(eISSN:3083-8231)
Review
Volume 2, Issue 1, 2025
https://doi.org/10.51200/susten.v2i1.5220

A Review of Nanotechnology Applications in Sustainable Cementitious Materials

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Received: 04 July 2024, Accepted: 11 Sept 2024

Abstract: The application of nanotechnology in the construction industry can solve numerous environmental issues by effectively utilizing large quantities of waste materials generated during construction. Applying nanotechnology in the cement concrete industry has led to numerous benefits. The adoption of nanotechnology in cement mortar and concrete results in significant improvements in properties such as workability, compressive, flexural, and tensile strengths, as well as water absorption, chloride ion permeability, and shrinkage. Hence, the utilization of nanotechnology can enhance environmental sustainability in construction materials by incorporating significant amounts of waste materials as supplementary cementitious materials. This practice not only reduces the emissions of carbon dioxide (CO_2) resulting from the accumulation of waste in landfills but also reduces cement production. By advocating for the integration of nano-waste materials in significant volumes within construction materials, this research underscores the pathway towards sustainable construction practices. Such endeavors align with the broader objective of contributing to the attainment of the sustainable development Goals (SDGs), particularly in protecting the environment and combating climate change.

Keywords: Nanotechnology; Supplementary cementitious materials; Sustainable construction materials; Cement concrete

1. Introduction

In recent years (2018-2023), cement-based waste materials were extensively accepted in the construction industry due to their favorable properties and low cost [1, 2]. Generally, ordinary Portland cement (OPC) is the main active material. In 2019, the total cement production was around 4.1 billion tons, which recorded an increment of 1.2% compared to the production rate in 2018, as reported by American Geological Survey [3]. The previous studies stated that the production of 1 ton of cement leads to the release of around 0.7 to 1 ton emissions of carbon dioxide (CO_2) into the atmosphere, causing climate change and environmental pollution [4, 5]. Hence, decreasing the cement consumption is tremendously significant for reducing emissions of CO_2, and thus enhancing environmental and sustainability in construction materials [6]. Existing literature highlights that high-pozzolanic materials like silica fume and fly ash can be used as cement replacements in different replacement levels, thus it contributes in reducing CO_2 emission and cement production [7, 8]. Moreover, these materials can enhance the mechanical, durability, and microstructure properties of cement mortar and concrete when utilized up to a certain level [7, 9].

Nanotechnology is an innovative approach focused on modifying the performance and behavior of materials at the nano-scale, typically ranging between 1-100 nanometers (nm) [10]. Thus, this process facilitates the creation of new materials characterized by enhanced durability and performance through molecular-level modifications [11]. This process can also achieve significant environmental and sustainability benefits within the construction industry [12]. Also, nanomaterials have gained widespread acceptance among researchers in the construction materials field due to their potential to modify material properties at the nanoscale. The most widely used nano-materials in construction materials include carbon nanotubes, nano-$CaCO_3$, nano-TiO_2, nano-Al_2O_3, and nano-SiO_2 [13, 14]. While these nanomaterials have shown a significant potential for enhancing the mechanical and durability properties of cement concrete, the widespread of their applications in the construction industry is hindered by their high cost. Hence, a new approach has emerged, focusing on modifying and treating waste materials, transforming their particles into the nanoscale to enhance their properties at that level. The next section introduces some of these materials and their impact on the performance of cement composites in detail.

