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pp. 49 –63

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## Research paper

# Comparative analysis of natural nanosilica versus commercial nanosilica on compressive strength and durability of high-performance concrete

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**Abstract:** This study provides a comparative analysis of natural nanosilica (NSn), which is an extract of natural silica sand processed into nanosilica with commercial nanosilica (NSc) derived from semiconductor industrial waste, in 80 MPa high performance concrete (HPC). The percentage of using nanosilica is (3%, 5%, 10%, 15%) by weight of cement used directly and combined with 5% silica fume. Analysis was carried out through compressive strength test, durability through permeability test, rapid chloride penetration test (RCPT), and microstructure test through scanning electron microscopy (SEM). The results of the analysis show that natural nanosilica is equivalent to commercial nanosilica, in applications it is better to use silica fume incorporation. The optimum percentage of using NSn10% and (SF) 5%, while 5% NSc and 5% SF, in these proportions shows the best compressive strength and durability. It's just that the use of natural nanosilika is 5% more than commercial nanosilika. The benefit of this research is that natural materials such as silica sand with high SiO2 content, can be processed into nanosilica as an advanced material, which can be used as an eco-friendly construction material.

**Keywords:** Natural nanosilica, commercial nanosilika, compressive strength, durability, High Performance Concrete, eco-friendly

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## 1. Introduction

A.R.I. TJAHJANI, et al.

The use of cement as the main construction material continues to increase, it is estimated that by 2017 world cement production will reach 4 billion tons [1–4]. But unfortunately, cement production has an impact on the environment because producing 1 ton of cement will release 1 ton of CO2 into the air [5–7]. Various attempts have been made to overcome this problem, one of which is by using materials supplementary cementitious materials such as fly ash, silica fume, and slag to replace cement [8,9].

The development of materials in the field of construction is increasingly advanced, triggered by the application of nanotechnology and knowledge of cement-based nanoscience. Nanotechnology produces nanomaterials with nanometer size, has a very large surface area and is very reactive [10–14].

Several researchers have carried out nano-engineering for cement base materials, it turns out that it has tremendous potential in applications. The results showed that the compressive and tensile strength of concrete will increase with the use of nanosilica, especially at the initial age and the initial strength of the concrete will be smaller with the use of silica fume, after which it will increase as the age of the concrete increases. This shows that the reactivity of nanosilica pozzolan is greater than that of silica fume pozzolan [15, 16]. The effect of different sizes of SiO<sub>2</sub> nanosilica affects the compressive strength, for example, nanosilica used 15 nm and 80 nm, the compressive strength of concrete with nanosilica 15 nm produces a greater compressive strength compared to 80 nm nanosilica [17, 18].

Nanosilica is a relatively new material, therefore research on its use is increasingly widespread. One of the efforts is to use a combination with other materials such as silica fume or other materials and the effect of using various types of nanosilica [19–21]. One of the materials developed by nanotechnology is natural nanosilica material (NSn) derived from natural materials, namely silica sand natural.

This research is to carry out a comparative analysis using natural nanosilica compared to commercial nanosilica (NSc), when used alone and when combined with silica fume in high performance concrete, on the compressive strength and durability of concrete. In the concrete mixture, a polycarboxylic ether type superplasticizer is used which can increase the compressive strength [22].

The benefit of this research is to understand the effect of using natural nanosilica on commercial nano so that it can be used as a basis for the use of natural nanosilica materials.

## 2. Material and methods

The material used is the ordinary portland cement used by the tiga roda brand comes from PT. Indocement Tunggal Perkasa. In the present study, Ordinary Portland Cement (OPC) conforming to ASTM Type-I (ASTM 2015) [23], natural nanosilica (NSn) is nanosilica derived from natural silica sand which is processed into nanosilica through the Polishing Milling Technology process with a particle size of 40–80 nm [24] Nanosilica Industry Waste (NSc) brand HDKN 20 from Bratacem with a particle size of 20 nm, Silica

fume (SF) and superplasticizer (SP) brand Sika. Table 1 shows the chemical composition of the materials used, while Table 2 shows the proportions of the material for 1 m<sup>3</sup>. BR is the reference concrete used as a control. NSn3 is natural nanosilica as much as 3%, NS5 as much as 5%, NSn10 as much as 10% and NSn15 as much as 15%. Codes NSc3-NSc15 and NSc3SF5 to NSc15SF5 are the same, only nanosilica is used for commercial nanosilica.

