



Performance and applications of lightweight geopolymer and alkali activated composites with incorporation of ceramic, polymeric and lignocellulosic wastes as aggregates: A review

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ABSTRACT

As the construction industry moves towards greater sustainability, the application of more durable and environmentally friendly materials, capable of providing comfort in buildings and infrastructure, is a key element to consider. In this context, the use of alkali-activated binders (AAB) and geopolymers (GP), which have a lower carbon footprint than ordinary Portland cement (OPC), has emerged as an important alternative. Moreover, the addition of waste-based lightweight aggregates (LWA) to AAB and GP matrices produces lightweight composites that offer enhanced mechanical performance and improved comfort as building materials, while offering an alternative use to the increasing number of waste materials from diverse sources. This paper presents a comprehensive review of the literature on the above-mentioned topics (waste LWA in an AAB/GP matrix) published between 2012 and 2023, mainly indexed in the Scopus database. The waste-based LWA reported in the literature were categorized, and their properties and morphology presented. Then, the influence of the size, quantity, and nature of the LWA on the composite's properties and performance was analyzed. Fresh state performance, mechanical performance, density, and thermal and acoustic insulation were considered. This review is complemented by a bibliometric analysis, where keyword correlation and co-authorship networks on this field are established. The review highlights the potential of cementitious composites including waste-based LWA as a sustainable building material for structural and non-structural applications. However, more studies are required to further understand the behaviour of these composites under innovative manufacturing processes, such as extrusion and 3D printing.

1. Introduction

Concrete is the most widely used building material in the world [1] and, along with water, is one of the most consumed materials on earth [2]. Every year, 30 billion tons of concrete are used worldwide [3,4], which is three times more per capita than 40 years ago. Global concrete needs continue to rise [5], at a rate even higher than for steel and wood demand [4]. This trend is due to its low price, versatility, strength and durability as a building material, and the availability of its constituents [1,2]. However, the growing demand for concrete has significant negative environmental impacts, mostly related to the high consumption of energy and natural resources in

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Abbreviations

AAB	Alkali-activated binder
BA	Bottom ash
CCB	Cellular concrete block
CDW	Construction and demolition waste
EPS	Expanded polystyrene
FA	Fly ash
FCB	Fired clay brick
GBFS	Ground-granulated blast furnace slag
GP	Geopolymer
ITZ	Interfacial transition zone
LWA	Lightweight aggregate
MK	Metakaolin
OPS	Oil palm shell
PET	Polyethylene terephthalate
PIR	Polyisocyanurate
POC	Palm oil clinker
POFA	Palm oil fly ash
PP	Polypropylene
PSF	Polystyrene foam
PU	Polyurethane
PUR	Rigid Polyurethane
PVC	Polyvinyl chloride
RCB	Recycled concrete block
RHA	Rice husk ash
SF	Silica fume
SH	Sodium hydroxide
SS	Sodium silicate
WFC	Waste fibre cement

its production. The cement industry is responsible for about 8% of anthropogenic carbon dioxide emissions [6]. Indeed, one ton of ordinary Portland cement (OPC) releases a ton of carbon dioxide [7]. This alarming situation means that urgent CO₂ emissions reduction from the concrete industry is imperative [8]. Alternatives to the use of Portland cement are required to improve the sustainability of the construction industry and reduce its environmental impact. In this context, the development of alkali-activated binders (AABs) and geopolymers (GPs) has gained relevance in recent years.

Developments of AABs and GPs have been recorded since the 1940s, initially based on the activation of blast furnace slag using sodium hydroxide. In the 1960s, Glukhovskiy developed a type of binder from ground calcium-rich aluminosilicate and alkali-rich industrial waste, which he called “soil cement”. However, research in this field increased significantly following the work of Davidovits, who developed binders derived from the alkali activation of metakaolin and coined the term “geopolymer” in 1978 [9].

Alkali activation is the generic term applied to the reaction of a solid aluminosilicate or calcium silicate source (called “precursors”) under alkaline conditions induced by an “alkali activator”, usually a solution containing sodium hydroxide (SH) and sodium silicate (SS). During this exposure, they undergo a chemical process in which the raw materials are dissolved [10], and an amorphous to semi-crystalline structure is formed, giving the material greater strength [11].

The term “geopolymer” is widely used to describe alkali-activated aluminosilicate binders with an amorphous or 3D microstructure [12,13], which are considered a subset of the wider domain of alkali-activated binders [2,5]. Moreover, alkali-activated calcium-rich cementitious binders have a 2D microstructure modified with aluminium atoms. The source of aluminosilicates varies in different studies, but some materials used extensively for this purpose include class F fly ash (FA) [14–16], and metakaolin (MK) [11,17,18]. In some cases, raw materials such as rice husk ash (RHA) [19,20] and other biomass ashes [21–23] have been used as precursors to adjust the silica or alumina content. The most used calcium-rich raw material was ground-granulated blast furnace slag (GBFS) [24–26].

The greatest advantage of alkali-activated materials is probably their reduced environmental impact. Apart from the aforementioned emissions, the current cement industry places a burden on natural resource deposits and ecosystems [10], whereas alkali-activated binders offer the possibility of using waste products such as ashes [15,20] and slags [26–28], making them ideal for green buildings. Other benefits of AABs include faster strength gain, lower permeability [9], better resistance to acids and sulphates, better heat resistance and lower drying shrinkage [5]. These properties contribute to a more resilient material, which should be considered when discussing the sustainability of the material.

In addition to the environmental impacts of cement itself, the mining and production of conventional aggregates have their own impacts, such as the depletion of non-renewable natural resources and the degradation of ecosystems [29]. Rock-based aggregates are a non-renewable material, and their excessive consumption can lead to the depletion of aggregate sources [30]. For example, concrete

uses 20 billion tons of coarse aggregate per year [31], and 60% of sand mined worldwide is destined for the construction industry [32]. This represents a compelling argument for the use of alternatives to natural aggregates. In this regard, one potential alternative that has gained importance as a replacement to aggregates without compromising the quality of the material is the use of lightweight concrete [33].

Lightweight concrete is designed to have a unit weight below 1900–2100 kg/m³ [34–37]. In most cases this is achieved by using low-density aggregates such as vermiculite, pumicite, diatomite, perlite, expanded clay and expanded glass, which contain voids in their structure [38]. This material is used in the construction of high-rise walls, slabs and precast panels. The lower density of lightweight concrete gives it several advantages over conventional concrete, such as a reduction in dead loads in structures, improvements in thermal and acoustic insulation, fire resistance and ease of transport [39–41].

The above-mentioned properties can be achieved using by-products and wastes from various streams as aggregates [41] to develop cementitious composites. This could represent an opportunity to complement current waste management systems with circular economy practices. The transformation of industrial wastes and surpluses as substitutes for materials extracted from quarries or rivers points is one step in this direction. In this way, it is possible to mitigate the degradation of ecosystems caused by the exploitation of natural resources and the inappropriate final disposal of useable waste. This is especially significant as a growing number of industrial, commercial, agricultural and urban sources are producing waste materials, amounting to an estimated 19 billion tons of waste by the year 2025, which makes waste management activities increasingly difficult [7]. The incorporation of waste aggregates complements the documented use of industrial wastes such as FA, GBFS and biomass ashes as raw materials in the production of alkali activated matrices.

These aggregates can be classified according to their structural nature as ceramic, polymeric and lignocellulosic. The right combination of different technical and environmental properties of the waste can create a functional composite material with a low carbon footprint. Following the macrotrends of creating sustainable construction materials and revalorising waste materials, since 2012, several papers have been published on composites made from GP or AAB matrices that have been lightened by the addition of different waste lightweight aggregates (LWA) [42–44].

Although some reviews of lightweight geopolymer concrete or recycled aggregates are available in the literature, no publications have focused on the classification of waste-based LWA and their use with AABs to produce composite materials. There is even less emphasis on the origin and properties of the aggregates used in the available literature [10]. One review emphasizes the effect of the binder on the performance of lightweight concretes incorporating mostly commercial and some waste-based LWA but does not investigate the potential applications of these materials [10]. Another describes examples of the incorporation of recycled aggregates in these kinds of matrixes but does not specifically address their addition to lightweight materials [7]. This paper presents a thorough literature review of these composites published between 2012 and 2023. The effect of the type, quantity, and size of the waste LWA on the mechanical and physical (water absorption, density, thermal and acoustic) properties of the composites was investigated. In addition, the effect of LWA incorporation on the fresh state behaviour of the materials is included. Furthermore, the potential applications of the reported lightweight composites in the construction industry are investigated. The analysis of the effects of the use of different precursors of residual origin, such as FA and GBFS, on the properties of the compound are left out of the scope of the present paper, as they have been extensively documented a priori.

This review is complemented by a bibliometric analysis of keyword correlation and co-authorship networks constructed using VOSviewer 1.6.18 software. The review shows that aggregates such as oil palm and coconut waste have been found to form structural concretes. Others, such as cork waste, sawdust, and polymer foams, can produce non-structural composites with acoustic and thermal insulation properties that reduce energy consumption and improve thermal comfort in buildings. This paper seeks to elucidate the performance and applications of lightweight geopolymer and alkali activated composites with incorporation of ceramic, polymeric and lignocellulosic wastes as aggregates in the construction sector. This complements the understanding of the behaviour of these materials during linear and 3D extrusion processes, which are the subject of the authors' current research.

2. Methodology

Firstly, a systematic literature review was carried out to study the properties, morphology and pre-treatments of waste-based LWA from different origins. Moreover, the properties of the composites incorporating said LWAs in a GP or AAB matrix were studied, emphasizing the way in which they are influenced by the LWA's own properties and quantity. Secondly, a bibliometric analysis was conducted using the Scopus database and analysis tools to understand author and country research trends on the topic. The analysis also considered the variations of the physical and mechanical properties in both the LWA and the lightweight composites. The searches in Scopus were conducted on January 30, 2023 and focused on the use of published research or conference articles featuring composite materials formed by any type of waste-based LWA or cementitious matrix formed by a GP or AAB binder with possible construction applications, and densities of up to 2100 kg/m³, complying with the standard definition of lightweight concrete [37]. This review did not consider materials incorporating foaming agents, such as hydrogen peroxide or aluminium powder, an alternative method used to decrease the density of these composites. LWAs produced by the industrial transformation of a waste material were not considered either, as the relevant properties of the resulting material can differ significantly to the ones of the original waste.

The selected results, related to publications between 2012 and 2023, were divided into three categories according to their properties: ceramic, polymeric and lignocellulosic. These categories are chosen because they reflect shared characteristics of the aggregates included in each, which in turn have common effects on the behaviour of the composite. The physical properties and morphological characteristics of the LWAs reported in the reviewed papers were collected and summarized in this review. The LWA characteristics considered in the analysis were particle size, morphology, porosity, density, water absorption and pre-conditioning methods.

In addition, the physical and mechanical properties of the composites reviewed were collated. The influence of the considered properties and morphological characteristics of the LWA on the behaviour of the composite in the fresh and hardened state were studied. The properties considered in the analysis of the hardened composite were density, water absorption, mechanical strength and thermal and acoustic insulation of the material. Subsequently, the possible applications of the different composites analyzed according to the referenced studies are also summarized. VOSviewer 1.6.18 software was used to analyse the co-authorship and keyword relationships to this research topic. With this analysis, it is expected to find clusters of authors, and what type of aggregates each one works with, to determine whether the study seeks to respond to a specific problem of a local industry, or the valorisation of a common waste at a global level. Table 1 details the results of the Scopus searches and the strings used.

3. Characteristics of waste-based lightweight aggregates (LWA)

From the bibliographical review, a wide variety of residual materials fragmented into particles, which can be considered lightning agents in cementitious matrices, were identified. According to their nature and general properties, the waste-based LWA found to be used in research settings can be divided into three groups: i) ceramic; ii) polymeric, and iii) lignocellulosic waste.

3.1. Ceramic LWA

Fig. 1 shows some of the waste LWA of ceramic nature considered in this review. It can be observed that these aggregates have a porous appearance, which reflects their physical structural properties, as well as the nature of the material. This category is comprised of different forms of construction and demolition waste (CDW) and bottom ash (BA).

CDW comprise masonry, fired clay brick, stoneware, concrete, glass, and mortar wastes. Concrete-based wastes, such as recycled concrete block (RCB) [43,49,50] are the most studied CDW as aggregates in alkali-activated concretes, followed by fired clay brick (FCB) [51–53] (Fig. 1b). These aggregates have occasionally been impregnated with a phase change material (kerosene) to reinforce the physical and thermal properties of the cementitious composite [45]. Cellular concrete block (CCB) fragments (Fig. 1a) are another low-density CDW with a water absorption greater than 50% [49]. Another CDW-based LWA whose use was recorded is waste fibre cement (WFC) (Fig. 1c) [47].

BA (Fig. 1d and e) is a by-product of coal combustion in power plants and boilers. To a lesser extent, it also originates from the incineration of complex sanitary management solid waste [54]. Compared with stone aggregates, its irregular and porous particles capture more water but have less resistance to abrasive wear. Its low density allows it to be incorporated into construction materials as particles smaller than 9.5 mm in order to lighten them [48,55].

3.2. Polymer LWA

Fig. 2 shows some of the waste LWA used with polymeric properties. Due to their excessive consumption, there is a wide range of residual polymers that can be used as LWA in construction materials based on geopolymer and alkali-activated concretes and mortars.

Table 1
Results of Scopus searches.

