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Review Article

A review of the effect and optimization of use of nano-TiO₂ in cementitious composites

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Abstract

There are some problems and weaknesses related to cement-based materials, such as their very low tensile strength, low chemical resistance and the huge contribution of cement production to industrial CO₂ emissions. One possible method to reduce the impacts of such problems is the partial replacement of cement in cementitious materials with nano materials. This work provides a detailed review of incorporation of one of the most widely used nano materials, namely nano-titanium dioxide, and its effect on the properties of cementitious composites. Different properties have been considered in the current study, such as fresh properties, mechanical properties (compressive strength, split tensile strength and flexural strength), durability (permeability, ultrasonic pulse velocity (UPV), electrical resistivity, carbonation resistance, freeze and thaw resistance and sulfate attack resistance) and microstructural properties. This paper also investigates the optimum content of nano-TiO₂ in cement-based materials. Moreover, the cost effectiveness of use on nano-titania in cementitious composites has been discussed. Nano titania reduces the workability and setting time of cement-based materials. It can be very effective in improving the mechanical properties, durability and microstructural properties of cementitious composites.

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1. Introduction

Cement-based materials (i.e., cement paste, mortar and concrete) are the most widely and commonly used construction materials for different types of infrastructure. These cementitious composites are quasi-brittle and vulnerable to cracking, and possess low tensile strength and low chemical resistance, and have no functional properties as structural materials [1–3]. Moreover, cement industry is one of the main contributors to industrial CO₂ emissions. It is responsible for 5% to 8% of global anthropogenic CO₂ emissions [4–11]. One method to overcome these problems is by the incorporation of nanomaterials into cement-based materials. This solution can lead to the production of high performance cementitious composites with novel properties [1,3,12–17].

One of the most commonly utilized nano-additives in cement-based materials is nano titania (nano-TiO₂ or NT) [18–20]. Titanium dioxide is a noncombustible and odorless powder that has been produced abundantly and used frequently in various productions [21–24] because of its high chemical stability, non-toxicity, low cost, anticorrosion, electrical and superior photocatalytic property [22,23,25–27]. It exists in three different phases; brookite, rutile and anatase [23,24,27–29]. Although most of the TiO₂ used up to date was not in nanosize state, trends to use titania nanoparticles has increased significantly and is expected to even exceed the use of conventional titanium dioxide in the few following years [26]. Compared to conventional TiO₂, NT experiences a 500% increase in surface area [30]. It also can be available in extremely pure form ($\geq 99.9\%$) [11,31–35].

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This paper reviews the influence of use of titania nanoparticles on different properties of cement-based materials. The properties researched in this work are fresh properties, mechanical properties (compressive strength, split tensile strength and flexural strength), durability and microstructural properties. In addition, a guideline for the optimum content of nano-TiO₂ in cementitious materials is provided. The cost effectiveness of use of this nano material is also discussed.

2. Influence of Incorporation of Nano-Titania into Cementitious Composites

The effect of incorporating nano-titania into cement-based materials has been reported by many researchers. Due to its photocatalysis capacity, nano-TiO₂ can impart cementitious materials with new functionalities, such as self-cleaning, anti-microbial and smog-abating characteristics [11,14,25,36–44]. In addition, application of nano-TiO₂ photocatalysis on cement-based materials in urbanized areas can lead to air pollution reduction and air quality improvement in these areas [39,45–47]. It has been found that concentration of organic, such as volatile organic compounds, and inorganic pollutants, such as NO_x and SO₂, can be reduced with the use of nano-TiO₂ [19,29,36,38–40,44,45,47–50]. Such functionalities have the potential to improve the sustainability of construction materials [45]. Furthermore, despite being chemically inert in terms of its potential to directly react during cement hydration [19,24,40,44,45], nano-titanium dioxide can generally improve the mechanical performance and durability, as well as decrease the rigidity of cementitious materials [20,28,31,35,37,44,51–56]. Moreover, the flexural fatigue performance and abrasion resistance of concretes was reported to be remarkably improved with the use of TiO₂ nanoparticles [31]. Gain in strength in this case might be related to the microstructural modification and the hydration acceleration effects of nano-TiO₂ by providing additional surface area for product nucleation (i.e. the boundary nucleation effect) [20,31,35,40,44]. Addition of TiO₂ nanoparticles leads to greater homogeneity, better compaction, reduction in the pore volume and the pore size of cementitious materials, which results in remarkable reduction in permeability [20,31,35,44,56–59]. Titania nanoparticles has been also found to behave as an activator to accelerate pozzolanic reaction, to increase the rate of cement hydration, to increase the intensity of the heat peak, and to reduce the initial and final setting time [14,24,28,37,44,45,52,58,60,61]. The reduction in the setting time was found to be proportional to the content of the added nano-TiO₂ [20,40,52,58]. Reduction in setting time could lead to reduction in total time of construction which is economically beneficial [45]. Incorporation of TiO₂ nanoparticles can result in reduction in the water loss of cementitious materials and increase in the hydrophilicity of the pastes leading to mitigation in drying shrinkage [20].

On the other hand, like other nano-size materials, nano-titanium dioxide has some drawbacks that might limit the large-scale commercial applications. There are many concerns about the effect of titania nanoparticles on human wellbeing. Some reports have showed that nano-titania exhibits concerned cytotoxicity in different cultured cell models. TiO₂ nanoparticles have been also found to possess significant genotoxic activity in mammalian cells [21]. TiO₂ nanoparticles exposure has negative effects on the respiratory system or the metabolic circle system of organisms and on cellular function of human dermal fibroblasts [22]. Huang et al. [21] reported that long-term exposure to these particles affect cell cycle progression and duplicated genome segregation, resulting in cell transformation and chromosomal instability. Furthermore, nano-TiO₂ particles were observed to have the tendency to agglomerate [44,45,62], and hence an additional deagglomeration step is necessary when separate nano-TiO₂ particles are required [25]. Moreover, due to its high specific surface area (SSA), nano-titania reduces the workability of the mixture. Such reduction gets greater with increase of nano-TiO₂ content in the

mixture [5,20,51,55,63,64]. Although much research has been done related to the incorporation of titania nanoparticles into cementitious materials, there is still big lack of information, which necessitates further investigation. There are only few published reports related to the effect of nano-titania on different properties of hardened cement-based materials [5,20]. While some research works, such as [5,20,31,34,65,66], reported that of TiO₂ nanoparticles increases the compressive strength of cementitious materials, some others, such as [67], stated that it leads to reduction in the compressive strength. In addition, it is not fully understandable yet what is the best way to use the titania photocatalyst in cement-based materials [39].

