/a	28	FI: 7.64 CM: 89.4	Amalia <i>et al.</i> (2017b)
Fly ash (Class F)	12 M NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Abaca fiber	8 wt%	0.1 to 0.2	75 °C for 1 day	CM: cubic 50 × 50 × 50 mm <sup>3</sup> FI: prism, 50 × 50 × 180 mm <sup>3</sup>	n/a	28	FI: 27 CM: 50	Malenab <i>et al.</i> (2017)

\* Adapted from Korniejenko *et al.* (2020b) and Silva *et al.* (2019), \*\* FI: Flexural Strength, CM: Compression Strength, n/a: information not available



Differences between AA cements and geopolymers consist of raw materials composition, the formation chemistry and properties of the final product (Waldmann and Thapa 2018). In particular, precursors of AA cement are usually high-calcium aluminosilicates (blast furnace slag and in some cases fly ash with high amounts of calcium), while geopolymers are produced by the activation of raw materials with low calcium content such as fly ash (Class F) and metakaolin (Li *et al.* 2010). Figure 6 shows an illustration of the classification of various cementitious products based on the major cation content. Another difference is related to the pH of the activator. In the case of geopolymers, the pH is slightly alkaline, while for AA cement it is considerably higher. In addition, AA cement has higher mechanical strength but lacks long-term durability compared to geopolymers (Davidovits 2017). More research needs to be undertaken regarding the specific network structure of the mineral phases (Li *et al.* 2010). This section covers only composites obtained from calcium-rich raw precursors of AA cement and various types of lignocellulosic fibers.



**Fig. 6.** Classification of different subsets of various non-conventional cementitious materials in comparison to OPC

#### *Alkali activated (AA) cement – lignocellulosic composites*

Alike other cementitious products, the properties of AA cement composites were in most cases improved when natural fibers were incorporated. Generally, the addition of a small amount of fibers (1 to 3 wt%) positively influenced mechanical properties, density (lower), and other physical characteristics such as water sorption. Poletanovic *et al.* (2020) studied the effect of hemp fibers in AA cement. Their raw materials for two formulations consisted of fly ash and a mixture of fly ash with ground granulated blast furnace slag (GGBFS). In general, the addition of fibers did not have a great effect on the composites' mechanical properties, but it increased its energy absorption capacity and thus toughness. With the increment of the fiber proportion, the density decreased, while water absorption increased. Detailed information on raw materials and production parameters can be found in Table 8.

**Table 8.** Production Parameters and Mechanical Strength Properties of Lignocellulosic Fiber Reinforced with AA Cement Composites

Precursor	Activator	Type of fiber	Amount of fibers (%)	Length of fibers (mm)	Initial curing details	Type of casting	Density of the composite (kg/m <sup>3</sup> )	Curing time (Days)	Maximal strength (MPa)*	Reference
Furnace slag	NaOH	Wood fibers	3.5, 6.3, 8.3, 10.1	0.06 to 8 mm	60 °C for 1 day	<b>CM:</b> cubic 40 x40 x40 mm <sup>3</sup> <b>FI:</b> Metallic prism, 4 x 4 x 16 mm <sup>3</sup>	1000 to 1800	28	<b>FI:</b> 10.1 <b>CM:</b> ~21	Kielè <i>et al.</i> 2020
Furnace slag	KOH, NaOH + Na <sub>2</sub> SiO <sub>3</sub>	Wheat Straw	1	20 to 30	20 °C for 1 day	<b>CM:</b> cubic 40 x40 x80 mm <sup>3</sup> <b>FI:</b> Metallic prism, 4 x 4 x 16 mm <sup>3</sup>	n/a	7, 28	<b>FL:</b> 12.4 <b>CM:</b> 108.4	Zhu <i>et al.</i> 2019
Furnace slag		Rice straw	1	20 to 30					<b>FI:</b> 7.8 <b>CM:</b> 81.6	Zhu <i>et al.</i> 2019
Furnace slag		Corn stalk	1	20 to 30					<b>FI:</b> 9.6 <b>CM:</b> 108.1	Zhu <i>et al.</i> 2019
Furnace slag and Ca-rich fly ash	Na <sub>2</sub> SiO <sub>3</sub>	Hemp	0.5, 1	10	60-80°C for 1 day	<b>CM:</b> cubic 40 x40 x40 mm <sup>3</sup> <b>FI:</b> Metallic prism, 4 x 4 x 16 mm <sup>3</sup>	1800 to 2000	28	<b>FI:</b> 6.39 <b>CM:</b> ~32	Poletanovic <i>et al.</i> 2020
Furnace slag	4.8 M NaOH + 6.5 M Na <sub>2</sub> SiO <sub>3</sub>	Rice straw	1, 2, 3	15 to 20	20 °C for 1 day	<b>CM:</b> cubic 50 x50 x50 mm <sup>3</sup> <b>FI:</b> Metallic prism, 4 x 4 x 16 mm	1700 to 1900	14, 28	<b>FI:</b> 6.2 <b>CM:</b> 45	Nguyen and Mangat 2020

