



Review

Effect and optimization of incorporation of nano-SiO₂ into cement-based materials – a review

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ABSTRACT

Incorporation of nanomaterials into cement-based materials has great potentials to improve their performance to great levels and to produce construction materials with superior and unique properties. Various nanoparticles have been utilized in cementitious composites to improve their properties. This paper provides a detailed review about the effect of the most widely incorporated nanomaterial into cement-based materials, namely nano-silica, on different on properties of cement-based materials. The investigated properties are mechanical properties (compressive strength, split tensile strength and flexural strength), durability parameters (permeability, freeze and thaw resistance, high temperature resistance, fire resistance and sulfate attack resistance) and microstructural properties of mortar and concrete. The cost effectiveness of use of nano-silica in cement-based materials is also discussed. The optimum replacement percentage of cement with this nanomaterial to improve the performance of mortar and concrete is also investigated. The investigation showed that nano-silica has the ability to enhance the mechanical properties, durability and microstructural properties of concrete and mortar to a remarkable level. It also showed that the optimum content of nano-silica in concrete and mortar is 1.0-4.0% by weight of binder materials.

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1. Introduction

Research on use of nano-additives in cement-based materials began with nano-silica (Nano-SiO₂, NS) and then was extended to other nanoparticles (Juenger and Siddique 2015). Nano-silica is an amorphous material that consists of nano-sized particles of silicon dioxide (SiO₂) that have a high surface area to volume ratio, which can lead to a great pozzolanic reactivity (Biricik and Sarier 2014; Zhuang and Chen 2019). NS is one of the materials advancing the field of nanocomposites. It is the most widely incorporated nanomaterial into cementitious composites to enhance their performance due to its pore-filling effect and high pozzolanic reactivity (Aggarwal et al. 2015; Amin and Abu el-Hassan 2015; Ardalan et al. 2017; Behfarnia and Salemi 2013; El-Gamal et al. 2018; Givi et al. 2011; Pengkun Hou et al. 2013; Kawashima et al. 2013; Lazaro et al. 2012). In addition, nano-SiO₂ is becoming a progressively important

component of special concretes and other advanced cementitious composites.

This work presents the advantages and disadvantages of use of NS in cementitious composites, and gives a detailed review of the effect of NS on the mechanical properties (compressive strength, split tensile strength and flexural strength), durability and microstructural properties of cement mortar and concrete. It also presents a discussion about the cost effectiveness of us of these nanoparticles. This work also gives a guideline for the optimum content of this nanomaterial in mortar and concrete to improve their performance.

2. Types and Production Methods of Nano-silica

Depending on its produced form, nano-silica has two types: powdery nano-silica and colloidal nano-silica (Biricik and Sarier 2014; Chithra et al. 2016). The SEM

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images of nano-silica in its two forms are presented in Fig. 1. The powdery NS shown in Fig. 1(a) is of an average size of 15 nm (Biricik and Sarier 2014), whereas, the average size of the colloidal NS shown in Fig. 1(b) is 7nm (Chithra et al. 2016). As it can be seen from the SEM images of NS, the particles can be available in a spherical

shape. TEM images in diluted form of the two forms of dispersed nano-silica are shown in Fig. 2. As shown in the figure, NS particles are present in an agglomerated form with a number of linked particles. The figure also shows that the particle size and dispersion state of nano-silica vary over a wide range (Bolhassani and Samani 2015).

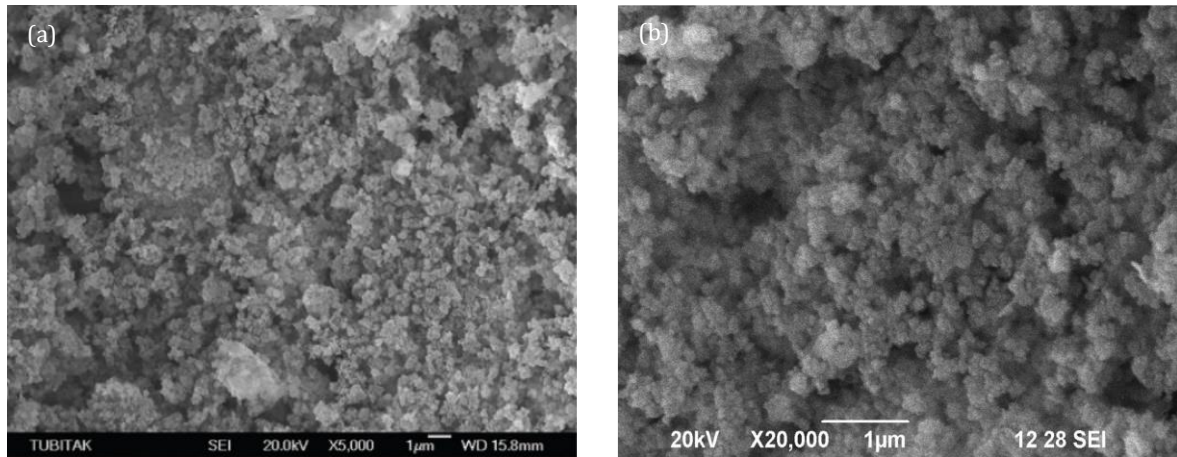


Fig. 1. SEM images of nano-silica: a) Powdery NS (Biricik and Sarier 2014); b) Colloidal NS (Chithra et al. 2016).

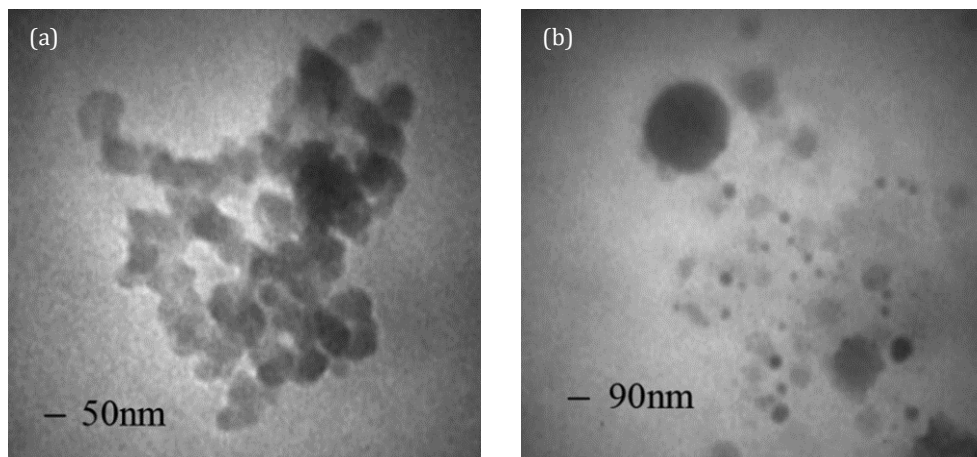


Fig. 2. TEM images of dispersed nano-silica in diluted form: a) Powdery NS; b) Colloidal NS (Bolhassani and Samani 2015).

The investigation of incorporating nano-silica into cementitious materials has become more common because of the successful results of using silica fume to improve the performance of cement-based materials and because of the ability of producing highly pure NS particles compared to silica fume (Bolhassani and Samani 2015). While silica fume, which is a by-product of the ferrosilicon industry, can be produced with a specific surface area (SSA) of 20 m²/g and 90% purity (Bolhassani and Samani 2015; Li et al. 2017), nano-silica can be produced with a higher specific surface area ranging from 60 to over 600 m²/g and with a purity up to 99.9% (Beigi et al. 2013; Bolhassani and Samani 2015; Givi et al. 2011, 2010; Jalal et al. 2015, 2012; Janković et al. 2016; Khanzadi et al. 2010; Li et al. 2004; Mohamed and Ragab 2014; Shebl et al. 2009). Due to this high surface area to volume ratio and since the pozzolanic reaction is related to the amount of surface area available for reaction (Givi

et al. 2011; Shebl et al. 2009), nano-silica has high pozzolanic activity compared to micro-silica (Behfarnia and Salemi 2013; Ghafari et al. 2014; Givi et al. 2010; Huang et al. 2020; Ji 2005; Seifan et al. 2020). For these reasons, it has been found that nano-SiO₂ is much more effective in improving strength and durability and reducing permeability of cementitious composites than micro-silica, especially at early ages (Ardalan et al. 2017; Behfarnia and Salemi 2013; Bolhassani and Samani 2015; Givi et al. 2011; Jalal et al. 2012; Seifan et al. 2020; Zhang and Islam 2012). It should be mentioned that incorporation of a composition of both nano-SiO₂ and silica fume has been found to be more effective in improving different properties of cementitious composites than the individual use of either one of the two materials (Amin and Abu el-Hassan 2015; Ardalan et al. 2017; Arif et al. 2020; Bi et al. 2012; Jalal et al. 2015; Li et al. 2017; Maheswaran et al. 2013; Nili et al. 2010).