3. Effect of Nano-TiO₂ on Different Properties of Cementitious Composites

In this section, the influence of use of NT in cementitious composites on their fresh properties, mechanical properties and durability is illustrated. Furthermore, the microstructural properties of nano-titania incorporated mortar and concrete are investigated by studying some SEM images examples. Since the final performance of cementitious composites is directly related to their strength and permeability [31,68], the focus in this section will be on the influence of nano-TiO₂ on the mechanical properties and permeability of cement mortar and concrete.

3.1. Effect on Fresh Properties

The fresh properties of cement-based materials are affected by the distribution of the particle size of its constituents. For this reason, nano materials can significantly change the fresh properties of the cementitious composites due to their nano size [55]. Reports that have discussed the effect of NT on fresh properties of cementitious composites show that NT can remarkably decrease the workability. Casagrande et al. [51] investigated the effect of NT on the workability of mortar using spread on table test. The results revealed that NT could remarkably decrease the workability of mortar. The results showed that in general as the content of NT increased the spread on table decreased. This was attributed to the high specific surface area of nanoparticles that adsorbs part of the water and superplasticizer. Gopalakrishnan et al. [52] reported a reverse relationship between the content of NT and flow of mortars. Joshaghani et al. [35] tested the effect of 3.0 and 5.0 wt% (by weight of the binder) nano-TiO₂ on the fresh properties of self-compacting concrete using slump flow, L-box, V-funnel and column segregation tests. The results indicated that while 3.0 wt% NT slightly increased the workability properties of concrete, 5.0 wt% resulted in a reduction in the workability. This was indicated to the higher water demand due to higher content of NT, which consequently reduced the workability. Siang Ng et al. [55] evaluated the workability of mortar incorporating 1.0, 3.0 and 5.0 wt% NT and 30 wt% fly ash using hydraulic flow. The results revealed that up to the optimum content (3.0 wt%), no significant effect of NT on the workability was observed. However, exceeding the optimum content led to a reduction in the workability of around 11%. Shaaban et al. [69] stated that NT inclusion in mortars dramatically decreased their workability and made the mixture sticky and dry as NT content increased. Dantas et al. [62] reported that the flow depended on the type of nano-titania incorporated and on the dispersion technique used to disperse the nanoparticles in the mixture. They reported that using NT in a dispersed form was more effective in controlling the fresh properties of mortar than the undispersed form. Patel and Mishra [28] reported a slight reduction in the workability, which was evaluated by slump test, due to the use of NT. Rao et al. [70] evaluated the workability of self-compacting mortars incorporating 30 wt% fly ash and 0.5-1.0 wt % NT using mini slump flow and mini V-funnel tests. The results showed a decrease in the workability due to the use of NT with a less workability for higher content of NT. Zhang et al. [20] reported a reduction in the fluidity of mortar due to the use of titania nanoparticles. The higher the content of NT the higher the reduction in fluidity. They found

that 5.0 wt% NT reduced the slump flow by around 21%. Salemi et al. [71] reported that 2.0 wt% NT reduced the slump of concrete by 50%. Noorvand et al. [5] studied the effect of a combination of untreated black rice husk ash and NT on the flowability of mortars. They reported a decreased in the flowability due to the use on NT when incorporated with rice husk. However, when the content of rice husk was increased to 30 wt%, NT was observed to enhance the flowability compared to mortar without NT. This was attributed to the effect of NT in filling up the surface pores of rice husk resulting in a less porous materials, which required a less mixing water.

NT has been also reported to reduce the setting time of cement-based materials. This reduction has been reported to increase as the content of NT increased, due to higher hydration rate [52,58]. Gopalakrishnan et al. [52] reported that NT remarkably reduced the initial and final setting time proportionally to its content. It was found that 10 wt% NT reduced the initial and final setting time by 10 and 15 min, respectively. Daniyal et al. [60] revealed that nano-TiO₂ decreased the setting time of cementitious composites due to its role as an accelerator. Wang et al. [58] studied the setting time of cement pastes incorporating 1.0-5.0 wt% NT cured at different temperatures (0-20°C). Setting time was found to decrease with the increase in the curing temperature and with the increase in content of NT. Similarly, Zhang et al. [20] found that while 1.0 wt% NT reduced the initial and final setting time by 37.9% and 15.7%, respectively, 5.0 wt% reduced them by 76.5% and 46.2%, respectively.

3.2. Effect on Mechanical Properties

Although nano-TiO₂ is considered to be one of most widely researched nano-additives, its effect on mechanical properties of cementitious materials has not been sufficiently reported. Some of the reported data related to the effect of NT on the compressive strength, split tensile strength and flexural strength of mortar and concrete are shown in Table 1. The table also presents some of the factors that have impact on the behavior of nano-TiO₂ based modified cementitious composites, such as NT particles size, SSA etc. The optimum content found in each reference and the improvement percentage in the 28-day strength due to the incorporation of such content are also shown in the table. The discussed data has been arranged as follows: First the effect on concretes has been discussed and then the effect on mortars. For concretes, first plain concrete then self-compacting concrete (SCC) and after that concrete incorporating other additives such as ground granulated blast furnace slag (GGBFS), fly ash (FA) and rice husk ash (RHA) have been studied. The same organization method has been also followed for mortars.

From the table it can be said that generally NT has the potential to improve different mechanical properties of concrete and mortar with even a small content. However, there is a lack of data and there are some inconsistencies between some results which emphasize the importance of conducting more research related to this topic. The effect of NT on the mechanical properties increases with the increase in its content in the mixture up to some point (optimum content) and then starts to decrease. As can be seen from Table 1, generally, the same optimum content for all the mechanical properties has been reported in different references.