\* FI: flexural strength, CM: compression strength, n/a: information not available

Zhu *et al.* (2019) produced AA cement composites incorporating 1 wt% of three types of fibers (wheat straw, straw, and corn stalk). Wheat straw-AA cement composites reached the highest strength because of the more homogenous distribution of these fibers compared to others. The strength values were equivalent to those of OPCs. Furthermore, the composites maintain their flexural strength even at extreme temperatures (200 ~ 400 °C). In a similar approach, small amounts (1 to 3 wt%) of rice husk were used to prepare AA cement composites (Van Nguyen and Mangat 2020). The embodied fibers improved mechanical performance and physical properties such as drying shrinkage and water absorption. The authors attributed the change of these properties to the reduced porosity because of the reduction in liquid activator/binder ratio caused by the hydrophilic nature of rice straw and the internal curing induced by the moisture held in the straw. In contrast with other studies, the water absorption of composites was reduced compared to pure AA cement concrete. The authors attributed this finding to the reduction in “bleeding” as fibers increase mix stiffness and reduce the settlement of aggregates (sand).

AA cement composites were produced containing various amounts (3.5 to 10.1 wt%, higher than other studies) of wood shavings (Kielè *et al.* 2020). Flexural strength of the composites containing up to 6.3 wt% shavings nearly doubled compared to that of pure AA cement concrete. Further addition of wood particles worsened the mechanical properties. Increasing contents of fibers in the composites reduced the thermal conductivity up to 84% compared to reference specimens. A content of up to 6.3 wt% wood shavings was recommended to achieve the balance between thermal and structural properties.

### **Influence of Production Parameters on the Performance of Geopolymer- and Alkali-Activated Cement-Bonded Lignocellulosic Composites**

The foregoing has clearly shown that the performance of both geopolymer- and alkali-activated cement-bonded lignocellulosic composites depend on the production parameters, most especially the raw materials and on the precursors, activators, and curing conditions as well as the type of surface treatment of the lignocellulosic aggregate.

#### *Precursors, activators, and curing conditions*

The curing of freshly prepared geopolymer concrete is of critical importance in the entire geopolymerization process. Hence, proper curing has a positive effect on the final properties of the geopolymer materials (Patil *et al.* 2014; Yaprak *et al.* 2019). The preparation of a geopolymer composite involves the addition of an activator solution, which comprises of a chemical base (NaOH or KOH), water glass (aqueous alkali-silicates  $\text{SiO}_2:\text{M}_2\text{O}$ , where M = Na or K), and water, to the solid materials (*i.e.*, precursor and/or aggregates). These are mixed according to a prescribed mixing scheme to form a paste, which is then placed in molds of specific dimensions. The cast pastes in the mold are then covered or sealed with a plastic film/sheet and cured at ambient temperatures, or a specified temperature for a certain period. At ambient temperatures, however, the reaction of the geopolymer precursors such as fly ash is very slow and the resulting geopolymers correspondingly show a slower setting and strength development (Patil *et al.* 2014; Azevedo *et al.* 2019). Previous studies (Tables 4, 5, 6, and 7) have shown, that higher temperatures increase the reactivity of aluminosilicate phases in the geopolymer precursors. Therefore, they are generally cured at elevated temperatures between 40 and 90 °C but can also be cured at a higher temperature of 105 °C for a short duration.