Aggregate	Search String	Number of results		
		Unfiltered	Filter 1	Filter 2
All	((“geopolymer*” OR “alkali activ*”) AND (“lightweight”) AND (“concrete” OR “mortar” OR “aggregate” OR “composite”) AND (“waste” OR “recycl*” OR “sustainable” OR “eco-friendly”)) AND (LIMIT-TO (DOCTYPE, “ar”) OR LIMIT-TO (DOCTYPE, “cp”))	174	82	36
Aggregate	Search String - (“geopolymer*” OR “alkali activ*”) AND ...	Number of results		
		Unfiltered	Filter	Filter
			1	2
Palm Oil	“palm oil waste” OR “OPS” OR “oil palm shell” OR “POC” OR “palm oil clinker”	53	26	17
Paper	“paper sludge”	20	6	1
Sludge				
Wood	“sawdust” OR “waste wood” OR “wood shavings” OR “wood chips” OR “wood aggregates”	41	18	5
Cork	“cork”	13	9	4
Coconut	“coconut shell”	18	4	3
Shell				
Biomass	“biomass” AND “aggregates”	29	13	3
Rubber	“rubber” AND (“lightweight” OR “aggregate”)	87	46	10
Polymers	(“PET” OR “plastic waste” OR “PVC” OR “EPS” OR “polyurethane” OR “recycled plastic” OR “packaging foam”) AND (“lightweight” OR “aggregate”)	59	28	13
Bottom Ash	(“bottom ash”) AND (“lightweight” OR “aggregate”)	78	25	4
CDW	(“recycl* concrete” OR “recycl* lightweight” OR “concrete waste” OR “recycl* block” OR “clay brick” OR “recycl* brick” OR “CDW” OR “construction waste” OR “demolition waste” OR “waste ceramic” OR “waste tile” OR “waste fiber cement” OR (“waste” AND “aerated block”)) AND (“lightweight”)	51	11	11

Filter 1: Title and Abstract reading.

Filter 2: Full-text reading.

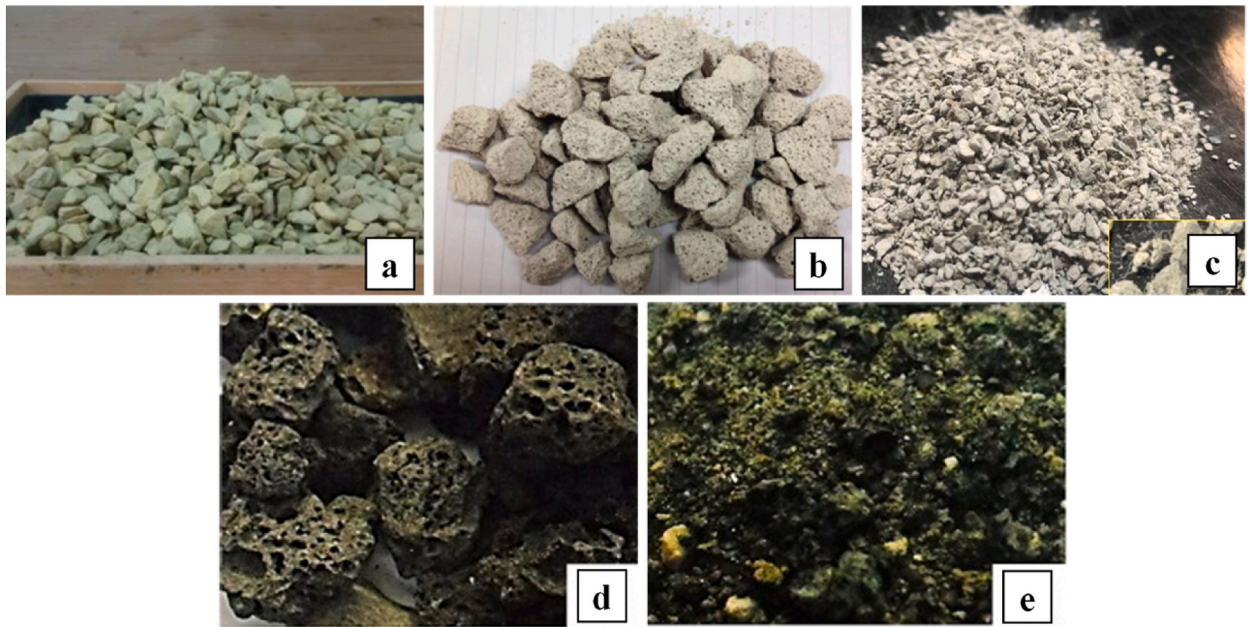


Fig. 1. Morphology of ceramic LWA: a. waste cellular concrete block [45]; b. clay brick waste [46]; c. fibre cement waste [47]. d. coarse bottom ash; e. fine bottom ash [48].

Among the thermoplastic polymers evaluated as LWA for concrete, polyethylene terephthalate (PET) from post-consumer packaging was particularly noteworthy [42,57,62–64]. Research on PET as an aggregate highlights the variety of shapes such as flakes (Fig. 2a), strips (Fig. 2b) and particles in which it can be used (Fig. 2c) [42,56,56,57,63]. Other thermoplastics used as aggregates were polyvinyl chloride (PVC) waste from insulation coating for energy transmission wires [65] and polypropylene beads (Fig. 2d) [66]. Instances of the use of assorted scraps originating from different kinds of plastic waste as aggregate have also been documented [67].

However, the polymeric waste-based LWA that has attracted the most research is rubber from discarded tires (Fig. 2e). This interest is justified, as annually more than half of the 500 million discarded tires are buried in landfills [68,69]. This problem is becoming increasingly complex as world tire production reached 2.9 billion units in 2017 [70]. Shredded waste rubber has been incorporated in asphalt and concrete mixes. However, when added in large quantities, it deteriorates the properties of concrete, since its compressive strength and elastic modulus are lower than those of stone aggregates [71]. Rubber is hydrophobic; one possible solution for this is to modify its surface conditions with alkaline treatments to improve its adherence to cement pastes [72].

Other materials considered were porous thermoplastics recycled from food packaging, industrial products, thermal insulation, such as expanded polystyrene (EPS) (Fig. 2f) [73] or polystyrene foam (PSF) (Fig. 2g) [60], polyisocyanurate (PIR) and polyurethane (PU)

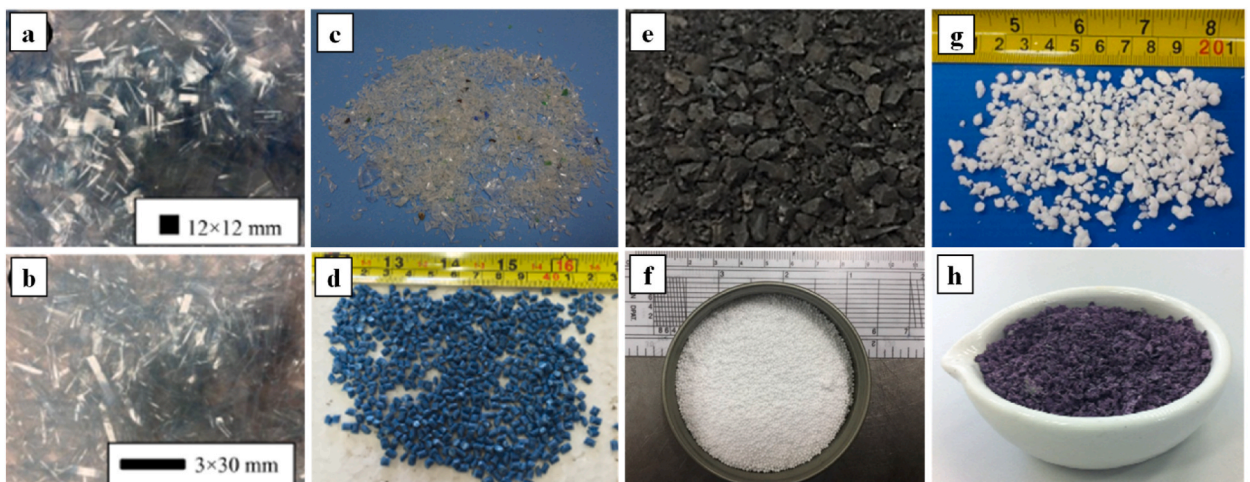


Fig. 2. Morphology of polymer LWA: a. PET flakes, b. PET strips [56]; c. waste PET granules [57]; d. recycled plastic beads; e. shredded rubber [58]; f. EPS beads [59]; g. recycled polystyrene foam [60]; h. PU/EVA [61].

foam have all aroused great interest [67,74]. In order to decrease both the density and the thermal conductivity in concrete, mixtures of polyurethane (PU) and ethylene vinyl acetate (EVA) (Fig. 2h) from footwear manufacturing surpluses [61,75] have been added.

Most polymeric aggregates were shredded until particles with a size of less than 5 mm were obtained. They were considerably less dense than stone aggregates but absorbed more water when they had open porosity. The exceptions to this are PET in the mentioned shapes, and shredded rubber, which was used in sizes up to 7 mm [56,71].

3.3. Lignocellulosic LWA

Agricultural and wood industry residues with lignocellulosic structures have been grouped in this section. The origin and characteristics of those lignocellulosic residues used as LWA in geopolymer, and alkali-activated cements are described below. Fig. 3 shows some of the lignocellulosic waste LWA used.

According to the selected literature, waste from the palm oil industry is the most used LWA. *Elaeis guineensis* is widely cultivated in Indonesia and Malaysia, which are the largest and second largest producers respectively of oil palm in the world [81]. The increasing demand for palm oil has resulted in the generation of large amounts of waste such as oil palm shell (OPS) and palm oil clinker (POC) [22]. OPS is derived from the hard endocarp that surrounds the kernel (Fig. 3a and b). POC is a waste (Fig. 3c–e) obtained from the calcination process of OPS and other by-products to produce energy [82,83]. It has a porous nature because of the combustion of the essential oils in the oil palm waste. Both OPS and POC have been used in Portland cement concrete as an alternative to reduce the harmful effects of landfill disposal [82,84,72]. Both OPS and POC are crushed and screened to adjust their particle size as aggregates. As a result of the pyrolysis process, the oil residues are removed and the POC acquires a higher density and specific gravity than the OPS. The oil that coats the OPS particles is removed by washing and air-drying processes; this favours adhesion with cementitious pastes, and therefore increases the resistance of cementitious composites [85,86]. The water absorption of OPS is 25%, compared to 1% for granite [85]. To reduce this, OPS particles can be immersed in polyvinyl alcohol (PVA) for 30 days to form a hydrophobic coating [86].

Other residues used include coconut shells, cork, wood waste, paper sludge and ground coffee waste. World production of coconut (*Cocos nucifera*) [87], exceeded 61 million tons in 2020 [88]. The slow decomposition of coconut waste degrades the soil around the harvesting site. To make use of this discarded material and reduce the pollution it causes, hard coconut shell has been used as an LWA in concrete [89]. It is crushed into particles up to 20 mm (Fig. 3f) [90,91]. As a pre-treatment, it can be immersed in water and partially dried by solar radiation, so that the moisture retained in the shell prevents the absorption of water from the cementitious binder, thereby improving concrete workability [89,90]. The low content of metal oxides in the shell suggests a lack of pozzolanic actions, making it suitable for use as LWA [90].

Wood waste, such as sawdust, shavings and bark, represent 5–10% of wood production losses and can be considered recoverable biomass [92]. Its low thermal conductivity [44] has encouraged its application in the thermal insulation of buildings and homes [93]. Wood sawdust (Fig. 3g and h) up to 8 mm has been used as LWA in alkali-activated binders and geopolymers [79,94]. As shown in Table 2, the wood residues used as LWA vary in morphology, which gives different properties to the cementitious compounds that contain them.

Cork is obtained from the cork oak (*Quercus Suber* L.) by partially removing its bark, which allows the tree to regenerate [125]. In Portugal, the main producer, up to 201,000 tons are processed annually for various applications [23]. Surplus cork from the manufacture of products such as cork slabs and wine stoppers has been converted into LWA, which can be shredded into particles of up to 6 mm and added to a composite mix [23,109]; or pyrolyzed to reduce its weight by 70% and then expanded in volume in an autoclave at



Fig. 3. Morphology of lignocellulosic LWA: a. Uncrushed OPS; b. Crushed OPS [76]; c. POC fragment taken from the factory, d. POC coarse aggregate, e. POC fine aggregate [77]; f. coconut shell [78]; g. fine sawdust; h. coarse sawdust [79]; i. spent coffee ground [80]; j. expanded cork granules [23].

Table 2
Waste-based LWA found in Scopu, classified by type of aggregate.