There are two commonly used commercial methods for nano-SiO₂ production; the neutralization of sodium silicate (water-glass or Na₂SiO₄) solutions with acid, such as sulfuric acid (the wet route or sol-gel process) and flame hydrolysis (the thermal route) (Kooshafar and Madani 2020; Lazaro et al. 2013, 2012; Quercia et al. 2013; Quercia and Brouwers 2010). However, because of the energy needed in the processes and the price of the raw materials, nano-SiO₂ produced by these two methods is expensive (Lazaro et al. 2013, 2012). Furthermore, due to the huge amount of CO₂ emitted during these processes, these methods are environmentally unfriendly (Lazaro et al. 2013). Researchers have been trying to overcome such problems through the development of new industrial, low cost, production processes. One example of such processes is the production of nano-SiO₂ by dissolving olivine in acid at low temperatures (50-95°C) (Lazaro et al. 2013, 2012). Using this method, nano-SiO₂ with primary particles of 10-25nm, a specific surface area of 100-400 m²/g and an impurity content below 5% can be produced (Lazaro et al. 2013, 2012). However, the produced NS was noticed to be agglomerated in 3D chains leading to the reduction of the packing factor of the aggregate mix, and to have a lower pozzolanic activity compared to the standard micro-silica at the same replacement level (Lazaro et al. 2012; Quercia and Brouwers 2010). This process is a convenient alternative method to the existing one because olivine dissolution can be done in a low temperature, making this process cheaper (even cheaper than contemporary micro-silica (Quercia and Brouwers 2010)) and more environmentally friendly (Lazaro et al. 2013). Moreover, used olivine in nano-silica production can be a waste material (Lazaro et al. 2013). Low-cost methods also include the production of nano-SiO₂ from rice husk (Crucho et al. 2018; Mor et al. 2017), barley grain waste (Akhayere et al. 2019), Equisetum arvense (Carneiro et al. 2015), Sorghum vulgare seed heads (Balamurugan and Saravanan 2012), fly ash (Manchanda et al. 2017) and waste glass powder (Asadi and Norouzbeigi 2018). The production of nano-silica using low-cost methods will reduce its price and lead to more production of nano-silica, leading to more use of it in the construction industry and other industries.

3. Advantages and Disadvantages of Incorporation of Nano-SiO₂ into Cement-based Materials

The effect of using nano-silica in improving mechanical, physical and microstructural properties of cement-based materials has been widely reported (Aggarwal et al. 2015; Arif et al. 2020; Behzadian and Shahrajabian 2019; Biricik and Sarier 2014; Givi et al. 2011; Haruehansapong et al. 2014; Lazaro et al. 2012; H. Li et al. 2004; Rashad 2014; Sikora et al. 2015; Zhang and Islam 2012). These reports have shown that nano-silica leads to significant enhancement in compressive, flexural strength, split tensile strength and elasticity modulus of mortar and concrete. As a result of the high reactivity of these nanoparticles, nano-SiO₂ accelerates the hydration process of dicalcium silicate (C₂S) and tricalcium

silicate (C₃S) and increases the amount of calcium silicate hydrate produced in the matrix through intense reaction with crystals of Ca(OH)₂, which have adverse effects on strength and durability, leading to exceptional enhancement in different properties of cement-based materials (Aggarwal et al. 2015; Bahadori and Hosseini 2012; Behfarnia and Salemi 2013; Beigi et al. 2013; Biricik and Sarier 2014; Kewalramani and Syed 2018; Ngo et al. 2020; Niewiadomski et al. 2018; Senff et al. 2013; Singh et al. 2013; Zhuang and Chen 2019). Nano-silica addition also fills the voids and improves the microstructure of the final products leading to reduction in porosity and increase in density and in resistance to ion penetration and water permeability, and hence improve the durability (Amin and Abu el-Hassan 2015; Arif et al. 2020; Barbhuiya and Qureshi 2015; Behfarnia and Salemi 2013; Biricik and Sarier 2014; Givi et al. 2011; Kewalramani and Syed 2018; Lazaro et al. 2012; Liu et al. 2020; Quercia et al. 2014; Ying et al. 2017). It also reduces bleeding water, enhances the bond between aggregates and cement paste and increases the cohesiveness of mixtures in the fresh state (Aggarwal et al. 2015; Behfarnia and Salemi 2013; Biricik and Sarier 2014; Carmo et al. 2021; Ji 2005; Lazaro et al. 2012; Nima et al. 2011; Zhang and Islam 2012). Nano-silica has been proved to be beneficial in controlling the leaching of calcium, which is one of the main reasons for various types of concrete degradation (Bahadori and Hosseini 2012; Givi et al. 2011; Nima et al. 2011; Quercia et al. 2013; Singh et al. 2013). Furthermore, NS decreases the setting time of cement-based composites (Aggarwal et al. 2015; Behfarnia and Salemi 2013; Biricik and Sarier 2014; Choolaei et al. 2012; Givi et al. 2011; Rashad 2014; Senff et al. 2009; Zhang and Islam 2012; Zhuang and Chen 2019). The reduction in setting time of cement could increase the throughput of precast plants and also reduce formwork removal time and total time of construction leading to direct economic benefits (Jayapalan et al. 2013). In addition, replacement of cement with nano-SiO₂ should have a great effect on the environment through the considerable reduction of CO₂ emissions caused by cement production (Lazaro et al. 2013; Quercia and Brouwers 2010). Thus, by incorporating NS, cementitious composites with remarkable high performance can be produced (Lazaro et al. 2012).

Despite all the great effects of addition of silica nanoparticles to cement-based materials, use of nano-silica has also some drawbacks. Currently high price of various NS products available in the markets is one of the reasons that limit its commercial applications (Biricik and Sarier 2014). However, some latest studies have focused on solving such problem through developing new less energy consuming methods of production of NS, such as production of NS from olivine (Biricik and Sarier 2014; Lazaro et al. 2013, 2012; Quercia and Brouwers 2010). Furthermore, increasing NS amount more than a specific limit has reverse effects on different properties of cementitious materials due to agglomeration of NS particles and unsuitable dispersion of the nanoparticles (Amin and Abu el-Hassan 2015; Biricik and Sarier 2014; Quercia and Brouwers 2010). Inappropriate dispersion of NS in the mixture affects the rate and extent of hydration

of cement as well as other mechanical and physical properties (Biricik and Sarier 2014). For these reasons, some researchers have suggested to use small amount of NS (1.0-5.0 wt%) unless an appropriate method of dispersion is used (Biricik and Sarier 2014; Senff et al. 2009). Due to its high specific surface area that can absorb some of the water content, NS also reduces the workability of cementitious materials and increases water demands. Every added kilogram of NS requires 0.4 kilogram of water to preserve the same workability (Aggarwal et al. 2015; Al Ghabban et al. 2018; Amin and Abu el-Hassan 2015; Bahadori and Hosseini 2012; Bolhassani and Samani 2015; Givi et al. 2011; Niewiadomski et al. 2018; Rashad 2014; Senff et al. 2009; Siang Ng et al. 2020; Sikora et al. 2015; Singh et al. 2013; Zhuang and Chen 2019). In addition to the fact that the effect of application of NS into cement-based materials is not fully understood yet, due to the insufficient available data related to this topic.

4. Effect of Nano-SiO₂ on Different Properties of Cement-based Materials

Since the final performance of cement-based materials depends to a great deal on the strength assessments and permeability of such materials (Nazari and Riahi 2011a, 2011b), in this section, focus will be on the effect of nano-SiO₂ on the mechanical properties (compressive strength, split tensile strength and flexural strength) and permeability of mortar and concrete. Moreover, to have an idea about the effect of NS on the durability of cementitious composites, some of the main durability parameters have been discussed. In addition, to better understand the changes that happen in the cementitious materials due to the incorporation of nano-additives, the effect of such materials on the microstructural properties have been illustrated through evaluation of some examples of SEM images.

To better understand the effect of nano-silica on the mechanical properties and permeability of concrete and mortar, some examples from the literature have been discussed in this section. The gathered research results have been organized in terms of type of cement-based materials (first for concrete and then for mortar). For concrete, the effect of nano additives on the properties of plain concrete, self-compacting concrete (SCC) and concrete incorporating other additives such as silica fume (SF), fly ash (FA) and copper slag have been respectively discussed. The same has been done for the effect of nano additives on the properties of mortar.

4.1. Effect on mechanical properties

The main characteristic of cement-based materials are mechanical properties, since they directly express the performance of different structural members to the applied loads. Effect nano-SiO₂ on the mechanical properties depends to a great deal on the properties of the used nano-silica, such as specific surface area, particle size etc. (Givi et al. 2010; Haruehansapong et al. 2014). Results found on the literature on the effect of nano-silica on the

mechanical properties of cement-based materials vary to some extents from one to another. However, results in some research works were observed to have some similarities. In general, the results show that the incorporation of SiO₂ nanoparticles into cement-based materials can significantly increase the overall compressive strength, split tensile strength and flexural strength at all ages.

Data collected from different research papers on the effect of NS on the compressive strength, split tensile strength and flexural strength of mortar and concrete materials are shown in Table 1. As shown in the table, researchers have applied different variables in their research to study the effect of NS on the mechanical properties of cementitious materials. While some researchers investigated the effect of NS on mechanical properties of plain mortar and concrete, some others investigated its effect on other types of cement mortar and concrete, such as mortar and concrete with their cement partially replaced with some other additive, such as silica fume (SF), fly ash (FA) and copper slag etc. The table also shows the optimum percentages of the replacement of cement with NS reported in different research work and their optimum effect on mechanical properties.

