Plastic-fly ash waste composites reinforced with carbon nanotubes for sustainable building and construction applications: A review

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ABSTRACT

The building and construction industry is the largest consumer of natural resources and contributes towards high volumes of carbon emission. To achieve sustainability in the industry, waste materials have been applied in construction composites such as bricks and cement/concrete mortar to reduce the dependency on non-renewable resources. This review focuses on the application of waste plastics and coal fly ash in building and construction composites, especially bricks. The properties (e.g., mechanical strength, water resistance and thermal stability) of the waste-derived composites were evaluated to ascertain their applicability in the construction industry. Nanomaterials such as carbon nanotubes (CNTs), which are known to possess high mechanical strength have been identified as suitable filler materials to provide the desired strength to the final products such as bricks. The dispersion of the CNTs in the composite is at a finer scale, compared to conventional fillers. Thus, extremely low contents of CNTs can make a substantial difference in enhancing the properties of the composites. The waste-derived composites enhanced with CNTs are lightweight strong materials that can withstand harsh conditions of weather, fire or earth tremors. Moreover, the environmental concerns on the release of CNTs from the composite are reviewed to ensure their sustainable application.

Introduction

Bricks are a large and important part of the building and construction industry. Bricks have been used as masonry units due to the strength and durability they provide in structural applications [1]. The properties of each brick produced depends on the raw materials used and their preparation methods [2]. The commonly used bricks are those produced from clay, cement concrete and recently fly ash, with clay bricks being the most ancient and widely used [3]. Clay is made up of alumina and silica, with the latter being the promoter of particle fusion at low temperatures and it also determines the colour of the brick produced [4]. A typical brick manufacturing process involves: i) raw material preparation and mixing, ii) brick formation, iii) drying, iv) firing and v) cooling [4]. Clay bricks are either burnt (fired) or unburnt (unfired) during the

production process. Burnt clay bricks obtain their strength through the sintering process, where the crystalline mineralogical phases in clay are fused into the pores to strengthen the microstructure of the brick. Their mechanical and durability properties are also dependent on the type of clay, water ratio, burning and curing processes [5]. Unburnt clay bricks are normally incorporated with lime stabilizers to foster the pozzolanic reaction, which ensures effective particle bonding and thus aids the stiffness and mechanical strength in the bricks [6]. Furthermore, the size and geometry of the bricks have an effect on the mechanical performance of wall panels [7].

Concrete bricks are another type of conventional brick made of Ordinary Portland Cement (OPC), water, aggregates and additives [8]. Aggregates in concrete bricks comprise of 65–70% of the total volume of a brick [9]. Thus, the construction industry is considered to be among

the largest consumer of natural resources. The process of cement production is an energy intensive one using about 4 GJ energy per ton of cement produced [10]. Furthermore, 622 kg of CO_2 per ton of cement is produced as well as other hazardous greenhouse gases during fuel combustion and limestone-calcination [10]. Similarly, clay bricks use great amounts of natural resources which causes ecological degradation due to excessive movement/removal of the brick constituent from the environment. The firing process to make burnt clay is normally performed at temperatures between 1000 and 1200 °C and can take up to 5 days [11]. This process is almost 300% more energy demanding as compared to the manufacturing of concrete bricks [10].

It is evident that the conventional brick production methods are not environmentally benign as they contribute a great deal to carbon emissions and are also not economically sustainable. The building and construction industry is the backbone of the economy for many countries, and as such, alternative ways of producing bricks in the future that are eco-friendly and affordable is crucial. This is also important in addressing the UNESCO sustainable development goals (SDGs), such as SDG 13 on climate action; which has an agenda of reaching net zero carbon by 2030 [12]. Furthermore, sustainable building materials speaks to SDG 9 and 11 relating to industry innovation and infrastructure and sustainable cities and communities respectively [12]. The application of sustainable building materials has a positive impact on both the quality of people's lives and the environment.

The incorporation of waste into bricks as sustainable construction materials is among the studies that have gained considerable attention. This is due to rapid industrialization that results in the production of large volumes of waste which causes disposal and handling problems. Moreover, significant volumes of construction materials are necessary for the creation of infrastructure, such as used in buildings for housing and industries as well as facilities for water and sewage storage [13]. Thus, sustainable approaches are required to meet this ever-growing demand. Zero-cement products known as alkali-activated bricks have been developed as sustainable construction materials [14]. These products utilize waste such as rice husk, fly ash, bottom ash, cement kiln dust, blast furnace slag and mine tailings [15-17]. These bricks are produced by alkali activation (sodium hydroxide (NaOH) or potassium hydroxide (KOH)) of the aluminosilicate source material. Alkali-activated bricks are developed through a process of geopolymerization. The process relies on the particle size, loss on ignition and the alumina-silica content of the source materials [18]. The process results in creation of high strength eco-friendly products with a compressive strength between 5–60 MPa and a density of 1400–1500 kg/m^3 [19]. For example, Gavali et al., produced alkali activated bricks using co-fired blended ash and stone with a compressive strength of 5–15 MPa, water absorption of 6–14% and thermal conductivity of 0.35–0.40 W/(m.K) [17]. Similarly, in another study Gavali et al., used the same industrial waste to make sustainable bricks. The alkali activated bricks resulted in bricks with a reduced cost (7%), embodied energy (22%) and lower operation energy (13%) compared to bricks used in conventional buildings [19].

Reusing construction concrete waste materials (i.e. ceramics, bricks, blocks, pillars) as pozzolanic admixtures in the production of new products is another sustainable approach to minimize the depletion of natural aggregates, and to reduce energy usage and environmental pollution. These waste materials mainly come from demolitions, renovations and products with defects. They effectively substitute or even replace cement and aggregates in construction materials [20]. The waste is crushed and used as either small or large aggregates. The replacement ratio is normally between 1 and 50 wt% of cement or aggregates [20]. Gencel et al., replaced clay with up to 15 wt% concrete waste and obtained a compressive strength of about 7 MPa, which met the required building standard [21]. There are other waste materials that have been incorporated into bricks to reduce the reliance on non-renewable resources. These include paper processing residues, cigarette butts, textile effluent treatment plant (ETP) sludge, mill scale, marble slurry, polystyrene foam, straw, polystyrene fabric, cotton waste, dried sludge, rubber, wood sawdust and waste plastic [22,23].

When hybrid composites, consisting of reinforcement and matrix materials are made, the former impart their distinct mechanical and physical properties to the brick to enhance the matrix properties [24]. The incorporation of waste materials in construction composites can compromise some of the brick properties. For example, waste plastic is known to have a high linear burning rate which renders it flammable [25]. Its incorporation in construction materials compromises the thermal stability of the construction material, which is a set standard requirement.

This review seeks to evaluate the application of waste materials, in particular, coal fly ash and plastics, their typical replacement ratios and their physicochemical properties, in construction composites (bricks). The influence of nanomaterials, especially CNTs, as a reinforcement filler in the brick composite made of waste, is also described. As will be seen, the success of hybrid composites is based on strong linkages between the individual materials to ensure that the nanomaterials do not agglomerate in the composite and facilitate load transfer. Further, a mechanism is proposed to explain the interaction of waste materials and CNTs within the composite. Lastly, the potential release of CNTs from brick composites to the environment is reviewed to comment on the safe application and end of life cycle of these composite types.

Application of waste materials in construction composites

The generation of high volumes of waste is both an environmental and economic challenge. It has become unsustainable to manage this waste. Thus, recycling of waste into valuable products has become part of the sustainable development goals for many countries. Plastic waste and coal fly ash are among the largest contributors of solid waste generated by the plastics (polymers) and mining industries respectively [26]. The rate at which these waste materials are generated requires large industrial applications, such as in the building and construction industries, to utilize them. The application of waste plastics and coal fly ash in construction composites is reviewed below.

Waste plastics

Plastics are among the largest contributors of the total global solid waste generated, with low-density polyethylene (LDPE) being the most produced [27] (Fig. 1). LDPE plastics mainly include household items
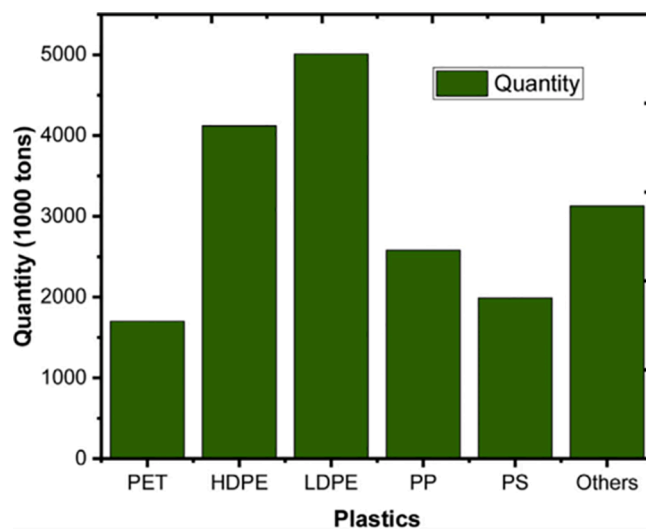

Fig. 1. Estimated quantities of waste plastic generated per annum (PET = polyethylene terephthalate, HDPE = high density polyethylene, LDPE = low density polyethylene, PP = polypropylene, PS = polystyrene) [33].

such as plastic bags, squeezable plastic bottles, frozen food containers, and cosmetic and detergent bottles [23]. These items are generally used once and discarded, typically in disposal sites. They also later find their way into the ground water, riverbanks and ultimately the sea. South Africa ranks as the 11th country (out of 192 countries) with the highest level of mismanaged plastic waste entering the ocean [28]. Plastic waste pollution in the costal environment has an adverse effect on aquatic life and poses a health risk to humans [29]. Thus, recycling and reusing plastic is a sustainable solution for reducing the negative impact the waste has on both the environment and the quality of life. The recyclability of plastics is dependent on their type and properties. Plastics are categorized as either thermosets or thermoplastics according to their characteristic properties [30]. Thermoset plastics have polymers that combine to form irreversible chemical bonds during the curing process. Their strength is reinforced when heated, however, following the heating they cannot be reshaped or reheated [31]. Thermoplastics are the most common type of plastic and are easier to recycle and reuse due to their ability to be molded and remolded. Current thermoplastics soften in the range of 65 °C-200 °C [32]. The prominent characteristics of typical plastics are shown in Table 1.