Ceramic				
LWA	Particle size (mm)	Density (kg/m ³) or SG ^a	Water absorption (%)	Reference
Bottom Ash	0.3–2.5	SG: 1.87	5.45	[95]
Bottom Ash	Fine	SG: 2.05	7.24	[96]
Bottom Ash	0.075–9.50	1116–1371	3.32–3.53	[48]
Bottom Ash	4.75–9.50	1230	3.83	[55]
Concrete + FCB	0.5–2.0	1210–1290	–	[97]
FCB	0–4.75	SG: 2.12	16	[52]
FCB	Coarse	870–970	27.7	[51]
FCB	0.075–10.0	1008–1040	13.04–15.34	[53]
Recycled Concrete	0–4.75	1449	–	[50]
RCB	0–4.75	1468	–	[50]
CCB	0–12.5	360–720	56–76	[49]
CCB	0–12.5	360–720	56–76	[43]
CCB	0–12.5	360–720	56–76	[98]
CCB	9.5–19	400	71 (5–8.5 w/PCM)	[45]
WFC	0.125–4.75	1124	12.24	[47]
Polymeric				
LWA	Particle size (mm)	Density (kg/m ³) or SG ^a	Water absorption (%)	Reference
EPS	0–5	16	–	[73]
EPS	1.0	30	–	[59]
PET	Coarse	138	–	[42]
PET	0–4.75	552	–	[64]
PET	0–30	–	–	[56]
PET	0.315–1.25	163	–	[63]
PET	0–4	127	–	[57]
Plastic Scraps	–	–	–	[67]
PIR foam	Powder	110	–	[74]
PP beads	Fine	546	0.7	[66]
PSF	2.36–4.75	215	225	[60]
PU foam	Powder	85	–	[74]
PU + EVA	0.15–6	283–314	–	[75]
PU + EVA	0.15–6	283–314	–	[61]
PUR foam	–	–	–	[67]
PVC	0–2	–	–	[65]
Rubber	0.15–4.75	620	–	[99]
Rubber	–	–	–	[67]
Rubber	–	–	–	[100]
Rubber	0–2.36	1187	0.63	[101]
Rubber	2.0–7.0	–	17.20–22.60	[71]
Rubber	0–2	–	–	[65]
Rubber	0–1	380	–	[102]
Rubber	1.0–7.0	SG: 0.54	85	[70]
Rubber	0–3	1202	–	[103]
Rubber	0–4	487	3.27	[104]
Rubber	0.14–1.92	–	–	[105]
Rubber	0–4.75	487	3.27	[106]
Lignocellulosic				
LWA	Particle size (mm)	Density (kg/m ³) or SG ^a	Water absorption (%)	Reference
Coconut Shell	1.2–17	1270	21.6	[91]
Coconut Shell	0–12.5	550	22.0	[78]
Coconut Shell	18–20	695	–	[90]
Coffee ground	Median: 0.244	400	–	[80]
Cork	0.8	–	–	[107]
Cork	6	70	–	[108]
Cork	0.5–6	64–76	–	[109]
Cork	6	70	–	[23]
OPS - Crushed	Coarse	SG: 1.32	24.74	[110]
OPS - Crushed	Fine	–	–	[111]
OPS - Crushed	0–9	655	24.7	[85]
OPS - Crushed	0–9	655	24.7	[76]
OPS - Crushed	5–9	638	24.3	[86]
OPS - Crushed	1.18–14	596	22.78	[112]
OPS - Crushed	1.18–14	596	22.78	[72]
OPS - Crushed	2.36–9.5	652	24.39	[113]
OPS - Crushed	2.36–9.5	652	24.39	[22]
OPS - Crushed	2.36–9.5	652	24.39	[21]

(continued on next page)

Table 2 (continued)

Ceramic				
LWA	Particle size (mm)	Density (kg/m ³) or SG ^a	Water absorption (%)	Reference
OPS - Crushed	2.36–9	SG: 1.35	–	[114]
OPS - Uncrushed	0–14	633	25.7	[85]
OPS - Uncrushed	0–14	633	25.7	[76]
Paper Sludge	0–3.36	–	–	[115]
POC	5–14	780	–	[116]
POC	0–4.75	SG: 1.94–2.11	3.30–5.69	[117]
POC	0–4.75	SG: 1.92	3.30	[77]
POC	0–14	823–1096	4.43–14.65	[35]
POC	0–14	823–1096	4.43–14.65	[118]
POC	0–18	693–1015	–	[119]
Pyrolyzed Cork	0–0.075	140	–	[120]
Sawdust	Fine	SG: 0.41	4.5	[121]
Sawdust	0–6	174–182	–	[79]
Sawdust	Length: 0-15	790	67	[122]
Sawdust	0–0.8	650	–	[123]
Sawdust - Fine	0.07–0.15	140–200	–	[94]
Sawdust - Coarse	3–5	150–190	–	[94]
Wood Chips	0.5–2	320	–	[124]
Wood Fiber	Length: 3-7	100–230	–	[94]
Wood Shavings	0.25–8	153.7	–	[44]
Wood Splinters	–	–	–	[107]

^a SG: Specific gravity. The value provided corresponds to density unless stated otherwise.

300 °C [120] (Fig. 3i).

Paper sludge is a by-product of wastewater treatment processes, typically those of the paper recycling industry. In some cases, this material is incinerated to produce paper sludge fly ash. To enable its use as a fine aggregate, it is necessary to dry the sludge in an oven so that it can be crushed and screened to the desired size [115]. Ground coffee waste is ubiquitous, as coffee is one of the most consumed beverages in the world and requires no further pre-processing for use in mortars [80] (Fig. 3j).

Table 2 summarises the main physical properties of the aforementioned LWA when incorporated into geopolymeric or alkali activated matrices. In addition, Fig. 4 shows a summary of the aggregates found in the literature reviewed.

The aggregate type with most publications was lignocellulosic LWA, followed by polymeric and ceramic (Fig. 4). Of the lignocellulosic LWA, and indeed of all the LWA types, oil palm shell (OPS) is the most studied, with 13 publications. Rubber, with 11 publications, and concrete aggregates, with 6 publications, are the polymeric and ceramic LWA with the highest number of publications respectively.

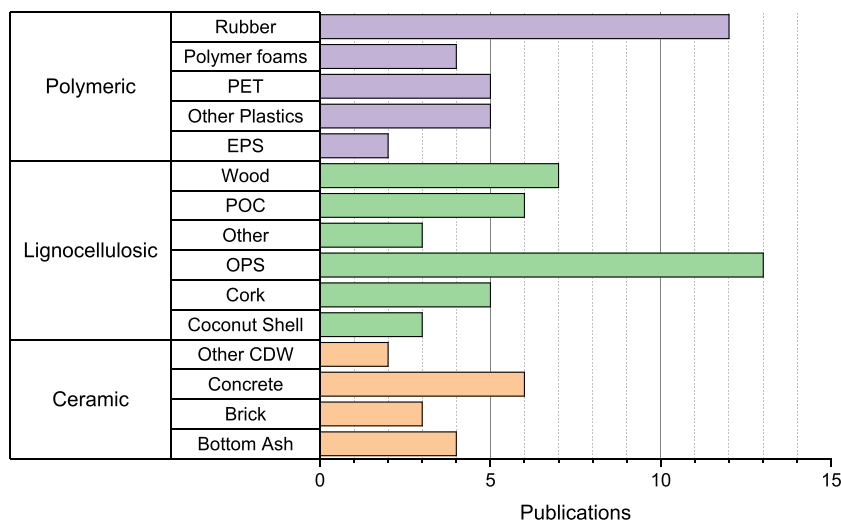


Fig. 4. Distribution of publications according to the types of LWA found in literature.

Table 3
Composites containing waste-based LWA found in Scopus.

Ceramic							
LWA	Precursors	Alkaline activators	Density (kg/m ³)	28d Compressive strength (MPa)	Water absorption (%)	Thermal conductivity (W/m-K)	Reference
Bottom Ash	FA + GBFS	SH	1620–1740	1.0–15.0	–	–	[95]
Bottom Ash	FA	SH + SS	869–1891	0.20–12.27	–	–	[96]
Bottom Ash	FA	SH + SS	1661–1688	14.3–18.1	–	0.43–0.47	[48]
Bottom Ash	FA	SH + SS	1466–1502	5.7–8.6	–	0.30–0.33	[55]
Concrete + FCB	Biomass FA + MK	SH + SS	1780–1820	22–42	12.1–12.3	–	[97]
FCB	FA + GBFS + OPC	SH	1800–2100	17–40	–	–	[52]
		SH + SS					
FCB	FA + Glass Powder	SH + SS	1745–1835	19.7–24.9	4.79	–	[51]
FCB	FA	SH + SS	1685–1749	8.2–18.3	–	0.62–0.65	[53]
Recycled Concrete	FA	SH + SS	1650–1740	4.5–9	11.1–16.5	0.56–0.64	[50]
RCB	FA	SH + SS	1700–1750	2.5–8	10.4–13.6	0.6–0.7	[50]
CCB	FA	SH + SS	860–1400	1–16	9.5–32.5	–	[49]
CCB	FA + OPC	SH + SS	1100–1350	2.0–14.5	–	–	[43]
CCB	FA	SH + SS	860–1495	2.0–17.7	–	–	[98]
CCB	FA + SF	SH + SS	1670–1825	4.9–8.3	–	–	[45]
WFC	FA	SH + SS	1750–1913	13–37	8.9–12	0.38–0.83	[47]
Polymeric							
LWA	Precursors	Alkaline activators	Density (kg/m ³)	28d Compressive strength (MPa)	Water absorption (%)	Thermal conductivity (W/m-K)	Reference
EPS	MK + Marble Powder	SH + SS	516–827	1.8–6	–	0.12–0.21	[73]
EPS	FA + Calcium Carbide Residue	SH + SS	705–990	1.5–3.4	40–50	–	[59]
PET	GBFS	SH + SS	1120–1930	1,8–29,6	8–25	–	[57]
	GBFS + MK						
PET	FA	SH + SS	1333–1837	3.54–10.39	8.56–19.65	–	[42]
PET	FA	SH + SS	1585–1838	12.8–42.7	7.9 - 8.4	0.64–1.01	[64]
PET	FA	SH + SS	–	10.5–14.2	–	–	[56]
PET	FA	SH + SS	1565–1790	13.0–14.0	6.73–8.18	0.38–0.83	[63]
Plastic Scraps	FA	SH + SS	1450	6.5–7.1	–	0.16–0.34	[67]
Plastic Scraps	FA + GBFS	SH + SS	1500	18.2	–	0.16–0.34	[67]
PIR foam	MK	SH + SS	1030–1149	8–11	Porosity: 10.5	0.36	[74]
PP beads	FA	SH + SS	1000–1500	15.1–35.8	7.8–11.7	0.32–0.81	[66]
PSF	FA	SH + SS	1050–1350	7–10	–	0.27–0.35	[60]
PU foam	MK	SH + SS	885–1126	6.5–11	Porosity: 12.7	0.30	[74]
PU + EVA	FA + MK	SH + SS	2100–2120	1.42–3.07	27.5–30.5-	–	[75]
PU + EVA	FA + MK	SH + SS	2100–2120	1.42–3.07	27.5–30.5-	–	[61]
PUR foam	FA	SH + SS	950–1250	5.4–8.3	–	0.16–0.34	[67]
PUR foam	FA + GBFS	SH + SS	1050–1120	6.3–15.5	–	0.16–0.34	[67]
PVC	FA + GBFS	SH + SS	1790–1870	13–36	1.5–3.95	1.20–1.39	[65]
Rubber	FA	SH + SS	1880–2000	9.5–12	–	–	[99]
Rubber	FA	SH + SS	1500	4.1–4.3	–	0.16–0.34	[67]
Rubber	FA	SH + SS	1400–1800	10–33	4–15	–	[100]
Rubber	FA + GBFS	SH + SS	1250–2000	7.0–11.5	Porosity: 19–25.5	–	[101]
Rubber	FA + GBFS	SH + SS	1716–2010	1.3–25.4	3.8–7.7	–	[71]
Rubber	FA + GBFS	SH + SS	1700–1970	9–28	3.2–4.55	1.3–1.39	[65]
Rubber	MK	SH + SS	1810–2010	9.00–42.93	–	–	[102]
Rubber	FA + GBFS	SH + SS	1980	11.7	–	–	[70]
Rubber	FA + GFBS + SF	SH + SS	1480–1750	4–17	7.8–13.5	–	[103]
Rubber	FA	SH + SS	1067–1275	2.07–3.29	7.8–17.1	0.237–0.298	[104]
Rubber	GFBS + Glass Powder	SH	1500–1950	15–39.5	–	0.75–1.80	[105]
Rubber	FA	SH + SS	1075–1697	2.7–12.3	9.9–18.4	0.187–0.576	[106]
Lignocellulosic							
LWA	Precursors	Alkaline activators	Density (kg/m ³)	28d Compressive strength (MPa)	Water absorption (%)	Thermal conductivity (W/m-K)	Reference
Coconut Shell	FA + GBFS	SH + SS	1820	22.3	–	0.56	[91]
Coconut Shell	GBFS	SH + SS	1980	32.60	–	–	[78]
Coconut Shell	GBFS	SH + SS	2013–2078	40.4	–	–	[90]
Coffee grounds	MK + Biomass FA	SH + SS	1648–1832	10.63–21.66	13–16.49	0.47–0.575	[80]
Cork	GBFS	SH + SS	1000	5.3	–	0.154	[107]
Cork	Biomass FA + MK	SH + SS	200–900	0.04–4	–	0.068–0.1870	[108]
Cork	Biomass FA + MK	SH + SS	300–1200	0.8–19	–	0.06–0.28	[109]

(continued on next page)

Table 3 (continued)