Table 1. Optimum contents of nano-titania in cementitious composites and improvement percentages of the mechanical properties

Ref.	Mortar/ concrete	NT properties				Optimum content (wt%)	Enhancement in 28-day strength (%)		
		Size (nm)	SSA (m ² / g)	Phase	Invest. contents (wt%) (by weight of the binder)		Comp. strength	Split Tensile strength	Flexural strength
[5]	mortar ⁽¹⁾	15	240	anatase	0.5, 1, 1.5	1.5	19.1	-	-
[20]	mortar	25	-	-	1, 3, 5	5	21	-	-
[28]	concrete	15-30	-	anatase	0.5, 1, 1.5	1	8.2	-	8
[31]	concrete (self-compacting) ⁽²⁾	15	155	-	1, 2, 3, 4	3	36.4	33	27.8
[32]	concrete	15	150	-	0.5, 1, 1.5, 2	-	-	-	-
[34]	concrete (self-compacting)	20	165	-	1, 2, 3, 4, 5	4	22	36.1	-
[52]	mortar	15±2	153±10	anatase	2, 4, 6, 8, 10	10	30.65	-	-
[55]	mortar ⁽³⁾	15	60	-	1, 3, 5	3	36	-	11
[57]	concrete ⁽⁴⁾	25 ± 5	-	anatase	1, 2, 3	2	17	-	-
[58]	mortar	15	200	-	1, 2, 3, 4, 5	2	4	-	51
[60]	mortar	30	-	-	1, 3, 5	5	11.67	-	-
[64]	concrete	50-200	60	anatase	0.5, 0.75, 1, 1.25, 1.5	1	85	-	-
[65]	mortar (self-compacting) ⁽⁵⁾	15	200	-	1, 3, 5	5	23	-	-
[66]	mortar (self-compacting) ⁽⁶⁾	15	200	-	1, 3, 5	5	16	-	-
[67]	concrete	20	40	rutile	1, 2, 3, 4, 5	3	-16	-	-
[72]	concrete	15	155	-	0.5, 1, 1.5, 2	1	17.9	-	-
[73]	concrete	15	240	anatase	1, 3, 5	1	18.03	-	10.28
[74]	mortar	21	58.8	anatase	5, 10	10	10	-	-
[75]	mortar ⁽⁷⁾	21	35-65	-	1, 2, 3,	2	16	-	-
[76]	concrete (self-compacting)	20	165	-	1, 2, 3, 4, 5	4	-	-	44

[77]	concrete	<10 0	-	-	1, 2, 3, 4, 5	1	18	-	-
[78]	mortar	-	50	-	1, 2, 3, 4, 5	3	-	68.15	61.1
[79]	mortar concrete	21	-	-	1, 2, 3,	3	11	-	-
[80]	(self- compactin g)	<25	50	-	0.5, 2, 4	2	3	-	(NT 0.5%) 5
[81]	mortar	10- 30	50- 150	-	0.5, 1, 1.5, 3	1.5	33	40	-

Notes:

- (1) Black rice husk ash was used by 35 wt% of cement in all mixtures.
- (2) Ground granulated blast furnace slag (GGBFS) was used by 45 wt% of the cement in all mixtures.
- (3) Incorporating 30% FA ash by weight of cement in all mixtures.
- (4) Recycled aggregate concrete.
- (5) Incorporating 0-15% rice husk ash (RHA) by weight of the total binder in all mixtures.
- (6) FA was used by 25 wt% of cement in all mixtures.
- (7) SF was used by 5.0 wt% of cement in all mixtures.

3.2.1 Effect of NT on Compressive Strength

The influence of nano-titania on the compressive strength of concrete and mortar has been studied by some researchers. The results have shown that NT can have a very good effect on the compressive of such materials. Nazari et al. [72] conducted an experimental study on nano-titania effect on the compressive strength of plain concrete. Their results demonstrated that NT had a good effect on the compressive strength of concrete. Maximum enhancement of around 18% in strength was observed with 1.0 wt% nano-TiO₂. Nazari et al. reported that the effect of NT on compressive strength got reduced with higher content, however, NT up to 2.0 wt% still had better performance than that of the plain concrete. These findings are consistent with what Zhang and Li [73] reported. However, Zhang and Li studied contents of NT up to 5.0 wt% and reported that such content didn't show any remarkable influence on the strength compared to the plain concrete. On the other hand, the findings of Behfarnia et al. [67] contradict the above findings. They stated that incorporation of NT into plain concrete led to reduction in the strength of plain concrete compared to mixes without NT. Their results showed that the least reduction in 28-day compressive strength was -16% and was observed in samples incorporating 3.0 wt% nano-TiO₂, while the maximum reduction was around -27% and was observed for 2.0 wt% nano-TiO₂. They ascribed this to the negative effect of titania nanoparticles on dicalcium silicate (C₂S) hydration which contributes to properties of hydrated concrete at later ages. Some other scholars have investigated NT effect on the compressive strength of SCC. Jalal et al. [34] found that the use of NT increased the compressive strength of SCC at all studied ages and for all investigated contents of NT. The maximum enhancement in the compressive strength was observed for 4.0 wt% NT content. This content led to an increase of 22.5%, 22% and 26.9% in the 7, 28 and 90-day compressive strength, respectively.

NT effect on the compressive strength of mortar has been also investigated by some researchers. Chen et al. [74] reported that higher content of NT (10 wt%) led to negligibly higher performance compared to lower content (5.0 wt%). On the other hand, [5,20,65,66,75] reported small content of NT (≤5.0 wt%) to be the optimum content. This content was observed to improve the strength by more than 15%. Mohseni et al. [66], who conducted an experimental study on the effect NT on the compressive strength of self-compacting mortar incorporating a constant amount of fly ash (FA), demonstrated that NT increased the strength at all ages and for all studied replacement percentages. The best results were observed with 5.0 wt% replacement percentage, which caused an

improvement of about 28%, 16% and 23% in the compressive strength in the 7th, 28th and 90th curing days, respectively.

As stated before, enhancement in strength in this case could be attributed to the fact that titania nanoparticles enhance the microstructure properties by improving the homogeneity, enhancing the compaction, reducing the pore volume and size of cement-based materials. It might also be related to the hydration acceleration effect of NT through increasing the surface area for product nucleation [20,31,32,35,40,44,52,56,60,63,72,81]. It has to be noted that while some papers, such as [31,32,72] stated that TiO₂ nanoparticles lead to more formation of hydrated products, some others, such as [19,40,45,63,67,74,79,82] stated that NT is chemically inert and hence does not increase the amount of hydration products. On the other hand, the observed reduction in the compressive strength by increasing the content of nano-TiO₂ more than the optimum content may be related to the agglomeration and bad dispersion of nanoparticles that lead to weak zones [31,34,44,55,72,78,81].