The application of plastics is vast due to their inherent characteristics such as high impact resistance, their light weight and flexibility to be molded into different shapes. They are also resistant to bacteria [33]. However, they are not biodegradable. Further , there are benefits to incorporating waste plastics in construction materials (Fig. 2). According to Fig. 2, the usability lifespan of plastics is shorter than its non-usability life span and thus the plastic persists for years as unusable waste rather than as a valuable product. Similar to coal fly ash, it is a waste by-product which persists longer as waste than a valuable product due to its limited applications. In contrast, concrete has a longer usable lifespan than its non-usable lifespan [35]. Therefore, the insertion of waste such as plastic and fly ash in concrete materials is a sustainable approach for recycling and safely disposing waste products. Importantly, the limitations of labour intensive sorting, which is a strict requirement in other industries, is exempted in some construction applications [26]. This makes the recycling process easier and less costly. Furthermore, the utilization of waste in construction materials not only reduces pollution but also the number of natural aggregates used, which are responsible for environmental degradation and the depletion of natural resources.

Processing plastics for application in construction composites

Like any composite material, the controlling factor towards the successful application of the composite is the interfacial transition zone between different materials. The substitution of natural aggregates such as sand with plastic reduces the density and compressive strength of a composite at high substitution ratios, resulting in a loss of strength [36]. This may be as a result of the hydrophobic surface of the plastic leading to the retention of excess water, which may lead to poor bonding between the plastic and the matrix. Appropriate mixtures as well as the choice of waste plastic can limit the reduction of strength [8]. Furthermore, reduction in compressive strength can be minimized by improving

the chemical and physical bonding in the concrete mix. Different physical and chemical approaches exist to achieve this in construction mixtures.

Physical approaches, such as shredding plastic to smaller particle sizes is normally the first treatment step. During shredding, the size of the plastic is reduced to a range of 2.36–4.75 mm. This process has been shown to improve blending in concrete mixes and to minimize the loss of compressive strength [37]. Tulashie et al., used shredder blades to obtain 3 mm waste plastics. The shredded plastic, together with the pit sand or sea sand, were placed in an extrusion machine in batches. The batches were melted at 175 °C until there were no longer flakes [37]. In another study, LDPE was ground after being melted at 120 °C into a semi-liquid form using a pestle and mortar. Then a 4.75 mm sieve was used to achieve the desired particle size. Additionally, a rotary grinder was used for shredding plastics to fine aggregates of less than 500 μm [38]. Chen et al., proposed a simple method of processing waste plastic in concrete mixes. The waste plastics were directly placed in a screw extruder and pelletized without any mechanical size reduction. Together with the filler and coupling agents, the pellets were again passed through the extruder machine. The initial size of the plastics was not important as the plastics melted and different segments stuck together to form homogeneous pellets [39]. It is noteworthy that mechanical size reduction, shredding or pelletization makes it easier for the fine plastic particles to mix with solid material before heating. When the plastic composition, on melting, becomes viscous, it becomes a challenge to mix with other aggregates such as concrete, stone and sand [34]. Thus, the mix design is important to ensure plastics mix homogeneously throughout the composite to give a good matrix and effectively improve the mechanical strength.

Chemical compounds such as sodium hydroxide and sodium hypochlorite have been used to treat the surfaces of plastics prior to mixing with construction mixtures [40]. Plastics are expected to mix physically with concrete mixtures rather than form chemical bonds. However, oxidising agents such as sodium hydroxide enables a chemical reaction to occur between the polymer chains and the concrete by modifying the polymer surface functional groups. The polymers can also contain ionic groups (hydroxide, oxide) that will bind to the concrete [40]. Choi et al., treated shredded PET bottles between 5 and 15 mm with ground granulated blast furnace slag (GGBS). The aim was to solidify the aggregate with GGBS and foster a reaction which resulted in formation of a pozzolanic material due to the strengthening of the interfacial zone between a cement paste and the aggregate [41].

Properties of construction composites with waste plastics

Various types of plastics such as LDPE, high density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), polycarbonate (PC) have found application in different construction materials (Table 2). The waste plastics are incorporated as aggregates, binders, or molds in construction materials. As aggregates, waste plastics were found to be favorable in reducing the cost of concrete, improving resistance to impact loading and changing the modes of failure of concrete from brittle to ductile [8]. Furthermore, waste plastics can be incorporated in bricks to improve their porosity while firing, to reinforce bricks under ambient temperatures [23]. PET waste plastic was reported to improve resistance to corrosion especially against sulfuric acid and also found to make the concrete lighter [42]. In plastic-related studies waste materials were prepared by cutting or shredding them into narrow strips in a range of a few millimeters [23]. Limami et al., used HDPE and PET as fine aggregates (1–6 mm) in unburnt clay bricks [43]. Xu et al., used expandable polystyrene (3 mm) to manufacture lightweight concrete and bricks [44].

In brick and concrete production, waste plastics partially replace one or more of the conventional materials. The replacement ratios are typically from 0.1 to 15%, although in some studies up to 80% waste plastic has been used as a binder material [45]. For example, Hannawi et al., replaced sand with 3%, 10%, 20% and 50% of PC and PET waste
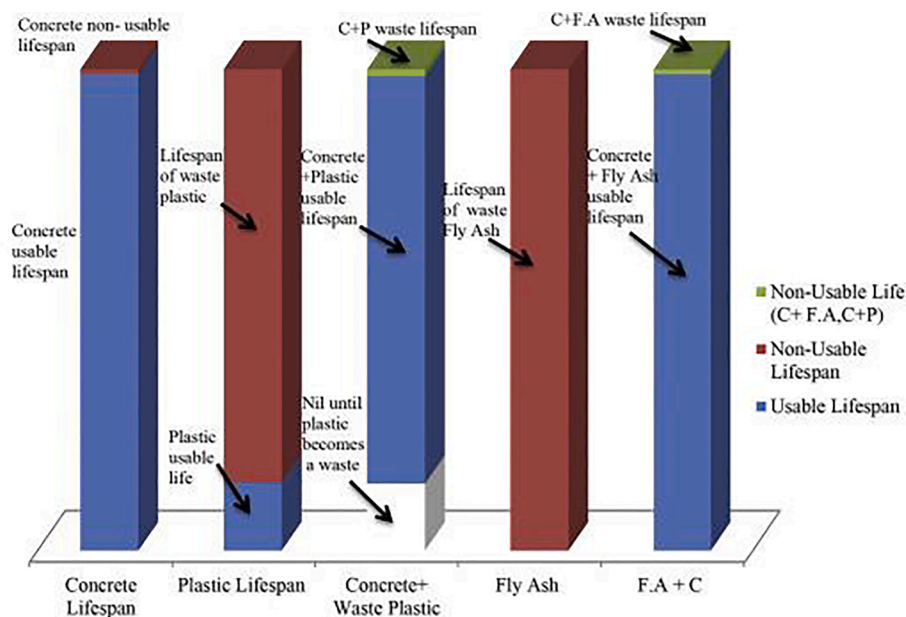
Table 1
Properties of different plastics commonly found as waste in the environment [34].

Properties of plastics	Types of plastics				
	LPDE	HDPE	LDPE	PP	PET
Density (g/cm^3)	0.92	0.94–0.96	0.92	0.905	1.37
Melting point (°C)	105–111	126–135	130	160	225
Max service temperature (°C)	80	80	50	80	70
Tensile strength (MPa)	0.2–0.4	0.2–0.4	0.2–0.4	0.95–1.3	0.25
Notched impact strength (kJ/m^2)	No break	No break	No break	3–30	1.5–3.5

Fig. 2. The benefits of using waste in concrete, C = concrete, FA = fly ash, P = plastic [35].

Table 2
Physical properties of recycled plastics and their possible construction applications [33].

Plastic	Physical properties	Possible construction application
HDPE	Rigid	Plastic lumber, table, chairs
LDPE	Flexible	Bricks, blocks, stepping stones, pillars
PP	Hard and flexible	Aggregate in asphalt mixtures
PS	Hard and brittle	Insulation material
PET	Hard and flexible	Fibres in cementitious materials
PC	Hard and rigid	Aggregates in cementitious materials

plastic in mortar [46]. Natural aggregates were partially replaced with PET in burnt clay bricks [47]. It was observed that concentrations above 15% of PET led to disintegration of the bricks at high temperature. Thus, concentrations less than 15% PET were optimum for fired clay bricks using controlled conditions [47].

The incorporation of plastic waste into construction composites has environmental benefits of reducing pollution and producing new construction materials with distinct properties compared to conventional materials. Belmokaddem et al., studied the mechanical and physical properties of concrete containing plastic waste aggregates. Three types of plastic waste were used (i.e. HDPE, PET and polyvinyl chloride (PVC)) to substitute natural aggregates in different amounts (25, 50 and 75%) [48]. It was noted that the use of plastic waste reduced the density of the composite and the lowest dynamic elastic modulus was recorded for composites containing 75% HDPE, which corresponds to a more ductile composite. The lowered dynamic elastic modulus resulted in more deformable concrete composites which could be used in flexible structures. An improved thermal insulation was detected using composites with 75% PVC and all the composites containing plastic aggregates showed a low heat capacity. This suggests the use of these materials as durable concrete pavements that alleviate the night-time island effects due to high temperature differences between daytime and nighttime. However, a decline in the compressive strength upon an increase in plastic aggregate substitution was noted [48]. It is noteworthy that the concrete and bricks with waste plastics still showed a relatively high compressive strength which is acceptable according to building and construction standards. Mondal et al., observed a decline in the compressive strength of bricks from 34 MPa to 17 MPa upon addition of 0–10 wt% polycarbonates, polystyrenes and mixed plastics [49].