Ceramic							
LWA	Precursors	Alkaline activators	Density (kg/m ³)	28d Compressive strength (MPa)	Water absorption (%)	Thermal conductivity (W/m-K)	Reference
Cork	MK	SH + SS	230–830	0.23–2.8	–	0.072–0.156	[23]
OPS - Crushed	POFA + MK	SH + SS	1782	30	–	–	[110]
OPS - Crushed	FA	SH + SS	–	11–32	–	–	[111]
OPS - Crushed	GFBS + POFA	SH + SS	1918–1935	29–33	–	–	[85]
OPS - Crushed	GFBS + POFA	SH + SS	1873–1978	21–29	–	–	[76]
OPS - Crushed	POFA + GFBS + MK	SH + SS	1900	25	10.84	–	[86]
OPS - Crushed	FA	SH + SS	1744–1824	13.0–32.7	–	–	[112]
OPS - Crushed	FA	SH + SS	1434–2052	9.61–32.63	–	–	[72]
OPS - Crushed	FA + POFA	SH + SS	1791	30.1	–	0.58	[113]
OPS - Crushed	FA + POFA	SH + SS	–	30.1	4	–	[22]
OPS - Crushed	POFA + FA	SH + SS	1861.1–1997.3	9.7–30.1	–	–	[21]
OPS - Crushed	FA	SH + SS	–	25.9	–	–	[114]
OPS - Uncrushed	GFBS + POFA	SH + SS	1900–1915	27–32	–	–	[85]
OPS - Uncrushed	GFBS + POFA	SH + SS	1843–1994	20–27	–	–	[76]
Paper Sludge	FA	SH + SS	2025	28	9	–	[115]
POC	FA	SH + SS	–	20–27	–	–	[116]
POC	FA	SH + SS	1805–1900	55–64	7.8–9.9	–	[117]
POC	FA + GBFS						
	FA	SH + SS	1710–1753	19.8–53	8.8–11.0	–	[77]
POC	FA + GBFS						
	FA	SH + SS	1821–1942	32.7–46.4	1.8–3.2	–	[35]
POC	FA	SH + SS	1821–1942	32.7–46.4	1.8–3.2	–	[118]
POC	FA + RHBA	SH + SS	1573–2038	–	0.8–0.83	–	[119]
Pyrolyzed Cork	Biomass FA + MK	SH + SS	1180–1200	15–16	–	–	[120]
Sawdust	FA + GBFS	SH	1650–2050	1.2–6.5	8–13	–	[121]
Sawdust	FA + GBFS	SH + SS	890–1980	48.6–61.1	10.1–16.9	0.09–0.24	[79]
Sawdust	FA	SH + SS	1580–1980	62–68	–	–	[122]
Sawdust	MK	SH + SS/RHA	700–1600	Flex: 1.8–12.5	–	0.15–0.57	[123]
Sawdust - Fine	FA + MK	SH + SS	1417–1471	36.2–38.4	–	–	[94]
Sawdust - Coarse	FA + MK	SH + SS	1215–1329	20.2–20.8	–	–	[94]
Wood Chips	GBFS	SH + SS	1010–1250	4–13	4–13	0.15–0.23	[124]
Wood Fibre	FA + MK	SH + SS	1329–1426	29.1–29.9	–	–	[94]
Wood Shavings	GFBS + Phosphogypsum	SH	1050–1800	3.0–23	10.7–25.3	0.203–0.411	[44]
Wood Splinters	GBFS	SH + SS	1100	6.1	–	0.073	[107]

4. Performance of composite materials featuring alkali activated cements with waste-based LWA

Table 3 summarises the composition and properties of various composites. It shows the precursors, alkaline activators and aggregates forming each composite, as well as some of their discussed properties such as density, 28-day compressive strength, water absorption, and thermal conductivity.

4.1. Fresh state behaviour

The fresh state characteristic that describes the capacity of cementitious composites lightened with residual LWA is called workability. This feature, directly related to fluidity and slump, is affected by the quantity, size, and surface condition of the residual LWA particles. In this respect, it was found that increasing the volume of OPS or POC added to the cementitious agents resulted in a reduction to workability [35,76,77]. In other studies, the same effect was observed when stone aggregates were substituted with wood sawdust. Reduction of up to 43% in the slump of the composite was achieved when the substitution reached 100% [79]. Similar behaviour was reported for composites incorporating increasing amounts of other residual lignocellulosic LWA such as coconut husk and cork. Another cause of the loss of workability of composites is related to the absorption of water into the residual LWA particles. This is possibly a consequence of the increase in the specific contact surface area as the LWA particle size decreases. A plausible explanation for this could be the increase in inter-particle friction due to the decrease in particle size [79,85].

In various studies, the workability of composites has been shown to be hindered due to the decrease in the initial and final setting times of the cementitious materials. Curing times were significantly reduced in composites with lignocellulosic LWA. This was because these LWA absorbed part of the alkaline solution added to form the cementitious paste into their capillary structure [79]. A 69–76% reduction in setting times was also observed when 40% of the stone aggregates were replaced by residual fibre cement particles (WFC) [47]. Likewise, a 79% decrease in the flowability of the cementitious material was reported because of the rapid formation of gels induced by the CaO contained in the WFC. It was also found that hydrophilic character of the cellulose fibres contained in the WFC and the rough morphology of its particles reduce the workability of the composite.

Studies about composites with shredded tire rubber show that workability reduction is induced both by the amount of these

particles and their angular and rough morphology [104]. It was possible to form composites with acceptable workability by replacing up to 30% wt. of stone aggregates with tire rubber [71]. Similarly, it was observed that the flowability did not vary significantly in composites with substitution of up to 40% of the stone aggregate with PET from shredded containers [63]. However, the flowability decreased in the range of 7.0–13.4% when the substitution with PET was between 60 and 100%. In both cases, it should be considered that the amount of PET rubber needed to provide adequate workability could vary depending on its particle size.

Unlike lignocellulosic LWA, rubber is hydrophobic, a characteristic that, together with the roughness of the particles, promotes the nucleation of air bubbles on its surface. This is undesirable because it generates a porous interfacial transition zone (ITZ) between the rubber particles and the cementitious material that covers them. Porosity in the ITZ decreases the compressive and flexural strengths of composites [58,71,104]. Furthermore, it enhances the water absorption of composites in their hardened state [57,66,75], limiting their application in construction products.

In composites incorporating bottom ash as LWA, cohesive forces develop between the particles and the cementitious material that impede the slump test [85]. The loss of workability in these composites can be compensated by immersing the LWA for 24 h in water, as the saturated LWA releases water into the cementitious material, increasing its flowability. To achieve the same effect in composites lightened with tire rubber, it was recommended to immerse the particles of this LWA in water [70] or in sodium hydroxide (SH) solution [71]. While the first method increased the binding energy in the ITZ, the second method introduced surface roughness in the rubber particles, providing greater anchorage with the cementitious material. In composites made up of lignocellulosic LWA such as wood sawdust, it was recommended to incorporate superplasticizers to compensate flowability losses [79]. All these methods had the added value that the water or alkaline solution absorbed by the saturated LWA was gradually released, promoting internal curing [47, 108]. The benefit of this is that the mechanical strength of the composites in the hardened state can be increased.

4.2. Hardened state behaviour

4.2.1. Density

The purpose of adding LWA to any concrete is to reduce its weight and thereby optimize the construction processes of buildings and infrastructure. The density of cementitious composites was affected by the nature, size, and morphology of the LWA [94]. Density of the composite was reduced according to the nature of the LWA in the following order: ceramics, polymers (thermoplastics and elastomers) and lignocellulosic.

By incorporating between 40 and 60% of fragments (9.5–19 mm) of cellular concrete blocks (CCB), composites of 1792 and 1670 kg/m³, respectively, were obtained. When the particles of this LWA were impregnated with paraffin, the densities increased between 1825 and 1688 kg/m³ [45]. This slight increase in density was related to the sealing of the surface pores of the particles. The substitution of stone aggregates with a mixture of pumice fragments and clay bricks (FCB) produced composites of 1685 and 1749 kg/m³, respectively [53]. Since the proportions of both aggregates were equal, it can be assumed that the difference in density was associated with the porosity of both LWA. In composites incorporating CCB particles, the density ranged between 860 and 1400 kg/m³ [49]; while another study reported a variation between 1200 and 1500 kg/m³ [98]. In addition, a decrease in composite density was observed as LWA particle size increased. Different LWA dosing methods were used in both studies and LWA porosity was not reported. Therefore, it should be considered that LWA porosity has a possible influence on composite density. However, in bottom ash lightened composites, the density was reduced by 4.6% when fine particles were used and by 9.3% with coarse particles. This occurred because a higher proportion of coarse particles was incorporated into the cementitious material [126]. In both cases the density of these composites was less than 2000 kg/m³. In another study, where no discrimination was made between fines and coarse bottom ash, the density of the composite ranged from 1661 to 1688 kg/m³ [48].

In some composites, LWA of ceramic nature was replaced by LWA extracted from waste polymers. Substitution of 60–80% of slag granules by PET flakes (same granulometry) produced composites of less than 1850 kg/m³. These composites were classified as lightweight structural concretes according to ACI 213R [57]. Replacing quartz sand with PET particles (0.315–1.25 mm) produced composites with densities below 1565 kg/m³ [63]. The density was reduced by 14.7% when 100% of the sand was replaced by PET. Similarly, with the substitution of 25 and 50% wt. of river sand with recycled polypropylene (PP) pellets, composites of 546 kg/m³ were produced [66]. These composites were also classified as lightweight structural concrete according to ASTM C330.

Composites in which stone aggregates were replaced by rubber at between 15 and 60% vol had densities ranging from 1067 kg/m³ [104] to 2125 kg/m³ [70]. Composites of 2125 kg/m³ and 1980 kg/m³ were obtained when the fine (1–3 mm) and coarse (5–7 mm) aggregates were replaced with 15 and 30% vol of rubber, respectively [70]. A different study with 15% wt. rubber produced composites from 1900 to 2000 kg/m³ and 1700 kg/m³ when the substitution reached 30% wt [71]. Here, the fine rubber fraction was larger (3–5 mm), while the coarse fraction was in the same range (5–7 mm). This difference in density could be due to the characteristics of the cementitious materials; but it could also be modified by the size of the rubber particles. This effect was evidenced in composites where 40% vol of the stone aggregate was replaced with fine (1–5 mm) and coarse (5–10 mm) rubber particles [58]. Composites made with fine rubber were found to be denser (approx. 2150 kg/m³) than those incorporating coarse rubber (approx. 2100 kg/m³). The density was further reduced (approx. 2060 kg/m³) by replacing the aggregate with 20% fine rubber +20% coarse rubber. The authors do not explain this behaviour, which could have been induced by a higher content of entrapped air in the fresh state.

It is evident that the molecular weight and porosity of the polymeric LWA contributed to decreasing the density of the composites. Further lightening of matrices (geopolymeric and alkali activated) was achieved with the addition of foamed thermoplastics. This is due to their cellular structure that stores air or gas. By incorporating particles (5 mm) of expanded polystyrene foam (EPS), composites of 516–840 kg/m³ were obtained [73]. With the addition of 14–20% wt. of rigid polyurethane foam (PUR), metakaolin (MK) composites

of 923 to 885 kg/m³ were formed [74]. In contrast, 40% substitution of sand with PU/EVA pellets produced metakaolin (MK)-flying ash (FA) composites of 2100 kg/m³ [75]. The increase in density was due to degassing of the pellets during extrusion. Likewise, fly ash mortars yielded densities of 1000–1300 kg/m³ when 0.85–1.05% wt. of river sand was substituted with particles (2.36–4.75 mm) of polystyrene foam (PSF) [60]. Polystyrene foams such as EPS (16 kg/m³) [73] and PSF (215 kg/m³) [60] may differ in porosity. None of the above-mentioned studies characterized the cell structure of both foams.

Lower densities were reported in composites incorporating lignocellulosic LWA. Replacing 100% of the stone aggregates with fine (0–5 mm) and coarse (5–14 mm) POC particles resulted in composites of 1821 kg/m³, which represents a density reduction of 22.3% [35,77,86,118]. In other composites, the density reduction was 17% when sand was replaced with 4.75 mm POC particles [77]. A higher density reduction was reported for composites in which 50% wt. of stone aggregates were replaced with fine (0–7 mm) and coarse (7–18 mm) POC particles [119]. Therefore, it seems to be confirmed that larger size of LWA leads to lower densities. However, in one study it was found that with 70% fine POC +30% coarse POC the density of the composites was 1573 kg/m³, while with 30% fine POC +70% coarse POC it was 1618 kg/m³ [119]. The reasons for this discrepancy were not explained by the authors of the study. Small POC particles exhibit a more porous surface, which promotes the formation of bubbles in their interface [22,35]. In composites where 100% of stone aggregate was replaced by coconut shell (4.7–7 mm) the density was 1820 kg/m³. The density of the composite increased to 1945 kg/m³ when using 100% coconut particles previously coated with the matrix cementitious agent [91]. This pre-treatment was performed to improve the compatibility between the matrix and the coconut husk LWA.

By adding 10% wt. of wood chips to a phosphogypsum and slag cementitious agent, a composite of 1027 kg/m³ was obtained [44], equivalent to a 44% reduction in density. Replacing 100% of river sand with sawdust (6 mm), composites of 890 kg/m³ were developed [79]. Composites made with LWA from wood showed lower densities than those made with POC. This may be associated with the higher porosity of wood LWA, which makes them less dense. Consequently, the density of the composites was further decreased by the incorporation of lignocellulosic LWA with a porous cellular structure such as cork. This was evidenced in composites that replaced up to 90% volume of stone aggregates with cork particles [109,108]. The cork fractions reproduced the proportions and sizes of the stone aggregates (sand: 0–4 mm, gravels: 4–8 mm and 8–16 mm). The density of these composites with 100% substitution was 716.5 kg/m³, which was a 70% decrease with respect to the matrix (2383.7 kg/m³). When the aggregates were substituted with cork (5.7 mm) at 65–92% vol, the composites showed densities between 830 and 260 kg/m³, respectively [23]. The addition of 92% cork resulted in a 77% reduction in density with respect to the matrix (1150 kg/m³). Likewise, adding 45–90% vol of expanded cork (6 mm) to the cementitious materials resulted in composites of 168 kg/m³. Substitution of 90% led to a decrease of 86.62% in density with respect to the cementitious matrix (1168 kg/m³) [108]. It is inferred that the density of the composites is determined by both the size and the amount of LWA and the density of the cementitious matrix. Also, composites of 750 kg/m³ were formed when 2.5–3.75% wt. of pyrolyzed cork (75 µm) was added to the cementitious materials [120]. It is not possible to establish whether greater lightning is provided by virgin or polarized cork LWA, since the former was dosed in volume, and the latter in mass.