2. Materials and Methods

In recent years, there has been a growing interest in nanomaterials technology, driven by advancements in the construction industry and the potential advantages of applying this technique to modify construction materials [15]. Furthermore, nanomaterials have demonstrated considerable achievements in enhancing the mechanical strength and durability of construction materials [16]. Besides that, nanotechnology also improved the environment through the treatment of waste materials instead of accumulating them in landfills that might be causing health problems for humans and resulting in generating large amounts of greenhouse gases (GHG) such as CO_2 [17]. While not all waste materials can be treated to nanoscale and used in concrete, some common examples of nano-waste materials that can be treated and utilized in concrete mixtures include nano-silica, nano-alumina, nano-clay, nano-palm oil fuel ash, nano-slag, nano-glass waste, and nano-fly ash as they are compatible with existing construction practices and regulations. Therefore, the use of waste materials in concrete production, especially as cement replacement, will mainly contribute to reducing cement production, thus reducing the release of CO_2 emissions into the atmosphere, thereby fostering the creation of a cleaner environment with highly sustainable construction materials. The reduction of cement consumption in the construction industry can be achieved using nanoparticles technology. This technology enhances pozzolanic activity, resulting in the formation of calcium silicate hydrate (C-S-H) gel, which contributes to the strength and durability of concrete. Additionally, it reduces permeability and, in some cases, improves workability, thus allowing for a reduction in the amount of cement needed to achieve the desired properties. The following subsections present findings from previous studies on waste materials modified by nanotechnology, which have been employed as sustainable construction materials.

2.1. Nano clay

Nano-clay refers to nanoparticles of layered mineral silicates, whose properties are influenced by both nanoparticle morphology and chemical composition [18]. Nano-clay is prepared in different categories according to their morphology and chemical composition like halloysite, hectorite, kaolinite, bentonite, and montmorillonite [19]. Nano-clay made of montmorillonite mineral is the most widely spread due to its unique structure, characterized by dimensions ranging from 70–150 nm in width and 1 nm in thickness [20]. The unique structure of nano-clay made it more stable, with high chemical reactivity, and high hydration [21].

The primary source material for the production of nano-clay is abundantly available clay, which can be collected from various locations at no cost. This factor makes nano-clay the most effective option compared to other nanomaterials [22]. Even though many studies were conducted on the benefits desired from using nano-clay in construction materials, the use of nano-clay is still limited. For this purpose, this review provides insights into the advantages of utilizing nano-clay in concrete production and its impact on the performance of cement concrete. These details are summarized in table 1.

Table 1. Effect of nano-clay on the properties of cement mortar and concrete.

Ref.	Type of clay mineral	Nano-clay content (kg/m^3)	Cement content (kg/m^3)	Effect on the concrete properties
Mokhtar et al. [23]	kaolinite	15 - 27	450	The high pozzolanic activity and large surface area of nano-clay contribute to enhanced performance when added to concrete samples, resulting in increased compressive, flexural, and tensile strengths
Mehrabi et al. [24]	-*	3.15-9.45	350	Owing to their micro-filling ability and pozzolanic reactivity, using nano-clay leads to an increase in the density of concrete. In addition, the high pozzolanic reactivity and specific surface area of nano-clay can minimize the porosity and increase the density of the cement mortar.
Hamed et al. [25]	-	22.5 - 45	450	The particles of nano-clay improved all mechanical properties of concrete due to the use of the optimal proportion of 7.5% nano-clay. SEM images revealed four key benefits of nano-clay in concrete mixtures: Pozzolanic reactivity effect, filling effect, needle effect, and nucleation effect.
Allalou et al. [26]	halloysite	0-27	450	Increasing the calcined nano-clay leads to an increase in the percentage of water for standard consistency, while significantly reducing the setting times. This increase also promotes the production of extra hydrated products, mostly C-S-H gels, due to their highly pozzolanic reactions.

*Not mentioned in the literature.

2.2. Nano-fly ash

Nano-fly ash can be prepared using a mechanical process that minimizes their particle sizes from the micro to nano scale. Alobaidi et al. [27], prepared the fly ash by sieving the clean particles at size 75 µm then dehydrated the particles at a temperature of 100°C for about 24 hours. They then exposed the dried-fly ash to grinding machines for two phases. High energy ball milling machine was used at first stage, while the second phase comprised of using a local custom fine-tuning electrical milling machine. They achieved a small particle size of approximately 60 nm with a high specific surface area for the fly ash. In addition, the researchers observed that the addition of 10% nano-fly ash recorded the highest compressive strength of self-compacting concrete (SCC) among other mixtures, as illustrated in Figure 1.