Interestingly, Herki et al., detected no change in the concrete compressive strength up to 30 wt% waste using expanded polystyrene replacement [50]. In another study, 3 wt% polypropylene waste plastic was found to be the maximum for sand replacement in burnt clay bricks [51].

Plastic replacement optimization studies are required to ensure that the properties of the construction composite meets those of the desired application. Table 3 shows the application of waste plastic in bricks using various replacement ratios. In sand bricks, waste plastics are used as binders without the use of cement or pozzolanic aggregates.

Bricks are classified according to their properties, which informs their application. First-class bricks refer to bricks with a compressive strength of about 14 MPa, with a water absorption rate of not more than 20% when immersed in water for 24 h. These bricks are used for flooring and external walls applications [51]. Bricks with compressive strength of not less than 7 MPa and water absorption of not more than 22% are classified as second class. This type is not necessarily preferred for flooring, but is good for walls that do not require plastering. The third class of bricks has a compressive strength that ranges between 3.5 and 7 MPa, with water absorption of not more than 26% and are used in ordinary construction. Class four bricks have low porosity and strength of about 15 MPa. This type is not good for building purposes and is usually applied in road construction [51].

Alaloul et al., investigated the effect of polyethylene terephthalate (PET) and polyurethane (PU) on the mechanical strength and thermal stability of interlocking bricks. A compressive strength of 5.3 MPa was obtained for a PET/PU brick with a 60:40 ratio, suitable for a non-load bearing masonry brick wall such as a partition wall. A low thermal conductivity of 0.153 W/mK was achieved for a PET/PU brick with a 80:20 ratio which signifies a good thermal insulating material [52]. Bharathi et al., obtained a compressive strength of 55.91 MPa which is 88.59% higher than the standard clay bricks, upon employment of 60 wt % waste plastic in sand bricks. The water absorption was also reduced from 19.8% for conventional bricks to 0.452%. The properties of waste plastics thus show their potential application in different types of innovative building and pavement bricks.

Coal fly ash

Coal fly ash (FA) is a cementitious coal combustion by-product

Table 3
Recent applications of waste plastic in bricks.

Polymer	Replacement %	Type of replacement	Other materials	Application	Brick dimensions (mm)	Test performed	Ref
PP	1–5	Shreds	Crushed glass, clay	Aggregate	150 × 100 × 63	Water Absorption, Shrinkage, Mechanical strength	[51]
PET& PU	60:40	Shreds	–	Binder	64 × 13 × 3.2	compressive strength, impact, flexural strength and thermal conductivity	[52]
LDPE	20	Pellets	Sand, cement	Aggregate	40 × 20 × 10	Compressive strength Water absorption Drag strength	[30]
PET	30, 20	Shreds	Sand	Binder	222 × 106 × 73	Durability through acid, Water absorption Compressive Tensile strength	[53]
PP & PE	10–30	Shreds	Sand	Binder	200 × 195 × 100	compressive strength, efflorescence, water absorption, soundness, fire resistance test	[54]
LDPE	33.33–66.67	Shreds	Sand	Binder	200 × 150 × 100	Thermal conductivity Bending stress	[55]
HDPE	100	Shreds	–	–	240 × 120 × 60	Compressive strength Thermal properties Fire resistance test	[56]
LDPE	5–20	Shreds	Sand, cement, coal fly ash	Aggregate	190 × 90 × 90	Compressive strength Water absorption Efflorescence test Soundness test Crushing test	[57]

produced by thermal power plants in large volumes [58]. Fly ash dumps take up space and cause contamination of the environment in the long term. Its disposal by these means is not sustainable because it is costly and soon will be forbidden. Although fly ash is considered a waste product, it has been used in the production of valuable products. This is due to its cementitious, alkaline and pozzolanic properties [59]. The utilization of fly ash as an environmentally benign product has received great attention. This arises since it is freely available, cheap, and would reduce its environmental impact (pollution and wasted land) [59]. In South Africa, fly ash is mainly used in the cement and concrete industry. Other limited applications of fly ash are in soil stabilization, agriculture, low-cost adsorbents and for the synthesis of zeolites and geopolymers [60].

Properties of coal fly ash

The appearance of fly ash varies from grey to black, and its colour gives information about its properties and composition [60]. Fly ash consists of small particles (0.5–200 μm) informed by the combustion process, coal source etc. and is mostly alkaline and refractory in nature [59]. It is spherical in shape and also contains irregularly shaped minerals and unburnt carbon. The hollow cenospheres and irregularly unburned carbon content tends to have larger sizes as shown in Fig. 3. Moreover, fly ash with lower calcium levels is normally coarser than high-calcium ashes. The specific surface area of fly ash is 150 m²/kg for mechanically collected fly ashes and 1200 m²/kg for ones collected by electrostatic precipitators. Its specific gravity is in the range of 1.90–2.96 for subbituminous ash and iron-rich bituminous ash respectively [61].

The main components in fly ash are silicon (Si) and aluminium (Al) with minor amounts of sulphur (S), sodium (Na), iron (Fe), titanium (Ti), phosphorus (P) and calcium (Ca) [63]. Fly ash also contains trace amounts of elements, such as chromium (Cr), barium (Ba), nickel (Ni),
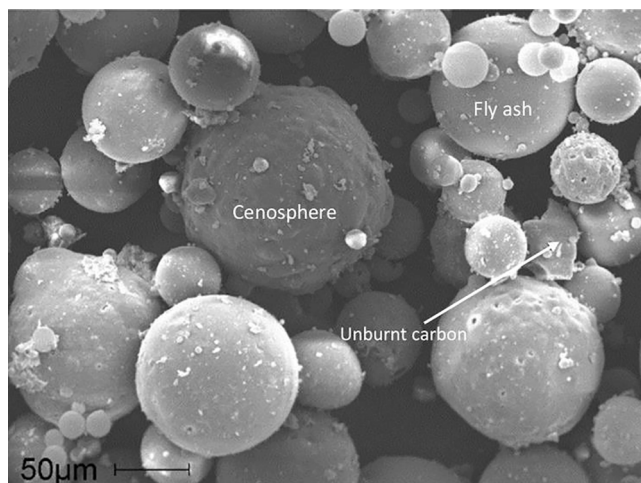

Fig. 3. SEM image of a representative sample of coal fly ash particles in the range of 10–100 μm [62].

palladium (Pb), strontium (Sr), vanadium (V) and zinc (Zn). The major mineral compound in fly ash is amorphous aluminosilicate (glass) and the major crystalline minerals are quartz and mullite. Other crystalline minerals present in fly ash include, but are not limited to, anhydrides, hematite, feldspars, periclase, wollastonite, cristobalite and lime [64]. The composition of fly ash depends on factors such as the type of coal burnt, the process the coal undergoes before combustion and the combustion process [65]. Mullite is produced during coal combustion of decomposed clays, and does not normally contribute in the cementing reaction because of its low reactivity. Similarly, quartz is generally unreactive during the fly ash hydration. Classification in terms of surface

chemistry, mineralogy, composition, and reactivity is significant for fly ash applications [63,66]. Notably, South African fly ash is reported to have high concentration of silica (SiO_2) and alumina (Al_2O_3), making it suitable in the construction industry and as a zeolite precursor.

According to the American Society for Testing Materials (ASTM), fly ash can be classified based on its chemical composition (i.e., SiO_2, Al_2O_3 calcium oxide (CaO) and iron oxide (Fe_2O_3) content) into two categories, namely Class C and Class F (Table 4). Class C fly ash is normally produced from the combustion of lignite or sub-bituminous coal. It contains between 50 and 70 wt% of SiO_2, Al_2O_3 and Fe_2O_3 and the fly ash also has a high lime content. Class F fly ash if formed from the combustion of harder, older anthracite and bituminous coal. It contains more than 70 wt% of SiO_2, Al_2O_3 and Fe_2O_3 and has a low content of lime.

A cement agent like cement, hydrated lime or quicklime is required for Class F coal fly ash to possess cementitious properties. The incorporation of sodium silicate as a chemical activator to Class F coal fly ash may result in geopolymerization. The combustion of younger sub-bituminous coal results in a high calcium fly ash with pozzolanic and self-cementing properties. Furthermore, higher contents of sulphur were also found to be beneficial in enhancing the self-cementitious strength of fly ash [67]. Class C coal fly ash has increased strength over time in the presence of water. More than 20% lime (calcium oxide) is possessed by Class C coal fly ash. Thus, unlike Class F fly ash, an activator is not required for self-cementing Class C coal fly ash [68]. However, coal fly ash with low calcium content has been used in the production of geopolymers or as an admixture in concrete and has shown good performance [61]. High calcium fly ash has been suggested to contribute to premature distress in concrete. The instability in concrete caused by high levels of calcium oxide is a major concern which hinders fly ash applications [61].

Application of coal fly ash in construction composites

In the construction industry, fly ash has been applied as either a raw material or additive in cement products. It is useful in the substitution or replacement of cement in concrete and other building applications due to its pozzolanic properties [69]. The incorporation of fly ash as an aggregate enhances workability and minimizes the bleeding of freshly mixed concrete. Class F fly ash replacement of cement reduces the heat of hydration and the possibility of cracking in concrete in its early stage of formation [70]. Hardened fly ash composites have great strength and low permeability [71]. Furthermore, the addition of fly ash could save production costs.