It is deduced that the density of the composites was influenced by their porosity, which determines their density and hydrophilic character. This characteristic, together with the surface roughness of the LWA, controls the adhesion with the cementitious material. In addition, it was evident that increasing the amount of aggregate decreases the density of the composites. On the other hand, as the size of the LWA is reduced, the density of the composites increases due to higher compactness between the particles [94].

Fig. 5 shows the change in composite density as a function of the amount of waste LWA that replaced the stone aggregate. The data shown in the graphs corresponds to some of the densities recorded in the referenced articles.

It is confirmed that, regardless of their nature, the incorporation of waste LWA decreases the density of the composites. From the slopes it can be inferred that some LWA cause a greater reduction in the density of the composites than others. These were bottom ash and FCB (ceramic LWA, Fig. 5a); PET (polymeric LWA, Fig. 5b); and cork and coffee (lignocellulosic LWA, Fig. 5c). In the case of PET, one composite departs from this trend [63], which could be associated with the incorporation of finer particles [58,119]. It was possible to reduce the density of the composites to 1000 kg/m³ with ceramic LWA, 1300 kg/m³ with polymeric LWA and 260 kg/m³

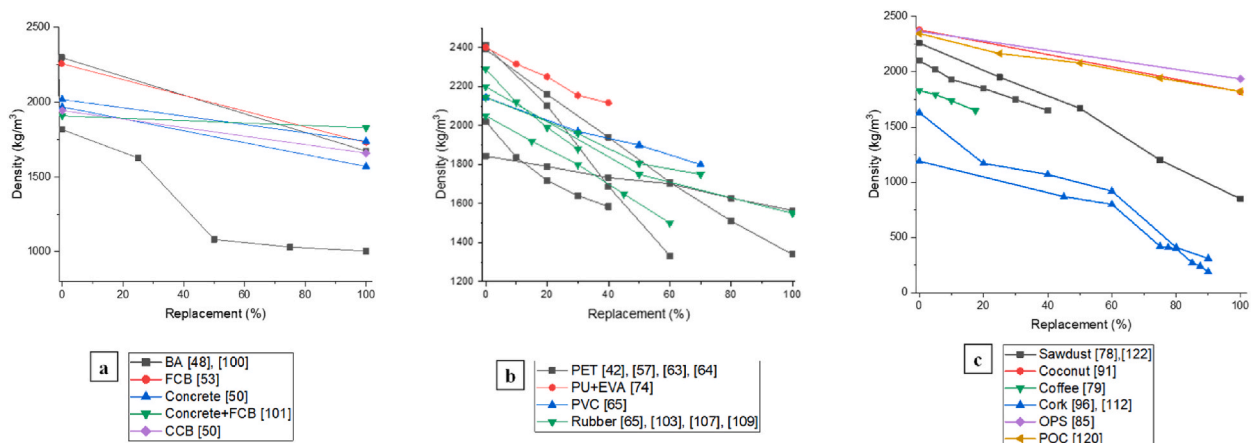


Fig. 5. Density of composites replacing conventional aggregate volume with a. ceramic LWA; b. polymeric LWA; c. lignocellulosic LWA.

with lignocellulosic LWA. The influence of the matrix density (1500–2400 kg/m³) on the density of the composites, with equal incorporation of residual LWA, is also evident.

4.2.2. Mechanical performance

Most authors reported drops in the compressive strength of composites as the proportion of different LWA increased. This behaviour has been associated with the lower mechanical strength of the LWA with respect to the cementitious matrix. Therefore, a careful formulation of the cementitious matrix is needed to achieve the strength required by the specific application where the composite will be used.

Composites incorporating FCB fragments developed 24.9 MPa compressive strength, exceeding the 21.4 MPa achieved for composites made with commercial expanded clay (Leca) [51]. Composites containing FCB also exhibited higher compressive, flexural, and tensile strengths. Further, they demonstrated better performance after being exposed for the same amount of time at 800 °C. Such differences may be associated with the surface morphology and porosity of these two LWA. Replacing 100% of the stone aggregate with FCB caused the compressive strength to decrease by 50% [53]. Likewise, it was found that the strength dropped 63% when the LWA/cement ratio went from 2.0 to 2.6 [49]. With the substitution of 15% vol of stone aggregate by FCB, the density of the composites was 18.3 MPa [53]. Composites made with LWA/cement ratio of 1.6 achieved 17.5 MPa compressive strength [98]. In this case, fine particles of cellular concrete (CCB) were used to plug the pores of the matrix, increasing the density and the elastic modulus of the composite [49]. Composites made with limestone + river sand developed compressive strengths of 34.1–38.2 MPa. Substitution of both aggregates with bottom ash (BA) of equal grain size produced composites of 14.3–18.1 MPa compressive strength. In addition, the tensile strength went from 2.2–2.7 MPa to 1.2–2.0 MPa when the aggregates were substituted with BA. This behaviour was associated with the low density of BA aggregates that results from their high porosity and abrasion losses [48]. The compressive strength of the composites decreased with the progressive increase in WFC fragments. The incorporation of 10% vol of WFC produced the highest compressive strength (38.3 MPa), flexural strength (6.1 MPa) and splitting tensile strength (2.0 MPa). This increase was related to an increase in the CSH gel caused by the geopolymerization between the SiO₂ of the cementing agent and the CaO contained in the WFC [47]. However, exceptions to this trend have been found, where the total replacement of sand with a mixture of concrete and FCB has resulted in an improvement in compressive strength of up to 80%, especially when using a mixed fraction of particles in the range of 0.5–2 mm, resulting in a better packing density, which favours the development of strength [97]. This is the reason for the improvement of the compression strength of the composites containing CDW.

Composites formed by adding polymeric LWA to the cementing agents showed lower mechanical resistance than the composites lightened with ceramic LWA. This was evidenced in composites in which an aggregate of blast furnace slag (GFBS) was replaced by PET flakes [57]. The composite containing GFBS developed a compressive strength of 70 MPa; while the composites that incorporated 60 and 80% vol of PET reached 17.2 and 13 MPa. The decrease in resistance was due to the low adhesion of the cementing agent to the PET, which is hydrophobic and has a smooth texture, in contrast to GFBS which is porous and rough. Other composites developed 14 and 13.7 MPa of compressive strength for the same substitutions (60 and 80% vol) of aggregates by PET [63]. The best results were recorded for 40% PET, in terms of workability and resistance to compression (14.1 MPa), splitting tensile (1.3 MPa) and flexion (2.9 MPa). For these composites, a linear dependence was found between the compressive and tensile strengths described by a correlation coefficient of 0.91 [57]. On the other hand, it was deduced that the laminar morphology of PET decreased the cracking of the matrices, providing them with a ductile failure mode. With the replacement of 50% wt. of stone aggregate by PP pellets [66], composites with compressive strength (27.4 MPa), splitting tensile strength (2.30 MPa), and flexural strength (3.25 MPa) were formed, classified as lightweight concrete according to ASTM C330/C330M – 14 [24–26].

The addition of foamed polymers produced greater drops in the mechanical properties of the composites than those obtained with dense polymers. The substitution of 65–72.5% vol of sand by EPS particles produced composites with compressive strength of 6 and 2.4 MPa and flexural strength up to 0.6 MPa [73]. Composites with between 4.0 and 11.2 MPa were also formed, by substituting 0.85–1.05 % wt. of river sand for EPS [60]. Adding between 9 and 20% wt. of polyurethane foam (PUR) to the cementing agent produced composites with compressive strength of 7–11 MPa and flexural strength of 1.5–3.6 MPa [74]. Much lower compressive strengths (1.42 and 3.07 MPa) were reported for composites in which between 10 and 40% vol of sand was replaced with polyurethane (PU) and ethylene-vinyl acetate (EVA) residues [61].

FA and FA + GFBS matrices experience decreases in their compressive strength of up to 92% when the stone aggregate is replaced by crumb rubber [58,71,104]. This behaviour was related to a weak adhesion in the ITZ, resulting from its hydrophobic nature, and the lower elastic modulus of rubber compared to the stone aggregate [58]. Despite this, the rubber aggregate prevented cracking during curing due to volumetric shrinkage of the cementitious matrices [104]. In addition, the impact toughness of the rubber-lightened composite increased [70].

Composites with total replacement of aggregates by crushed OPS developed 33 MPa of resistance after 28 days of environmental curing [85]. However, the drop in resistance due to the increase in the amount of this LWA was corroborated [76]. The pre-treatment of the OPS particles with alkaline solutions or PVA increased the resistance of the compounds by up to 32.69 MPa [112]. Another study reported a slight increase (32.94 MPa) in the strength of the composites with the addition of OPS particles saturated in water for 48 h [72]. Composites that incorporated fine POC particles developed up to around 53 MPa with 100% replacement of aggregates [77] (Table 3). After 90 days of curing, a 49.3% decrease in resistance was evidenced due to the substitution of stone aggregates with POC [35]. The tensile strength was 75% of the flexural strength and the measured elastic modulus values were comparable to those of Portland cement lightweight OPS concrete. The strength drop was associated with the formation of a porous ITZ between these LWAs and the cementitious material, caused by low workability and poor compaction in the fresh state. However, it was found that the physical anchorage in the ITZ was strengthened as the cement filled the pores of the POC [86]. It was found that the gradual release of

activator solution and water stored in the pores of the POC promoted the formation of strong gels during curing.

Similar compressive strengths were achieved in composites lightened with fine sawdust (38.4 MPa), shavings (30.0 MPa) and coarse sawdust (20.2 MPa) [94]. The finer the wood LWA, the more uniform its dispersion within the matrix, which increased the resistance of the composites. The decrease in strength was attributed to the lower properties of the wood LWA compared to the cementitious matrix [44,79]. The resistances also decreased due to the weak adhesion of the ITZ of the wood particles with the matrix [44]. This occurred because the clustering of the sawdust particles prevented their complete coverage with the cementing agent. The elongated morphology of the wood chips restricted the propagation of cracks in the tension zone due to bending loads [44,94]. In particular, the composites containing up to 6.3% wt. of chips reached flexural strength of 10.7 MPa, which contrasts with the 5.1 MPa presented by the matrix.

The composites lightened with coconut shell developed resistances between 22.3 MPa [91] and 40.4 MPa [90], which equates to those reported in composites made with OPS and POC. This is because both LWAs are tough, dense fibrous rinds, in contrast to wood sawdust that has porous and less dense capillary structures. Lignocellulosic LWAs such as cork are low-density cellular structures whose contribution to the strength of composites is minimal. This was evidenced in composites that reached resistances of 15.3–22.2 MPa with the addition of 2.5–3.75% wt. of cork [120]. Cork additions greater than 90% vol decreased the resistance of the composites up to 0.04 MPa [108] and 0.23 MPa [23].

The mechanical behaviour of the composites described above can be improved according to the recommendations of the authors. Regardless of their nature, the smaller the size of the residual LWA, the greater the compressive strength and elastic modulus of the composites [49,94]. Higher roughness and porosity of LWAs strengthens the ITZ and increases the mechanical properties of composites [53]. Pre-treatment with paraffin [45] or alkaline solutions and PVA [112] slightly increases the resistance of the composites. Furthermore, the combined addition of LWA and fibres improves the flexural performance of composites [77,110].

Fig. 6 shows the change in composite compressive strength as a function of the amount of waste LWA that replaced the stone aggregate. The data shown in these graphs originates from the various research articles referenced.

With one exception [97], the graphs confirm that strength tends to decrease with increased LWA content. The decrease in strength appears to be more pronounced for polymeric aggregates (Fig. 6b) such as rubber and PET, for the previously stated reasons. This indicates the importance of pre-treating these aggregates to improve their surface roughness. Another aspect to consider is the influence of the matrix composition on the mechanical properties of the composite. This can result in different matrix formulations containing a similar aggregate exhibiting vastly different compressive strength value. This is observed in ceramic aggregates such as bottom ash, FCB, PET (Fig. 6a) and sawdust incorporated into FA-based matrices. It is observed (Fig. 6c) that LWA of lignocellulose nature present higher compressive strength than other aggregates, reaching up to more than 50 MPa with a 100% replacement of conventional aggregate.

4.2.3. Water absorption

Along with mechanical strength, water absorption is another key property in determining whether concrete is classified as structural or not. As in any type of concrete, water absorption in composites increases with the amount and size of the incorporated LWA particles [86]. The decrease in water absorption was most noticeable in composites incorporating LWA whose pores of less than 1.18 mm were plugged by the cementitious binder [49]. At early ages, water absorption was higher but was reduced until matrix curing was complete [119].