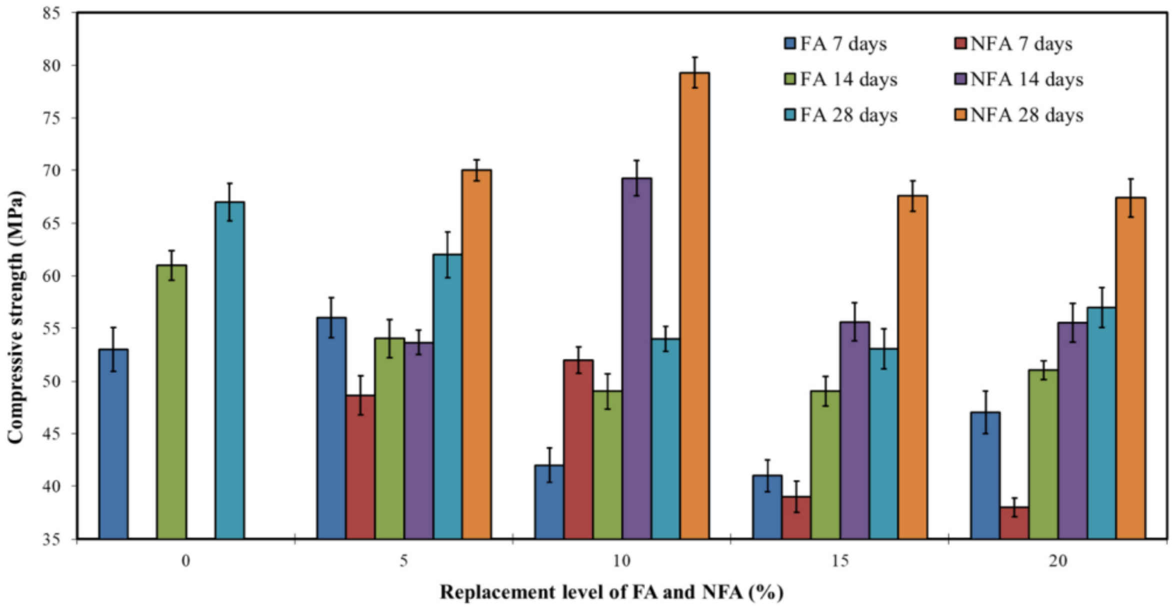

Figure 1. Effect of nano-fly ash on the compressive strength of SCC at various curing ages [27].

Hamada et al., [28] investigated the effect of nano-fly ash on concrete through a comparative study between nano-fly ash and nano-palm oil fuel ash (POFA). The study concluded that nano-fly ash exhibits favorable pozzolanic behavior, attributed to its high specific surface area and high silica content. Using the nano-fly ash can reduce the amount of cement used in concrete production by up to 80% and consequently decrease the environmental pollution.

2.3. Nano-palm oil fuel ash (POFA)

Huge amounts of POFA have been generated due to the production of crude palm oil, most of these wastes have been disposed into landfills and open areas causing the release of GHG like CO_2 [29-33]. Some of the previous studies used POFA in large scale (micrometer scale) up to certain extent to reduce consuming cement and/or fine aggregate [34]. But this particle size offer limited benefits as they result in lower pozzolanic reactions and restrict the production of C-S-H gels, which are crucial for enhancing the mechanical and durability properties of cement mortar and concrete [33]. Few studies have showed the effect of nano-POFA on the mechanical and durability properties of concrete [28, 35-38]. Therefore, it is crucial to conduct further studies to discover the potential use of nano-POFA as a cement replacement in concrete production, thus contributing to reducing environmental pollution by reducing cement consumption and incorporating these waste materials into concrete production. Table 2 shows the effect of nano-POFA on the cement mortar and concrete properties.