According to the ASTM the maximum loss on ignition (LOI) of fly ash should be 6% (Table 4) for application in concrete. LOI gives an indication of the residual carbon in the fly ash. Furthermore, LOI values also provides an indication of the presence of carbonates and water in clay minerals. However, the main contributing factor is the carbon combustion [72]. LOI of the carbon content and the fineness of the material are two properties of concern in using fly ash. They are often interdependent because a high carbon content (high LOI) results in a coarser fly ash due to the presence of coarse carbon particles [73]. The residual carbon and coarser particles present in the fly ash can affect its

pozzolanic properties by absorbing water and chemical admixtures (e.g., superplasticizers), thus not reducing their efficiency [74]. Subsequently, the strength development of concrete is negatively affected by low quality fly ash with high LOI and large particle sizes [75]. Typically, fly ash-based composites show quasi-brittle behavior as a result of their ceramic-like properties, which includes low tensile strength and fracture toughness [76].

The replacement of fly ash varies according to its intended purpose. Typically, the additive amount can range from 15 to 35 wt%. The replacement can go up to 70 wt% for pavements, walls and parking lots and even up to 80 wt% for autoclaved aerated concrete [77]. The higher replacement of fly ash in large structures helps to avoid temperature rise of concrete. Recent applications of coal fly ash in bricks and the tests performed on the specimens are shown in Table 5. Like clay, fly ash is rich in Al and Si, and has a great potential to be used as a precursor for the synthesis of sustainable geopolymer bricks. Iftikhar et al., replaced 0–100 wt% of clay with fly ash to develop sustainable green bricks. The bricks with 30–60 wt% fly ash were observed to have low density, allowable water absorption and high compressive strength and could be applied for load bearing purposes [11]. Furthermore, the resultant geopolymer bricks had enhanced physical and mechanical properties relative to traditional clay bricks [11]. The ASTM data suggests that a good brick should possess compressive strength of 3.5 MPa and its water absorption should not be more than 20% of its weight. Evarasan et al., incorporated 40 wt% of fly ash in clay bricks and observed a water absorption of 13.28% and compressive strength greater than 3.5 MPa for his synthesized bricks [78]. In another study, fly ash/lime bricks were produced in a steam autoclave. Lime was used as a binding agent and tobermorite and katoite were formed as binding phases in bricks. The optimum brick produced from 88% of ash, 12% lime and 20% water exhibited a compressive strength of 7.5 MPa [79].

Fly ash, bottom ash and cement were used at an optimum ratio of 1:1:0.45 in the production of bricks. Compressive strengths of between 7.13 and 17.36 MPa were obtained with an ultra-velocity pulse ranging from 2.20 to 2.96 km/s. Fire resistance tests revealed that the bricks did not show any spalling, and, after heating the strength increased up to 30% [80]. In another study, high volumes of fly ash of above 50% were used to replace clay. The bricks showed high compressive strength, low water absorption, no cracking due to lime, no frost effects and a high resistance to frost-melting of the brick [81].

Other waste materials

Apart from plastic and coal fly ash discussed above, there are many other waste materials that have been used in construction composites to reduce the amount of natural aggregates required and to beneficiate waste (Table 6). These waste additives used in bricks are typically classified as sewage sludge, ash, inorganic residues and organic wastes [92]. The use of waste materials with silica in clay bricks to act as fluxing agents has been of particular interest. The silica binds clay particles at relatively low temperature and reduces the energy consumption in brick manufacturing [92].

Waste glass has been employed in burnt clay bricks as an additive. Typically, waste glass is added to brick samples in the range of 0.5–94 wt %. However, most studies usually focus in the range of 5–20 wt%, with the glass particle size between 45 and 600 μm [93]. The glass incorporated bricks are typically shaped through three methods: moulding, pressing and extruding. A pressure of about 1–40 MPa is applied during the pressing method to produce samples. Once, the samples are shaped, they are fired and dried at 100–110 °C for between 3 h to 3 days [93]. It was found that shrinkage increased as the percentage glass by mass increased, and as the firing temperature increased. For larger particles (greater than 132 μm) the shrinkage decreased due to incomplete sintering [93]. The incorporation of waste glass has allowed bricks with enhanced compressive strength and low water absorption to be obtained [94].

Table 4
Classification of coal fly ash according to ASTM.

Property	Fly ash class	
	Class C	Class F
Silicon dioxide (SiO_2) + aluminium oxide (Al_2O_3) + Iron oxide (Fe_2O_3), min. (%)	50	70
Sulphur trioxide (SO_3), max. (%)	5	5
Moisture Content, max. (%)	3	3
Loss on ignition, max. (%)	6	6
Available Alkalis (as NaOH) max. (%)	1.5	1.5

Table 5
Recent applications of coal fly ash in bricks.

Class	Replacement %	Other materials	Brick dimensions (mm)	Test performed	Ref
F	10–50	Clay	30 × 10 × 60	Compressive strength Linear shrinkage Loss on ignition Water absorption Leaching Microstructure	[82]
F	10–30	Waste slug, cement, polypropylene fibers	80 × 40 × 80	Compressive strength Bending strength Shrinkage	[83]
	10,12,16	Cement, lime, wood	–	Compressive strength Water absorption Shrinkage Density	[84]
	10	Cement, sand, palm oil clinker	–	Compressive strength Microstructure Flexural strength Water absorption	[85]
C	2–8	Clay, glass	140 × 65 × 40	Compressive strength Porosity Shrinkage Density	[86]
F	7	Stone dust, lime	190 × 90 × 90	Compressive strength Density Water absorption Percentage voids Efflorescence Modulus of elasticity Shrinkage Impact energy	[87]
F	10–100	Black cotton, granite waste	210 × 100 × 70	Compressive strength Efflorescence Water absorption Weight analysis	[88]
F	85–95	Cigarette butts	50 × 50 × 50	Compressive strength Density Water absorption Microstructure	[89]
C	80–90	Waste brick powder	50 × 50 × 50	Compressive strength Absorption Flow Microstructure	[90]
F	80	Sand	240 × 115 × 53	Porosity Microstructure	[91]

Agricultural waste such as rich husk and sugarcane bagasse have been employed in the manufacturing of bricks. Even though these waste products compromise the strength and increase the water absorption of clay bricks; they result in lighter bricks that produce more energy efficient structures due to the enhanced insulation properties [95]. Gorhan et al., [96] studied the effect of rice husks on the porosity and thermal properties of clay fired bricks. It was observed that the bricks with 5% and 10% addition of rice husk exhibited a compressive strength of 7–10 MPa which is smaller than the reference clay bricks. However, these results still satisfy the building and construction requirements. Samples made using finer rice husks (up to 10%) had higher mechanical strength and thermal conductivity than coarser samples [96]. Similar observations of reduced compressive strength were made by Kazmi et al. [97], upon the addition of agro-waste (sugarcane bagasse ash and rice husk) to clay bricks. The waste was incorporated at 5%, 10% and 15% by clay weight. It was found that 5% addition of sugarcane bagasse ash and rice husk was the optimum addition to produce bricks that met the ASTM requirements. Furthermore, the bricks showed resistance to efflorescence and microscopic analysis showed the specimens to be porous, consistent with the formation of lighter bricks [97]. Interestingly, several waste materials incorporated in bricks produces light bricks which are preferred in earth quake areas [98].

Hybrid construction composites with engineered nanomaterials

Engineered nanomaterials are manufactured materials having at least one dimension (length, breath or height) between 1 and 100 nm [108]. The nanomaterials are more attractive than their bulk counterparts due their excellent inherent properties resulting from their small nanoscale sizes or dimensions. The nanoscale sizes result in a high percentage of surface atoms (relative to bulk atoms) which gives novel properties and reactivity compared to larger size materials with the same chemical composition [109]. At the nanoscale, materials exhibit unique optical, magnetic, electrical and mechanical properties [110]. These properties allow the nanomaterials to be applied in various fields such as electronics, coatings, adhesives and reinforcement polymers [111-114].

In recent years, nanomaterials have gained popularity as reinforcement materials in building and construction composites. This is especially the case for fly ash geopolymers or waste-infused composites that are characterized by brittleness, low tensile strength and fracture toughness [61]. These nanomaterials include nano-silica (nano-SiO_2), nano-titanium dioxide (nano-TiO_2), nano-aluminium oxide (nano-Al_2O_3), nano-zinc oxide (nano-ZnO), CNTs and graphene-based materials (graphene oxide and reduced graphene oxide) [115]. Nanomaterials improve the durability, thermal, mechanical, and electrical properties of composites. Table 7 shows the mechanical improvements of concrete composites enhanced with nanomaterials at loadings as low at 0.5 wt%. Furthermore, studies have shown the hydration kinetics of cement can be significantly influenced by nanoparticles, which reduces the setting time, dormant period, and increases the early strength of the cement [116].

Various techniques are used to test and characterize nanocomposites in order to understand their application properties. These include scanning electron microscopy (SEM) and transmission electron microscopy (TEM) . These techniques assist in determining the morphology of the composite and the binding interaction between the nanomaterials and aggregates [117]. Mechanical properties of the nanocompostie such as tensile strength, compressive strength, bond strength and bend strength are usually tested using universal testing machine procedures [118]. Furthermore, efflorescence (salt content), flame retardancy, moisture content and density tests provide important strength data on construction composites, especially bricks. The test results obtained must comply with a country's materials test standards or international standards (i.e. ASTM) for applicability in building and construction use [119].

Table 6

Recent applications of waste additives in bricks.