In composites lightened with ceramic LWA water absorption increased as follows, depending on the added aggregate: FCB (4.79%) [51], WFC (8.9–12%) [47] and CCB (9.5–32.5%) [49]. In composites lightened with thermoplastic polymeric LWA, absorption increased in the following order: PP (7.8–11.7%) [66] and PET (6.73–20%) [57,63]. For rubber-lightened composites, water absorption ranged between 4.7 and 13% [58,71]. In composites that incorporated PUR, the absorption varied between 10.3 and 12.7%

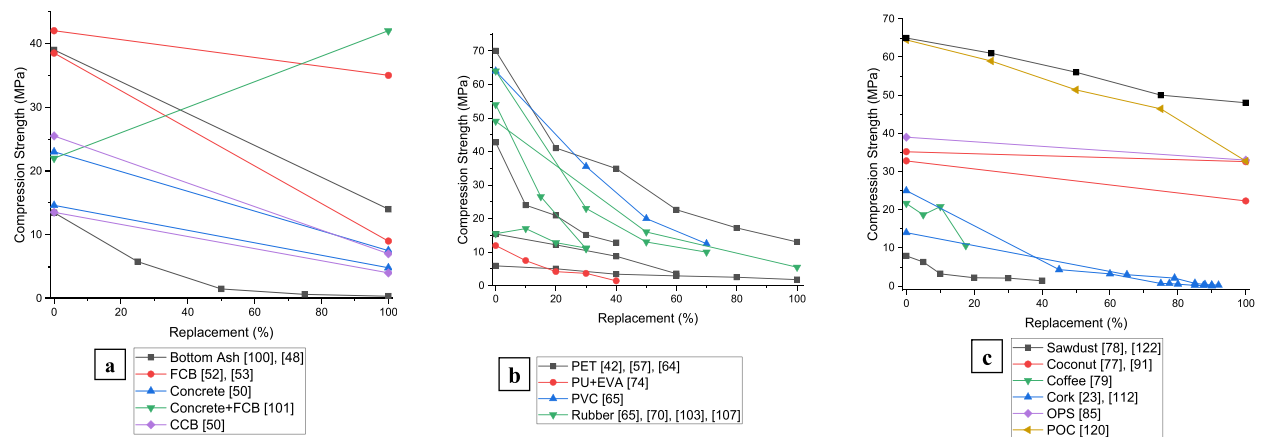


Fig. 6. Compressive strength of composites replacing conventional aggregate volume with a. ceramic LWA; b. polymeric LWA; c. lignocellulosic LWA.

[74]; while with the addition of PU + EVA it changed from 7.8 to 30.5% [61,75,104]. Water absorption increased by more than 14% in composites with incremental substitutions of stone aggregate with PU, PS and PET particles [57,66,75]. This was related to the porous ITZ formed because of the weak bond between the cementitious paste and the hydrophobic LWA. Volumetric shrinkage during curing may induce the growth of voids in the ITZ, thereby increasing absorption. Composites incorporating rubber particles absorbed between 5 and 11 times more water than conventional mortar [104]. Replacements of up to 30% wt. of stone aggregate with rubber increased the absorption between 2.7 and 4.7% [71]. The greater absorption in this type of composites was explained by the increased porosity caused by air nucleation on the surface of the rubber particles [58]. Both the roughness and the superficial micropores in the rubber promote the nucleation of air bubbles trapped in the cementing paste [57,66,75].

It seems that the capillary structure of lignocellulosic LWAs increases water absorption in composites that incorporate them. Water absorption in composites lightened with POC particles was 4.3 times higher than in composites containing granite (8%) or stone (7.5%) aggregates [35]. Water absorption of 10.8% was found for composites lightened with OPS, but when 50% of the OPS was replaced with POC the water uptake was reduced to 10% [86]. The lowest water absorption in composites lightened with lignocellulosic LWA was recorded when POC was used as an additive (0.8–7.76%) [86,119]. The incorporation of fine POC particles increased water absorption (1.8–10.8%) [35,77,86,118]. OPS-lightened composites absorbed up to 10.8% water, while with additions of POC + OPS the absorption was reduced (9.4–9.6%) [86]. In composites lightened with wood LWA, the absorption ranged between 10.1 and 25.2% [44,79]. High water absorption limits many applications of these composites in the construction industry. However, it was identified that cork particles can gradually release the water or activator solution retained in their pores, increasing the formation of resistant gels in the matrices at early ages of curing [108].

From these results it can be deduced that water absorption in the composites was increased by the porosity of the LWA rather than by its nature. However, both the amount and size of the LWA and the porosity of the matrix influenced the water absorption.

4.2.4. Thermal performance

The ASTM C332-09 standard establishes a limit of 0.43 W/m-K for the coefficient of thermal conductivity for insulating lightweight concrete [104]. The thermal conductivity of the lightened composites decreased with increasing amount and porosity of the residual LWA [79,104]. This was observed regardless of the nature of the LWA that each composite incorporated.

In the composites lightened with CDW, the thermal conductivity was lower (0.62–0.65 W/m-K) than in those of the same matrix made with stone aggregate (1.41–1.58 W/m-K) [53]. The impregnation of aerated concrete block (CCB) particles with paraffin significantly improved the thermal insulation of the composites [45]. The replacement of 40% vol of sand with WFC reduced the conductivity of the composites by 43.3% (0.702 W/m-K) [47]. The conductivity of the composites that incorporated bottom ash varied between 0.43 and 0.7 W/m-K [48]; but when rubber was incorporated it decreased to 0.237 W/m-K [104]. Both studies reported that the composition of the precursors and the alkaline solution used to make the matrix did not significantly modify the thermal conductivity of the composites.

The replacement of stone aggregates with PET flakes decreased the conductivity of the composites by up to 58.7% (0.38 W/m-k) [63]. When river sand was replaced by PP pellets, the conductivity of the composite was reduced by 76% [66]. The decrease in conductivity was greater in composites lightened with rigid polyurethane (PUR) and polyisocyanurate (PIR) foams, for which values of 0.30 and 0.36 W/m-K, respectively, were recorded [74].

The replacement of stone aggregates with PET flakes decreased the conductivity of the composites (0.38 W/m-k) by up to 58.7% [63]. When river sand was replaced by PP pellets, the conductivity of the composite was reduced by 76% [66]. The decrease in conductivity was greater in composites lightened with rigid polyurethane (PUR) and polyisocyanurate (PIR) foams, for which values of 0.30 and 0.36 W/m-K, respectively, were recorded [74]. With additions of 0.85–1.05% wt. of PSF, the conductivity oscillated between 0.27 and 0.35 W/m-K, reaching a reduction of up to 65% compared to the matrix [60]. In composites that incorporated EPS, the conductivity varied between 0.12 and 0.21 W/m-K [73].

The thermal conductivity of composites lightened with coconut shell (0.56 W/m-k) was 60.4% lower than that of the matrix that contained stone aggregates (1.49 W/m-k) [91]. The conductivity of composites lightened with wood sawdust was reduced by 77% and 84% with the partial and total replacement of the stone aggregate [44,79]. This reduction in conductivity was related to the high porosity of the wood LWA. In composites with additions of 65–75% vol. cork, the measured conductivity was 0.156 and 0.068 W/m-K, respectively [23,108]. Depending on the amount of virgin cork added, the thermal conductivity of the composite can be reduced up to a quarter of the original conductivity of the cementitious matrix [108].

4.2.5. Acoustic performance

Studies exploring the acoustic performance of alkali-activated and geopolymeric matrices confirmed the increase in acoustic absorption with increasing amount of residual LWA. These results correspond to composites made with lignocellulosic LWA whose weak mechanical performance classifies them as non-structural concrete.

In composites where stone aggregates are completely replaced with wood sawdust, the acoustic absorption coefficient (α) varied from 0.43 to 0.74; showing greater soundproofing in the range from 500 to 3000 Hz [79]. Composites that incorporated less than 60 vol% cork showed weak acoustic attenuation between 400 and 3150 Hz [108]. With 75% vol of cork, the absorption peak ($\alpha = 0.2$ –0.6) was recorded for the range of 1000 and 2000 Hz. In the composites with replacement of 40% vol of stone aggregate by WFC, the α coefficient varied between 0.055 and 0.103 [47].

All these studies associated the low values of α coefficient with the attenuation of the acoustic waves between the pores of the residual LWA and the interfacial transition zone (ITZ).

5. Discussion

The composites analyzed in this review can be classified as lightweight concrete for structural and non-structural use according to their properties and performance. The literature reviewed consistently included evaluation of the density and compressive strength of the composites studied. This trend reflects the importance of these two properties in defining a lightweight material and its suitability for structural or non-structural applications. The evaluation of compressive strength is, in many cases accompanied by that of other properties such as flexural and tensile strength, which are important for specific applications such as panels. For thermal insulating materials, thermal conductivity is extensively investigated. For sound insulation properties, a variety of measurements are considered. The closest thing to a catch-all property in this respect is the sound absorption coefficient (α), but this is not consistently used in all sound insulation measurements. Water absorption values are inconsistently reported across publications, especially considering the importance of this physical property for the durability of composites. However, it is the presentation of aggregate properties that shows the most inconsistencies. Several publications fail to differentiate between important characteristics such as the range and distribution of particle sizes, which are sometimes considered critical to the mechanical strength of the composite. Density and water absorption of the aggregates are also under-reported, and it is recommended that future research on this topic pays more attention to these properties. Inconsistencies are also found in the description of waste LWA additions, with some articles not specifying whether the addition is a percentage by volume or by weight.

Fig. 7 shows a summary of the applications for the different composites studied.

The “Insulation” bar (Fig. 7) groups all papers that studied the thermal conductivity or sound absorption of composites. These are also included in the papers that report composites for non-structural use. Geopolymeric and alkali-activated composites lightened with residual LWA have potential for weight reduction, energy savings, increased thermal comfort and soundproofing in buildings. Many of the analyzed publications suggest that geopolymeric or alkali-activated composites incorporating residual LWA have potential use in the production of precast elements for the construction industry [21,22,35,47,53,76,77,72,85,86,110,112,114,118]. It is feasible to form non-structural elements from alkali-activated cements containing rubber and coconut shell [89]. The replacement of stone aggregate with tire rubber in geopolymers enables the manufacture of lightweight and sustainable non-structural masonry [104]. PET [57] and PSF [60] are other residual LWAs that allow the manufacture of composites for non-structural use. In lightweight composites with recycled PP beads, the amount incorporated determined the use as of lightweight concrete as structural and non-structural, in accordance with the requirements of ASTM C330/C330M – 14 [66].

It was recommended to manufacture low-weight non-structural partitions and panels using MK and cork composites, whose thermal inertia would reduce energy consumption in buildings [23]. Likewise, FA and MK composites lightened with cork regulate humidity inside buildings, reducing energy consumption and increasing comfort [108]. FA + MK composites lightened with PU-EVA are applicable as sealants and finishing mortars [75].

According to their mechanical properties and densities, some of the reported composites were classified as lightweight structural concrete. The number of recommendations for residual LWA to elaborate geopolymeric or alkali-activated structural concretes were in the following order: OPS [22,76,77,82,72,85,110,112] and POC [35,77,86,118]. A smaller number of studies on lightweight composites with coconut shell [90,91], FCB [52,53], PET [57], rubber [58] and WFC [47] were found.

The reduction they achieve of up to 15% of the unit weight enables rubber-lightened MK composites to be considered as lightweight concrete according to EN 1992-1-1. The reduction of the weight in buildings contributes to reducing dead loads, the size of structural elements and the total cost of construction [58]. Structural and non-structural elements such as precast concrete blocks, slabs, nailed concrete and stone backing can be manufactured with this composite. On the other hand, the elastic deformation of rubber increases the absorption and dissipation of desirable energy on roads and runways and take-off at airports [83].

Depending on the amount of wood sawdust incorporated, slag + FA [44] and slag + phosphogypsum [79] composites can be used to manufacture structural and non-structural elements. These elements can include floor formwork, suspended ceilings, screeds, and interior masonry blocks combining flexural strength and thermal insulation.

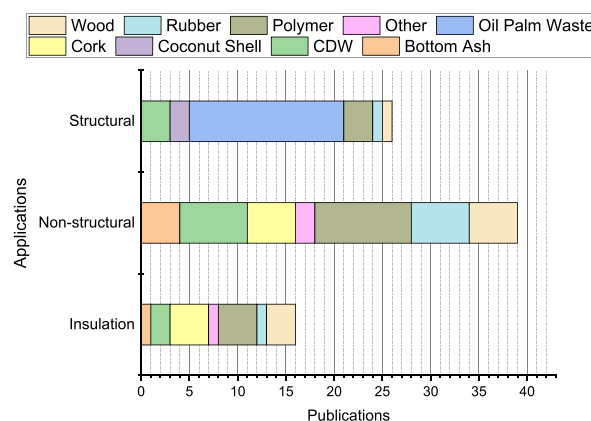


Fig. 7. Applications of composite materials.

Lightweight composites with paraffin-CCB [45] or EPS [73] were considered appropriate for manufacturing structural panels with thermal insulation. Also, it is considered feasible to manufacture MK insulating panels lightened with residual PU and PIR particles [74]. PET-lightened FA compounds can serve to form elements that support bending and require thermal insulation [63]. For the construction of non-structural walls and partitions [49] and the manufacture of heat insulating elements [45,47,73], compounds lightened with CDW could be used. Meanwhile, composites lightened with pyrolyzed cork could be used to manufacture protection panels against electromagnetic interference, because this LWA dampens the incident waves [120].

Considering the potential of these materials in different instances and the visible macro-trends, research and development opportunities were identified.

First, further studies are needed to identify optimal combinations of waste-based LWA and AAB or GP matrices, in order to obtain mechanical and physical composite properties fitted to specific applications. This research should also explore the influence of different forming techniques, such as compression molding, linear extrusion and 3D printing, on the composite properties. In addition, it is proposed to further investigate the effect of particle size, shape and surface texture of waste-based LWA on the properties of composite materials.