It's worth mentioning that when nano silica is incorporated in cement-based materials, the mix design is the same as for normal cement-based materials with one major different. Since nano-silica is usually used as a partial replacement of cement in the mixture, the content of cement is reduced by the chosen replacement percentage and nano-silica is added to the mixture by a weight equal to the replacement percentage. Another important point that should be taken into consideration in the mix design is that nano-silica dramatically reduces the workability of cement-based materials, as mentioned before. Thus, to have a desirable workability, the use an adequate amount of superplasticizer might be needed (Amin and Abu el-Hassan 2015; Chithra et al. 2016; Du et al. 2014). In addition, it has been found that superplasticizer can lead to a better distribution of the nanoparticles in the mixture (Behfarnia and Salemi 2013). Some authors suggested the premixing of the nanoparticles with some quantity of the required superplasticizer and water using a high-speed mixer for few minutes to help distribute the nanoparticles into the mixture (Amin and Abu el-Hassan 2015; Behfarnia and Salemi 2013). While some authors have premixed nanoparticles with the whole required quantity of water and superplasticizer (Behfarnia and Salemi 2013; Kooshafar and Madani 2020), some others have premixed them with only half of the quantity (Amin and Abu el-Hassan 2015). Some other authors such as (Chithra et al. 2016) have premixed nano-silica with 60% of the required water. The rest water quantity was mixed with superplasticizer. It should be noted that some other researchers such as (Du et al. 2014) have not premixed nano-silica with water, and instead, they directly added it to the mixer after mixing the dry materials and adding water to the mixer. Since this paper tries to determine the optimum content of nano-silica in mortar and concrete, only the type of cement-based material, the content of nano-silica used in each study and the optimum content of nano-silica in the related study are mentioned in Table 1.

Table 1. Optimum content of nano-silica in cementitious materials and enhancement percentage of the mechanical properties.

Reference	Mortar/ concrete	NS properties				Optimum content (wt%)	Enhancement in 28-day strength (%)		
		Size (nm)	SSA (m ² /g)	Form	Investigated contents (wt%)		Compressive strength	Split tensile strength	Flexural strength
Li et al. (2004)	mortar	15	-	-	3, 5, 10	10	26	-	-
Givi et al. (2010)	concrete	15	160	powder	0.5, 1, 1.5, 2	1	18.5	83.3	31.8
		80	560			1.5	12	72.2	22.7
Nili et al. (2010)	concrete	50	80	colloidal	1.5, 3, 4.5	4.5	22	-	-
Givi et al. (2011)	concrete	15	160	powder	0.5, 1, 1.5, 2	1	18.5	83.3	31.8
Nazari and Riahi (2011a)	concrete (self-compacting)	15	165	-	1, 2, 3, 4, 5	4	74.37	118.75	64.29
Oltulu and Şahin (2011)	mortar ⁽¹⁾	12	200	powder	0.5, 1.25, 2.5	1.25	11.77	-	-
Zhang and Li (2011)	concrete	10	640	-	1, 3	1	12.3	-	4.21
Ibrahim et al. (2012)	mortar	-	-	colloidal	2.5, 5, 7.5	7.5	37	-	22
Said et al. (2012)	concrete	35	-	colloidal	3, 6	6	25	-	-
Stefanidou and Papayianni (2012)	mortar	14	200	-	0.5, 1, 2, 5	2	15	-	6
Behfarnia and Salemi (2013)	concrete	20	220	-	3,5,7	5	30.1	-	-
Beigi et al. (2013)	concrete (self-compacting)	15	160	-	2, 4, 6	4	18	35	40
Oltulu and Şahin (2013)	mortar ⁽²⁾	12	200	powder	0.5, 1.25, 2.5	1.25	19.24	-	-
Biricik and Sarier (2014)	mortar ⁽³⁾	15	640	powder	5, 10	10	83.4	-	31.8
Ghafari et al. (2014)	concrete ⁽⁴⁾	15	160	powder	1, 2, 3, 4	3	7.5	-	-
		12	200			9	23.9	-	-
Haruehansapong et al. (2014)	mortar	20	90	-	3, 6, 9, 12	9	52.9	-	-
		40	50			9	54.4	-	-
Shaikh et al (2014)	mortar	25	160	-	1, 2, 4, 6	2	14.29	-	-
Bolhassani and Samani (2015)	mortar	7-25	80 - 380	powder/ colloidal	0.5, 1.5, 3, 5, 7	7	13.1	-	-
Du et al. (2015)	concrete ⁽⁵⁾	12.4	220	colloidal	1, 2	2	5.6	-	-
Li et al. (2015)	concrete ⁽⁶⁾	20	-	-	0.5, 1, 1.5, 2	1	3.74	-	16.2
Madandoust et al. (2015)	mortar (self-compacting) ⁽⁷⁾	15	200	-	1, 2, 3, 4, 5	4	15	-	-
Mohseni et al. (2015)	mortar (self-compacting) ⁽⁷⁾	15	200	powder	1, 3, 5	3	7	-	-
Rong et al. (2015)	ultra-high-performance cementitious composites ⁽⁸⁾	20	-	-	1, 3, 5	3	22	-	9
Chithra et al. (2016)	concrete ⁽⁹⁾	5-40	-	colloidal	0.5, 1, 1.5, 2, 2.5, 3	2	18.2	25.4	36.5
Wu et al. (2016)	concrete ⁽¹⁰⁾	5-35	160	-	0.5, 1, 1.5, 2	1	10	-	11

Ma and Zhu (2017)	concrete	<20	>600	powder	0.6, 1.2, 1.8	1.2	7.07	6.6	-
Ying et al. (2017)	concrete ⁽¹¹⁾	15 ± 5	-	colloidal	1, 2, 3	2	17	-	-
Al Ghabban et al. (2018)	concrete	40	-	-	1, 2, 3, 4	3	47.17	68.75	63.64
Niewiadomski et al. (2018)	concrete (self-compacting)	10–20	-	powder	0.5, 2, 4	4	8	-	(NS 0.5%) 4
Behzadian and Shahrajabian (2019)	concrete ⁽¹²⁾	20-30	-	-	1, 3, 5	3	43.42	48.1	20.34
Arif et al. (2020)	concrete	17	640	-	2.5, 5, 7.5	5	8.5	9	(NS 2.5%) 12.5
Siang Ng et al. (2020)	mortar ⁽¹³⁾	20-30	180–600	powder	1, 3, 5	3	38	-	19
Ngo et al. (2020)	concrete ⁽¹⁴⁾	13	-	powder	0.5, 1, 1.5, 2, 2.5, 3	1.5	6.09	-	14.83
Seifan et al. (2020)	mortar	20	-	powder	5, 10, 15	5	31	22	-
Vivek et al. (2020)	concrete	-	202	-	1, 2, 3, 4	4	9.9	17.48	-

Notes:

- (1) SF was used by 5 wt% of cement in all mixtures.
- (2) FA was used by 15 wt% of cement in all mixtures.
- (3) Mortar with good dispersion of NS was studied.
- (4) Concrete incorporating SF of around 27 wt% of the total binder content in all mixtures.
- (5) Light weight concrete.
- (6) Ultra-high-performance concrete (contains silica flour, SF and FA).
- (7) FA was used by 25 wt% of cement in all mixtures.
- (8) FA was used by 35 wt% of cement in all mixtures.
- (9) Concrete with copper slag as 40 wt% partial fine aggregate replacement in all mixtures.
- (10) Ultra-high strength concrete (UHSC).
- (11) Recycled aggregate concrete.
- (12) Concrete incorporating 10 wt% of waste PET aggregates.
- (13) FA was used by 30 wt% of the cement in all mixtures.
- (14) High performance concrete (SF was used by 5 wt% of the cement in all mixtures.)

4.1.1. Effect of NS on compressive strength

NS effect on the compressive strength of mortar and concrete at different ages has been thoroughly reported in the literature. The results demonstrate that incorporation of NS into mortar and concrete can remarkably enhance their compressive strength at all curing ages. They also show that with the increase in NS content up to a specific limit in the mixture, its effect on the compressive strength increases. This limit is referred to as the optimum content. When the content of NS exceeds the optimum content, its effect starts to decrease. To understand the effect of NS on compressive strength of concrete and mortar at different ages some examples have been provided in this section. More examples can be found in the related references.

Nili et al. (2010) experimentally studied the effect of NS on the compressive strength of concrete. They reported the NS optimum content to be 4.5 wt%, which led to an enhancement in the 7-day, 28-day and 90-day compressive strength of around 20%, 22% and 7%, respectively. Their results also demonstrated that better effect was observed when both SF and NS were used in the same mix. Using 6 wt% SF and 1.5 wt% NS enhanced the 28-day compressive strength by about 29%. Behfarnia and Salemi (2013) found a maximum improvement in the compressive strength with the replacement of cement with 5.0 wt% NS. They reported that this content caused an increase of 15%, 30% and 45% in the 7, 28 and 120-day compressive strength, respectively. Beigi et al. (2013) stated that 2.0, 4.0 and 6.0 wt% NS content led to an increase of 3.0%, 18% and 17%, respectively in the compressive strength after 28 days of curing. Unremarkable

difference in the effect of NS on the compressive strength was found for 4.0 wt% and 6.0 wt% content cases.