Type of waste	Replacement %	Specimen dimensions (mm)	Preparation conditions	Properties tested	Ref
Industrial slug	10 & 20	190 × 90 × 50	Mechanical mixing for 5 min at 8–11 moisture content, 40 MPa applied pressure, firing at 1000 & 1100 °C, soaking time of 2 h at a heating rate of 10 °C/min	Compressive strength Efflorescence Porosity Water absorption	[99]
Biomass (wheat straw, olive stone flour)	6–9	75 × 80 × 75	Dried at 105 °C, fired in an electric furnace 92 °C for 1 h	Compressive strength Density Porosity Thermal conductivity Shrinkage Plasticity Loss on ignition	[100]
Waste marble	Up to 35	–	Dried and then fired at 950 °C and 1050 °C for 2 h	Shrinkage Loss on ignition Bulk density Porosity Water absorption, Compressive strength Thermal conductivity Microstructure Phase content	[101]
Agro-waste (sugarcane bagasse, rice husks)	5–15	228 × 114 × 76	Manual mixing with 20.8–22.9 % moisture content for 3 h, sun-dried for 2 day and then burnt in industrial brick kiln plant at 800 °C for 36 h, conventional air cooling for 45 days	Compressive strength Thermal conductivity Water absorption Porosity Leaching toxicity Specific gravity Microstructure	[95]
Zeolite, steel slag, expanded perlite	15, 15, 50 respectively	150 × 150 × 45	Mechanical mixing at 17–21% moisture content, dried in ambient temperature for 24 h, then oven at 110 °C for 24 h, fired in an electrical furnace at 5 °C/min until 1000 °C for 1 h	Density Weight loss Shrinkage Apparent porosity Water absorption Resistance to freezing and thawing Compressive strength Thermal conductivity Microstructure	[102]
Paper pulp		–	Samples extruded through a square die (45 × 45 mm) at 10 MPa with a length of 160 mm, Oven dried at 105 °C at a rate of 5 °C/min for 24 h, firing 900 °C and then soaking for 5 h	Compressive strength Porosity Thermal conductivity	[103]
Slate tailings	5	Cylinder: 36 × 36	Molding pressure of 5 MPa, 7 days curing at room temperature and 1 day curing in oven, moisture content of 15%	Compressive strength Microstructure Water absorption Durability	[104]
Cigarette butts	1–8	Cylinder: 25 × 50	Mechanical mixing for 25 min with a 15.5% water content, compaction pressure of 240 KPa, dried at 105 °C for 24 h, fired at 1050 °C for 3 h	Thermal properties Energy savings Porosity Micro and macro structure	[105]
Tea waste	2.5–12.5	12 × 40 × 80	Mixed for 30 min with 15% moisture content, oven drying at 40 °C and 100 °C for 24 h, firing temperatures of 950 and 1050 °C	Compressive Thermal properties Porosity Microstructure	[106]
Glass wool	10	20 × 20 × 80	Mechanical mixing at 500 rpm for 2 min, drying for 60 °C for 24 h, demolded and further dried at 10 0 °C, sintered at 950 ° C	Compressive strength Thermal properties Microstructure Flexural strength Water absorption Apparent density	[107]

Table 7
Nanomaterials in cementious construction composites.

Nanomaterial	Diameter Nm	wt.% loading	Mechanical improvements	Ref.
nano-SiO$_2$	100–250	3	Compressive strength: 14%	[124]
nano-TiO$_2$		5	Tensile strength: 35.6% Flexural strength: 30.3%	[125]
nano-Al$_2$O$_3$	3–8	0.5	Compressive strength: 10%	[126]
nano-metakaolin	20	6	Compressive strength: 18%	[127]
nano-TiO$_2$/ nano-Al$_2$O$_3$	10–20	0.5–1	Compressive strength: 42% Splitting strength: 24% Flexural strength: 28%	[128]
nano- Fe$_2$O$_3$	30	2–4	Compressive strength: 167% Flexural strength: 158%	[129]
nano-CuO	15	4	Compressive strength: 237%	[130]
nano-CaCO$_3$			Flexural strength: 39% Fracture toughness: 41%	[131]
nano-ZnO/nano-SiO$_2$	60–100	1.5–3	Compressive strength: 6.6%	[132]

A comparison of fillers with different particle sizes and surface areas used in concrete composites is illustrated by Fig. 4. For the longest time, cement was deemed the finest component to bind aggregates to form composites. However, the need for better performing composites introduced the need for supplemental pozzolanic materials such as fly ash, silica fumes and metakaolin to the cement. Fibers such as carbon, glass and steel are also used in construction composites as reinforcement materials [120]. They increase the ability of composites to resist cracks by postponing the beginning of microcracks, although they are unable to stop their initiation. This is because the spacing between the fibres is large and microcracks are still able to grow [121]. However, carbon nanomaterials such as graphene and CNTs have been extensively studied as reinforcements in concrete and cement composites [117,122,123]. The small particle sizes of the materials reinforces the cement matrix as

their sizes are close to that of the calcium silicate hydrate (C–S–H) gel and this leads to less cracking at the nanoscale. Nanoparticles exhibit pozzolanic properties that contribute in reducing the calcium hydroxide crystals to produce C-S-H crystals which improves their interface structure [122].

Zhang et al., studied the influence of 25 nm nano-TiO$_2$ on the hydration and shrinkage of cement-based materials [133]. It was observed that 5 wt% TiO$_2$ increased the compressive strength of cement mortar through the acceleration of cement hydration and a pore-filler effect. However, a 5 wt% loading of the nanomaterials decreased the cement workability, resulting in the requirement of high volumes of water being required [133]. In another study, nano-TiO$_2$ and nano-ZnO particles (up to 1 wt% loading) were shown to have a more limited effect on mortar fluidity as compared to nano-SiO$_2$ and nano-Al$_2$O$_3$. This was due to their relative lower surface areas. To overcome this, water reducing agents such as superplasticizers were used [134]. Farzadnia et al., added nano-SiO$_2$ to cement mortar and observed an increase in the strength development rate by 15% over 7–28 days of curing. The nano-SiO$_2$ affected the shrinkage by modifying the microstructure, increasing the hydration volume and lowering the free water content in the cement matrix [135]. Furthermore, nano-SiO$_2$ provided a filler effect for the calcium silicate hydrate (Ca–Si–H) particles in the cement and acted as a strong binding agent, thus improving the interaction between the cement and the aggregate [135]. Similarly, nanoparticles enhance the fresh and hardened behavior of fly ash binders, sequentially improving the mechanical strength of the composite [116]. The presence of graphene nanoplatelets (GNP) in a fly ash geopolymer composite improved its resistance towards cracking, as depicted in Fig. 5b). When a crack was indented on the composite surface, micro-cracks were formed. The cracks grew until they met the GNP which bridged the crack and resisted crack widening. This arose since high energy was required to open the crack, and this led to material toughening. A branching and deflection mechanism was observed, which instead of widening the crack, increased its effective length to absorb more energy and release stress [136]. This phenomenon was related to the high Young's modulus (2 TPa) of graphene which allowed its composite to resist the applied load and deflect the crack [136]. Furthermore, as depicted in Fig. 5a, GNPs create a bridging effect which enabled load distribution across the composite.

Bricks remain an important construction material due to their versatility, durability and mechanical stability. Nanomaterials incorporated into bricks or their surfaces modified and enhanced the
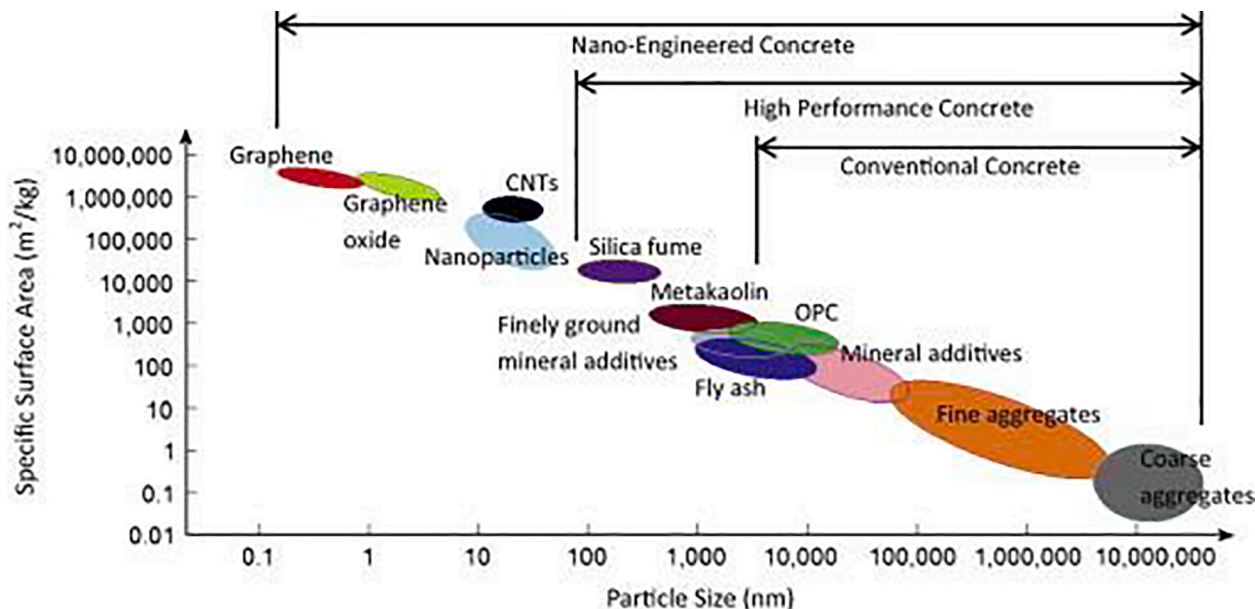