Second, the authors recommend further research on the long-term durability of these alternative cementitious composites. Tests on resistance to chlorides, sulphate and acid attack, as well as the influence of the carbonation process should be included. In this regard, the development of surface treatments or coatings to improve durability of composites could be evaluated.

Third, the environmental impact of implementing these composites in construction should be measured. This would require life cycle analysis assessments to evaluate the environmental performance of the materials and identify opportunities to further reduce their carbon footprint. In addition, research could focus on developing more techniques to recycle or reuse materials at the end of their useful life by implementing circular economy strategies.

Finally, further research into the economic feasibility of using LWA from waste in composite materials is recommended. This would involve conducting cost analyses to assess the economic feasibility of using these materials in large-scale construction projects. This research could also explore the potential for creating new markets for these materials and the economic benefits that could be derived from their use.

In summary, future research should focus on developing composite materials that are sustainable, durable, and cost-effective, and that have the potential to contribute to the circular economy in the construction industry. By addressing the gaps in knowledge identified in this review, materials researchers could contribute to the development of a more sustainable construction industry.

6. Bibliometric analysis

This section presents the results of the bibliometric analysis of keyword correlation and co-authorship networks about composites made from waste-based LWA in AAB or GP matrices.

Firstly, a search focused on finding articles with GP/AAB matrices and waste-based LWA in general was carried out using the search strings in Table 1. Secondly, a series of searches focused on finding publications using GP/AAB matrices and a specific type of waste or lightweight material were carried out. The selected aggregates were defined based on those found in the first search. Afterwards, two filters were applied to the results: one by reading the titles and abstracts of the articles and the other by full text reading. In both cases, the aim was to discard articles that do not focus on composite materials like those mentioned in the aim of the paper, or do not provide enough relevant information about the material. Examples of this are materials using a foaming agent such as aluminium powder or hydrogen peroxide, synthetic aggregates manufactured by alkali activation, or composites with densities higher than 2100 kg/m^3 , which do not comply with the standard definition of lightweight concrete. After applying the filters to the results, 72 articles were selected. Table 2 lists the LWA found, and Table 3 lists the composites that include them, classified by aggregate category. Fig. 4 shows a summary of the waste-based LWA found in literature.

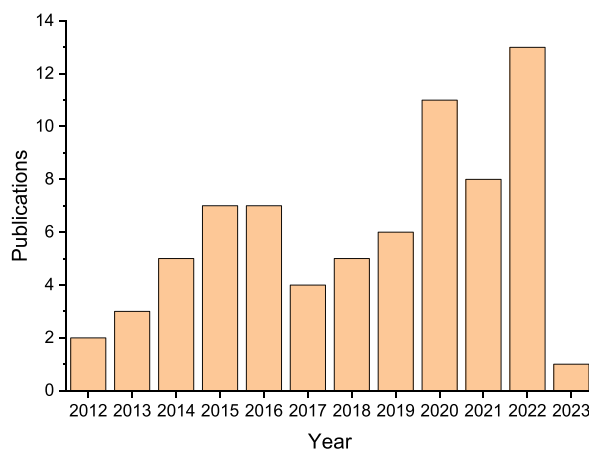


Fig. 8. Evolution of the number of publications on lightweight cementitious composites.

Fig. 8 shows the selected articles according to their year of publication. It is important to highlight that the selected articles span between 2012 and January 2023, the month that the searches took place. It can be observed that the number of relevant articles published rose from 2 to 13 between 2012 and 2022, with a significant rise since 2020. This tendency can be attributed to a growth in interest in the circular economy and reduction in waste disposal in landfills, as well as greater environmental awareness in society

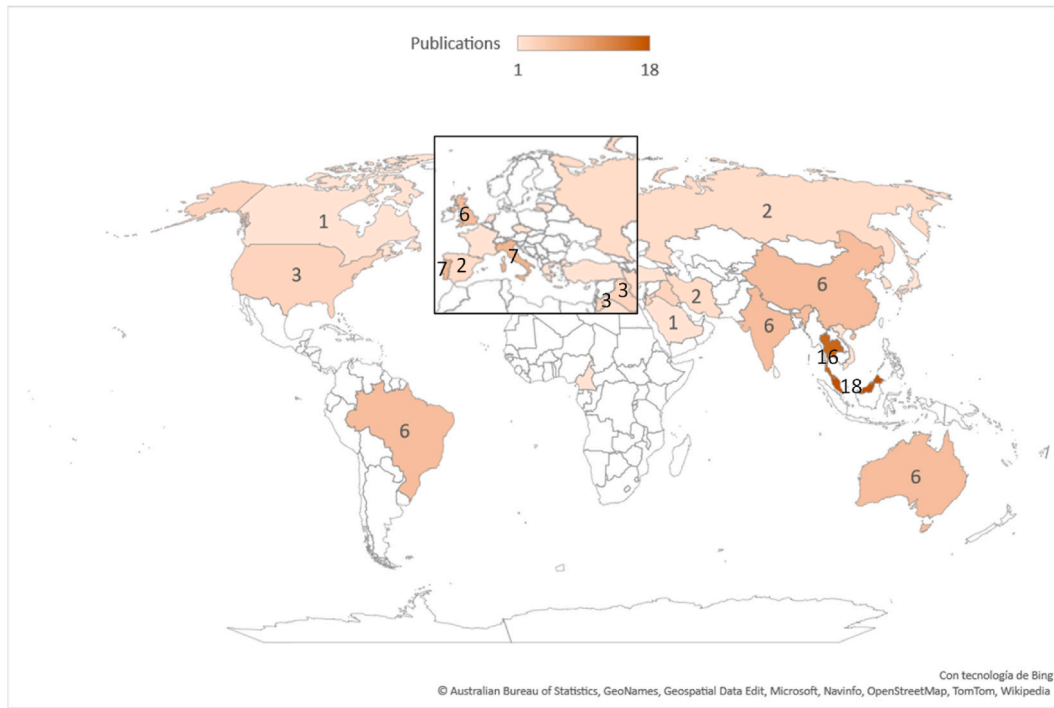


Fig. 9. Global distribution of publications between 2012 and 2023.

Table 4
Distribution of publications per category of aggregate.

Country	Publications			
	Total	Ceramic	Lignocellulosic	Polymer
Malaysia	18	1	16	1
Thailand	16	10	–	6
Italy	7	–	2	5
Portugal	7	1	4	2
Australia	6	1	2	3
Brazil	6	1	3	2
China	6	–	3	3
India	6	–	5	1
United Kingdom	6	1	–	5
Iraq	3	2	–	1
Jordan	3	–	2	1
United States	3	–	–	3
Iran	2	–	1	1
Russian Federation	2	–	–	2
Spain	2	–	2	–
Cameroon	1	–	1	–
Canada	1	–	1	–
Czech Republic	1	–	1	–
France	1	–	1	–
Greece	1	–	–	1
Japan	1	1	–	–
Lithuania	1	–	1	–
Netherlands	1	–	–	1
Saudi Arabia	1	–	1	–
South Korea	1	1	–	–
Turkey	1	–	–	1
Vietnam	1	–	–	1

Table 5
Authors with most publications on alkaline activated matrices with lightweight aggregates.

Author	Papers	Citations	Affiliation	Country	h-index	Total papers	Total citations
Chindapasirt, P.	15	614	Khon Kaen University	Thailand	70	409	18902
Alengaram, U.J.	12	852	University of Malaya	Malaysia	51	164	7161
Jumaat, M.Z.	10	767	University of Malaya	Malaysia	61	280	10619
Sata, V.	10	505	Khon Kaen University	Thailand	36	69	4852
Wongsa, A.	9	374	Khon Kaen University	Thailand	16	25	1265
Bashar, I.I.	6	335	University of Malaya	Malaysia	10	15	645
Labrincha, J.A.	5	96	University of Aveiro	Portugal	58	375	11734
Novais, R.M.	5	96	University of Aveiro	Portugal	30	74	2363
Posi, P.	5	219	Rajamangala University of Technology Isan	Thailand	10	20	473
Zaetang, Y.	4	166	Kasetsart University	Thailand	7	9	469

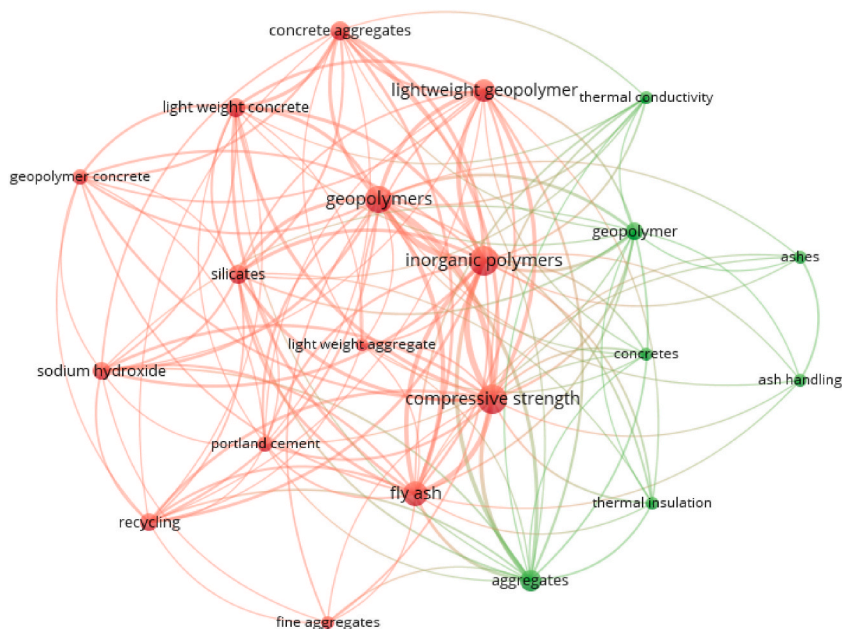


Fig. 10. Keyword network for composites containing ceramic LWA.

generally.

Fig. 9 highlights the countries contributing the highest number of publications between 2012 and 2022. Malaysia (18 publications) and Thailand (16) are the biggest contributors. Table 4 shows a distribution of the numbers of publications in each country classified by the type of aggregate used. It is noteworthy that, in the case of lignocellulosic aggregates, the specific wastes used tended to be from industries where the country is a world leader. The most prominent examples are the use of palm oil waste in Malaysia, cork in Portugal and coconut shell in India. The same tendency is observed regarding the concentration of investigations on these LWA in countries with high production of them. In contrast, there are no such trends regarding studies on polymeric and ceramic-type wastes coming from synthetic materials without a strong geographic identity, such as rubber tires, PET bottles and concrete blocks.

Table 5 identifies the authors with the most publications about composites of interest for this review. Consistently with the trends regarding publications by country, most authors come from Malaysia or Thailand. Prinya Chindraprasirt (Thailand, 15 publications) and U. Johnson Alengaram (Malaysia, 12 publications) stand out as the authors with the highest production in this line of research. Another aspect to consider is that the top 10 authors on the subject are concentrated in 5 universities in 3 countries, which suggests the existence of a cluster of authors in each country responsible for the research on the subject. Regarding the journals publishing this research, those with the most publications are Construction and Building Materials (24), Journal of Cleaner Production (6), Key Engineering Materials (4), Cement and Concrete Composites (3), Energy and Buildings (3) and Journal of Building Engineering (3).

Once selected, the listed publications were exported to the VOSviewer software to build keyword networks around the publications of all three categories listed in Tables 2 and 3. The networks for composites containing ceramic, polymeric and lignocellulosic LWA are shown in Fig. 10, Figs. 11 and 12, respectively. In every network, the results are limited to the keywords that are among the top 30 with the highest link strength, and have at least 4 occurrences, and only links with a minimum strength of 3 are shown. The size of the circle represents the number of articles in which a keyword is used. As expected, the networks share some general terms such as “compressive strength”, “geopolymers” and “inorganic polymers”, which are among the most used and share the most links in all three categories.

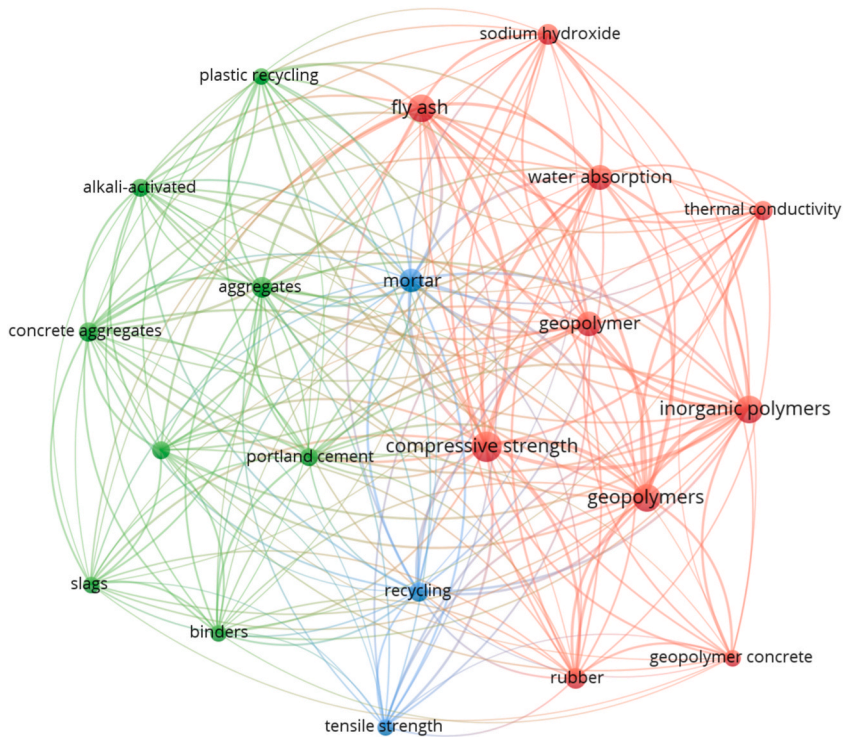


Fig. 11. Keyword network for composites containing polymeric LWA.

