Journal of Building Pathology and Rehabilitation (2020) 5:21
https://doi.org/10.1007/s41024-020-00089-9

RESEARCH ARTICLE

Nanomaterials in cementitious composites: review of durability performance

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Received: 28 April 2020 / Accepted: 16 June 2020 / Published online: 18 June 2020

Abstract
Cementitious composites have been a preferred building material for different rehabilitation and construction purposes due to its strength, durability and versatility. However, the permeability of the composites poses a pathway for the ingress of moisture alongside detrimental ions which might subject it to various durability threats. Several methods have been developed over the decades about ways to reduce the permeability properties of cementitious composites and improve its overall durability. One of the effective ways to improve the durability of these cementitious composites gaining attention recently is the incorporation of nanomaterials to densify the composite's matrix. As the application of nanomaterials in cementitious composites is relatively new, it is of great importance to understand how different nanomaterials affect the durability performance of cementitious composites. Therefore, this comprehensive overview is carried out to explore solely the durability properties investigated by various studies where nanomaterials were incorporated into various cementitious composites (i.e. paste, mortar and concrete). The durability properties explored include permeability, volumetric changes and resistance to various chemical and physical attacks. Discussions presented in this paper showed that the incorporation of nanomaterials into cementitious composites at certain dosages can be used to improve the durability performance. However, there is a critical need to not exceed the optimum dosages of nanomaterials as this will result in a detrimental effect on the durability performance of the cementitious composites.

Keywords Cementitious composites · Durability · Nanomaterials · Permeability · Nano-silica

1 Introduction

The continuous dominance of cementitious composites as a building material for different infrastructure rehabilitation and construction is a result of its enhanced strength, durability and versatility compared to other building materials such as steel, glass, wood, etc. In addition, the raw materials used to make cementitious composites are relatively cheap and readily available. However, the subjection of structures made with cementitious composites to various aggressive environments has called for a need to enhance the durability performance of these structures. The evolution of the cement-based materials technology has resulted in various ways to improve the performance of cementitious composites. One of the effective methods to improve the performance of cementitious composites gaining attention recently is the use of nanomaterials. Nanomaterials are generally materials with sizes ranging from 0.1 to 100 nm. The durability enhancement of cementitious composites with the use of nanomaterials can be associated with its smaller size which refines the nano and microstructure of the composite. Also, nanomaterials used in cementitious composites possess pozzolanic properties which result in more product formation and corresponding densification of the microstructure [1, 2]. The reaction of nanomaterials with the calcium hydroxide in the pore solution of cementitious composite results in the formation of more calcium silicate hydrate. The hydration reaction of Portland cement has also been found to be expedited in the presence of nanomaterials and resulted in the formation of more calcium hydroxide [3–5]. The increase in the formation of hydration products can be attributed to the nanomaterials serving as a nucleation area for the formation of hydration products as a result of their high surface area. Several other studies have also shown that the incorporation of nanomaterials into cementitious composites results in an

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increase in the formation of calcium hydroxide especially at an early age [4, 6]. The formation of silicate chain length as a result of the presence of nanomaterials in cementitious composites has also been found to result in increased resistance of the composites to chemical attacks [7]. In addition to the durability enhancement of cementitious composites associated with the use of nanomaterials, the use of nanomaterials has also been found to result in a reduction in cement content. For example, the use of about 1 kg of nanomaterials will result in a reduction of about 4 kg of cement to achieved enhanced properties [6, 8]. Though several types of nanomaterials exist for various applications, the common types used in cementitious composites are nano silica, nano titanium and nano alumina [2]. Ozyildirim and Zegetosky [9] reported significant improvement in the permeability properties of concrete mixtures incorporating nano silica and nano clay at a very low dosage. The improvement of the durability properties of cementitious composite with the incorporation of nanomaterial is generally attributed to the pore filling and nuclei activating ability of the nanomaterials. Despite the benefits of the use of nanomaterials in cementitious composites, it is essential to ensure that these nanomaterials are well dispersed in the mixtures when used. The improper dispersion of nanomaterials in cementitious composites has been found to result in consequential creation of weak areas and voids in the cementitious matrix [10]. In order to propel more application of nanomaterials in enhancing the durability performance of cementitious composites, this overview was carried out to explore the effect of nanomaterials on the durability properties of cementitious composites. The durability performance of the cementitious composites was explored in terms of permeability and resistance to physical and chemical attacks. It is anticipated that this overview will be useful resources in understanding the effect of nanomaterials on cementitious composites and will propel more research and development in this field.