Natural Commercial Silica Cement Composition nanosilica nanosilica (OPC) (%) Fumes (%) NSn (%) (NSc) (%) SiO<sub>2</sub> 99.60 99.99 19.0-21.0 87.74 4.0 - 6.0 $Al_2O_3$ 0.720 Fe<sub>2</sub>O<sub>3</sub> 0.32 2.5 - 3.51.63 0.520 CaO 62.0-67.0 MgO 1.0-4.0 55-64  $C_3S$ 9-20  $C_2S$ 

7 - 11

9-11

 $C_3A$ 

 $C_4AF$ 

CuO

BaO

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0.08

Table 1. Chemical analysis of materials

Table 2. The proportions of the material for 1 m<sup>3</sup>

0.01

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No	code	Cement (Kg)	NSn (Kg)	NSc (Kg)	Ratio water/ binder	Sand (Kg)	Coarse aggregate (Kg)	SP (Liter)
1	BR	900		-	0.23	638	1094	15
2	NSn3	900	27	_	0.23	638	1094	15
3	NSn5	900	45	_	0.23	638	1094	15
4	NSn10	900	90	_	0.26	638	1094	20
5	NSn15	900	135	-	0.3	638	1094	22
6	NSn3- SF5	900	27	-	0.24	638	1094	15
7	NSn5- SF5	900	45	-	0.25	638	1094	15

Continued on next page

52

No	code	Cement (Kg)	NSn (Kg)	NSc (Kg)	Ratio water/ binder	Sand (Kg)	Coarse aggregate (Kg)	SP (Liter)
8	NSn10- SF5	900	90	_	0.27	638	1094	21
9	NSn15- SF5	900	135	_	0.3	638	1094	24
10	NSc3	900	_	27	0.23	638	1094	15
11	NSc 5	900	_	45	0.23	638	1094	15
12	NSc10	900	_	90	0.26	638	1094	20
13	NSc15	900	_	135	0.3	638	1094	22
14	NSc3- SF5	900	-	27	0.24	638	1094	15
15	NSc- SF5	900	ı	45	0.25	638	1094	15
16	NSc10- SF5	900	_	90	0.27	638	1094	21
17	NSc15- SF5	900	_	135	0.3	638	1094	24

# 2.1. Compressive strength testing

The compressive strength test of concrete specimens at the age of 1, 3, 7 and 28 days was carried out following ASTMC109/C109M (ASTM 2016) [25], an average of three specimens for each mixture.

## 2.2. Concrete permeability testing

Concrete permeability testing was carried out according to DIN 1045 [26]. The specimens used were concrete blocks measuring  $200 \times 200 \times 120$  mm and the concrete was aged 28–35 days.

# 2.3. RCPT testing

Rapid chloride ion permeability test (RCPT) was conducted as ASTM C1202 [27] at the curing ages of 28 days. An average of three specimens of each mix was considered as the charge passing value, with a cylindrical specimen size of 100 mm in diameter and a height of 50 mm.

### 3. Result and discussion

COMPARATIVE ANALYSIS OF NATURAL VERSUS COMMERCIAL NANOSILICA . . .

# 3.1. Effects of natural silica nano on concrete compressive strength

Figure 1 the following shows the change in compressive strength at various ages of concrete up to 28 days as a result of using nanosilica. Based on the figure, the compressive strength of reference concrete is 87.7 MPa, using NSn5 increases the compressive strength to 93.8 MPa (7%), and NSn10 increases the compressive strength to 89.6 MPa (2%), while NSn3 does not increase the compressive strength.

In the case of using NSn15, the compressive strength decreased to 48.1 MPa (45%), this was due to agglomeration as shown in Figure 1. The agglomeration effect resulted in the nanoparticles not being dispersed with the cement matrix which caused pores to occur, so the concrete was not homogeneous, in line with researchers [28–30].

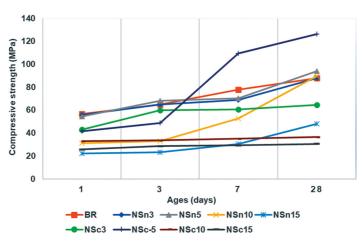


Fig. 1. The effect of using NSn and Nsc on compressive strength

# **3.2.** Effects of commercial nanosilica (nsc) on concrete compressive strength

Figure 1. shows the effect of using NSC on the compressive strength of concrete at various ages of concrete. Based on the figure, the use of NSC–5 increased the compressive strength to 126.1 MPa (43.8%), while NSC–3, NSC–10, and NSC–15 each decreased the compressive strength to 64.6 MPa (26.3%), 34.4 MPa (58.5%) and 30.5 MPa (65.2%). This shows that the smaller the NSC particle size is 20–40 nm, compared to NSN = 36–80 nm, the greater the agglomeration effect occurs, resulting in a decrease in the compressive strength of concrete [31,32].