Trying to find the optimum content of NT in cementitious materials to improve their mechanical properties is hard, due to the lack of information related to its effect. However, to have an idea about the range of the optimum content of NT in concrete and mortar to improve their mechanical properties, we can compare the collected data from different publications, regardless of the different characteristics found in each one of them. Since the most investigated mechanical property is the compressive strength and to make the comparison much easier, the authors presented the data related to the compressive strength graphically in Fig. 1. Comparing the results presented in Table 1 and Fig. 1, we can roughly say that the optimum content of NT to enhance the mechanical properties of mortar and concrete is 1.0-5.0 wt%.

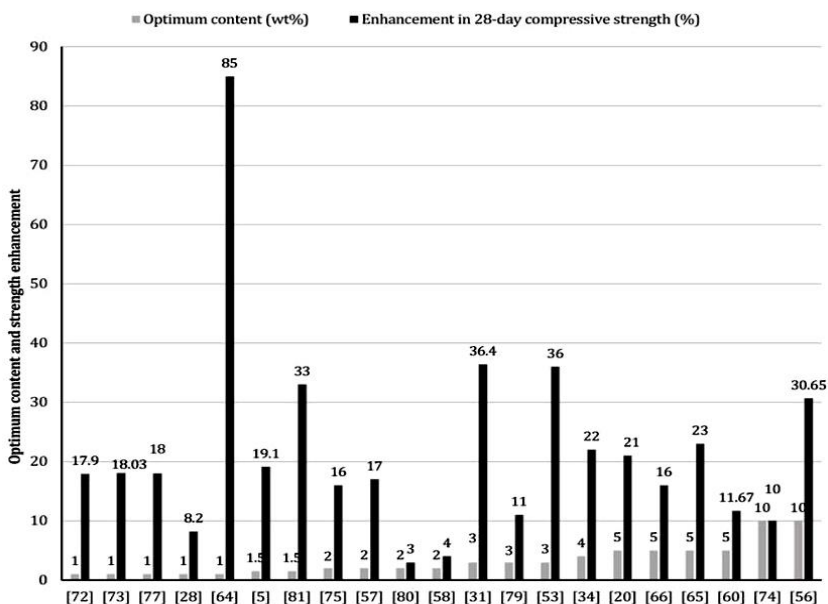


Fig. 1 Different optimum contents of NT to improve the compressive strength of cementitious composites and their enhancement percentages

3.2.2 Effect of NT on Split Tensile Strength

Only few reports have been published until now on the effect of NT on the split tensile strength of concrete and mortar. Jalal et al. [34] studied the split tensile strength of SCC

incorporating titania nanoparticles. They found that incorporating NT into SCC greatly improved its split tensile strength at all ages and for all content of NT. The best performance was observed for 4.0 wt% NT, which improved the split tensile strength by 34.5%, 36.1% and 46.2% after 7, 28 and 90 curing days, respectively. These results were supported by the findings of Nazari and Riahi [31], who evaluated the effect of NT on the properties of SCC incorporating 45% of GGBFS by weight of cement. Nazari and Riahi demonstrated that replacement of cement with 3.0 wt% NT led to approximately 80%, 33% and 38% increment in the 7, 28 and 90-day strength, respectively. They also showed that although higher content of NT (up to 4.0 wt%) showed less effect, it was still remarkably higher than that of the reference specimen. The split tensile strength of mortar incorporating NT was researched by Ma et al. [78], who showed that NT incorporating samples had importantly higher split tensile strength than the reference sample. They also reported that 3.0 wt% NT, which presented the optimum content in their research, could enhance the split tensile strength of mortar by 68.15%. These findings prove that titania nanoparticles have the potential to overcome some of the problems related to cementitious materials, such as possessing small tensile strength. However, since there is great lack of reported data related to this topic, more research is of a great importance to confirm such results.

3.2.3 Effect of NT on Flexural Strength

Similar to the split tensile strength, only few reports on the effect of nano-titania on the flexural strength of concrete and mortar have been published. Zhang and Li [73] and Patel and Mishra [28] studied the influence of incorporating NT on the flexural strength of concrete. They concluded that the best performance occurred when a small content (1.0 wt%) of NT was used. Their results showed that this content increased the flexural strength by 10.3% and 8%, respectively. It was observed that higher content of NT caused reduction in its effect on the flexural strength. Zhang and Li [73] stated that 5.0 wt% NT content caused a reverse effect on the flexural strength and that a reduction of around -3% was observed. On the other hand, Patel and Mishra [28] stated that 0.5 wt% NT led to less flexural strength compared to the plain concrete. Jalal et al. [76] studied strength development of SCC incorporating NT. The results showed that incorporation of NT up to 4.0 wt% by weight of cement improved the flexural strength significantly. However, for higher content of NT the flexural strength started to decrease. This behavior might be ascribed to the same reasons stated in the case of compressive strength. Jalal et al. reported an improvement of around 32%, 44% and 33% in the 7, 28 and 90-day strength, respectively, compared to the reference samples due to the addition of 4.0 wt% NT. It was observed that mixtures with 5.0 wt% NT, although had less flexural strength compared to those with 4.0 wt% NT, still had higher strength compared to mixtures with 1.0, 2.0 and 3.0 wt% NT. The effect of NT on flexural strength of mortar was investigated by Ma et al. [78]. They demonstrated that NT could greatly improve the flexural strength of mortar. They showed that 3.0 wt% NT had better performance than that of the other content percentages. This content was reported to cause 61.1% increment in the flexural strength compared to the control sample. Although more research is needed, it can be said from the results above that use of titania nanoparticles is a suitable solution to increase the flexural strength of mortar and concrete.

3.3. Effect on Durability

Reports on the effect of NT on the durability of cementitious materials have revealed that NT importantly improves the durability of cement-based materials. Use of nanoparticle has been found to lead to a denser and more homogenous microstructure due to their role as nano fillers and as nucleation sites due to their high surface area. This leads to smaller pore sizes and as a result to a lower permeability and a higher durability [35,65,83,84].

However, higher content of NT has been reported to lead to a reverse effect on the durability [85,86]. In this section, the effect of NT on durability is evaluated using some of the commonly investigated durability parameters; namely, permeability, ultrasonic pulse velocity, electrical resistivity, carbonation resistance, freeze and thaw resistance and sulfate attack resistance.