2 Influence of nanomaterials on durability

The durability of cementitious composites is its ability to resist the detrimental forces in the environment to which it is subjected. The durability performance of cementitious composites is dependent mostly on the ease and amount of penetration of deteriorating ions and fluids into the composite. Due to the ultrafine nature of nanomaterials, they can act as pore fillers between the grains of the cement thereby resulting in a more densified structure and higher resistance to the penetration of any deleterious materials. In addition, the pozzolanic reactivity of most nanomaterials makes them react with the calcium hydroxide in the pore system of the cementitious composites and produce more calcium silicate hydrate which will result in the densification of the composite's microstructure. Table 1 presents the effect of various types of nanomaterials used in cementitious composites on its corresponding performance. It can be seen from Table 1 that the incorporation of nanomaterials generally results in pore refinement and a corresponding lower permeability. Nonetheless, the incorporation of these nanomaterials also has other benefits such as the improvement of mechanical performance, self-cleaning, thermal performance improvements, etc. The effects of different types of nanomaterials on the durability performance of cementitious composites are discussed in the following sections.

2.1 Permeability properties

The permeability properties of cementitious composites are dependent on the corresponding pore structure. These properties indicate the ease at which various deleterious materials can penetrate the composite. Generally, the incorporation of nanomaterials has been found to reduce the permeability of the corresponding composite due to the pore refinement as a result of the filler ability of the nanomaterials alongside its pozzolanic reaction [19–21].

Table 1 Effect of nanomaterials on properties of cementitious composites

Type	Effects	References
NMT	Pore refinement, enhanced mechanical properties, self-sensing	[11]
NMK	Lower permeability	[12]
NS	Enhanced durability and low permeability	[13]
NT	High photocatalytic activity, self-cleaning, self disinfection	[14]
NCLy	Enhanced thermal properties and pore refinement	[15]
NA	Pore refinement and lower permeability	[16]
NCC	Accelerated hydration reaction and pore refinement	[17]
CNT	permeability reduction, resistance in an aggressive environment	[18]

CNT carbon nanotubes, *NA* nano alumina, *NCC* nano calcium carbonate, *NCLy* nano clay, *NMK* nano metakaolin, *NS* nano silica, *NT* nano titanium, *NMT* nano magnetite

2.1.1 Water absorption

Water absorption of cementitious composites is a good indication of its overall durability as most deleterious ions penetrate the composite using water as a pathway. The easier the water can penetrate cementitious composites, the higher its susceptibility to various durability threats. The water sorption of concrete has been found to be reduced significantly when nano titanium was used to replace cement up to 2% [22]. A similar study by Jalal et al. [23] also found out that the use of 2% nano silica resulted in an approximately 35% decrease in the water sorption of self-compacting concrete. The densification of the nanostructure and microstructure by the incorporation of up to 7% nano alumina into cementitious composites was found to reduce the sorption significantly [24]. The study by Ansari et al. [25] also showed that the use of nano silica and nano alumina resulted in a decrease in the water absorption of basalt fibre reinforced concrete mixtures. These observations are in agreement with the results reported by Behfarnia and Salemi [26] when nano silica and nano alumina were used as nanomaterials in concrete. Ehsani et al. [27] investigated the effect of nano silica on the characteristics of concrete mixtures. Results from the study showed a significant reduction in the concrete mixtures incorporating up to 1.5% nano silica alongside 3% micro silica. This observation corresponds to that of Diab et al. [12] where there was a reduction in the water absorption of concrete mixtures incorporating nano silica and nano metakaolin as shown in Fig. 1. It can be seen from Fig. 1 that the effect of both nano silica and nano metakaolin on the water absorption of the concrete mixture is similar. The lower water absorption in concrete incorporating these nanomaterials was also attributed to the pore filling effect of the nanomaterials coupled with enhancement of the interfacial transition zone between the aggregate and cement matrix. Similarly, Ji [28] reported a significant reduction in the water permeability of concrete incorporating nano silica as partial replacement of Portland cement. The decrease in the water permeability of concrete incorporating nano silica was attributed to the significant production of additional calcium silicate hydrates which results in refinement of the microstructure and a corresponding reduction in permeability. This observation strongly agrees with that of Kalhori et al. [29] where nano silica and nano clay were used to enhance the performance of shotcrete. However, results from the study indicate the use of nano silica is more effective in reducing the water absorption compared to when nano clay was used.