Nano-silica effect on the compressive strength of mortar at different ages has been also investigated by many researchers. Li et al. (2004) conducted an experimental study on the effect of nano-SiO₂ on compressive strength of plain mortar. They found that while NS of 5.0 wt% had better effect on the 7-day strength (20% higher than plain specimen) compared to 3.0 wt% NS, which only caused an increment of around 6%, higher content of NS (10 wt%) had the same effect as that of 5.0 wt%. However, in the case of 28-day compressive strength, 10 wt% had higher effect (26% higher than plain concrete) on compressive strength compared to 3.0 wt% and 5.0 wt% NS (13.8% and 17%, respectively). Nano-silica effect on the compressive strength of self-compacting mortar was also studied by some researchers such as Madandoust et al. (2015) and Mohseni et al. (2015). In both research work, the authors investigated the effect of NS on the properties of self-compacting mortar incorporating 25% fly ash by weight of cement. While Madandoust et al. (2015) reported that the optimum replacement of cement with NS was 4.0 wt%, and that it increased the 28-day compressive strength by 15%, Mohseni et al. (2015) reported it to be 3.0 wt%, and that it resulted in an improvement in the strength of about 28%, 7% and 15% after 7, 28 and 90 days of curing, respectively. A possible reason for the difference between the optimum replacement content found in these two research work is that Madandoust et al. (2015) have investigated more replacement percentages compared to Mohseni et al. (2015), who haven't investigated the effect of 4 wt% NS.

As can be noticed from the literature review above, replacement of cement with nano-SiO₂ up to the optimum percentage increases the compressive strength of cement-based materials. However, when more content of NS is used, its effect starts to reduce proportional to its content. The improvement in the strength of cementitious materials might be attributed to the behavior of nano-silica as a nano-filler to improve the microstructure and reduce the porosity of the mixture by recovering its particle packing density, as well as an activator to accelerate the pozzolanic reaction (Ghafari et al. 2014; Haruehansapong et al. 2014; Jalal et al. 2012; Kawashima et al. 2013; Siang Ng et al. 2020; Vivek et al. 2020). As a result of its high reactivity and high content of SiO₂, NS can accelerate the hydration process of cement paste causing faster consuming of Ca(OH)₂, formed mainly at early ages of the cement hydration process (Ghafari et al. 2014; Givi et al. 2011, 2010; Peng-kun Hou et al. 2013; Pengkun Hou et al. 2013; Niewiadomski et al. 2018). This leads to more production of C-S-H gel (Al Ghabban et al. 2018; Givi et al. 2011, 2010; Haruehansapong et al. 2014; Kawashima et al. 2013; Niewiadomski et al. 2018). On the other hand, reduction of strength that happens after exceeding the optimum content could be related to that the quantity of silica nanoparticles that exist in the mix is more than the quantity needed to react with the liberated Ca(OH)₂ particles leading to reduction in strength since it partially replaces the binding materials but doesn't contribute to the strength (Ghafari et al. 2014; Givi et al. 2011, 2010). In addition, large content of NS increases the weak zone in the concrete (Behfarnia and Salemi 2013; Ghafari et al. 2014; Seifan et al. 2020; Siang Ng et al. 2020). Reduction in the strength might also happen due to the agglomeration and unsuitable dispersion of silica nanoparticles (Behfarnia and Salemi 2013; Bolhassani and Samani 2015; Ghafari et al. 2014; Seifan et al. 2020). Another reason for the strength reduction might be the insufficient workability, since water available for lubrication is insufficient allowing free movement of particles (Ghafari et al. 2014).

As can be seen from Table 1, different optimum values for the percentage of replacing cement with nano-SiO₂ to enhance the compressive strength and other mechanical properties of mortar and concrete have been reported. Finding such optimum content accurately is of a great difficulty, due to the existence of various factors that play role in determining such value and due to the lack of data related to this topic. However, to have an idea about the approximate optimum content of NS, a comparison between all the collected optimum values, regardless of the different variables, can be of a significant help. As can also be seen from the table, mostly the same optimum value has been found for different mechanical properties. It is also obvious that the most investigated mechanical property is the compressive strength. Hence, to make the comparison easier, the collected data related the effect of NS on the compressive strength is shown graphically in Fig. 3. Comparing data in Table 1 and Fig. 3, it's clear that most authors have reported their optimum content to be between 1.0 wt% to 5.0 wt%. Hence, it can be said that the optimum content of NS to enhance different mechanical properties of mortar and concrete is between 1.0-5.0 wt%.

4.1.2. Effect of NS on split tensile strength

Nano-silica effect on the split tensile strength of mortar and concrete has been studied by some researchers. The results show that, similar to the compressive strength case, with the increase of NS content up to the optimum value, its effect on the split tensile strength increases and then starts to decrease. This behavior can be related to the same reasons stated above for the compressive strength. The results also demonstrate that NS can remarkably increase the tensile strength of cementitious composites, and hence solve the problem of low tensile strength related to such materials.

Givi et al. (2010) evaluated the effect of NS of 15nm and 80nm average particle sizes on split tensile strength of plain concrete. They observed that while smaller particle size of NS (15nm) had better performance at early ages, NS with bigger particles size (80nm) had better performance at later ages. The results showed that NS optimum content was 1.0 wt% in the case of the finer NS (15nm) and 1.5 wt% in the case of the courser one (80nm). They also showed that while the optimum content in the case of 15nm NS led to 100%, 83% and 56.5% enhancement in the 7th, 28th and 90th days split tensile strength, respectively, the optimum content in the case of 80nm NS resulted in 93.33%, 72.22% and 100% enhancement. The effect of NS on split tensile strength of SCC was studied by Beigi et al. (2013), who showed that 2.0, 4.0 and 6.0 wt% NS content caused an increase of 7.5%, 35% and 35% in the 28-day split tensile strength, respectively. They also reported that similar to the case of compressive strength the difference between the effect of 4.0 wt% NS and 6.0 wt% NS was negligible. Similarly, Chithra et al. (2016) stated that the development of split tensile strength was similar to the development of compressive strength at all curing ages. The results of their research showed an increment of 21.4%, 25.4% and 25% in the 7, 28 and 90-day strength of concrete incorporating copper slag as 40% by weight of fine aggregate was observed for 2.0 wt% NS content.

As stated before, the optimum content of NS to enhance different mechanical properties is to be taken the same. However, the enhancement percentage of split tensile strength due to the incorporation of the optimum content of NS was observed to be generally higher than that of the compressive strength in different references (see Table 1).

4.1.3. Effect of NS on flexural strength

The effect of nano-SiO₂ on the flexural strength of mortar and concrete has shown a similar trend to that of the compressive strength and split tensile strength. Givi et al. (2010) reported that smaller particles of NS tend to have better effect on flexural strength at early ages of curing while bigger ones tend to have more pronounce effect at later ages. They reported that 1.0 wt% NS of 15nm particle size improved the flexural strength by around 31%, 32% and 32% after 7, 28 and 90 curing days, respectively, compared to 17%, 16% and 45% in

the case of NS of 80nm particle size. However, the optimum content in the case of 80nm (i.e. 1.5 wt%) resulted in 32.8%, 22.7% and 55.3% enhancement in the 7, 28 and 90-day flexural strength, respectively. In their work, Beigi et al. (2013) studied the effect of nano-SiO₂ on flexural strength of SCC and showed that the optimum content (4.0 wt%) increased the strength by 40%. They also reported that while 2 wt% NS only increased the strength by 7.0%, 6.0 wt% NS had strength close to that of the optimum content (39%). Although NS was reported to enhance the flexural strength of concrete remarkably, some authors reported it to only have a small effect on the flexural strength. For instance, Zhang and Li (2011) stated that the optimum content of NS (1.0 wt%) led to only 4.21% increment in the 28-day flexural strength. Moreover, by increasing NS content in the mixture up to 3 wt%, its effect on the strength was found to be even much less. 3.0 wt% NS was found to reduce

the flexural strength of concrete by -1.87% compared to the plain concrete.

Nano-silica effect on the flexural strength of mortar has been also investigated by some researchers. Li et al. (2004) showed that by incorporating NS into mortar, the flexural strength could be remarkably improved. They reported that 5.0 wt% NS, which represented the optimum content in their research, improved the flexural strength of mortar by around 27%. These results are close to what Ibrahim et al. (2012) reported in their research. They stated that 7.5 wt% increased the flexural strength by 22%. On the other hand, some other researchers such as Stefanidou and Papayianni (2012) reported an unimportant effect of NS on the flexural strength. They stated that the optimum content (i.e. 2 wt%) caused an enhancement in the flexural strength of only 6%. The inconsistencies found in some references confirm the need for further research.