Fig. 4. Particle size and surface area comparison of nanofillers and other aggregates in concrete [122].
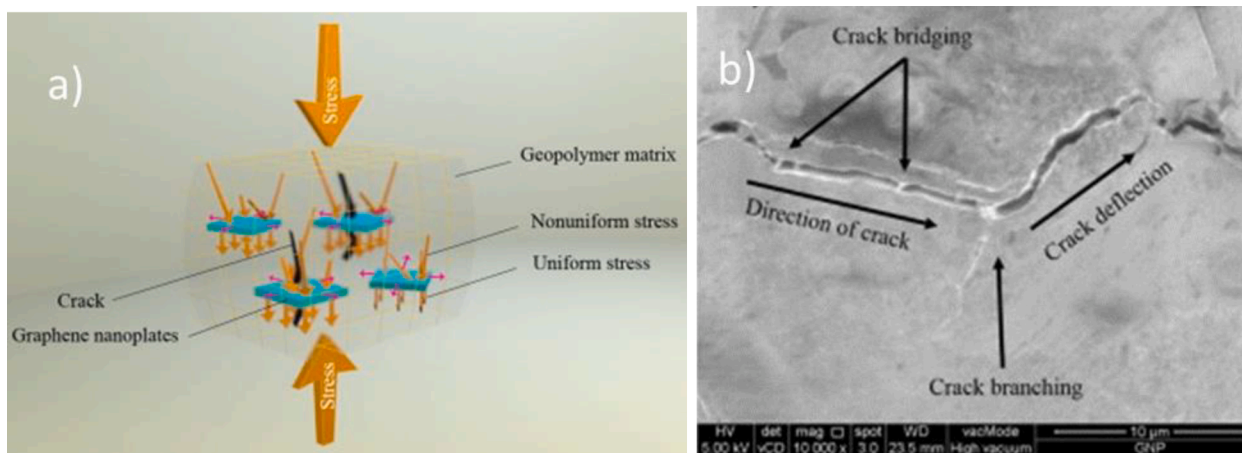
Fig. 5. a) Schematic diagram of graphene/fly ash geopolymer composite under compression load and toughening mechanism b) SEM image of graphene composite under stress [136].

properties of the brick. Cheraghcheshm et al., coated brick surfaces with ZnO nanoparticles and modified them with silver nanoparticles to create a hydrophobic surface as well as generate photocatalytic and anti-bacterial properties. The coated bricks showed an improvement of almost 1.8 times in flexural strength, reduced efflorescence and improved photocatalytic and antibacterial properties [137]. In another study, self-heating graphene oxide (GO) nanocomposite bricks were developed to reduce the energy associated with indoor heating which consumes energy [138]. The bricks showed superior electrical properties and improved mechanical strength from 3.15 MPa to 7.21 MPa when the GO concentration was 0.1 wt%. An increase in graphene loading led to an increase in the heating rate of the composite at a constant applied voltage. For example, at 1 wt% graphene loading, the temperature increased from 22 to 30 °C within 500 s and reached saturation. At a 5 wt% loading, the sample heated to a higher temperature of 150 °C in a shorter time (300 s) [138].

Various nanoparticles have been applied in construction composites. In this review the focus will be on CNTs, which are known to possess inherent properties that makes them suitable as reinforcing composite fillers for waste materials. Unlike, nano-SiO_2 and nano-TiO_2 which has been used in composites, CNTs have a higher aspect ratio which makes them suitable for hindering nano-size crack formation.

Carbon nanotubes

Carbon nanotubes (CNTs) are 1-dimensional (1D) cylindrical carbon allotropes characterized by excellent mechanical strength, thermal stability, high aspect ratio and a high surface area [139,140]. The filler-effect due to their nanosize makes CNTs exceptional reinforcement materials in polymers and concrete composites. CNTs are lightweight materials with a tensile strength of 150 GPa, (100 times stronger that steel), which significantly improves the mechanical properties of composites [141].

CNTs exist as either single walled (SWCNTs), double walled (DWCNTs) or multiwalled (MWCNTs) according to the synthesis procedure which informs the number of rolled-up graphene sheets that make the nanotube (Fig. 6) [142]. MWCNTs are more extensively used due to their relatively low cost, high production yield and facile synthesis procedure. They are commonly used in large application industries as they can be synthesized in large amounts [143]. For building and construction applications, industrial grade MWCNTs (low purity) compared to research grade MWCNTs (high purity) can be used. This makes the application of MWCNTs economically viable especially if the aim is towards creating affordable building materials using cheaper raw materials such as waste. SWCNTs and DWCNTs can also be applied in construction composites to improve their properties [144] but their higher production costs makes them less preferred than MWCNTs.

The surfaces of CNTs have been modified and treated (functionalized) using various procedures in order to enhance their properties for application. Functionalization of CNTs result in the introduction of functional groups, alteration of electrical properties, increase in adhesion/dispersion in composite matrix and an increase in hydrophilicity [140]. The functional groups also enhance various physicochemical properties such as chemical reactivity and solubility. Chemical functionalization of CNTs which involves covalent bonding is normally performed using strong acids (nitric or sulfuric acid) which introduces
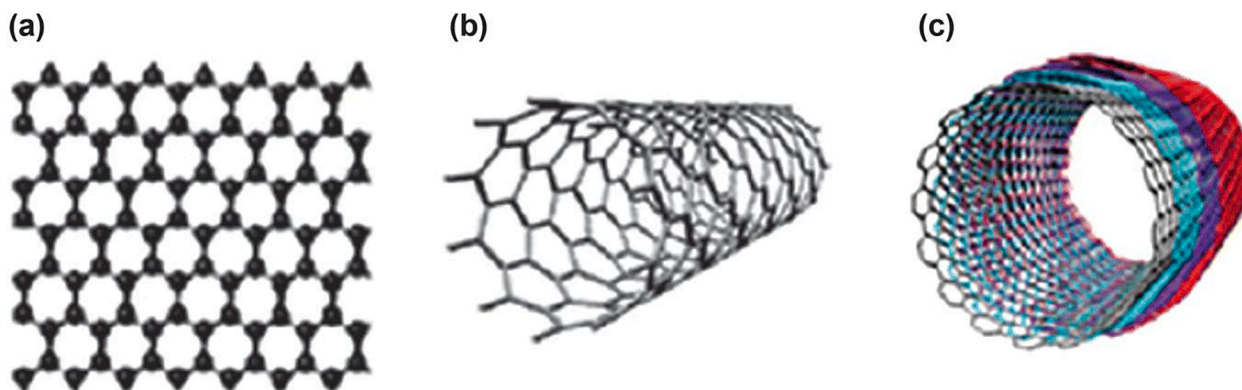
(a)   **(b)**   **(c)**

Fig. 6. Schematic diagram of a) graphene sheet b) SWCNTs c) MWCNTs [142].

oxygenated functional groups. Heteroatoms such nitrogen (N), fluorine (F) or boron (B), phosphorus (P) and sulfur as dopants can also be added [145]. Heteroatom-doping is a process in which heteroatoms replace carbon atoms within the graphitic structure of CNTs [146]. The CNTs functionalized by the covalent methods are dispersible in various organic solvents because the CNTs possess polar or non-polar functional groups. Non-covalent functionalization is used to modify the interfacial properties of CNTs. The CNTs are functionalized by surfactants, polymers and aromatic compounds. This is done by employing π-π stacking or hydrophobic interactions. Non-covalent functionalization ensures that the inherent properties of CNTs are not altered, while improving their dispersibility significantly [147].

CNTs have been shown to be effective fillers in composites [148]. However, they do have some limitations. CNTs have a large specific surface area, bending fiber-like shape, and strong van der Waals interactions, and this makes them agglomerate and entangle. CNTs are hydrophobic and thus difficult to mix with water, organic solvents, or polymers [148]. Furthermore, the long length of CNTs and high polarizability are considered to be the deciding factor that raises the energy required to disperse CNTs within a matrix [149]. The dispersion of CNTs in a composite matrix remains a challenge and this can compromise the strength of the composite especially at high CNT loadings [150]. This is due to difficulties of stress transfer from the matrix to the reinforced materials. Parameters that affect the quality of CNT dispersion are length, purity, entanglement, viscosity, surface modification such as functionalization, and graphitization [151]. Poor dispersibility of CNTs in a matrix can even lead to reduced mechanical strength and ultimately reduce the original properties of the composite [151]. Therefore, the reinforcement of composites by CNTs is dependent on a homogeneous dispersion in the matrix. Advances have been made in the dispersion of various nanomaterials in a polymer matrix for enhanced nanocomposite properties. However, much work is still required in the production of building and construction materials. The challenge lies in the difficulty of modifying the intermolecular interaction between nanomaterials and construction materials such as cement, clay and even fly ash, which have complex mineral and chemical structures [152]. The conventional admixtures used in the cement industry have been reported to act as dispersants and ensure that the nanomaterials are not agglomerated [120,153].

Dispersion methods of CNTs in composite matrix

The dispersion of CNTs in a matrix is crucial for obtaining the desired optimum mechanical strength of the composite. There are two main approaches taken to improve CNT dispersibility, namely, mechanical mixing and functionalizing of CNTs to promote interaction between the interfaces of CNTs and the composite matrix [138]. Ultrasonication and acid treatment of CNTs are the most commonly used dispersion methods.

Ultrasonication of CNTs in the presence of surfactants has been used to obtain better dispersion effects. This is because, ultrasonication aids temporary dispersion of CNTs which results in re-agglomeration [154,155]. Thus, surfactants are used concurrently with ultrasonication as stabilizers to ensure CNTs remain dispersed in solution. Surfactants work by reducing the surface energy of CNTs in a solvent. Sodium dodecylbenzene sulfonate (SDBS) is commonly used as a surfactant [156]. Fig. 7 shows the dispersion of CNTs at various standing times. Poor dispersion was observed when ultrasonication was performed alone, compared to when it was performed in the presence of a sodium dodecyl sulfate (SDS) surfactant. The CNTs remained dispersed in solution beyond 48 h. This was expected to improve their reinforcement performance in a composite matrix [157].