2.1.2 Chloride ion permeability

The use of nano magnetite (i.e. nano Fe_3O_4) up to 1.5% has been found to result in a significant reduction in the chloride ion penetration of cement pastes [30]. Li et al. [24] also reported a reduction in the chloride ion permeability of cementitious composites incorporating nano alumina. The reduction in the chloride ion penetration was attributed to the densification capability of the nano alumina. The study by Zhang and Li [20] showed that the chloride ion penetration of cementitious composites can be reduced with the incorporation of nanomaterials such as nano silica and nano titanium up to a dosage of 1%. At higher dosage above 1%, the effectiveness of the nanomaterials to reduce the chloride ion permeability was less effective. The reduction in

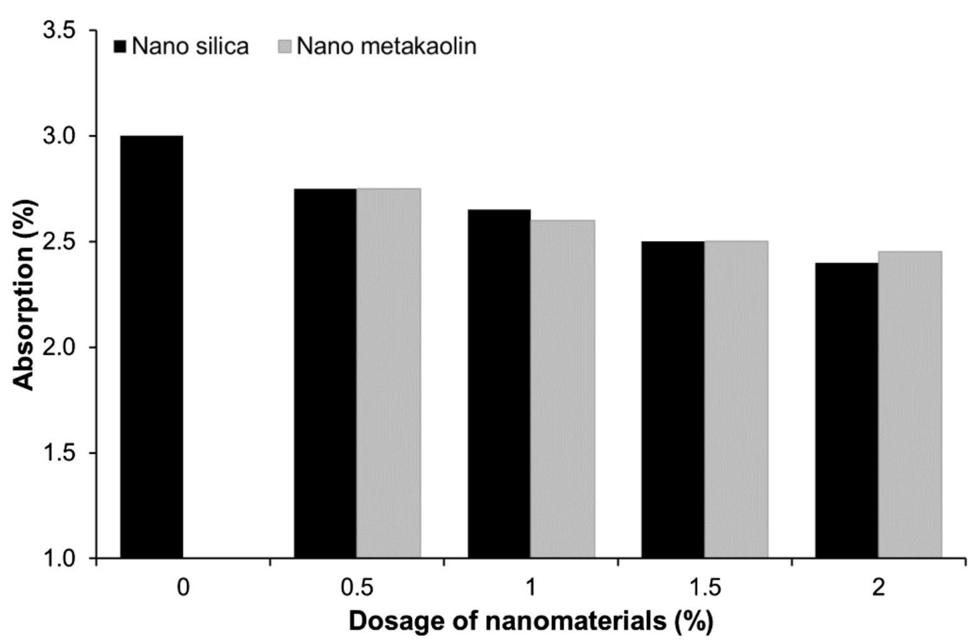

Fig. 1 Effect of nanomaterial dosage on absorption (adapted from [12])

the effectiveness of the nanomaterials at higher dosages can be associated with their poor dispersion which results in the accumulation of the nanomaterials and a consequential creation of voids in the matrix. Similarly, significant improvement in the chloride ion permeability was observed by Joshaghani et al. [31] when nanomaterials (i.e. nano titanium, nano alumina and nano maghemite) were incorporated at different dosages into self-compacting concrete. The enhancement in the chloride ion penetration with the incorporation of the nanomaterials was ascribed to the accelerating of the cement hydration coupled with the filler effect of the nanomaterials. However, compared to nano titanium and nano maghemite, the use of nano alumina resulted in higher improvement in the chloride ion penetration resistance of the composites.