# 3.3. Effects of natural nanosilica and sf 5 on concrete compressive strength

In this study, the use of silica fume was taken at 5% and the effect on the compressive strength of concrete in the NS and NSHD cases was presented in Figure 2 respectively. Based on this figure, the use of NSN–5 and SF5 increased the compressive strength to 91.6 MPa (4.4%) and the use of NSN–10 and SF5 increased the compressive strength to 131.7 MPa (50%). On the other hand, the use of NS 3 and SF-5 reduced the compressive strength to 84.4 MPa (3.8%) and the use of 15% NS and 5% SF reduced the compressive strength to 57.8 MPa (34%).

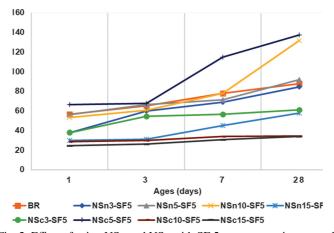


Fig. 2. Effect of using NSn and NSc with SF 5 on compressive strength

Based on the figure, the use of NSc5 and SF5 increased the compressive strength to 137.3 MPa (56.6%), and the use of NSc10 and SF5 increased the compressive strength to 131.7 MPa (50.2%). Nano-SiO<sub>2</sub> improves because it has pozzolanic properties and provides a filler effect. Nano-SiO<sub>2</sub> particles are a very pozzolanic material because they consist of SiO<sub>2</sub> in an amorphous form with a high specific surface, while the use of NSc3 and SF5 reduces the compressive strength to 61 MPa (30.4%), and the use of NSc 15 and SF 5 reduced the compressive strength to 33.7 MPa (61.6 %).

Figure 2 the following shows the change in compressive strength at various ages of concrete up to 28 days for the use of NSc5 and SF5. The highest compressive strength was achieved at 28 days of concrete. In the figure it can be seen that the use of NSc5 and SF 5 produces a compressive strength of 137.3 MPa [33–35].

# 3.4. Permeability testing

The results of the permeability test for BR using NSn, NSc and SF can be seen in Figures 3 and 4. As previously explained, the standard used is DIN 1045, which states that concrete is said to be watertight if water is pressed against the surface of the specimen

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with a pressure of 5 bar for three days, and only experienced penetration of less than 5 cm. The permeability value is measured based on the penetration of water into the concrete specimen at the age of 28 days.

COMPARATIVE ANALYSIS OF NATURAL VERSUS COMMERCIAL NANOSILICA...

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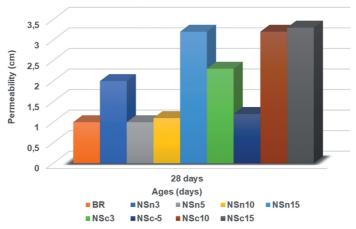


Fig. 3. Use of nanosilica NSn and NSc for water penetration in concrete

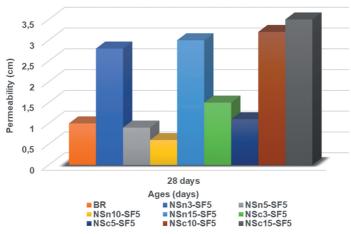


Fig. 4. Use of NSn and NSc with SF on water penetration in concrete

#### 3.4.1. Effect of NSN on concrete permeability

Based on Figure 3 the use of NS with the percentage of NSn3 increases the permeability value to 2.0 cm (100%), and 5% NS does not change the permeability value. The use of NSn10 increased the permeability to 1.1 cm (10%) and the use of NSn 15 increased the permeability value to 3.0 cm (220%). This shows that the use of nanosilica in general has practically no effect on the permeability of concrete and even tends to increase the permeability value. However, specifically for the use of NSn10 and SF 5, the permeability

can be reduced to 0.6 cm (40%), because the pozzolanic properties give rise to a better hydration phase and a denser microstructure [36, 37].

#### 3.4.2. Effects of NSN and SF on concrete permeability

The use of NSn used together with SF is shown in Figure 4. In NSn–SF5 and NSn–SF5, the permeability value increased to  $2.8~\rm cm$  (180%) and increased to  $3.0~\rm cm$  (200%). While the use of NSn5 and SF 5, NSn10 and Sf5, each lowered the permeability value to  $0.9~\rm cm$  (10%) and to  $0.6~\rm cm$  (40%). This shows that the use of nanosilica NSn used together with SF at the percentage of NSn10 and SF5 is the optimum value that can be used to reduce permeability.