In addition to the curing conditions, the type and composition of the alkaline activator are crucial for the material properties, since they contribute to the formation of

the structures in geopolymers during the polycondensation process (Van Jaarsveld *et al.* 2002; Shalbahfan *et al.* 2016). Currently, there is a dearth of studies on the influence of curing conditions (*i.e.*, curing duration and curing temperature) and activator solution composition on the strength performance of geopolymer composites from different types of precursors. Such information, however, is important because geopolymers exhibit a wide range of properties and characteristics (Heah *et al.* 2011; Rashad 2013). The precursor/alkaline liquid ratio is a governing factor in the activation process for AA cements. Almost all of the aforementioned studies utilized Ca-rich blast furnace slag as a precursor. The higher amount of CaO in the blast furnace can enhance the mechanical performance of AA cement (Li *et al.* 2017). In the systems with higher CaO contents, the general trend has been that a higher Si/Al ratio (>2.5) leads to a lower strength, *e.g.*, an Si/Al ratio of 1.9 led to higher strength (Li *et al.* 2017). Concerning the activator, higher strength was obtained when a mixture of sodium hydroxide and sodium silicate (added as an aqueous solution called “water glass”) at an optimal SiO<sub>2</sub>/Na<sub>2</sub>O ratio was used instead of sodium hydroxide alone (Živica 2007, Aydin and Baradan 2014), whereas potassium silicate was reported to be more effective than sodium silicate (Zhu *et al.* 2019). The water glass modulus (ratio of SiO<sub>2</sub> to M<sub>2</sub>O) and alkali dosage are other key factors for the hydration of AA cement. Higher flexural strength was achieved using a lower water glass modulus (Zhu *et al.* 2019).

#### *Influence of the lignocellulosic fiber treatment and compatibility*

Empirical evidence has shown that the natural fibers are less compatible with geopolymer matrix in terms of interfacial bonding than their inorganic synthetic counterparts such as glass or carbon fibers (Korniejenko *et al.* 2016). This implies that surface modification of the natural fibers may essentially improve the properties of the reinforced geopolymer composites (Malenab *et al.* 2017). Only a few studies, however, have been carried out to assess the effect of surface modification on the fiber-matrix interfacial interaction and compatibility of lignocellulose materials with geopolymer. One study investigated the effect of alkali-pretreatment using 2M NaOH solution on the mechanical properties of sweet sorghum fiber-reinforced fly-ash-based geopolymer. The pre-treatment induced a significant increase in unconfined compressive strength, the splitting tensile, and flexural strengths as well as the post-peak toughness with fiber content up to an optimum level of 2 wt% compared to respective composites without alkaline pretreatment (Chen *et al.* 2014). The chemical treatments of abaca fibers with sodium hydroxide (6 % by wt. NaOH solution for 48 h) and aluminum salt (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> solution for 12 h) revealed that the highest tensile strength among the treated fibers was achieved after soaking the abaca fibers in the latter solution and adjusting the pH to 6 (Malenab *et al.* 2017). The improvement of tensile strength is partly attributed to the removal of lignin, pectin, and hemicelluloses, and, partly, the roughening of the fiber surface due to the deposition of aluminum hydroxide (Al(OH)<sub>3</sub>). The interconnection between the geopolymer matrix and abaca fibers is thus improved, while the deposition of insoluble aluminum hydroxide protects the fibers from disintegration by the alkaline geopolymer matrix (Malenab *et al.* 2017).

The evaluation of the compatibility of metakaolin-based geopolymer paste with 15 different (wood and non-wood) plant fibers revealed a good interaction. No notable differences were found in the inhibitory effects of the fibers (Tan *et al.* 2019). Ye *et al.* (2018) studied the influence of the common chemical composition of natural fibers *viz.* cellulose, hemicelluloses, and lignin, on geopolymer properties and fiber-matrix

compatibility. Increasing the hemicelluloses content above 5 wt% lowered the degree of geopolymerization. The authors attributed this retarding effect to the presence of carboxylic acid moieties (Ye *et al.* 2018); these are present in glucuronic side-chains or may be generated by saponification of acetyl groups in hemicelluloses.