3.3.1 Effect on Permeability

Permeability is considered to be one of the primary factors that influence the durability and performance of concrete and other cementitious materials. It greatly affects the service life of concrete structures and accordingly their life-cycle costs [18,73,87,88]. The most critical factor in corrosion of the reinforcement steel in concrete members is the ingress of deleterious ions into the concrete. Therefore, by reducing the permeability of concrete, especially the outer layer, penetration of such ions can be prevented [18]. One way to significantly reduce the permeability of cement-based materials is through the incorporation of nano-additives, such as nano-TiO₂. Nanoparticles can improve the microstructure, reduce the porosity, increase the density and consequently reduce the permeability of cementitious materials [6,17,35,38,52,58,59,67,87–91]. As a result, they will significantly improve their durability, and thus improve the health of structures [1,92–95].

Incorporation of nano-titania into cement-based materials can importantly reduce their permeability [31,32,34,59,67,73,76]. This can be related to the high action and filler effect of NT particles, which result in refining the pore structures [34,66,67,76]. Titanium dioxide nanoparticles have been also observed to enhance the interfacial transition zone (ITZ) in concrete [31–34,76]. It has been found from previous works that permeability of cementitious composites decreases with the increase in the content of NT in the mixture up to the optimum content and then starts to increase. It should be noted that the optimum content of NT was observed to be mostly the same for both improvement of mechanical properties and reduction of permeability in different references.

Table 2. Optimum contents of nano-TiO₂ in cementitious composites for permeability reduction and the related reduction percentages

Ref.	Investigated contents (wt%)	NT optimum content (wt%)	Decrease percentage (%)			
			Water absorption	Chloride permeability	Capillary water absorption	Gas permeability coefficient
[31]	1, 2, 3, 4	3	45.7	-	-	-
[32]	0.5, 1, 1.5, 2	0.5	59.1	-	-	-
[57]	1, 2, 3	2	-	33	-	-
[65]	1, 3, 5	5	18	59	-	-
[66]	1, 3, 5	5	10	59	-	-
[67]	1, 2, 3, 4, 5	4	-	-	-	81
[73]	1, 3, 5	1	-	31	-	-
[75]	1, 2, 3	2	-	-	-	4
[76]	1, 2, 3, 4, 5	4	30.4	47.9	20.7	-
[81]	0.5, 1, 1.5, 3	1.5	17	-	-	-

The collected data related to the effect of NT on different parameters of permeability (water absorption, chloride permeability, capillary water absorption and gas permeability coefficient) of concrete and mortar is presented on Table 2. The table also shows the optimum usage percentages of NT in the mixtures to reduce such characteristics and the

resulted reduction percentage. It should be mentioned that the properties of NT in each related reference are as given in Table 1. It can be observed from the table that the same optimum content of NT has been reported for the different parameters of permeability. It can also be noticed that the most investigated parameters of permeability are water absorption and chloride permeability. However, there is still a big lack of information related to NT effect on the permeability of cementitious composites. Thus, more research is required to overcome such a problem.

Some researchers have evaluated the permeability of NT incorporating concrete and mortar by testing the percentage of water absorption of such materials. Water absorption is one of the main parameters for studying the durability of cementitious composites [96]. It gives an indication about the porosity volume in the final product [88]. Nazari and Riahi [32] carried out an experimental study on the NT effect on the water absorption of plain concrete. They found that small amount of NT led to higher resistance to water absorption. The maximum reduction in the water permeability, which was 98.7%, 59.1% and 81.7% in the 7th, 28th and 90th curing days, respectively, was observed when cement was replaced with NT by 0.5 wt%. Their results showed that although higher content of NT led to increase in the water absorption compared to 0.5 wt% NT, incorporation of NT up to 2.0 wt% still had less water absorption compared to the reference concrete. Jalal et al. [76] researched the effect of NT on the water absorption of SCC. Their investigation demonstrated that 90-day water absorption got decreased up to 30.4% by addition of NT up to 4.0 wt% and then started to increase. According to them, the reason for that might be the agglomeration and inappropriate dispersion of the nanoparticles in the mixture and the weakening of the pore structure due to high replacement of the binder with a filler material.

The effect of NT on the water absorption of mortar has been also studied by some scholars. Mohseni et al. [66] evaluated the changes in the water absorption of self-compacting mortar containing a constant amount of FA caused by incorporation of titania nanoparticles. They revealed that water absorption got reduced in all NT containing samples compared to the control samples. A reduction in the water absorption of mortar of around 10% was found in samples with 5.0 wt% NT compared to samples without NT. Similar findings were also reported by Mohseni et al. [65], who investigated the effect of NT on the water absorption of self-compacting mortar incorporating different percentages of rice husk ash (RHA) (5.0, 10 and 15 wt%). They showed that water absorption got reduced in proportion to the amount of NT in the mixture. The best results were observed in samples containing 15 wt% RHA and 5.0 wt% NT, which had water absorption 18% less than that of samples without NT.

Chloride permeability is considered to be one of the main inherent properties that affect the reinforced concrete durability [87]. In their researcher, Zhang and Li [73] carried out an experimental investigation to find the effect of TiO₂ nanoparticles on the chloride ion penetration of concrete. They showed that use of NT reduced the chloride permeability of concrete at all investigated content of NT. They found that while 1.0 wt% NT had the best performance in enhancing the resistance to chloride ion penetration, the other NT contents (up to 5.0 wt%) still showed better performance than that of the plain concrete. Incorporation of 1.0 wt% NT reduced the chloride permeability by around 31%. Mohseni et al. [66] researched the effect of NT on the chloride permeability of self-compacting mortar incorporating a constant amount of FA. The results revealed that the effect of nano-TiO₂ on reducing the chloride permeability increased proportional to the amount of NT in the mixture. The maximum obtained reduction in the permeability was 59% compared to the control samples and was found in samples with 5.0 wt% NT. These results were supported by the findings of Mohseni et al. [65].

To better understand NT effect on water absorption and chloride permeability of concrete and mortar the related results are presented graphically in Fig. 2. The figure presents the optimum replacement percentages of cement with NT to reduce water absorption and chloride permeability as found in different reports and the resulted reduction percentage.