Table 2. Effect of nano-POFA on the cement mortar and concrete properties.

Ref.	Nano-POFA content (kg/m^3)	Cement content (kg/m^3)	Effect on the concrete properties
Hamada et al. [36]	0-150	500	The use of 30% nano-POFA as a cement replacement has significant impact in improving the workability of concrete mixtures. Nano-POFA led to increasing the compressive strength up to 58.3 MPa as well as improving the durability of concrete.
Wi et al. [39]	0-405	450	The compressive strength increased with time due to creating further C-S-H gels. Consequently, the optimum proportion of nano-POFA is 30% which should be used to get the best performance of concrete.
Hamada et al. [35]	0-150	500	The nano-POFA features particle sizes smaller than those of cement, measuring only 524 nm. These particles exhibit angular and semicircular shapes, along with a porous texture.
Hamada et al. [37]	0-108	450	The addition of 15% nano-POFA has achieved higher compressive, tensile, and flexural strengths of lightweight aggregate concrete.
Lim et al. [40]	0-105	525	Nano-POFA was used at high replacement levels of cement, reaching up to 80%. The results indicated that the compressive strength remained higher in nano-POFA concrete than in the control sample without nano-POFA.
Hamada et al. [38]	30	450	The addition of 15% nano-POFA recorded the highest compressive strength of concrete. Additionally, the use of nano-POFA leads to reduced carbon emissions and overall costs of the concrete mixtures.

2.4 Nano-eggshell powder (ESP)

In the literature, several methods for preparing nano eggshell powder (nano-ESP) have been reported. Mosaddegh [41] prepared the nano-ESP using ultrasound irradiation and used this product as a biodegradable catalyst. Hamada et al. [35] outlined a multi-step process for preparing nano-ESP, as shown in Figure 2, involving heating the eggshells and mechanically crushing them. They investigated its use as a cement replacement ranging from 0 to 5% of the total cement weight. Their findings revealed that the nano-ESP exhibited a nanoparticle size of 524 nm with a high specific surface area which facilitates interactions with other materials to enhance concrete properties. Amin et al. [42] investigated the effect of nano-ESP on the high strength concrete properties. The researchers observed that the use of nano-EPS accelerates the setting time of high strength concrete. In addition to that, the use of 5% nano-ESP improves the microstructure of high strength concrete in terms of reducing cracks and pores. Besides, Hamada et al. [43] reported that nano-ESP can be used as a stabilizing binder [44], fine aggregate [45], and filler in hot asphalt [46], in addition to using it as replacement for cement [47]. Table 3 shows the effect of nano-ESP on the properties of cement mortar and concrete.