Ju et al., found a 1.5:1 surfactant to MWCNTs ratio to be effective for dispersion [158]. Dispersion of CNTs in surfactant solutions varied with sonication energy from 320 to 50400 J/mL, with the sonication time varying from 5 to 120 min [159]. In another study, CNTs were dispersed using sonication in the presence of SDBS. It was observed that the CNT dispersion was dependent on sonication energy and 2250 J/mL was found to be the optimum dispersion energy [159].

Ultrasonic treatment of CNTs was reported to cause formation of a considerable number of defects in the CNTs as well as CNT buckling, bending, and formation of dislocations in the carbon structures [155]. Arrigo et al., investigated the effect of ultrasonic treatment on the structural characteristics of CNTs. It was shown that the length of CNTs was reduced with increased sonication time. The reduced CNT length affected the rheological behaviour of the nanocomposite, which progressively changed from solid-like to liquid-like upon increased sonication time [155].

In cement composites it was noted that ultrasonication and the use of superplasticizers was effective in dispersing CNTs. A study to address concerns relating to the synergistic effects between the surfactants and cement was undertaken. In the study, superplasticizers such as polyacrylic acid were used in cement composites [160]. The authors reported that the surfactant acted as both a dispersing agent for the CNTs and a plasticizing agent for the cement paste [160].

Chemical surface modification was performed on CNTs by attaching functional groups onto their surfaces to improve hydrophilicity, thus, reducing their tendency to agglomerate for improved dispersion [140]. Chemical treatment of CNTs led to better interfacial bonding between the CNT and the matrix and also enhanced the load transfer [140]. Kanagaraj et al., treated CNTs with concentrated nitric and sulfuric acid in a 1:3 ratio respectively at reflux for 30 min. The treated CNTs were further sonicated for 1 h to give better dispersion in the composite matrix [161]. Li et al., also modified MWCNTs using a nitric and sulfuric acid solution. The acid modification introduced carboxylic acid groups which induced a series of chemical reactions along the interface that enhanced the reinforcement efficiency. It was observed that using surface-treated CNTs improved the dispersion and mechanical properties (flexural and compressive strength) of the composite [118]. However, the shortfall of chemical treatment is the creation of defects on the CNT surfaces which can compromise the CNTs intrinsic strength. Thus, less aggressive chemical treatment should be considered for application of CNTs in reinforcement composites.
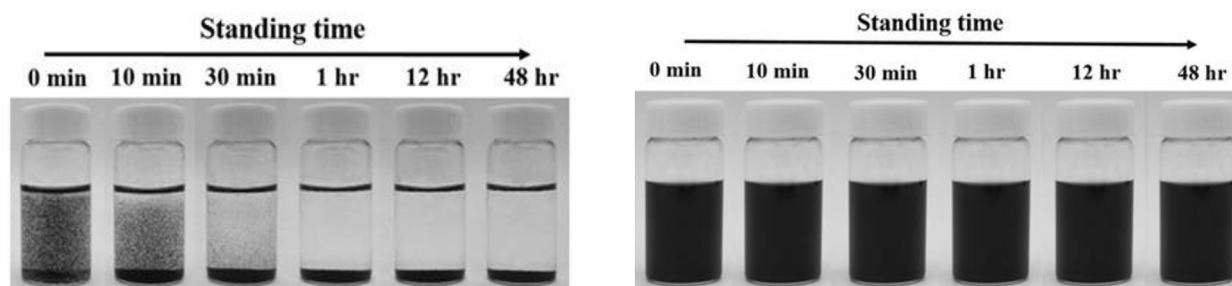

Fig. 7. Dispersion of CNTs a) ultrasonication b) ultrasonication and SDS surfactant [157].

Application of CNT composites in construction materials

CNTs have been used in construction composites for the production of high strength composites with materials such as cement. The reinforced composite matrix has a high aspect ratio and their large surface area increases reactivity at a nanoscopic level [122]. The distribution of the CNTs in the composite is at a finer scale compared to conventional fillers. Thus, extremely low contents of CNTs can make a significant difference in tailoring the properties of composites. Low additions of MWCNTs, ranging from 0.03 to 0.5 wt%, can present a significant improvement in the mechanical properties of construction composites. This is shown in Table 8, where 0.03 wt% of MWCNTs improved the flexural strength and module of elasticity by 25.1% and 13% respectively. It is anticipated that CNTs, when added to concrete, will increase the compressive strength beyond 200 MPa, thus enabling the construction of mile-high skyscrapers [162].

Amin et al., incorporated CNTs dispersed in surfactant (viscocrete) in cement composites at 0.02, 0.05, 0.1 and 0.2% loadings. It was observed that 0.1% was the optimum CNTs loading and this improved the mechanical strength from 80 MPa to 110 MPa [170]. Morsy, studied the hybrid effect of CNTs on cement mortar. The cement was partially replaced with 6 wt% nano-metakaolin and CNTs were added at 0.005, 0.02, 0.05 and 0.1 wt%. The CNT dispersion was assisted by nano-metakaolin which enhanced the interfacial interaction between the CNTs and the cement matrix. It was observed that the CNTs were fixed to the hydration products throughout the paste. They bridged the hydration products and resisted the formation of microcracks by creating networks to transfer the load in tension. The addition of CNTs up to 0.02

wt% improved the mechanical strength of the composite up to 11% but with additions above this level, a reduction in the mechanical strength was observed. This was attributed to CNT agglomeration which led to microvoids and consequential weak areas [127]. In a study performed by Makar et al., SWCNTs were used in cement composites. It was observed that the SWCNTs acted as nucleating sites, with the hydration products forming on the surface of the nanotubes instead of the cement particles. This demonstrated the reinforcing behavior achieved by the presence of the SWCNTs [144].

A denser concrete was obtained when 0.1 wt% CNTs was added to a concrete composite with waste glass and basalt fibre. An increase in compressive strength of up to 23% was recorded for the hybrid composite after 28 days of curing compared to the reference sample [171]. Chaipanich et al., studied the effect of CNTs on 20 wt% fly ash-cement composites. At 1 wt% CNT loading the compressive strength of the fly ash cement was improved to 51.8 MPa [172]. In a recent study, acid treated MWCNTs were added to a geopolymer of coal fly ash and granulated blast furnace slag (GBFS). The functionalized MWCNTs were homogeneously dispersed in an aqueous solution containing SDS dispersant. The geopolymer was incorporated with 0.1 wt% functionalized MWCNTs and 30% coal fly ash and GBFS. Lower water absorption, shorter stetting time and higher compressive and flexural strength of 70 MPa and 9.45 MPa respectively were reported [173].

The need to reduce the reliance on non-renewable resources such as cement, which contributes significantly to carbon emissions is rather an urgent one. The utilization of waste in construction composites is a possible solution to these problems. However, the compromise in the mechanical properties of the composite (using waste) is a problem that cannot be ignored. Nanomaterials such as CNTs compensate for the strength reduction and this could enable composites made from waste and CNTs to find application in the building and construction industry.

CNT-fly ash nanopolymer hybrids

The understanding of the interaction of the materials within a composite is very important as it informs its reinforcement efficiency. It has been noted that the dispersion of CNTs in a composite is the major contributing factor in reinforcement [164]. The strong Van der Waals forces that exists in CNTs makes them bundle together and agglomerate in a composite matrix [174]. This compromises the strength of the composite and introduces weak spots that easily crack when pressure/ load is applied. Chaturvedi et al., observed interfacial bonding to be the interaction mechanism in a fly ash-CNT nanocomposite. It was shown that the hydroxyl functionalized CNTs were well dispersed and no cracks or voids in the composite were observed (Fig. 8d), indicating strong interfacial bonding [175].

Salah et al., used xylene to disperse functionalized CNTs and to also dissolve HDPE. During sonication, the HDPE chains were broken and free carbon radicals might have been generated. The carboxylic (–COOH) and hydroxyl (–OH) groups on the surface of CNTs were probably attached on the generated radicals during the sonication and dissolution processes. The attached and generated radicals further reacted with the remaining radical groups in the CNTs to form covalent and non-covalent bonds (Fig. 9) [176]. In a hybrid composite containing coal fly ash, non-covalent (interfacial) bonding can exist within the matrix. To enhance bonding between coal fly ash and the polymer/CNT matrix, alkali activators (sodium hydroxide/sodium silicate solution) have been used [14]. This activates the SiO_2 and Al_2O_3 which are the major constituents in coal fly ash (Fig. 9). The silica and alumina species nucleate to form aluminosilicate oligomers which can experience interfacial adhesion to the polymer/CNT matrix to produce a hybrid nanocomposite.

Release of CNTs to the environment

Due to their wide application, the rate at which engineered nano-materials (ENM), especially CNTs are produced, is increasing. Thus,

Table 8
Carbon nanomaterials applied in cementitious composites.