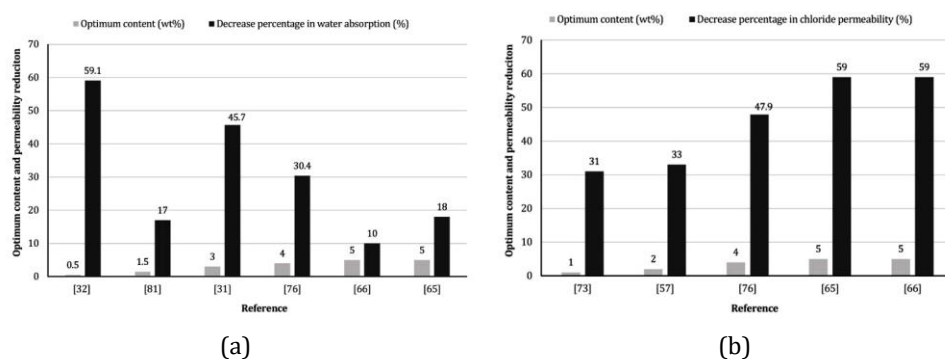


Fig. 2 Different optimum contents of NT to reduce (a) the water absorption and (b) the chloride permeability of cementitious composites and their related reduction percentages

The effect of titania nanoparticles on the permeability of concrete and mortar has been also investigated using other tests, such as capillary water absorption and gas permeability. Jalal et al. [76] researched the effect of NT on the capillary water absorption SCC. Their results showed that the height of absorbed water in the specimens decreased by increasing NT content up to 4.0 wt% and then started to increase. Such content of nano-titania resulted in a capillary absorption 21% less than that of the control sample. Farzadnia et al. [75] conducted a similar study on mortars incorporating 5.0 wt% SF in all the mixes. However, they reported an insignificant reduction in the capillary water absorption due to the incorporation of NT. The maximum reduction in the capillary water absorption was found to be 4% and was observed in samples containing 2.0 wt% NT. Behfarnia et al. [67] researched the effect of NT on the gas permeability of plain concrete. It should be mentioned that while the authors reported that NT had a reverse effect on the mechanical properties of concrete, they also stated that NT could reduce the gas permeability coefficient of concrete significantly. Their results showed that in general samples containing nano-TiO₂ had lower gas permeability coefficient compared to samples without NT. They demonstrated that the optimum content of NT (4.0 wt%) reduced the gas permeability by around 81% compared to the reference concrete.

Comparing the results presented in Table 2 and Fig. 2, it can be roughly said that the optimum content of NT for reduction of permeability of mortar and concrete is 1.0-5.0 wt%. To find the overall optimum content of NT, we should make a comparison between its result for the enhancement of the mechanical properties and its result for the reduction of permeability. Since the same optimum content was obtained in both the cases, it can be said that the optimum content of nano-TiO₂ for higher performance of concrete and mortar should be 1.0-5.0 wt%. However, more research should be conducted to confirm this result.

3.3.2 Effect on Ultrasonic Pulse Velocity (UPV)

UPV is an indicator of the uniformity, quality and durability of cement-based materials. As the microstructure becomes denser with less porosity, it reduces the pulse travel time and consequently increases the pulse velocity [65,85,97]. A higher value of UPV indicates a higher quality of cement-based material with a lower porosity [65,85]. Reports have

shown that NT improves the UPV of cement-based materials. This improvement has been found to increase as the content of NT increases up to the optimum content and then starts to decrease. Dezhampannah et al. [98] investigated the effect of NT on the UPV of heavy-weight concrete containing 0.6% polypropylene fiber and 2.0, 4.0, 6.0 and 8.0 wt% NT. Their results showed that use of NT up to 6.0 wt% increased the UPV. Further inclusion of NT was found to reduce the UPV. Xu et al. [84] stated that NT increased the UPV of concrete subjected to 0-50 sulfate dry-wet cycles. Nikbin et al. [97] investigated the ultrasonic pulse velocity of 2.0, 4.0, 6.0 and 8.0 wt% NT incorporated heavy-weight concrete. They found that the UPV increased as the content of NT increased. The optimum content was found to be 6.0 wt%, which led to an increase of 15% in the UPV compared to the reference concrete. Martins et al. [86] reported that NT increased the UPV of high performance concrete when it was used by 1.0 wt%. Higher content of NT resulted in a lower UPV. The results also revealed that incorporation of NT with 30 wt% fly ash showed much better performance than the use of only NT or fly ash. Similar results were also reported by Mohseni et al. [85]. Mohseni et al. [65] investigated the effect of combination of 0-5.0 wt% NT and 5.0-15 wt% rice husk ash on the UPV of mortars. Their test results revealed that the UPV increased with the increased in the NT and rice husk ash content. Noorvand et al. [5] conducted an experimental study to evaluate the UPV of mortar containing NT and untreated black rice husk ash. The results showed that NT was able to recover some of the loose of the UPV caused by the use of rice husk ash. However, in general, the combination of 1.5 wt% NT and 10-30 wt% rice husk ash showed a worse performance than that of the reference mortar. This was because rice husk ash remarkably decreased the UPV of mortars.

3.3.3 Effect on Electrical Resistivity

Electrical resistivity is often used as an index of initiation and propagation of corrosion of reinforcement and consequently as an index of durability. It is related to the porosity and permeability of cement-based materials. As the permeability decreases, the electrical resistivity increases and the durability increases [35,65,86]. Gopalakrishnan et al. [52] reported a proportional increase in the electrical resistivity of mortar due to the existence of NT. They found that 10 wt% NT increased the electrical resistivity of mortar by 21%. The effect of NT on electrical resistivity of SCC was researched by Joshaghani et al. [35]. Their work results showed that addition of NT remarkably increased the electrical resistivity of concrete, especially at the ages of 28 and 91 days. On the other hand, Martins et al. [86] revealed that NT resulted in a reduction in the electrical resistivity of high performance concrete at the ages of 7 and 28 days. This reduction was found to be proportional to the content of NT. However, with the use of 30 wt% fly ash, the electrical resistivity was remarkably increased. In their research, Mohseni et al. [65] investigated the combined effect of NT and rice husk on the electrical resistivity of cement mortars. They reported an increase in the electrical resistivity as the content of NT and rice husk increased.