## CONCLUDING STATEMENTS

1. In recent years, considerable progress has been made in the field of inorganically bonded lignocellulosic composites. Depending on the field of application and the desired characteristics of the final products, various points should be taken into consideration to come to the best possible solution.
2. Lime–lignocellulosic composites are generally low-strength materials compared to composites of conventional ordinary Portland cement; therefore, their application is intended for non-load-bearing and interior parts as well as insulation walls of buildings. Still, they have a promising future as sustainable alternatives in construction, owing to their low embodied energy, as well as their compatibility with a wide range of additives along with being tailorable for diverse applications. Lime enables the production of composites with high moisture buffer capacity by mixing biotic and hydrophilic porous materials with a high breathability matrix such as hydraulic lime. A wide variety of fibers can be incorporated, depending on the application. For improved insulation and moisture behavior rather than the mechanical performance of the composites, a preferable combination would be the mixture of hydrated lime with pith-containing lignocellulosic materials. Pith is a porous low-density spongy tissue with a microstructure that makes it interesting for certain building applications. Combining the permeable hydraulic lime binder with crushed vegetal pith, could result in a lightweight, humidity regulating, and insulating material.
3. Magnesium cement–lignocellulosic composites are carbon neutral and generally possess numerous advantageous properties. These include resistance to fire, chemicals, and temperature as well as low shrinkage and creep. They are easy to manufacture without the use of special equipment and high-energy input. The rapid fast curing nature of magnesium phosphate cement (MPC) and magnesium oxychloride cement (MOC) composites make them well suited for rapid repair/ and construction operations such as prefabrication. They allow for shorter compression time at temperatures close to ambient (near room) temperatures to yield materials having a wide range of functional properties. An interesting feature of MPC and MOC is their ability to accommodate and encapsulate materials. This feature can as well be exploited in the upcycling of out-of-service biocide-treated wood wastes. Magnesium oxysulfate cement (MOSC) and magnesium silicate cement (MSC) lack adequate mechanical strength for structural application. They are, however, more resistant to weathering and moisture than MPC and MOC and could therefore be used in lightweight non-structural applications where sound-, fire-, and water-resistance as well as thermal insulation properties are required.
4. Alkali-activated (AA) cements and geopolymer based–lignocellulosic composites have gained attention due to their high mechanical strength. They can be produced from a wide range of natural minerals or industrial byproducts and also show great potential

in terms of performance properties for application in several fields. Still, the arguable long-term durability of AA cements due to efflorescence and their brittleness raise the question of whether they should be used on a large scale and in exterior environments.

5. The main advantages of geopolymer materials include chemical stability, high compressive strength, low shrinkage, and resistance to corrosive environments. Additionally, geopolymer materials are non-combustible up to very high temperatures (1300 °C), which implies that anything it encapsulates is well protected in high-temperature environments. This feature can be exploited in applications that require high strength properties achievable by the inclusion of lignocellulosic fibers and high retention of strength after exposure to high temperatures. Similar to the MPC and MOC, geopolymers possess the capacity to accommodate a high waste loading, which can also be applied in the upcycling of large volumes of treated wood wastes reaching their end-of-life yearly. The main advantage of such composites is that the biocides are both physically encapsulated and often chemically immobilized. In addition, the composite products are fire-resistant, and therefore, unlike products based on organic polymer binders, hardly emit any harmful gases in the event/presence of fire.
6. The variability in performance properties of non-conventional mineral binder-bonded lignocellulosic composites is related to the manufacturing parameters. Consequently, there is an extensive need for research to optimize the manufacturing process parameters and thus improve their performance characteristics. Much is left to be understood about the influence of various production parameters on the mechanical properties of lime-based materials, especially in flexion. Future work should focus on improving the moisture stability of magnesium-based lignocellulosic composites from MPC and MOC, while enhancing the mechanical strength of MOSC and MSC materials is equally important. AA materials require further understanding to come to a clear conclusion regarding factors influencing the long-term performance of the cementitious matrix. For geopolymer-bonded lignocellulosic materials, there is also a significant need for research to optimize the production parameters, *i.e.*, the precursor type, activator composition, their ratio, and curing conditions to improve performance. In addition, the improvement of fiber-matrix interfacial bonding and the effect of various fiber surface modifications on the mechanical as well as other performance (*i.e.*, physical and water-related) properties of these inorganic mineral binder-bonded lignocellulosic materials requires further elucidation.

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