2.1.3 Permeable voids

The incorporation of nanomaterials into cementitious composite will result in a reduction in the porosity (i.e. permeable voids) in the composite. Sikora et al. [30] showed that the incorporation of nano magnetite into mortar mixtures resulted in up to 6% reduction in the porosity. The reduction in the permeable voids with the incorporation of nano magnetite can be attributed to the filler effect of the nano magnetite coupled with accelerating the formation of hydration products which leads to the densification of the microstructure. The effect of different dosages of nano magnetite on the porosity of mortar is presented in Fig. 2. A reduction in the porosity of cementitious composites was also reported when nano silica was used as the replacement of cement up to 7% [24]. This observation is in agreement with that of Zhang et al. [32] where the incorporation of 2% nano silica resulted in a 27% reduction in the porosity of mortar. Similarly, Hakamy et al. [33] showed that the use of nano clay in hemp fabric reinforced composites reduced the porosity significantly. The decrease in the porosity of the composites was associated with the formation of more hydration products which results in the densification of the microstructure. However, the optimum dosage to achieve lower porosity was deemed to be 1% nano clay as there is a consequential increase in the porosity at dosages higher than 1%. This observation corresponds to that of Jo et al. [34] where they reported an optimum dosage of 1% nano silica to reduce the porosity of mortar. Zhang and Li [20] also reported that nano titanium can be used to reduce the pore content in fibre reinforced cementitious composite as shown in Fig. 3. It can be observed that in contrast to the effect of nano magnetite on the porosity (Fig. 2), the optimum dosage of nano titanium to reduce the porosity of cementitious composite is 1%. This indicates that preliminary tests need to be carried out to evaluate the optimum dosage of nanomaterials before its use on a large scale.

2.2 Chemical attack resistance

2.2.1 Acid resistance

Structures made with cementitious composites and buried under the ground are susceptible to the acid attack. The acidic ions can penetrate the cementitious composites through the surrounding soil or moisture in the environment. Though cementitious composites can be subjected to various forms of acid attacks depending on the type of acid,

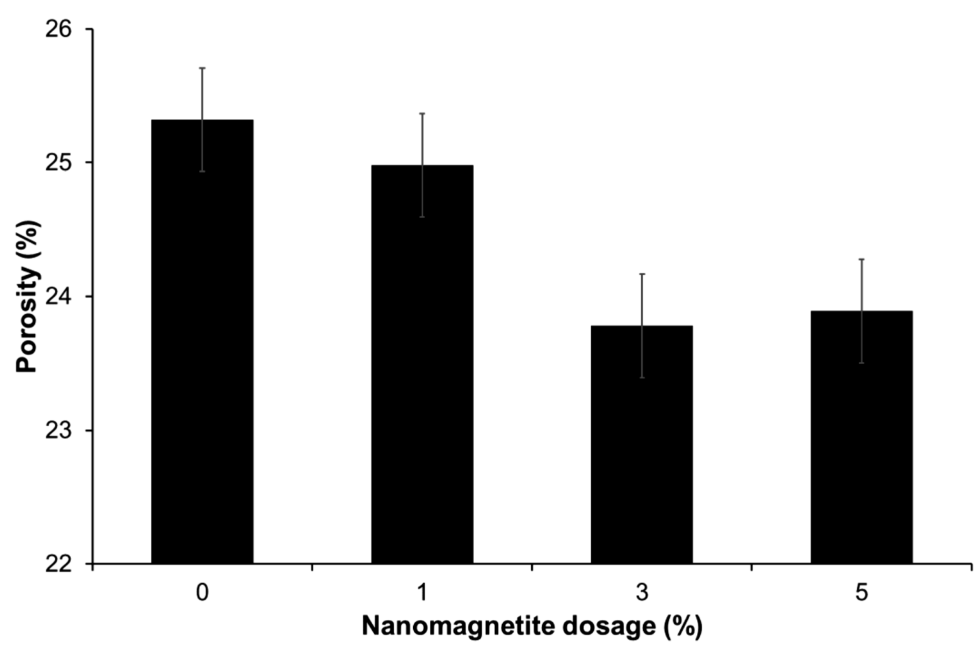

Fig. 2 Effect of nano magnetite dosage on porosity (adapted from [30])