3.3.4 Effect on Carbonation Resistance

Carbonation is considered to be the major cause of cement-based materials corrosion. The atmospheric CO₂ penetrates the cement-based materials and deteriorates their structure [99,100]. NT has been reported to reduce the carbonation resistance of cement-based materials. Moro et al., 2021 [101] reported an increase in the CO₂ uptake due to the use of NT in mortars. This was related to the role of NT in reducing the size of Ca(OH)₂ and increasing its surface area, which might promote the reactivity between Ca(OH)₂ and CO₂, resulting in a more consumption of CO₂. Moro et al. [102] found that NT increased the carbonation of normal and recycled cement mortars. Ramachandran et al. [99] investigated the carbonation depth of 40 wt% fly ash concrete incorporating 2.0 wt% NT and a combination of 1.0 wt% NT and 1.0 wt% nano-CaCO₃. The concrete was exposed to

three different environments; normal atmosphere, accelerated carbonation and sea water environment. Results revealed that the use of NT decreased resistance to carbonation. However, when used with CaCO_3 , it showed a remarkable improvement in the resistance to carbonation. This improvement was higher than the improvement resulted by the single use of either one of the two nano materials. Rao et al. [70] investigated the effect of NT on the carbonation depth of self-compacting mortars containing 30 wt% fly ash. The results revealed that the effect of NT on the resistance to carbonation of mortar depends on the binder:sand ratio. For binder:sand ratio equal to 1:1, NT didn't show any carbonation depth, similar to the reference mortar. However, when binder:sand ratio was 1:2, NT showed a reverse effect on the resistant to carbonation when its content was higher than 0.5 wt%. It's worth mentioning that although carbonation is a reason for corrosion of reinforcement, the effect of NT on increasing the ability of cementitious composites to adsorb CO_2 can be considered positive for cement-based materials without reinforcement. NT helps in reducing the quantity of CO_2 found in the environment.

On the other hand, Shaaban et al. [69] reported that NT increased the carbonation resistance of mortars. They investigated the effect of nano- TiO_2 on the carbonation depth of mortar exposed to CO_2 gas for different time periods (28, 56 and 90 days). Results showed that NT remarkably decreased the carbonation depth, especially at later ages. This was attributed to the NT filler effect, which resulted in a denser microstructure and consequently led to a reduction in the adsorption of CO_2 .

There is a lack of information about how titania nanoparticles affect the carbonation resistance of cement-based materials [103]. Moreover, contradictory results have been found in the available publications related to this topic. More research is needed to determine the behavior of NT modified cement-based materials exposed to CO_2 .

3.3.5 Effect on Freeze and Thaw Resistance

Frost resistance is considered as one of the main durability parameters, especially in cold climates [104]. Effect of NT on the freeze and thaw resistance of cementitious composites has not been sufficiently investigated. There is a limited number of reports available in the literature that have discussed this topic. More research is required to verify the frost performance of cementitious composites incorporating titania nanoparticles. Chunping et al. [83] conducted an experimental study to evaluate the freeze and thaw resistance of NT incorporated high performance concrete with and without applying flexural load. The samples were subjected to 800 cycles of freeze and thaw. The results revealed that NT didn't show any important difference on the mass loss and relative dynamic elastic modulus. A slight reduction in the mass loss due to the use of NT in the samples subjected to flexural load was observed. Salemi et al. [71] investigated the effect of nano-titania on the frost resistance of concrete. Normal and 2.0 wt% NT containing concretes were subjected to 300 freeze and thaw cycles. Their results revealed that NT remarkably increased the frost resistance of concrete. After 300 cycles of freeze and thaw, the reduction in strength, loss of mass, decrease in length and increase in water absorption were found to be around 100%, 84%, 28 % and 117%, respectively, for normal concrete, while they were found to be 11.5%, 5%, 2%, and 20%, respectively, for NT containing concrete. The improvement in frost resistance of concrete was ascribed to the denser microstructure which led to less water absorption and consequently lower effect of freeze and thaw.

3.3.6 Effect on Sulfate Attack Resistance

Nano-titania has been proven to increase the sulfate attack resistance of cement-based materials through the production of a more compacted and denser microstructure, which leads to reduction of ionic transport [60]. Xu et al. [84] investigated the sulfate resistance

of normal and nano-titania modified concrete subjected to 0-50 sulfate dry-wet cycles. They found that NT increased the resistance of concrete to sulfate attack and reduced the mass loss and compressive strength loss. After 50 sulfate dry-wet cycle, the mass loss percentage was 3.7% and 1.4% and the compressive strength loss percentage was 39.5% and 35.6% for concrete with and without NT, respectively. Shaaban et al. [69] studied the impact of incorporation of 3.0, 6.0 and 9.0 wt% titania nanoparticles into mortars on their sulfate attack resistance. They showed that as the content of NT increased, mass loss importantly decreased, which indicated a remarkable increase in the resistance to sulfate attack. They reported that 9.0 wt% NT containing samples shrank in sulfate solution. Daniyal et al. [60] conducted an experimental study on the compressive strength and corrosion resistance of mortar containing different percentages of NT (1.0, 3.0, 5.0 wt%). The mortars were subjected to different environmental exposures (tap water, saline water and 1% H₂SO₄ containing acidic solution). Their results showed that the 360-days compressive strength increased in all the situations due to the use of NT. Moreover, the corrosion resistance was increased due to the existence of titania nanoparticle. These effects were found to be proportional to the content of NT. Martins et al. [86] and Mohseni et al. [85] investigated the effect of NT on the sulfuric acid attack resistance of high performance concrete through mass loss test. The results showed 1.0 wt% NT increased the resistance to sulfuric acid attack. However, higher content of NT reduced this effect. They stated that 3.0 wt% NT had a mass loss more than the reference concrete.

3.4. Effect on Microstructural Properties

Some reports have discussed the effect of NT on the microstructure properties of cement mortar and concrete. These reports have demonstrated that nano-TiO₂ can improve the microstructure of concrete and mortar, and consequently improve the performance of such materials by reducing their permeability and increasing their strength [5,18,31–34,44,60,65,72,76]. NT improves the microstructure due to its role as a nano-filler to refine the pore structure and reduce the total specific pore volumes [31,33,34,55,57,59,72]. By addition of NT the distributed pores change to finer pores and become less harmful [18,33,34,59]. Incorporation of NT results in a better homogeneity, more compaction, lower porosity and reduction in the number of microcracks and their opening size [37,44,52,58,59,80,81]. Nano-TiO₂ can also notably improve the morphology of the ITZ by making it more homogenous and by enhancing the adherence between the cement paste and the aggregates [5,34,44,65].