Carbon nanomaterial	Dimensions (T: thickness/D: diameter/L: length)	Loading wt.%	Mechanical improvements	Ref.
Carbon nanofibers	D60–150 nm/ L30–100 μm	0.2	flexural strength: 14% module elasticity: 23%	[163]
MWCNTs (short)	D20–40 nm/ L10–30 mm	0.08	flexural strength: 25% module of elasticity: 62.5%	[121]
MWCNTs (long)	D9.5 nm /L1.5 μm	0.03 0.075	flexural strength: 25.1% module of elasticity: 13% flexural strength: 49.89% module of elasticity: 31.4%	[164]
SWCNTs		0.04	flexural strength: 4% compressive strength: 0.89%	[165]
Graphene sheets		0.025	compressive strength: 10% flexural strength: 16%	[166]
Graphene oxide	T0.8 nm/ D80–260 nm	0.05	compressive strength: 47.9% flexural strength: 35.8% module elasticity: 30.2%	[167]
Graphene nanoplatelets	T1.4 nm	0.16	compressive strength: 39% flexural strength: 38%	[168]
Reduced graphene oxide (rGO)		0.05	compressive strength: 29% flexural strength: 49%	[169]

Fig. 8. SEM images of a) fly ash, (b) CNT and (c–d) CNT-fly ash nanocomposites [175].
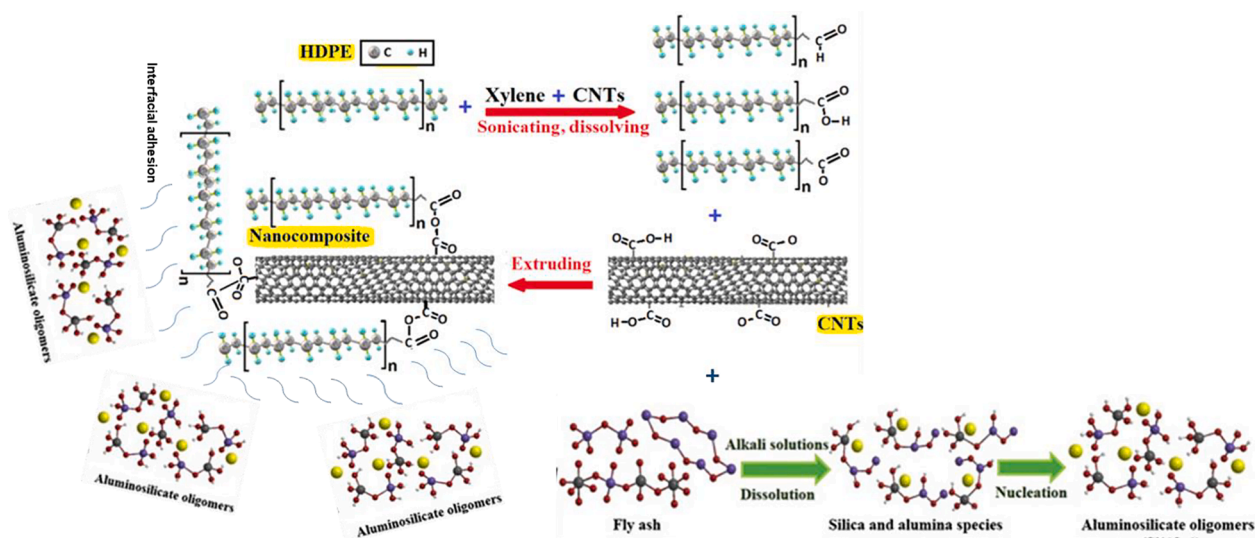
Fig. 9. Schematic diagram of the proposed mechanism of interaction between HDPE, functionalized CNTs and alkali activated coal fly ash [14,113].

their potential environmental release as either raw materials or from composite products must be considered. This is important because studies of their potential toxicity, which affects both the environment and human health, have gained importance [177-179]. A release assessment is critical for exposed workers, consumers and the environment. It is important for exposed individuals to protect themselves during the processing of the composites and during construction by following the set guidelines and regulations for handling nanomaterials [180]. Furthermore, to regard products as sustainable, their end-of life must also be understood.

The release of CNTs may occur at all stages in the life cycle of the product as shown in Fig. 10. Notably, most of the exposure and release of ENMs occurs during synthesis, while the potential release in other life cycle stages is considered to be low, but cannot be ignored [181]. Composites are likely to undergo some form of degradation when exposed to moisture, radiation and temperature. The alteration of the matrix by degradation likely exposes CNTs to the surface of the composite and they can be potentially released to the environment [182].

CNTs can be released from the nanocomposite as a result of various mechanical actions such as scratching, sanding, sawing and drilling. For
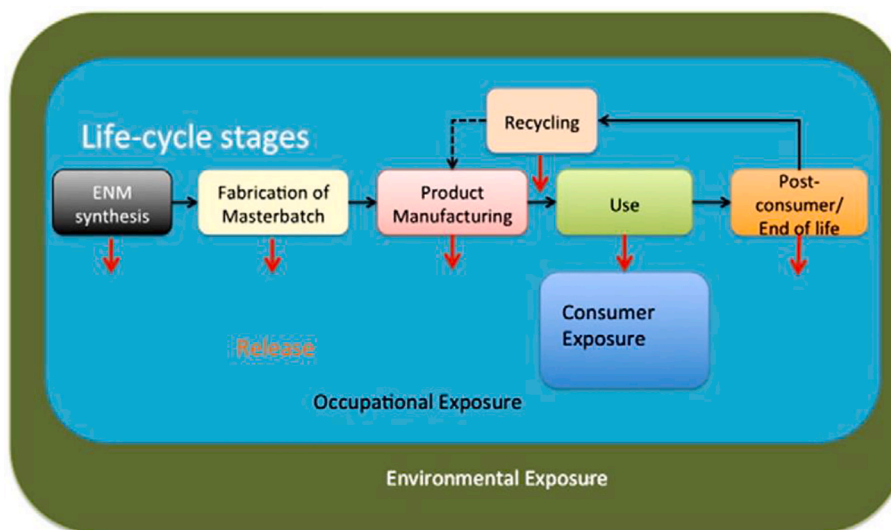
Fig. 10. Schematic diagram of the life-cycle stages of engineered nanomaterials [181].

example, the Taber Abraser is used to measure the release of particles during the lifespan of the product [183]. Golanski investigated the release of CNTs from CNT/cement composites with up to a 4 wt% CNTs loading. Low abrasion mechanical action was simulated and no release of CNTs was detected from well dispersed samples. However, with poorly dispersed CNT samples, free standing agglomerated CNTs particles were observed under a transmission electron microscope (TEM) [184].

Weathering is another pathway to release CNTs from a composite. Weathering sources include UV-light, elevated temperatures, water absorption or a combination of these processes. This effect weakens or even degrades the nanocomposite matrix and hence exposes the CNTs to the environment [183]. In a recent study, composites with CNTs and amino-functionalized CNTs were degraded with a combination of UV light and moisture [185]. The degradation of the matrix was observed by the formation of cracks shown in SEM images. The SEM images showed loose CNTs on the surface and within the cracks of the sample. The functionalized CNTs were better dispersed than the pristine CNTs but the matrix was not very homogeneous [183]. Thus, degradation and release of the CNTs from the composite was observed.

Lastly, CNTs can be released from the composite due to inappropriate disposal by fire or by accidental combustion. This is because the CNTs have a higher decomposition temperature than most composite matrix materials. Hence, they have been used as flame retardants in building and construction composites. Likewise, CNTs have been reported to form a protective layer in composites which could offer thermal insulation [186].

Furthermore, studies on the disposal or repurposing of CNT-reinforced building and construction composites must be undertaken to ensure sustainability at the end of their life-cycle.

**Summary and conclusion**

The application of nanomaterials in construction composites especially construction bricks presents an opportunity to transform waste materials such as plastics and fly ash into useful products. This is one sustainable way of cleaning up the environment while producing products for a wide range of applications within the built environment. The products produced from nanomaterial reinforcement materials such as CNTs have been shown to possess high mechanical, thermal, and water resistance properties that are desired in the industries.

Much research is still required to understand the mechanism of interaction between the waste materials and nanomaterials. This will assist in understanding the physicochemical properties of the waste-nanocomposite, which will in turn affect the quality and durability of the material. Concerns such as the environmental impact of incorporating engineered nanomaterials into construction materials must be addressed if the aim is towards sustainability. Greener methods for nanomaterial synthesis must be adopted lest the aim of reducing carbon emission from cement and concrete be counterproductive. It is noteworthy that little research has been conducted on the environmental effects of CNTs in construction composites. This is a point of concern considering the rate of their application as reinforcement materials. Their potential release to the environment and any harmful effects they may pose, is not fully understood. From the review, it is noted that well-dispersed CNTs in composites are unlikely to be released from the composite. Thus, efforts must be made to ensure homogeneity of CNTs in composites to reduce their likelihood of leaching to the environment.

In summary, the benefits of nanomaterials in construction composites cannot be ignored and their utilization must be in a sustainable manner. The effects of nanomaterials in construction composites are listed below:

- Nanomaterials allow the utilization of waste even at high replacement ratio by providing reinforcement properties which compensate the loss of mechanical strength from the waste.
- To maximize the intrinsic properties of nanomaterials, good dispersion is required to avoid agglomeration which compromises the strength of the composite. Functionalized CNTs and ultrasound-surfactant treated CNTs were shown to give a good CNT dispersion in composites. In cement composites, superplasticizers are mostly used as both a plasticizing agent and dispersing agent. However, the extent to which CNTs remain dispersed in a composite matrix needs to be better understood.
- Due to their large surface areas, nanomaterials are highly reactive and have been shown to accelerate hydration and provide nucleating sites that assist in reinforcing the composite. CNTs were observed to bridge the hydration products and to resist the formation of micro-cracks by creating networks for load transfer.
- Nanomaterials act as nanofillers and modify the microstructure of the composites which improves the overall mechanical strength.

Nanomaterials offer endless opportunities in the building and construction industry because they are engineered and can be tailored for purpose. The application of undesired waste such as coal fly ash, with high loss on ignition, is accessible to study and will aid in the utilization

of nanomaterials in the building industry. This will open more avenues for waste beneficiation and promote a more circular economy.

CRediT authorship contribution statement

Boitumelo Makgabutlane: Conceptualization, Writing – original draft. **Manoko S. Maubane-Nkadimeng:** Writing – review & editing, Supervision. **Neil J. Coville:** Writing – review & editing, Supervision. **Sabelo D. Mhlanga:** Conceptualization, Writing – review & editing, Supervision.

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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