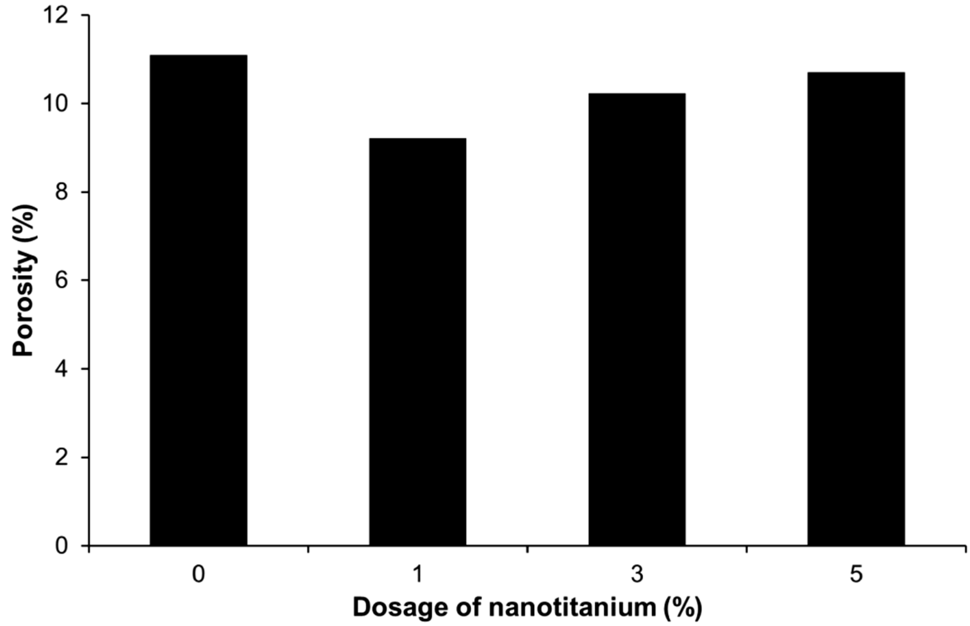

Fig. 3 Effect of nano titanium dosage on porosity (adapted from [20])

the mechanism of the acid attack is the same. Acid attack on cementitious composites results in deterioration of the composite due to the acid dissolving the hydration products leading to a weaker matrix and a corresponding exposure to other types of attacks. In contrast to the sulphate attack, deterioration of cementitious composites due to acid attack does not result in any notable expansions. Diab et al. [12] investigated the effect of nanomaterials on acid resistance of concrete. Results from the study showed that the resistance of concrete to acid attack can be enhanced significantly with the incorporation of nanomaterials such as nano silica and nano metakaolin. The enhancement in the resistance of the concrete mixtures to acid attack was evident in the lower loss in compressive strength and mass of samples subjected to nitric acid and sulphuric acid. The higher resistance of concrete mixtures incorporating nanomaterials was attributed majorly to pozzolanic contribution and pore-filling ability of the nanomaterials which inhibits the penetration of the acidic solution into the composite. This observation is similar to that of Senhadji et al. [35] and Chatveera and Lertattanaruk [36] where the incorporation of silica fume and rice husk ash respectively, resulted in improved resistance to acid attack.