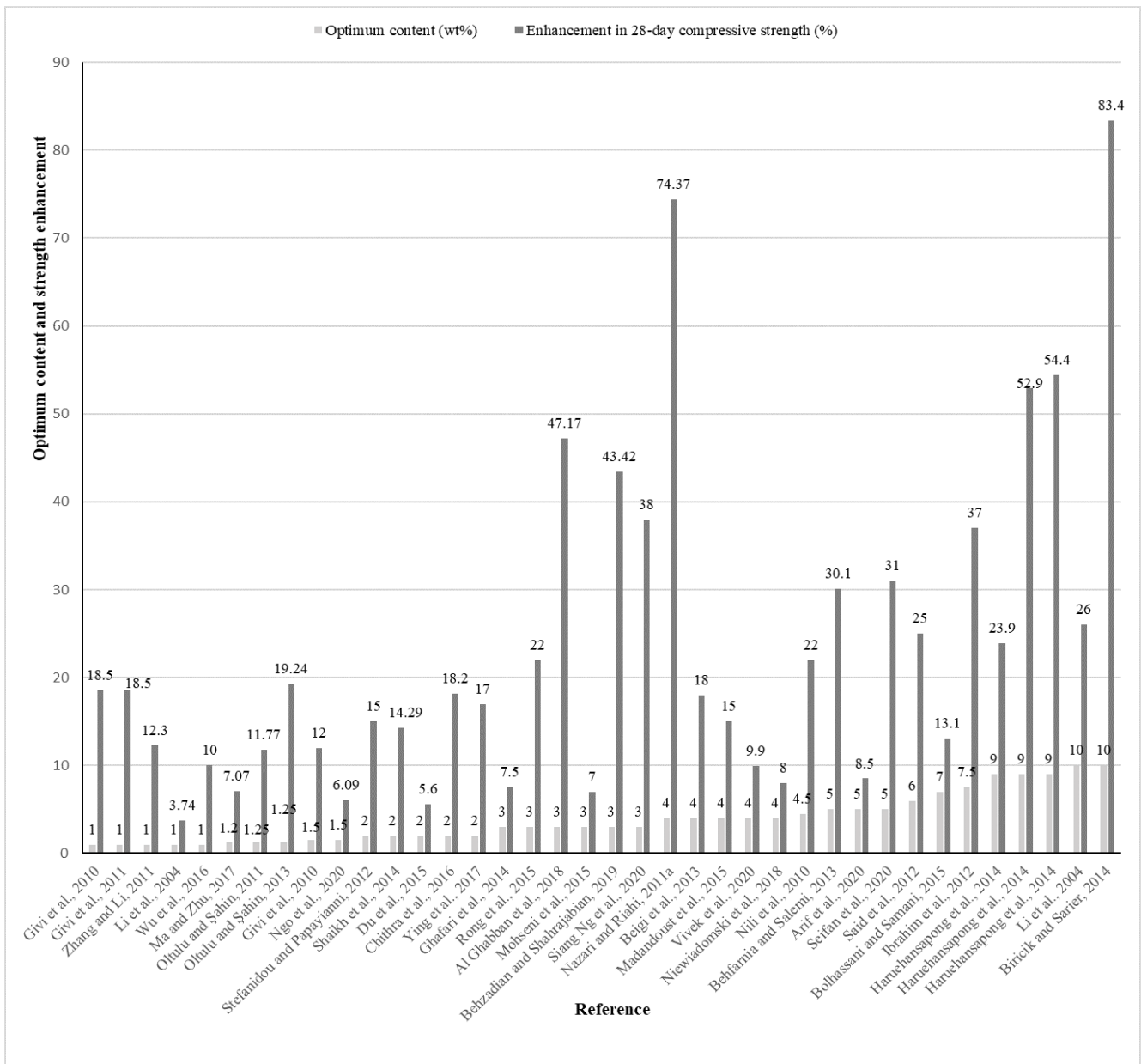


Fig. 3. Different optimum contents of NS to improve the compressive strength of cement-based materials and the related improvement percentage.

4.2. Effect on durability

Nano-silica has been found to improve the durability of cementitious materials due to its high pozzolanic reactivity and its effect on improving the microstructure through increasing compactness and reducing porosity. It also improves the hydration stability and works as a nucleus to bond strongly with C-S-H gel particles (Ashwini and Srinivasa Rao 2021; Du et al. 2014; Farajzadehha et al. 2020; Kooshafar and Madani 2020; Li et al. 2017; Shafiq et al. 2019; Sharaky et al. 2019; Vaz-Ramos et al. 2019). Durability can be evaluated using many parameters, such as permeability (water permeability, chloride permeability etc.), freeze and thaw resistance, wet and dry resistance, fire resistance, high temperature resistance, chemical attacks resistance (sulfate attack, $MgCl_2$ attack, $CaCl_2$ attack etc.), alkali-silica reaction, abrasion resistance, pore structure analysis etc. In this section, the effect of NS on the durability of cement-based materials is evaluated through discussing permeability, freeze and thaw resistance, high temperature resistance, fire resistance and sulfate attack resistance.

4.2.1. Permeability

Permeability of cement-based products can be defined as the penetration of external substances, such as liquids, gases, harmful ions etc., into a body of such product under a constant pressure gradient (Nazari and Riahi 2011c; Zhang and Li 2011). This property mainly depends on the geometric arrangement and properties of the different components of the materials. Permeability is directly affected by the porosity and solidity of the hydrated product (Jalal 2012; Khoshakhlagh et al. 2012; Nazari and Riahi 2011a). Permeability can be evaluated

by many parameters. However, in this section the effect of incorporating silica nanoparticles on the permeability of concrete and mortar are illustrated through studying some of the main parameters; namely percentage of water absorption, chloride permeability, capillary water absorption, water permeability coefficient and gas permeability coefficient.

Silica nanoparticles have been proved to enhance the microstructure of concrete and mortar to a great deal, and accordingly increase its resistance to penetration of different materials (water, chloride, gas etc.) (Aggarwal et al. 2015; Bahadori and Hosseini 2012; Ghafari et al. 2014; Givi et al. 2011; Madandoust et al. 2015; Nili et al. 2010; Quercia et al. 2014; Said et al. 2012; Sikora et al. 2015). It has been noticed that, similar to the effect of NS on the mechanical properties, the permeability of cementitious composites generally decreases with the increase of NS content in the mixture up to the optimum content and then starts to increase.

Table 2 presents some of the published data collected from different references on the effect of nano- SiO_2 on the reduction of different parameters of permeability. The table also shows the optimum percentages for partial cement replacement with NS to reduce the permeability of mortar and concrete and the related reduction percentage of different parameters of permeability. As can be seen from the table, generally, the same optimum content is applicable for different permeability measurements. Comparing Table 1 and Table 2, it can be observed that in some references this optimum content is different from that of the mechanical properties. It should be mentioned that different properties of NS used in the permeability tests are as given in Table 1. It can be realized from Table 2 that the most researched permeability parameters are water absorption and chloride permeability.

Table 2. Optimum content of nano- SiO_2 in cementitious materials for permeability.

Reference	Investigated contents (wt%)	NS optimum content (wt%)	Decrease percentage (%)				
			Water absorption	Chloride permeability	Capillary water absorption	Water permeability coefficient	Gas permeability coefficient
Nili et al. (2010)	1.5, 3, 4.5	3	-	-	40	-	-
Givi et al. (2011)	0.5, 1, 1.5, 2	2	21.25	-	-	51.4	-
Nazari and Riahi (2011a)	1, 2, 3, 4, 5	4	72.75	-	-	-	-
Oltulu and Şahin (2011)	0.5, 1.25, 2.5	1.25	-	-	15.79	-	-
Zhang and Li (2011)	1, 3	1	-	18.04	-	-	-
Said et al. (2012)	3, 6	3	-	70	-	-	-
Behfarnia and Salemi (2013)	3, 5, 7	7	23.01	-	-	-	-
Oltulu and Şahin (2013)	0.5, 1.25, 2.5	1.25	-	-	10	-	-
Ghafari et al. (2014)	0.5, 1, 1.5, 2, 2.5, 3	3	33.33	-	-	-	31.9
Madandoust et al. (2015)	1, 2, 3, 4, 5	3	(NS 4%) 6	52	57	-	-
Mohseni et al. (2015)	1, 3, 5	3	3	52	-	-	-
Chithra et al. (2016)	0.5, 1, 1.5, 2, 2.5, 3	2	34.8	38.6	-	-	-
Ying et al. (2017)	1, 2, 3	2	-	26	-	-	-
Al Ghabban et al. (2018)	1, 2, 3, 4	3	22	-	-	-	-

Numerous researchers have investigated nano-silica effect on the reduction of water absorption of cement mortar and concrete. Water absorption is considered to be one the primary tests in evaluating cement-based materials durability. In this test, water penetrates into the matrix voids and saturates them (Bahadori and Hosseini 2012). Percentage of water absorption is an indicator of the porosity volume of the hardened cement-based material (Givi et al. 2011). To have a better understating about the effect of nano-SiO₂ on the reduction of water absorption of mortar and concrete, some examples have been provided. Givi et al. (2011) experimentally evaluated the changes that occur in the water absorption of concrete due to the use of nano-silica. They found that partial cement replacement with nano-SiO₂ led to reduction in the water absorption proportional to the content of NS. When 2.0 wt% NS was used, water absorption was found to decrease by about 1%, 21% and 21% for 7, 28 and 90 curing days, respectively. The effect was observed to increase as the age of the test specimens increased up to 28 days. No difference was observed between the effect of nano-SiO₂ on the 28-day and 90-day waster absorption. Ghafari et al. (2014) demonstrated that incorporating both silica fume and NS together decreased water absorption as the nano-silica replacement percentage increased up to 3.0 wt% and then started to increase. The optimum content was stated to cause a reduction in the water absorption of around 33%. Such behavior has been also reported by Nazari and Riahi (2011a).