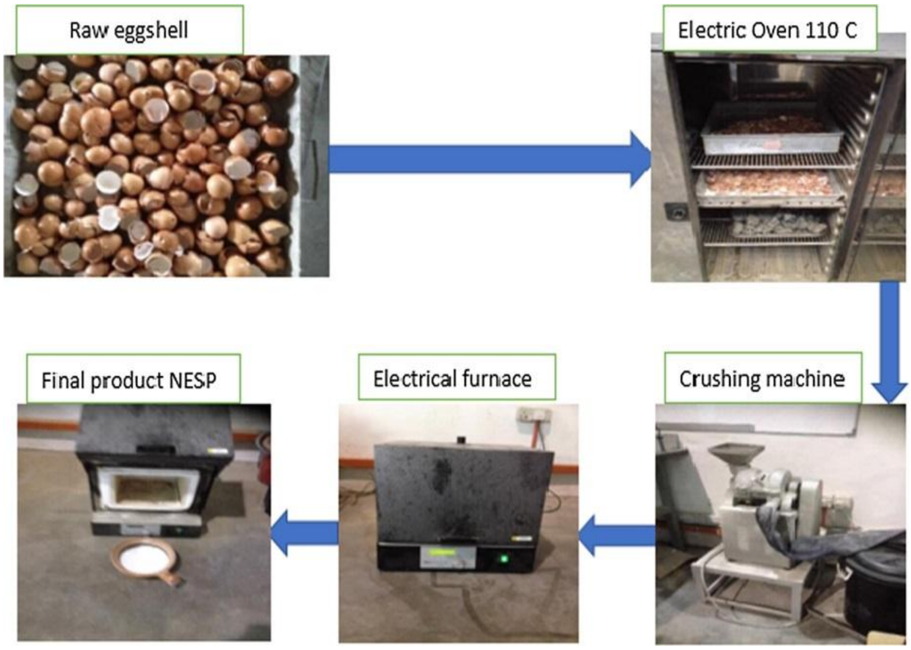

Figure 2. Process steps to prepare nano-ESP [35].

Table 3. Effect of nano-ESP on the cement mortar and concrete.

Ref.	Nano-ESP content (kg/m^3)	Cement content (kg/m^3)	Effect on the concrete properties
Shcherban et al. [48]	0-112.5	375	The addition of 10% nano-ESP proved to be the most effective, resulting in enhanced concrete performance. The compressive and tensile strengths of concrete increased by 9% and 11%, respectively, compared to the reference sample.
Amin et al. [42]	0-33.75	450	The use of nano-ESP delays the setting time of high strength concrete mixes. Additionally, the workability was reduced gradually due to the increase in nano-ESP content, leading to a reduction in slump value from 143 mm to 106 mm when 7.5% nano-ESP was used.
Hamada et al. [35]	0-25	500	The addition of 2.5% of nano-ESP with 10% nano-POFA leads to an increase in the compressive strength of concrete at all curing ages. Also, nano-ESP with nano-POFA resulted in a reduction in the water absorption of concrete, enhancing its durability against acidic and sulfate environments.

2.5 Nano-slag

Sahu and Bera [49] used high energy ball milling (HEBM) to reduce the particle size of slag from micro size to nanoparticle size. This method was used to modify the glassy and smooth slag surface into a further effective form. The grinding process in HEBM of microparticles took about 15 hours. Samples were collected at intervals of every 3 hours during the process for characterization purposes using SEM/EDX and XRD tests. Sharmila and Dhinakaran [50] investigated the influence of nano-slag on the properties of cement concrete. Nano-slag was used as cement replacement in different proportions of 0, 5, 10, and 15%. The results obtained showed that the best performance of concrete mixtures was recorded when 10%

nano-slag was used as a cement replacement. Table 4 shows the effect of nano-slag on the properties of cement mortar and concrete.

Table 4. Effect of nano-slag on the properties of cement mortar and concrete

Ref.	Nano-slag content (kg/m^3)	Cement content (kg/m^3)	Effect on the concrete properties
Sahu and Bera [49]	0-40	537	The addition of 20% nano-slag as a cement replacement resulted in higher compressive strength compared to other concrete mixtures, with an increase of 17% compared to normal concrete without nano-slag. However, the compressive strength reduced gradually as a result of adding more than 20% of nano-slag.
Sadawy et al. [51]	0-17.5	350	The addition of nano-slag can increase the formation of products of C–S–H, thus resulting in an increase in the early-age compressive strength of concrete. Besides, the production of further amounts of C-S-H gels and the filling effect of nano-clay lead to an increase in the density of concrete, as it becomes more compact than that of normal concrete without nano-slag.
Ab Atiyah et al. [52]	0-22.5	450	Addition of nano-slag results in an increase in the density by up to 1.43% and a reduction in the porosity and water absorption by around (9.9% to 55.2%) and (32.55% to 45.75%), respectively.