2.2.2 Sulphate resistance

Sulphate attack is one of the critical durability threats on cementitious composites, especially those in constant contact with moisture and soil. Sulphate attack on cementitious composites results in the formation of ettringite and gypsum within the composite which results in expansion and cracks development with time. Once these cracks are formed, the cementitious composite is subjected to other forms of deteriorating forces in the environment it is subjected to. As mentioned earlier, the sulphate attack is quite different from the acid attack as sulphate attack results in both expansions in the composites and possible dissolution of the hydration products. Therefore, it is paramount to ensure that the structures made with cementitious composite have resistance against sulphate attack in order to conserve its service life. As one of the effective ways to enhance the resistance of cementitious composites to sulphate attack is by reducing its permeability, the use of nanomaterials is expected to be useful. Evaluation of the sulphate resistance of concrete mixtures incorporating nano silica and nano metakaolin was carried out by Diab et al. [12]. The study showed that concrete mixture incorporated with nanomaterials exhibited higher resistance to sulphate attack higher resistance as evident in the lower strength loss, expansion and weight loss of the samples used. However, it was reported that the use of nano silica was more effective than nano metakaolin in terms of the resistance of cementitious composites to sulphate attack. The effect of nano silica on the sulphate resistance of concrete mixtures in terms of the loss in compressive strength and expansions is presented in Fig. 4. These observations correspond to that of Ghafooori and Batilov [37] and Jiang and Niu [38] where the incorporation of nano silica was found to enhance the resistance of cementitious composites to sulphate attack. Saloma et al. [39] also reported a significant enhancement in the sulphate attack resistance of concrete mixtures incorporating nano silica as a 10% replacement of Portland cement. The enhancement in the sulphate resistance of concrete mixtures incorporating nano silica was attributed to the formation of secondary products

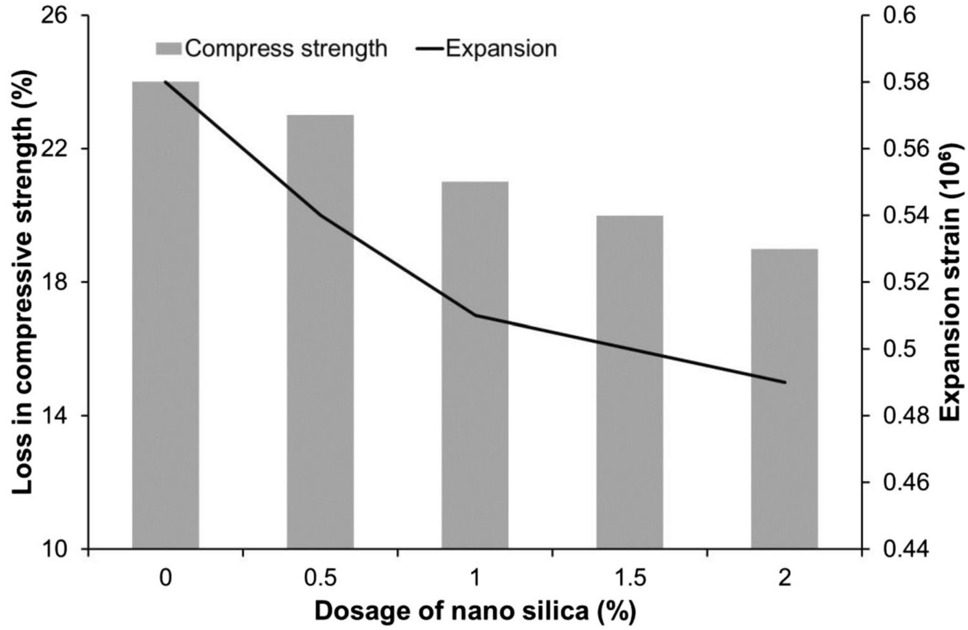

Fig. 4 Effect of nanosilica dosage on sulphate attack resistance (adapted from [12])

in the matrix due to the reaction of the nano silica with calcium hydroxide. The formation of these secondary products results in the densification of the microstructure and reduction in the calcium aluminate content of the concrete.