Nano-silica effect on the water absorption of mortar has been studied by Madandoust et al. (2015) and Mohseni et al. (2015). They demonstrated that NS positive effect on the water absorption increased in proportion to the increase in its content in the mixture up to the optimum value and then started to decrease. However, they found that NS led to unremarkable reduction in the water absorption of such mortars. While Madandoust et al. (2015) reported a maximum reduction in the water absorption of around 6%, which was observed for 4.0 wt% NS, Mohseni et al. (2015)

reported it as 3% and was observed for 3.0 wt% NS. Moreover, Madandoust et al. (2015) stated that higher content of NS caused a revere effect on the water absorption. They stated that 5.0 wt% NS led to more water absorption than that of the reference samples. NS effect on the water absorption of mortar and concrete is graphically shown in Fig. 4(a). The figure. also presents the optimum contents of NS to reduce water absorption collected from different references and the related decrement percentages.

Permeability of cement-based materials has been also evaluated by studying chloride ion penetration. Chloride permeability has been recognized as a main inherent property influencing the durability of reinforced concrete (He and Shi 2008). The results of different research work conducted on the effect of NS on the chloride permeability of concrete and mortar showed that NS had an important potential to increase resistance to chloride ion penetration. Zhang and Li (2011) conducted an experimental investigation on the effect of silica nanoparticles on the chloride permeability of plain concrete. They demonstrated that 1.0 wt% and 3.0 wt% NS could reduce chloride permeability by around 18% and 10%, respectively. Similarly, Said et al. (2012) confirmed that the least chloride permeability in plain concrete was observed in samples incorporating 3.0 wt% NS. This content was found to reduce chloride permeability by 70%. Madandoust et al. (2015) researched the effect of NS on the chloride permeability of self-compacting mortar incorporating a constant amount of FA in all the mixes. They showed that NS incorporation resulted in a significant reduction in the chloride penetration for all the investigated percentages (1.0-5.0 wt%). The best performance was observed for 3.0 wt% NS, which led to a reduction in the permeability of 52%. Similar results were also reported by Mohseni et al. (2015). The optimum contents for the incorporation of NS into cement mortar and concrete to reduce their chloride permeability are shown in Fig. 4(b). The figure also presents the decrement percentage in chloride permeability caused by such contents.

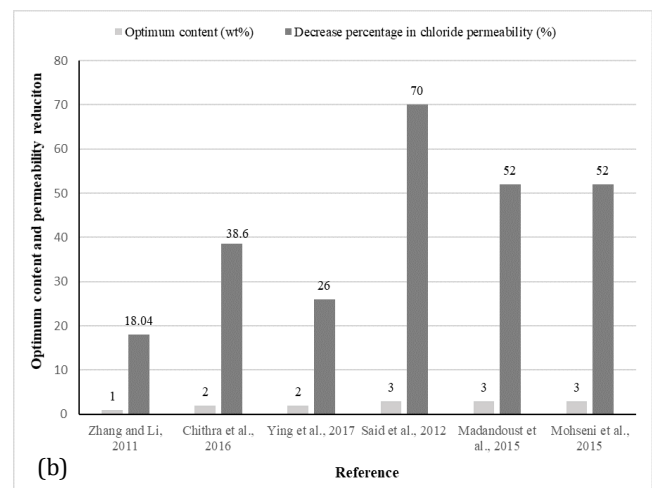
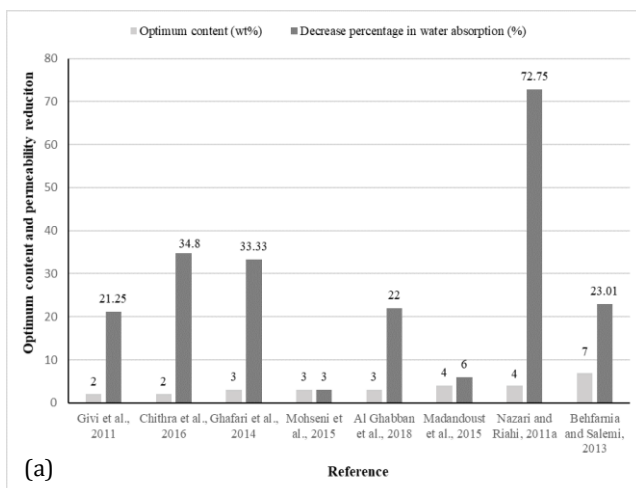


Fig. 4. (a) Different optimum contents of NS to reduce the water absorption; and (b) the chloride permeability of cementitious materials and the related decrease percentage.

Permeability have been also evaluated through finding capillary water absorption. Capillary water absorption is used to estimate the coefficient of water absorption due to the capillary action. In this test samples are partially immersed in water (Madandoust et al. 2015). Results of previous research showed that NS incorporation into cement-based materials could lead to an important reduction in their capillary water absorption. Nili et al. (2010) demonstrated that the capillary water absorption of concrete got reduced up to about 40% with the increase in the content of NS up to 3.0 wt%. However, higher content of NS (4.5 wt%) was observed not to cause much difference in the capillary water absorption compared to 3.0 wt% NS. Oltulu and Şahin (2011) investigated the effect of nano-SiO₂ on the capillary water absorption of mortar containing SF as 5.0 wt% of cement in all mixtures. The outcomes of their investigation showed that the capillary water absorption got reduced with the increase in the content of NS up to 1.25 wt% and then started to increase. When 1.25 wt% NS was used, the capillary water absorption was found to reduce by around 16% compared to the reference mortar.

Permeability has also been studied by finding water permeability coefficient and gas permeability coefficient of cement-based materials. However, the effect of nano-silica on these two parameters hasn't been sufficiently reported. Water permeability coefficient is a measure of permeability of water in cementitious materials (Nazari and Riahi 2011a). Givi et al. (2011) showed that partial cement replacement with nano-SiO₂ decreased the coefficient of water absorption of plain concrete. This effect was found to increase as the replacement percentage increased. The best results were observed in samples containing the highest investigated replacement percentage (i.e., 2.0 wt%). In this case a reduction in the coefficient of water absorption of around 8%, 51% and 30% at 7, 28 and 90 curing days, respectively, was observed. Gas permeability is primarily related to the percentage and proportion of the open porosity of the hydrated paste and the aggregates in the mixtures (Ghafari et al. 2014). Results reported by Ghafari et al. (2014) indicated that all mixtures incorporating NS (1.0, 2.0, 3.0 and 4.0 wt%) had lower gas permeability coefficient compared to the control samples. It was found that incorporation of 1.0, 2.0, 3.0 and 4.0 wt% NS decreased the coefficient of gas permeability of concrete containing copper slag by 7.5%, 24.2%, 31.9% and 25.7%, respectively.

Since the optimum value to reduce the different permeability parameters was found to be mostly the same, and comparing the results in Fig. 4 and Table 2, it can be said that the optimum content of nano-SiO₂ to reduce the permeability of the concrete and mortar is around 1.0-4.0 wt%. However, to find the overall optimum replacement percentage of cement with NS, we should compare the different obtained optimum values for both the improvement of mechanical properties and the reduction of permeability. Since the optimum content of NS for the improvement of mechanical properties was found to be 1.0-5.0 wt%, and the optimum content for permeability reduction was found

to be 1.0-4.0 wt%, it can be said that to get better performance in different properties of concrete and mortar, the content of NS should be 1.0-4.0% by weight of cement. This result is close to what Biricik and Sarier (2014) recommended. They stated that unless a good dispersion of nano-silica particles in the mixture is provided, the content of NS should be between 1.0-5.0 wt%.

4.2.2. Freeze and thaw resistance

Freeze and thaw is a critical problem that causes deterioration of cementitious materials, such as cracking and surface scaling. Freeze and thaw results in a remarkable adverse effects on permeability and mechanical properties of cementitious composites (Li et al. 2020). Nano-silica has been found to improve freeze and thaw resistance of cement-based materials. Ashwini and Srinivasa Rao (2021) investigated the effect of incorporation of 3% nano-silica and 15% alccofine by weight of cement on the freeze and thaw resistance of concrete. Their results revealed that concretes incorporating these additives had lower weight loss, reduction in compressive strength, reduction in density, reduction in ultrasonic pulse velocity and dynamic modulus of elasticity than normal concrete. Zhang et al. (2021) investigated the influence of NS on the freeze and thaw resistance of concrete containing coal fly ash. They found that low percentage of NS (up to 3 wt%) increased the freeze and thaw resistance of concrete remarkably. Higher content of NS showed a reverse effect on the freeze and thaw resistance. The optimum content was found to be 2 wt%. Zhao et al. (2021) conducted an experimental study on the effect of NS on the durability of concrete subjected to freeze and thaw and dry and wet cycles along with CaCl₂ attack. They found that nano-silica had a great effect on improving the environment resistance of concrete. 1 wt% nano-silica was found to be the optimum content to enhance durability. 1 wt% NS reduced the loss in mass, compressive strength and flexural strength by around 40%, 35%, 22%, respectively, compared to reference concrete, and increased the relative dynamic elastic modulus by around 10%. Shahrajabian and Behfarnia (2018) reported that nano-silica increased the freeze and thaw resistance of alkali-activated slag concrete. They found that smaller content of nano-silica performed marginally better than higher content. Behfarnia and Salemi (2013) studied the effect of nano-silica on compressive strength, mass loss, water permeability and change in length of concrete subjected to a number of freeze and thaw cycles. Their results indicated that nano-silica improved the frost resistance of concrete due to more compacted microstructure in the presence of nano additives.