2.6 Nano-glass waste

In recent years, the construction industry has shown increased interest in sustainable construction practices, particularly in the utilization of recycled waste materials to create new construction materials. Waste glass is among the materials being considered for such applications [53]. Elmoaty et al. [54] and Sancheti et al. [55] studied the potential use of bottle glass powder as a partial cement replacement at varying percentages of 5, 10, 15, 20, and 25% by cement weight. Glass powder was produced by crushing and grinding waste glass bottles using a ball mill, then filtering it through a 90 µm sieve. The results revealed that the compressive strength slightly increased when 5% of nano-glass powder was added. In addition to other properties such as chloride ion resistance, water absorption also improved with the addition of 5% nano-glass powder. Figure 3 shows the process used to prepare nano-glass, involving crushing glass using a roller machine, followed by grinding it with a ball mill and jet mill.

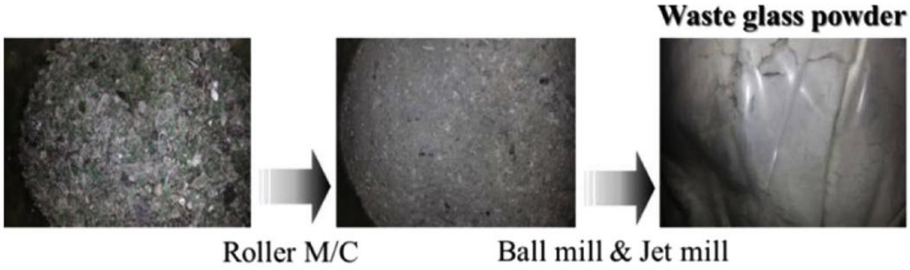

Figure 3. Process steps to prepare nano-glass [58].

Table 5 shows the effect of nano-glass on the properties of cement mortar and concrete.

Table 5. Effect of nano-glass waste on the properties of cement mortar and concrete

Ref.	Nano-glass content (kg/m^3)	Cement content (kg/m^3)	Effect on the concrete properties
Onaizi et al. [56]	0-46	460	The addition of nano-glass resulted in reducing the slump value by about 12.3% compared to the reference sample because of the shape and high specific surface area of nano-glass powder. Whereas the addition of 5% nano-glass powder led to an increase in the compressive strength of concrete from 12.46 MPa to 21.23 MPa.
Amran et al. [57]	0-46	460	The addition of nano-glass led to a reduction in the workability of concrete. Increasing the nano-glass powder from 0% to 6% led to enhancing the compressive, flexural, and tensile strengths as well as the elastic modulus of concrete.

3. Conclusions

This review paper reveals the effect of nanomaterials as sustainable materials on concrete properties such as workability, compressive strength, flexural strength, tensile strength, and water absorption. The results obtained from this paper can be summarized in the following conclusions.

- The addition of nano-clay and calcined nano-clay can improve the performance of cement concrete and prove to be optimal materials due to their availability, pozzolanic properties, high specific surface area, and cost-effectiveness when compared to other materials.
- Both nano-fly ash and nano-POFA exhibit a high pozzolanic activity and possess a high specific surface area. Utilizing them as partial cement replacements can lead to increased compressive strength and reduced water absorption. This is attributed to the additional production of C-S-H gels, which are crucial for enhancing the microstructure.
- The addition of 2.5 – 5% of nano-ESP can improve the mechanical properties and reduce the water absorption of concrete.
- The addition of nano-slag and nano-glass in small amounts can enhance the mechanical and durability properties of concrete, especially at later age.

From the conclusions mentioned above, this study recommends adopting other new techniques to obtain nano-waste materials, such as chemical treatment or using other physical methods to obtain nanomaterials with nano-particle sizes. Furthermore, it is advisable to utilize these nano-waste materials in the production of geopolymer concrete and ultra-high-performance concrete. This process aims to minimize cement usage while enhancing the properties of these materials.

Acknowledgments

The authors would like to express their gratitude to the American University of Sharjah for their unwavering support and continuous encouragement in fostering the development of our research towards sustainable development in the construction industry.

Conflicts of Interest

The authors declare no conflict of interest.

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