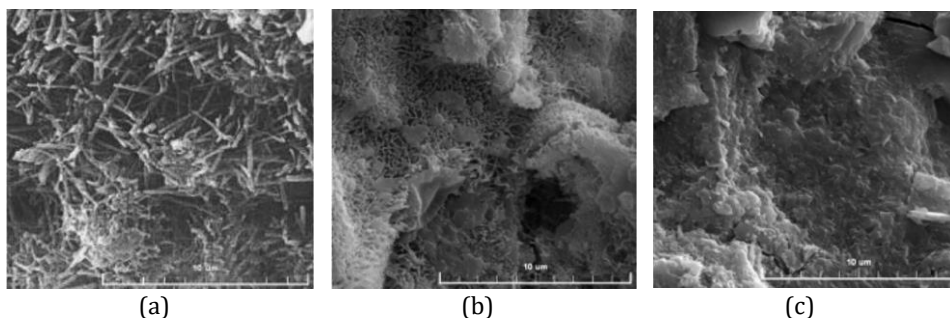


Fig. 3 SEM observations of SCC: (a) without nanoparticles, (b) with 1.0 wt% NT, (c) with 4.0 wt% NT [76]

Comparing SEM observations of samples before and after the incorporation of nanoparticles can provide a clear idea about how nanoparticles change the microstructure of cementitious materials. For this reason, here, the effect of NT on the microstructural properties of mortar and concrete are illustrated with the help of some examples of SEM images. Fig. 3 shows an example of the SEM observations of self-compacting concretes

after 7 days of curing. The figure presents the SEM images of concrete incorporating 0, 1.0 and 4.0 wt% NT. As can be seen from the figures, incorporation of NT leads to a more condensed and less permeable microstructure. It can also be observed that the sample containing 4.0 wt% NT has better microstructure than that of the sample with 1.0 wt% NT and the control sample. It should be mentioned that 4.0 wt% NT was concluded to be the optimum content of NT in the related reference [76]. These results coincide with the results found before in effect of nano-TiO₂ on the mechanical and permeability properties.

4. Cost Effectiveness of Use of NT in Cementitious Composites

The high price of nano materials has limited their use as construction materials, regardless of their huge benefits [19,86,105–114]. Cost of nanoparticles is roughly 1000× higher than the cost of the cement, which is the most expensive constituent of cement-based materials. Moreover, handling of nanoparticles might need special equipment and techniques, resulting in another increase in their cost [19]. Use of NT with even a small content results in a severe increase in the initial cost of cementitious composites [86]. However, if its effect over a long period of time is considered, the increase in cost of NT containing cement-based materials might be justified. Since NT increases the overall performance of cement-based materials along with the durability and sustainability, it will decrease the maintenance and repair cost of constructions. Moreover, NT provides cement-based materials with new properties such as self-cleaning properties, which leads to reduction in routine cleaning and maintenance costs [115,116]. Some reports have stated that NT can prolonged the service life of cement-based materials and consequently the constructions, resulting in a long-term economic benefits [117,118]. Some researchers have tried to reduce the cost of NT through the production of NT from waste materials such as sludge from sewage treatment [119].

Reports on the cost effectiveness of use of NT in cementitious composites are very scarce. Up to the best knowledge of the authors, no publication has thoroughly discussed the service life performance of cement-based materials containing NT, nor the service life cost effectiveness of NT. This indicates the lack of information related to the use of NT in cementitious composites, which needs to be overcome. Cost analysis of use of NT should take into consideration its long-term impact on the performance of cement-based materials. Moreover, new cost-effective techniques of production of NT should be researched, since they will reduce the cost of NT. This will also increase the production rate, resulting in another reduction in the cost of NT. The use of NT with a very small content in the cement-based materials will also reduce the cost. Reches [19] suggested the reduction of use of nanoparticles to less than 0.1 wt% for cost effectiveness of their use.

5. Conclusions

The current paper discusses the influence of use of NT on the properties of cementitious composites. Use of NT as an additive in cement-based materials can help them overcome some of their weaknesses, such as low tensile strength and low resistance to harmful chemical penetration. The outcomes of this work can be summarized as:

Incorporation of NT into cementitious composites can remarkably reduce their workability and reduce their initial and final setting time.

Use of nano-titania can importantly improve the compressive strength, split tensile strength and flexural strength of mortar and concrete, especially at early ages (1-7 days). This might be ascribed to the nano-filler effect and hydration acceleration effect of NT.

NT can remarkably increase the resistance of mortar and concrete to water, chloride and gas penetration. This can be related to the role of nano-TiO₂ in improving the

microstructural properties of mortar and concrete, which can be confirmed from the SEM observations.

Replacement of cement with an appropriate percentage of NT can improve the UPV, electrical sensitivity and sulfate attack resistance of cementitious composites. However, NT might lead to a reduction in the resistance of cement-based materials to carbonation.

Nano-titania can improve the sustainability and durability of structures and reduce their maintenance and repair costs by improving the overall performance of cement-based materials.

Incorporation of high percentage of NT might lead to a reverse effect on the properties of cementitious composites due to the unsuitable dispersion and agglomeration of nanoparticles.

Finding a specific optimum content for the incorporation of nano-titania into cement-based materials is very difficult. This is because there is still a big lack of information related to the effect of NT on different properties of cementitious composites. Moreover, some contradictory data has been found in the literature. However, as a guideline, the optimum content of NT can be considered as 1.0-5.0 wt%. More research in this regard is of a big importance.

Future research is encouraged to find solutions for the problems related to the use of NT, such as the potential impact on human wellbeing, high price and agglomeration of nanoparticles. More research is also recommended to increase our information about the effect of NT on properties of cementitious composites. The effect of NT on the freeze and thaw resistance and carbonation resistance should be further researched. Moreover, the service life performance and long-term cost effectiveness of use of NT in cement-based materials need to be investigated. More research on the effect of NT on dynamic properties of cement-based materials is also required. Future research for developing equations that illustrate the relationship between properties of cementitious composites and the content of NT is also needed. Studies similar to the current one for other nano-additives can also be topics for future work.

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