4.2.3. High temperature resistance

High temperature has a great impact on the properties of cement-based materials. It leads to irreversible changes and a probability of total failure. Elevated temperature results in a sharp reduction in the performance of cement-based materials as the temperature increases (Ibrahim 2017; Sikora et al.

2018). One method to increase the high temperature resistance of cementitious materials is through the use of nanoparticle. Nano-silica has been found to reduce the bad effects of high temperature on strength of cementitious materials. Elkady et al. (2019) studied the effect of 1.5, 3 and 4.5 wt% nano-silica on the compressive strength and bond strength of concrete subjected an indirect elevated temperature of 200 to 600°C. Their results have proved that nano-silica can increase the bond strength of concrete and reduce the lost in strength due to high temperature. Their results showed that while 3 wt% nano-silica was the optimum content for concrete subjected to room temperature, 1.5 wt% NS behaved better when concrete was exposed to a temperature of 600°C. Results also revealed that high percentages of nano-SiO₂ had better mechanical properties than low percentages when the temperatures was between 200°C and 400°C. Reddy Babu et al. (2019) showed that NS increased the resistance of mortar to elevated temperature by up to 94% when 10 wt% NS was used. El-Gamal et al. (2018) reported that NS showed better high temperature resistance than normal concrete. 2 wt% nano-silica was found to be the optimum content to increase the high temperature resistance of cement pastes. Results revealed that as the temperature increased, the effect of silica nanoparticles on high temperature resistance decreased. Horszczaruk et al. (2017) studied the effect of NS (1-5 wt%) on properties of mortar containing different types of aggregates and subjected to high temperature (up to 800°C). The results showed that incorporation of NS up to 3 wt% improved the resistance to high temperature of a range up to 200°C. However, for higher temperatures its effect was less pronounced and not important. Their results also revealed that the effect of NS depended to a great extent on the type of aggregates used. Ibrahim (2017) reported a better residual strength of NS based modified concrete compared to normal concrete. Heikal et al. (2015) stated that the effect of nano-silica on high temperature resistance increased with the use of superplasticizer. Bastami et al. (2014) investigated the effect of high temperature (up to 800°C) on the mechanical strength, spalling, and mass loss of high strength concrete incorporating NS. Their research revealed that use of NS could importantly increase the residual compressive strength and tensile strength, and reduce the spalling and mass loss of concrete.

4.2.4. Fire resistance

Fire is considered to be one of the most severe conditions that a building might be subjected to (Mussa et al. 2021). NS has been reported to have a good effect on the fire resistance of cement-based materials. Mussa et al. (2021) studied the fire resistance of high-volume fly ash RC slab incorporating 2.5 wt% NS. They found that structural members with superior fire resistance could be produced with the incorporating of NS. Nano silica incorporated RC slab was found to have a better thermal conductivity and a higher residual compressive strength than plain RC slab. Tobbala (2021) evaluated the fire influence on compressive strength of concrete

incorporating NS. Their results revealed that concrete incorporating NS had good fire resistance compared to normal concrete. 1, 2 and 3 wt% NS was observed to increase the residual compressive strength by around 28, 33 and 48%, respectively. Vaz-Ramos et al. (2019) studied the influence of NS on thermal conductivity of mortar in order to evaluate its effect on fire resistance of cementitious composites. The results showed that NS resulted in a decrease in the thermal conductivity of mortar, which was related to evaporation of free water. High temperature was found to reduce the effect of NS on thermal conductivity of mortar by around 50%. NS was concluded to have the potential to increase fire resistance of cement-based materials. Ibrahim et al. (2012) evaluated the fire resistance of cement mortar containing high-volume fly ash and nano-silica subjected to high temperature. NS led to a higher residual mechanical strength and a less pore size distribution than normal concrete. NS effect on the residual compressive and flexural strengths was found to decrease remarkably as the temperature increased from 400°C to 700°C.

4.2.5. Sulfate attack resistance

Chemical attacks can have a great impact on the overall performance of cement-based materials, since they cause spalling and corrosion. Chemical attacks usually happen to structures exposed to marine environments (Li et al. 2020). Nano silica has been found to increase the resistance of cementitious composites to chemical attacks. One of the most commonly found chemical attack is the sulfate attack. Huang et al. (2020) investigated the impact of NS utilization on the sulfate attack resistance of mortar. They evaluated the sulfate attack impact by means of linear expansion, loss in strength and mass. The results indicated that NS increased the resistance to sulfate attack through the refinement of pores and the reduction of pore connectivity. The effect of NS was reported to be more pronounced in higher content (up to 5 wt%) and in bigger average particle size of NS. Reddy Babu et al. (2019) researched the effect of nano-SiO₂ on the resistance of mortar to magnesium sulfate (MgSO₄). They observed that nano-SiO₂ showed better durability performance under chemical attacks than normal concrete. Vargas et al. (2018) conducted an experimental study to evaluate the influence of NS on the durability performance of light weight concrete subjected to magnesium sulfate attack. They reported a higher resistance to the sulfate attack due to the use of NS. However, they found that chemical compositions and porosity of lightweight concrete are the main parameters in defining the durability performance of light weight concrete. Li et al. (2017) found that nano-silica increased the resistance of mortar to sulfate attack. Results revealed that NS reduced the loss in strength due to sulfate attack. This reduction was found to increase as the percentage of NS increased. The results showed that a combination of nano-silica and silica fume increased the resistance of mortar to sulfate attack more than the single use of either one of them.

4.3. Effect on microstructural properties

Many reports have been published on the effect of NS on the microstructural properties of cementitious composites. The reports have shown that nano-silica incorporation can greatly improve the microstructure of cementitious materials resulting in a significant reduction in the permeability and an important enhancement in the mechanical and durability properties (Al Ghabban et al. 2018; Arif et al. 2020; Bahadori and Hosseini 2012; Choolaei et al. 2012; Gaitero et al. 2008; He and Shi 2008; Jalal et al. 2012; Ji 2005; Kawashima et al. 2013; Liu et al. 2020; Ma and Zhu 2017; Quercia et al. 2014; Rong et al. 2015). Tests conducted to investigate the microstructural properties have shown that incorporating nano-SiO₂ into cement-based materials results in denser and more compact products (Ardalan et al. 2017; Arif et al. 2020; Behzadian and Shahrajabian 2019; He and Shi 2008; Khalaf et al. 2020; Ma and Zhu 2017). The effect of NS on improving the microstructure of cementitious composites may be ascribed to the following; NS has the ability as a nano-filler to reduce the porosity and fill the voids that exist in the structure of C-S-H gel, leading to a denser matrix (Aggarwal et al. 2015; Bahadori and Hosseini 2012; Cassar et al. 2003; Choolaei et al. 2012; Givi et al. 2011; He and Shi 2008; Jalal et al. 2015, 2012; Ji 2005; Khalaf et al. 2020; Liu et al. 2020; Quercia et al. 2013; Senff et al. 2009; Singh et al. 2013; Zhuang and Chen 2019). Moreover, because of the great pozzolanic reaction and the high content of SiO₂ of nano-silica, it can behave as an activator to accelerate the hydration of cement paste and react with Ca(OH)₂ leading to more production of C-S-H products (Aggarwal et al. 2015; Ardalan et al. 2017; Bahadori and Hosseini 2012; Choolaei et al. 2012; Gaitero et al. 2008; He and Shi 2008; Pengkun Hou et al. 2013; Jalal et al. 2015; Ma and Zhu 2017; Reches 2018; Senff et al. 2009; Zhuang and Chen 2019). Pengkun Hou et al. (2013) stated that low stiffness C-S-H products decrease and high-stiffness C-S-H products increase by the incorporation of NS. Crystals of Ca(OH)₂, which are hexagonal type of crystals, are located in the ITZ between aggregates and cement paste. These crystals have bad effect on the performance of cement-based materials. Addition of SiO₂ nanoparticle reduces these products and makes the ITZ denser and more compact and improves the overall characteristics of this zone (Ardalan et al. 2017; Bahadori and Hosseini 2012; Du et al. 2015; Jalal et al. 2015; Ji 2005; Said et al. 2012). Additionally, NS can modify the C-S-H internal structure by increasing the silicate chains length (Aggarwal et al. 2015; Cassar et al. 2003; Pengkun Hou et al. 2013). These reasons result in a more stable and more strongly bonded cement paste (Pengkun Hou et al. 2013). It should be mentioned that the positive effect of NS on microstructural properties of cementitious materials increases with the increase in the content of NS up to the optimum value and then starts to decrease, due to the reasons stated before.