2.3 Physical attack resistance

2.3.1 Freeze and thaw

The incorporation of nano silica into concrete has been reported to enhance its resistance to deterioration from freeze and thaw cycles [40]. The enhancement of the resistance of concrete incorporating nano silica to freeze and thaw cycles was attributed to the additional formation of hydration products which densified the concrete and makes it less permeable to water. The study also reported a significant enhancement of the concrete to scaling due to deicers. These observations are in agreement with Behfarnia and Salemi [26] where concrete samples incorporating nano silica and nano alumina were found to have a higher resistance to freeze and thaw cycles. The percentage strength loss and reduction in length of samples incorporating nano silica and nano alumina after 300 cycles of freeze and thaw are presented in Fig. 5. As mentioned earlier, it can be seen from Fig. 5 that the optimum dosage for each type of nanomaterial varies. Kalhori et al. [29] also reported a significant enhancement in the freeze–thaw resistance of shotcrete mixtures with the incorporation of nano silica and nano clay. The findings from the study showed that shotcrete mixtures incorporating nanomaterials (i.e. nano silica and nano clay) exhibited a lower reduction in the compressive strength after subjecting to freeze–thaw cycles. The enhancement of the freeze–thaw resistance of the shotcrete with the incorporation of the nanomaterials was associated with the refinement of the composite's microstructure by the nanomaterials. However, the use of nano silica resulted in higher freeze–thaw resistance compared to when nano clay was used.

2.3.2 Abrasion

Cementitious composites used for applications such as pavements, flooring, etc., are subjected to constant moving load resulting in a corresponding abrasion of the surface. In order to ensure these types of infrastructures perform well, they must have good resistance to abrasion which is also a durability property. Abrasion performance of cementitious composites has been found to be related to the composition of the composites coupled with the finishing and curing method employed. The study by Li et al. [41] showed that concrete mixtures enhanced by incorporating nano silica have a higher resistance to abrasion compared to mixtures without nanomaterials. A similar observation was reported by Li et al. [24] where the found out that concrete mixtures modified with nano titanium have a higher resistance to abrasion compared to the mixtures incorporating nano silica at the same dosage as shown in Fig. 6. However, this resistance to abrasion as been found to reduce with an increase in the water to binder ratio of the composite and higher dosage of the nanomaterials. Nonetheless, it was concluded that the incorporation of nanomaterials into the concrete was more beneficial in increasing the abrasion resistance than the incorporation of fibres [41].

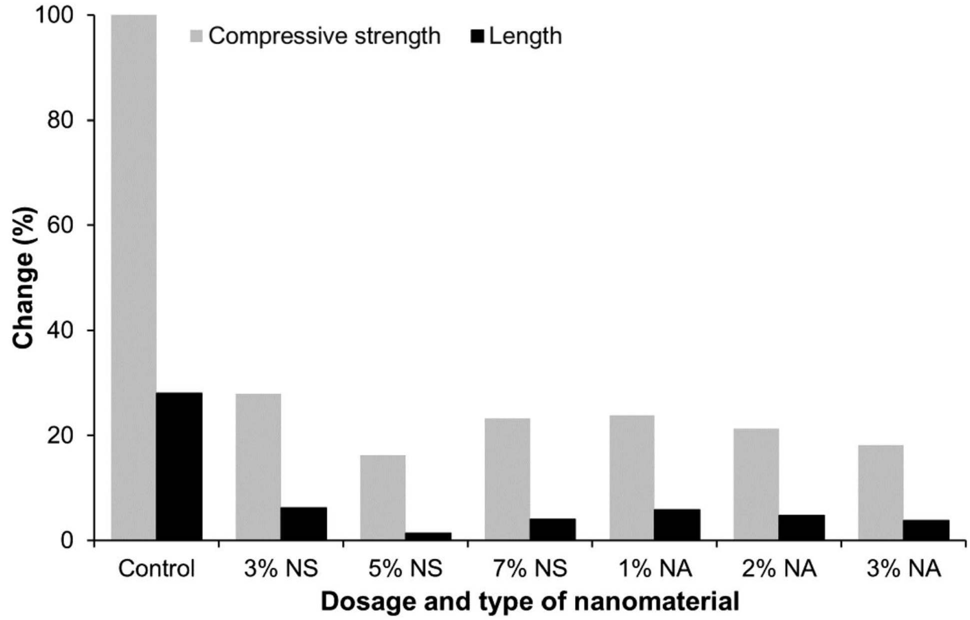

Fig. 5 Effect of dosage and type of nanomaterial on freeze and thaw adapted from [26]))