The increase occurs because NSn and SF will disperse and react with  $Ca(OH)_2$  crystals as a pozzolanic material, this will make the cement matrix more homogeneous and compact, reduce capillary pores so that the concrete is more impermeable and the compressive strength increases. [38, 39].

#### 3.4.3. Effect of NSC on concrete permeability

The use of NSc3 increased the permeability value to  $2.3~\rm cm$  (130%), 5% NSHD increased the value to  $1.2~\rm cm$  (20%), NSc 10 increased to  $3.2~\rm cm$  (220%) and NSc15 increased the value permeability to  $3.3~\rm cm$  (230%). This shows that the use of NSc has no effect on the permeability of concrete and even tends to increase the value of permeability, however, the resulting concrete is still considered water-resistant concrete because the penetration limit is less than  $5~\rm cm$ .

#### 3.4.4. Use of NSC and SF on concrete permeability

The use of NSHD which was used together with SF, for NSc3SF5, NSc10SF5 and NSc15SF5 there was an increase in permeability values of 180%, 220%, and 250%. In NSc5SF5 there is an increase of 10%. These results indicate that the use of NSc5 and SF5 is the optimum value. While the effect of using NSn, NSc and SF on permeability can be seen in Figure 4, in this figure it can be seen that the addition of NSn10 and SF 5 turned out to have a lower permeability value of 0.6 cm compared to the use of NSc5 and SF5 which had a permeability value of 1.1 cm. This is as [40], that the cement paste added with nanosilica will increase the CSH gel chain length and significantly increase CSH.

# 3.5. Rapid chloride penetration test

This test was carried out at the Structure and Materials Laboratory, Bandung Institute of Technology. RCPT test results for reference concrete and the use of NSn, NSc and SF for percentage variations, can be seen in Figures 5 and 6.

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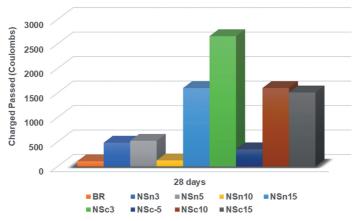


Fig. 5. Effect of using NS on charge passed concrete

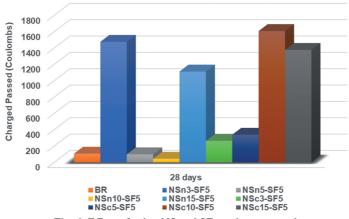


Fig. 6. Effect of using NS and SF on charge passed

## 3.6. Effect of NSN on charge passed concrete

The effect of using NSn on the charge passed in concrete at various ages is shown in Figure 5. In reference concrete, the charge passed was 107.1 Coulombs, so it is in the very low category. The use of NSn3 increased the charge passed by 353.8% to 486 Coulombs, including in the very low category. The use of NSn 5 increases the charge passed by 390% to 525 Coulombs, which is in the very low category. The use of NSn10% increased the charge passed by 16.8% to 125.1 Coulombs, including in the very low category. The use of NSn15 increases the charge passed by 1395%, to 1602 Coulombs, which is included in the low category. This shows that the use of nanosilica NSn tends to increase the charge passed in concrete, although in general, it is still in the very low to low category.

#### 3.6.1. Effects of NSN and SF on charge passed concrete

A.R.I. TJAHJANI, et al.

Figure 6 shows that the use of NS causes a decrease in the 107.1 Coulombs charge on the reference concrete, so it is included in the very low category. The use of NSn3 and SF5 increased the charge passed by 1278% to 1476 Coulombs, which is included in the low category. The use of NSn5 and SF5 reduced the charge passed by 7.6% to 99 Coulombs, including in the negative category. The use of NSn10 and SF5 reduced the charge passed by 58% to 45 Coulombs, including in the negligible category. The use of NSn15 and SF5 increases the charge passed by 938%, to 1112 Coulombs, which is included in the low category. It turns out that NS and SF can rupture capillaries in concrete, causing cavities to become more tortuous, disconnected and difficult to penetrate [41].

This shows that the use of nanosilica NSn and SF at the percentage of NS10 and SF 5 produces the optimum charge passed. This shows that NSn and SF can be used together effectively.