Comparing the SEM images of samples with and without nanomaterials can give us a clear idea about the changes that happen in the microstructure of cementitious materials due to the use of nanoparticles. SEM observations of nano-SiO₂ incorporating concrete and mor-

tar have been investigated by a number of researchers (Ardalan et al. 2017; Arif et al. 2020; Biricik and Sarier 2014; Haruehansapong et al. 2014). Fig. 5 presents an example of the SEM images of cement mortar at curing age of 7 days with and without nanoparticles. The used NS was of 40nm particle size and was used as 9.0 wt% replacement of cement, which resulted in the maximum enhancement in the mechanical properties in the related reference (Haruehansapong et al. 2014) (see Table 1). The figure reveals that in the case of the control sample, C-S-H products exist in stand-alone clusters form. These products appear to be lapped and linked by numerous needle hydrates. Whereas, in the sample containing nano-silica, porosity has reduced remarkably, the microstructure has become much denser and more compact. It can be also observed from the figure that by incorporating nano-silica into the mix, the quantity of Ca(OH)₂ has reduced and the quantity of C-S-H has increased sharply. The SEM results support the previously concluded findings about the effect of nano-silica on the mechanical properties and permeability and also illustrate the behavior of nano-silica. Similar results were also observed by Biricik and Sarier (2014), who found a heterogeneous dispersion of C-S-H gel and Ca(OH)₂ (CH) crystals and needle-like ettringite crystals in the reference mortar after 7 days of curing. Some micro cracks were also observed in the structure. They reported that incorporation of 10 wt% nano-SiO₂ resulted in a very condensed microstructure of mortar. They also stated that the Ca(OH)₂ grains in the samples containing NS were less visible than in the control samples. They demonstrated that after 28 days of curing, the microstructure of the specimens got denser and more compact. Large Ca(OH)₂ crystals weren't seen through the structure (Fig. 6).

5. Cost Effectiveness of Use of Nano-silica

Nano materials are expensive materials, which have minimized their use in the construction industry, despite of their numerous advantages (Adamu et al. 2021; Crucho et al. 2018; Janković et al. 2016; Mor et al. 2017; Reddy et al. 2020; Safiuddin et al. 2014; Sowjanya and Adishesu 2022; Varisha et al. 2021). The high price of nano materials is related to the expensive technology and equipment involved (Crucho et al. 2018). Nano-silica has been reported to cost \$185 per kg (Zidi et al. 2021). Despite the fact that nano-SiO₂ have been proven to improve the mechanical properties of cement-based materials, these improvements might not justify the huge increase in price of cement-based materials due to the use of nano-silica. However, a life-cycle cost assessment might justify the use of these materials, since nano-silica increases the durability and sustainability of cement-based materials and reduces the maintenance cost (Safiuddin et al. 2014). For this reason, a life-cycle cost analysis of NS incorporating cement-based materials is an important subject to be researched. The cost of nano-silica might be reduced through the production of NS using cost effective methods and through a higher production rate (Mor et al. 2017; Safiuddin et al. 2014). The cost of NS containing cement-based materials can be reduced

by reducing the content of NS in these materials, since higher percentages cause lower or even adverse effects on their properties (Adamu et al. 2021; De la Varga et al. 2019). More research is needed to assess the cost effec-

tiveness of nano-silica use in cementitious materials, and to develop new cheaper and more practical methods of production that may lead to more production and incorporation of such beneficial nanomaterials.

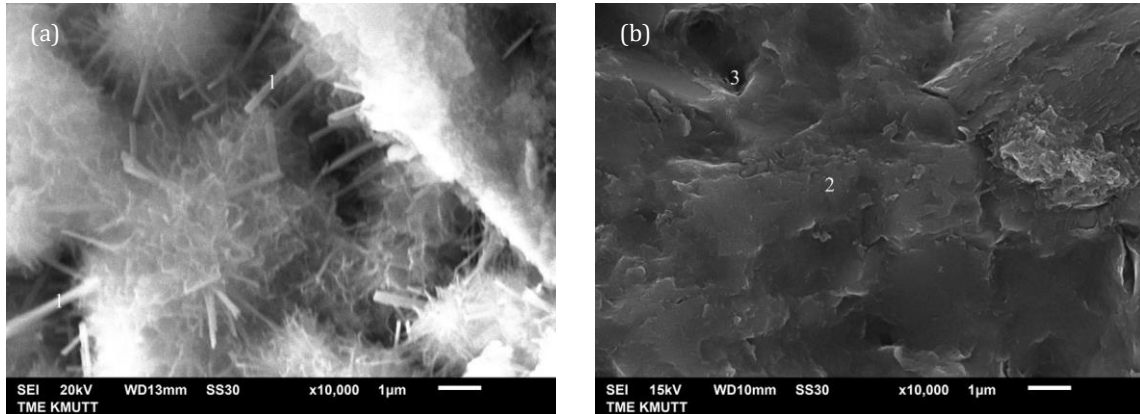


Fig. 5. SEM observations of mortar samples after 7 days of curing: (a) without NS; (b) with 9.0 wt% NS. 1=CH crystal, 2=C-S-H, 3=pore (Haruehansapong et al. 2014).

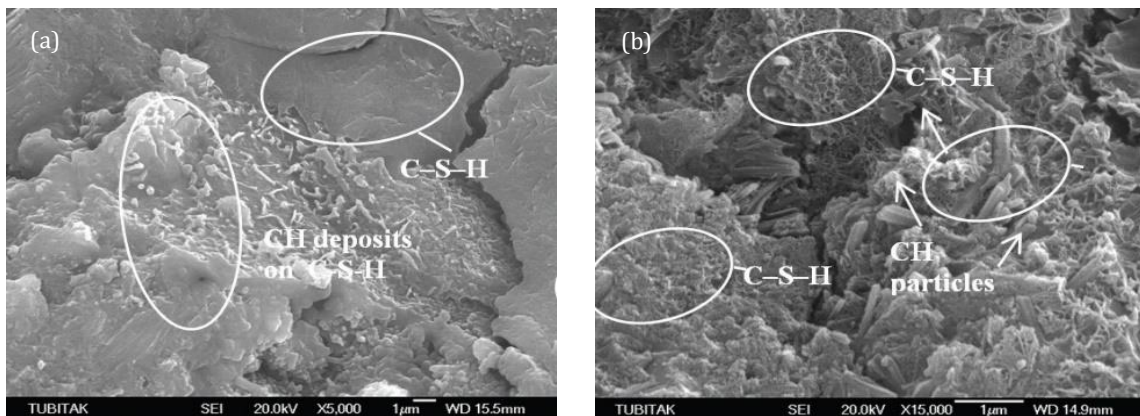


Fig. 6. SEM observations of mortar samples after 28 days of curing: (a) without NS; (b) with 10 wt% NS (Biricik and Sarier 2014).

6. Conclusions

The present paper discusses the pros and cons of use of NS and reviews the effect of its incorporation on different properties of cement-based materials. Some of the outcomes of the present study can be summarized as:

Nano-SiO₂ incorporation into mortar and concrete with even a small content can remarkably enhance their mechanical properties. This effect is generally more pronounced at early ages than at later ages. The enhancement in strength might be attributed to the behavior of nano-SiO₂ as a nano-filler to improve the microstructure, as well as to the hydration acceleration effect of such nanoparticles. In addition, due to its high pozzolanic reactivity, nano-SiO₂ reacts with Ca(OH)₂ and leads to more formation of C-S-H gel.

Nano-silica reduces the permeability of mortar and concrete and increases their resistance to water, chloride and gas penetration. Incorporation of nano-SiO₂ reduces the porosity by recovering the particle packing density, and leads to denser, more compact, less penetrable and more homogeneous microstructure. NS also

increases the resistance of cementitious materials to freeze and thaw effect, elevated temperature, fire and sulfate attack.

As a result of its effect in improving the performance of mortar and concrete, nano-silica can improve the durability and sustainability of structures, and reduce their repair and maintenance costs.

The positive effect of nano-SiO₂ on different properties of mortar and concrete increases with the increase in the content of this nanomaterial in the mixture up to the optimum content and then starts to decrease. This reduction in the effect of NS after exceeding the optimum content might be ascribed to the agglomeration and unsuitable dispersion of nanoparticles which lead to formation of weak zones. In addition, the amount of SiO₂ nanoparticles that exist in the paste might be exceeding the needed amount to react with the liberated Ca(OH)₂ particles.

Despite the large amount of research carried out on the effect of nano-SiO₂ on different properties of cementitious materials, there still exists a big lack of information related to this topic. In addition, contradictory

results have been reported in different research work. For these reasons, finding a specific optimum content for nano-SiO₂ is of a big difficulty. In spite of that, as a guideline, the optimum replacement percentage of cement with nano-SiO₂ in concrete and mortar to improve their overall performance can be considered as 1.0-4.0 wt%. However, more research is required to confirm these results.

Although use of nano-silica in cementitious materials has great advantages, it also has some disadvantages, such as the high price of nanoparticles that need to be considered in future research. More research is also needed to fully understand the effect of such nano-material on different properties of cement-based materials, and overcome the lack of information related to this topic.

More research on the relationship between the optimum content of nano-additives and the different factors that influence it, such as the properties of the used nano-SiO₂ (specific surface area, dispersion etc.) and the composition of the cement pastes (w/c ratio, content of cement, the addition of other additives etc.), is encouraged. Moreover, there is only little research on the effect of nano-silica on the dynamic properties of cementitious materials, which should be further researched. Future research should also focus on developing equations that describe the relationship between the content of nano-silica and the properties of cement-based materials. The optimum content of other nano-additives to best improve the properties of cement-based materials should also be investigated.

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