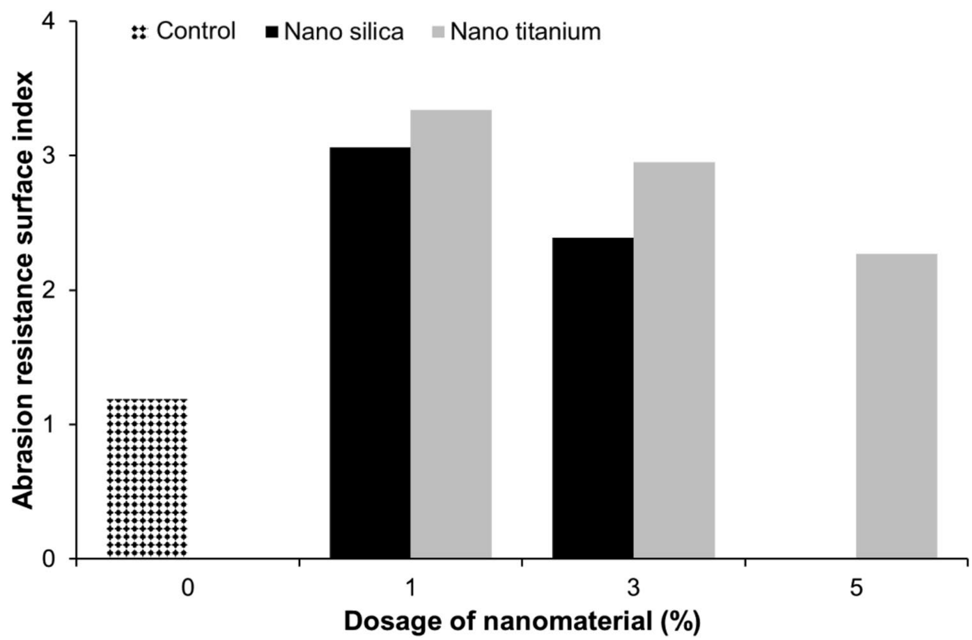

Fig. 6 Effect of dosage and type of nanomaterial on abrasion (adapted from [24])

2.3.3 Elevated temperature

There is limited study on the elevated temperature resistance of cementitious composites incorporating nanomaterials a. Nonetheless, cement paste incorporating nano silica has been found to have a higher resistance to elevated temperature as high as 500 °C. Ibrahim et al. [42] reported an increase in the mechanical performance of cement pastes subjected to elevated temperature in the range of 400 to 700 °C. This observation is in agreement with that of Lim and Mondal [43]. The study by Guler et al. [44] also concluded that the binary use of nano silica and nano alumina at a dosage of 1.5% is optimum to enhance the stability of concrete at elevated temperatures. However, with the limited study on the behaviour of cementitious composites incorporating nanomaterials in an elevated temperature environment, more research and development is imminent.

3 Conclusion

This paper presents an overview of the effect of various types of nanomaterials on the durability properties of cementitious composites. Generally, the incorporation of nanomaterials into cementitious composite will result in the enhancement of its durability properties due to the reduction in the permeability of the composites. The reduction in the permeability of the cementitious composites with the incorporation of nanomaterials can be attributed to the pore filling effects, pozzolanic effects and reactivity of the nanomaterials. However, it is necessary to determine the optimum dosage of nanomaterials to be used as there could be a consequential effect of the nanomaterials on the performance of cementitious composites when used at a higher dosage. Cementitious composites incorporating nano silica exhibited lower permeability properties compared to other types of nanomaterials due to their pozzolanic characteristics coupled with their ability to act as nucleation sites for cement hydration. However, in terms of resistance to physical attacks, cementitious composites with nano titanium exhibited higher abrasion resistance compared to their nano silica counterpart. As most studies on the effect of nanomaterials are focused on permeability properties, it is recommended that future study should also explore the effect of nanomaterials on the resistance of cementitious composites to physical attacks such as weathering and elevated temperatures.

Funding None.

Compliance with ethical standards

Conflict of interest None.

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