#### 3.6.2. Effect of NSC on charge passed in concrete

Figure 5 shows the use of NSc there is an increase in charge passed. In reference concrete the charge passed 107.1 Coulombs, very low category, the use of NS 3% increased the charge 2661 Coulombs (2384.6%) including the medium category, NSc5 charge passed 351 Coulombs (16.8%) very low, NSc10 charge passed 1602 Coulombs (1395.8%) was low, and NSc 15 experienced an increase in charge passed by 1512 Coulombs (1311.8%) in the low category.

#### 3.6.3. Effects of NSC and SF on charge passed in concrete

Figure 6 shows the use of NSc and SF, in reference concrete the charge passed 107.1 Coulombs, very low category, the use of NSc3 and SF5 increased the charge value of 264 Coulombs (146.5%), very low, NSc 5 and SF5 decreased charge passed 339 Coulombs (216.5%), very low, on NSc10 and SF5 there was an increase in charge passed 1608 Coulombs (1401.4%), low, on NSc15-SF5 there was an increase in the value of charge passed 1377 Coulombs (1185.7%), including the low category. It turns out that the use of NSc and SF tends to increase the charge passed in concrete, although in general, it is still in the very low to a low category, this is because nanosilica plays a more important role [42–45].

### 3.7. Scanning electron microscope

Based on the SEM results for reference concrete shown in Figure 7a, for NSn10-SF5 7b and NSc5-SF5 7c concrete, it appears that the use of natural nanosilica and silica fume becomes denser.

COMPARATIVE ANALYSIS OF NATURAL VERSUS COMMERCIAL NANOSILICA . . .

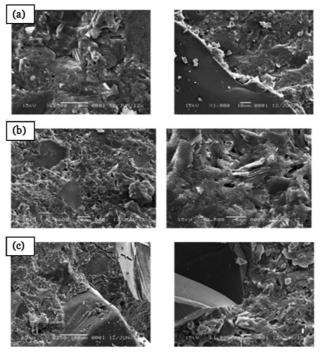


Fig. 7. SEM (a) BR, (b) NSn10-SF5 and (c) NSc5-SF5

# 3.8. Microstructure

Figure 8 shows the SEM results for BR. From the figure, it can be seen that the formation of CSH in the reference concrete is not as dense as in NSn10-SF5 and NSn5-SF5

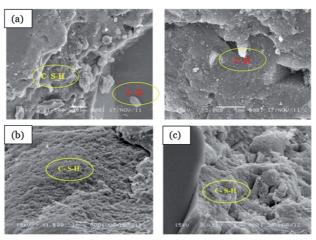


Fig. 8. SEM (a) BR (b) NSn10-SF5 and (c) NSc5-SF5

concrete (c). Thus, the use of natural and commercial nanosilica combined with silica fume can produce denser concrete [46–48].

Nanosilika reacts with calcium hydroxide crystals, reducing the size of the crystals in the interface zone and becoming weak calcium hydroxide crystals to become CSH crystals and increasing the Interfacial Transition Zone to become denser than cement paste and concrete in Figure 9.

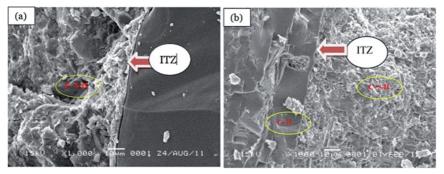


Fig. 9. SEM (a) BR, (b) NSn10-SF5

# 4. Conclusions

- 1. The combination of using natural nanosilika with silica fume is more effective than natural nanosilika without silica fume, whereas for commercial nanosilica, the combination of usage does not show a significant effect.
- 2. The optimum percentage for using natural nanosilica (NSn) is 10% and SF is 5%. This optimum percentage can increase the compressive strength by 50%, reduce the permeability value by 10% and become 40%, the value of the passed charge is 58%.
- 3. In commercial nanosilica the optimum percentage of (NSc) 5% with and without 5% SF increases the compressive strength by 56.6% reduces the permeability value by 10% and the value of Passed charge is 216.5%.
- 4. The percentage of using nanosilica which exceeds 10% decreases the mechanical properties and durability of concrete. This is due to agglomeration. Therefore, to avoid agglomeration effects, no more than 10% of nanosilica is used.
- 5. Observations through scanning electron microscopy (SEM), HPC which uses natural nanosilica (NSn) and commercial nanosilika (NSc) combined with SF produces denser concrete and denser or smaller ITZ.

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#### COMPARATIVE ANALYSIS OF NATURAL VERSUS COMMERCIAL NANOSILICA...

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