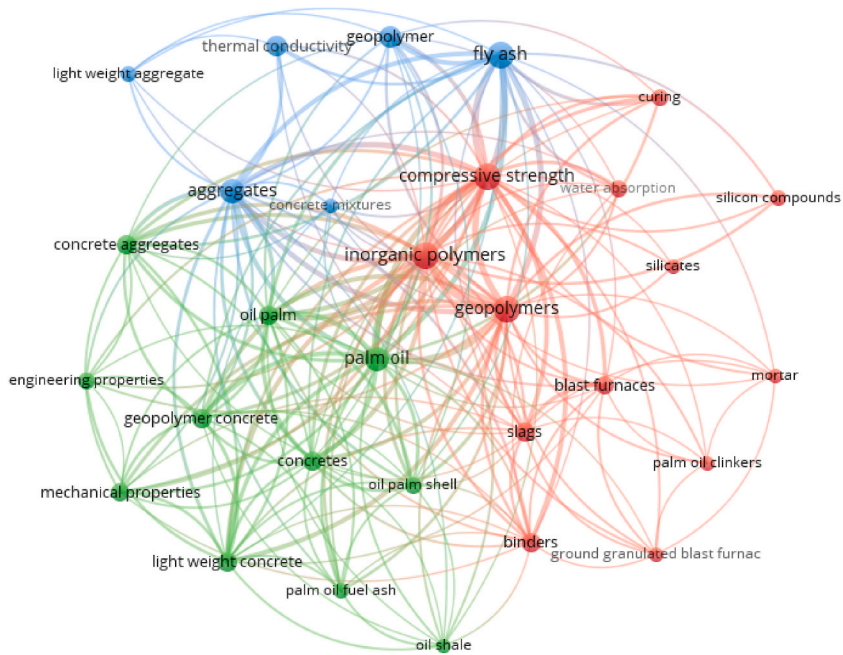


Fig. 12. Keyword network for composites containing lignocellulosic LWA.

This is a clear example of the importance of compressive strength as a property for the materials studied. Other terms, such as “aggregates”, “light weight aggregate” or “light weight concrete” show the emphasis made on the aggregates used in the studies and the lightweight nature of the materials made. Another main term is “fly ash”, which shows that it is the most used aluminosilicate for the binder. Other binder materials shown in lesser volume are “portland cement”, “sodium hydroxide”, “silicates”, “slags”, “palm oil fuel

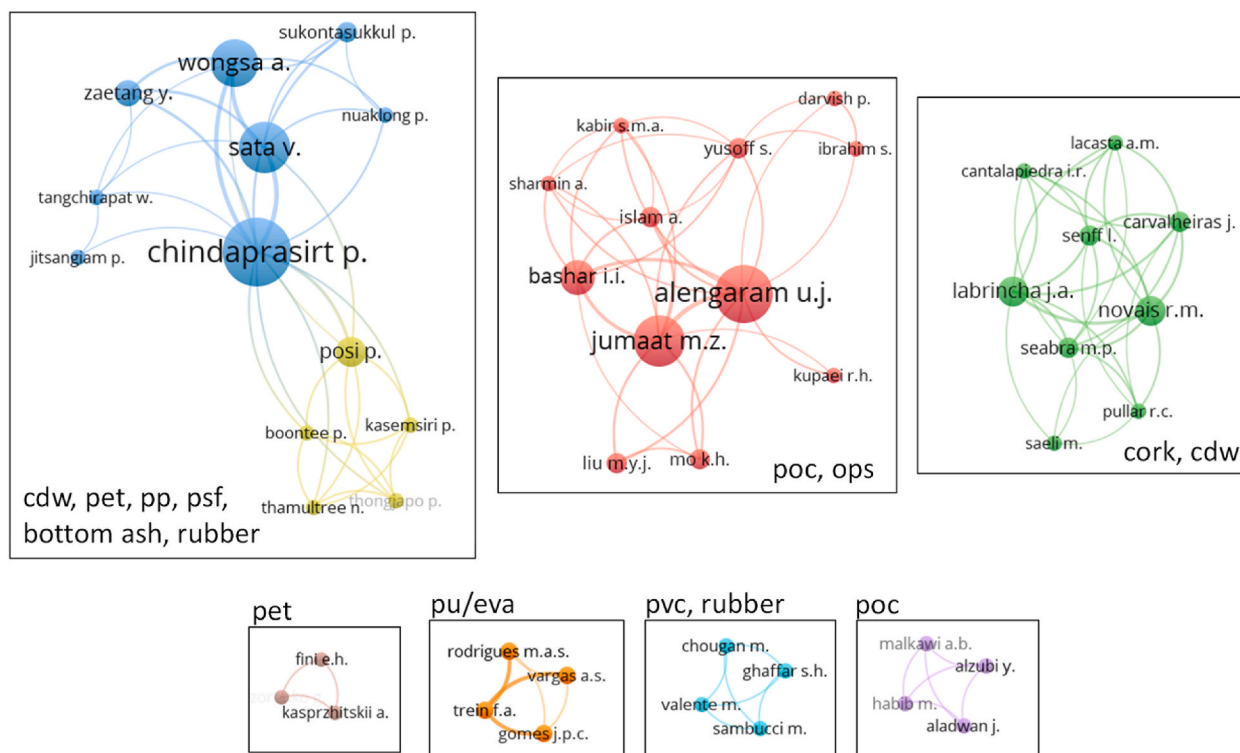


Fig. 13. Co-authorship networks of composites with waste-based LWA.

ash” and “ground granulated blast furnace slag”, which illustrates the range of binders commonly used. In terms of specific waste LWA, Fig. 10 features “concrete aggregates”; Fig. 12 shows the significant proportion of articles featuring palm oil wastes by featuring keywords like “palm oil”, “oil palm shale” or “palm oil clinkers”; while in Fig. 11 keywords such as “polyethylene terephthalates”, “plastic recycling”, “rubber” and “crumb rubber” can be seen. Regarding the properties presented in the articles, “water absorption”, “thermal conductivity”, “mechanical properties” and “tensile strength” can be seen across the maps.

The keyword maps give us an overview of the composition of the materials in the reviewed articles, as well as the properties that were prioritized in their characterization, such as water absorption, compression strength and thermal conductivity. These are all properties that must be considered when in further evaluations of the performance of any materials of this kind.

Fig. 13 shows the co-authorship networks found. The networks only feature authors with two or more publications on the subject, and the lines connecting two authors mean that they have co-authored a publication. With these conditions, seven different networks were found. The three most extensive of these are anchored by the authors in Table 5, and are based in Thailand (Chindraprasirt et al.), Malaysia (Alengaram et al.), and Portugal (Novais et al.). In each of them, a prominent type of LWA is used in their study, specifically: CDW [45,47,53] in Thailand, OPS [21,112,116,117] in Malaysia and cork in Portugal [23,109,120,108]. However, in addition to its main focus on CDW, the Thailand-based cluster has experimented with a wider variety of aggregates, such as rubber [106], bottom ash [48] and different polymers [60,64,66], while the Portugal-based cluster has also delved into using CDW aggregates [97].

7. Conclusions

This paper reviewed the state of the art of different ceramic, polymeric and lignocellulosic waste LWA incorporated in AAB/GP-based composites in research settings in the last decade. The influence of the LWA on the behaviour of the composites was presented, as well as the potential applications and challenges of these materials. These composites are a promising contribution to the sustainable development of the construction industry. The following conclusions can be drawn.

The waste-based LWA incorporated in cementitious matrices were found to be ceramic, polymeric (thermoplastics, elastomers) and lignocellulosic in nature. As precursors of cementitious matrices, fly ash (FA), ground blast furnace slag (GBFS), biomass fly ash and metakaolin (MK) were used. Activation was performed with solutions of sodium silicate (SS), sodium hydroxide (SH) or a mixture of both in different proportions.

It was found that the nature, size, quantity, and morphology of the LWA were determinant in the changes of the physical and mechanical properties of the cementitious composite.

The classification of these compounds for structural or non-structural use depends on the particular combination of these factors. However, it was also found that the properties of the cementitious matrix have an effect on the composite properties. It is well known that the properties of a composite material depend on a synergistic relationship between the specific phases.

Ceramic waste LWAs exhibit porous structures but tend to create composites of lower density and higher strength than other LWAs. Polymeric LWAs typically result in lower strength due to weak adhesion caused by the formation of a porous ITZ. This adverse effect can be mitigated by pre-treatment of these LWA to increase their surface roughness. Lignocellulosic LWAs lead to shorter setting times, due to their hydrophilic nature, as well as a higher water absorption.

Workability of cementitious composites is affected by the quantity, size, and surface condition of LWA particles. Increasing LWA volume reduces workability, due to water absorption by LWA causing increased interparticle friction and decreased flowability. Aggregates with a higher water absorption tend to affect workability by absorbing the activating solution. LWAs with hydrophobic nature hinder workability, due to the increase in porosity in the ITZ, which also reduces strength and increases water absorption in the composite. Various methods, such as immersion in water or superplasticizer use, can compensate for workability loss and enhance mechanical strength.

As expected, the addition of LWA reduces the density of the composites. However, the nature, size and morphology of the aggregates also affect the magnitude of this reduction. The compressive strength of composites generally decreases as the proportion of LWA increases, in a similarly to the density of the composite. Polymeric and lignocellulosic LWAs cause higher strength drops compared to ceramic ones. To enhance mechanical behaviour, approaches like using smaller-sized LWAs, improving roughness and porosity, pre-treating LWAs, and combining them with fibers can be employed. The choice of matrix composition is crucial for obtaining the desired mechanical strength, and it needs sufficient strength to compensate for the loss generated by the aggregate.

High water absorption into the porous LWAs was a limiting factor for the structural applications of the composites. High water absorption is related to both the hydrophilic character and the porosity of the LWA. One measure to reduce this is to impregnate the LWA with sealing agents. However, the porosity in the LWAs can contribute to the retention and gradual release of the water or alkaline solution, promoting the greater formation of gels that densify the cementitious matrices and prevent shrinkage cracking. Some studies showed that LWAs with smaller size tend to plug voids in the matrix, reducing water absorption. In addition, they showed a homogeneous dispersion in the matrix, achieving increases in the density and mechanical strength of the composites.

With the incremental incorporation of LWA, and the increase in porosity they bring, both the thermal insulation and sound absorption of the composites also increased. Both characteristics were determined by the nature, morphology and porosity of the LWA. The alkaline precursors and activators composing the cementitious matrix were not shown to have a significant impact on the thermal and acoustic performance of the composites.

Considering the requirements set forth in ASTM C330/C330M – 14, EN 1992-1-1 and ACI 213R code, mainly concerning density, compressive strength and percentage of water absorption, it was identified that ceramic (FCB, BA, WFC), polymeric (PET, PP, rubber) and lignocellulosic (OPS, POC, coconut shell) can be considered lightweight structural concrete. However, when the proportion of some of these residual LWA was increased, non-structural lightweight concretes were produced. In the manufacture of precast concrete elements (blocks, slabs, tiles), stone supports, roads and runways where energy absorption and dissipation are desirable properties, tire rubber composites demonstrate their viability.

In the case of composites incorporating wood sawdust, structural and non-structural elements can be manufactured depending on the amount of this LWA added. These elements can include floor formwork, suspended ceilings, screeds, and interior masonry blocks, combining flexural strength and thermal insulation. Composites lightened with polymer residuals and lignocellulose LWAs, which have lower densities and mechanical properties, were classified as non-structural lightweight concretes. Recommended applications include non-structural walls and partitions, heat insulating elements, sealants and finishing mortars.

Future research should focus on optimizing the combination of waste-based lightweight aggregates and alkali-activated or geopolymeric matrices to achieve desired properties for specific applications. Additionally, investigating the influence of forming techniques, particle properties, and the long-term durability of these composites is crucial. Evaluating the environmental impact, exploring recycling and reuse strategies, and assessing the economic feasibility of large-scale implementation are also important research directions. Ultimately, the goal is to develop sustainable, durable, and cost-effective composite materials that contribute to the circular economy in construction.

The bibliometric analysis found 72 papers relevant to this review's scope. Only a small group of countries, led by Malaysia and Thailand, has several papers focused on the development of alkali-activated geopolymers and cementitious composites incorporating LWA. The co-authorship networks show that research on the subject in each country tends to be dominated by a cluster of authors, each focused on different types of aggregates. A higher level of cooperation between the author clusters is encouraged, as well as the development of related research in more countries around the world. In the case of Malaysia, a focus on composites lightened with by-products from palm oil production (OPS and POC) is observed. This focus on a single LWA from key industries in each country (seen also in India and Portugal) highlights the need for better methods of waste management, as well as the potential presented by composites such as the ones reviewed contribution to resolving this issue.

Author contribution statement

A. Valencia Isaza: Performed the experiments; Analyzed and interpreted the data; Wrote the paper. J.M. Mejía Arcila: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper. J.W. Restrepo: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper. M.F. Valencia García: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data. L.V. Wilches Peña: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Data availability statement

Data included in article/supp. Material/referenced in article.

Declaration of Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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