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Optimizing Sonication Time and Solid to Liquid Ratio of Nano-Silica in High Strength Concrete

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Abstract— Nano-silica had been investigated as a partial replacement of cement by many researchers, regardless its high agglomeration specially when mixed to water. Sonication is one of the methods used to de-agglomerate nano-silica particles and to improve the dispersion of nano-silica in cement matrix. This research aims to reach the optimum indirect sonication time and solid to liquid ratio that enhances the dispersion of nano-silica and consequently increases its reactivity in high strength concrete. Particle size distribution of the sonicated nano-silica particles and the corresponding specific surface area were the main keys governing the optimization process of the studied parameters. Using the optimum solid to liquid ratio with the corresponding sonication time, slump and the compressive strength of concrete were examined and compared with those without nano-silica. In addition the microstructural analysis using SEM , and XRD helped in confirming the compressive strength results. The results revealed that the optimum solid to liquid ratio is 1:10. Moreover for every concentration, there is an optimum indirect sonication time. The optimum sonication time found to be 5 minutes for solid to liquid ratio 1:10. By using the optimum concentration an improvement in compressive strength of 32% and 24% after 7 and 28 days were reached as compared to the control mix. Where in terms of slump, there is no change in the flowability of concrete due to the incorporation of nano-silica in concrete at the same mix proportions.

Key Words: — Nano-Silica, Indirect Sonication, Solid to Liquid Ratio, Workability, Compressive Strength, Absorption, Microstructure Analysis.

1. Introduction

HIGH strength concrete is characterized by its high mechanical and durability properties. These superior properties can be achieved by using high cementitious content with very low water to cementitious materials and lowering maximum aggregate size. The use of pozzolanic materials (silica fume, ground granulated blast furnace slag, and fly ash) is essential for the production of high strength concrete [1-2]. Silica fume belongs to the category of highly pozzolanic materials because of its high purity and consists of silica in non-crystalline form with a high specific surface and could be effectively promot the strength development of concrete, compared to the other pozzolanic materials [3]. On the other hand the development of nanotechnology can produce silica in another form with a higher specific area, and more functions compared to conventional silica fume [4-5]. This can be explained that nano-silica can improve the strength and durability characterizes of cement and concrete [6-7-8]. This improvement was due to their fine size which can fill the pores of the cement matrix. Also, due to the nucleation site effect and the pozzolnaic reactivity of the nano-silica which improves the microstructure of the cement composites [9].

Nano-silica should be dispersed to de-agglomerate its particles, in order to optimize the utilization of the nano-silica particles within the cement matrix. Sonication is one of the dispersion methods used to improve the effectiveness of nano-silica to modify its granulometric distribution and to improve its densification [10-11]. Serag et al [12] had studied the reactivity of nano-silica using different techniques; homegenizier, stirrer and indirect sonication. The results indicated that the reactivity significantly enhanced by the use of indirect sonicator. Elkady et al [13] investigated the effect of changing the indirect sonication time on the dispersion and the reactivity of nano-silica as a cement substation in concrete mixes. The results showed that; 5 minutes of indirect sonication to nano-silica prior being added to concrete leads to an improvement in workability and gaining in compressive strength reached 33% compared to the control mix. The effect of solid to liquid ratio, and the time of sonication weren't taken into consideration as factors affecting the sonication process. Nazari and Riahi [14] had found an improvement in the compressive strength reaches 33% than the control mix when using 4% nano-silica as a cement replacement. However, E. Ghafari et al. [15] had studied the effect of using two different types of nano-silica in UHPC. When using nano-silica at a fixed dosage 5% of the cement weight, the compressive strength was improved by 11.4% and 5.9% after 7 and 28 days, respectively than mixes containing 0% nano-silica. The flexural strength was improved by 28.6% and 14.3% after 7 and 28 days, respectively, and the water absorption was reduced by 36% than without nano-silica.

This paper aims to find the optimum dispersion of Nano-Silica particles, through studying the effect of different solid to liquid ratios on the particle size distribution and specific surface area of the sonicated nano-silica particles. The effect of optimum solid to liquid ratio of nano-silica and sonication time on the fresh (workability), and hardened (compressive strength and absorption) properties of nano-silica high strength con-

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crete will be studied. In addition microstructure analysis (XRD) will be introduced to study in depth the effect of sonicated nano-silica on the micro structure of the cement matrix.

2. EXPERIMENTAL WORK

2.1 Materials:

Ordinary Portland Cement (OPC) complying with the requirements of ASTM C150 standard was used. Silica fume used was produced by the Egyptian ferro-alloys company in Aswan, Egypt. Chemical and physical properties of the used cement and silica fume are listed in table 1. The used Nano-silica was produced from the physic depatement in the national research and housing center in Egypt, with average particles size 30 nm. The properties of nano-silica are shown in table 2. Transmission electron micrographs (TEM) and powder X-ray diffraction (XRD) diagrams of nano-silica particles are shown in figs. (1 and 2). Natural silicious sand free of alkali-reactive materials from Suez quarries, in Egypt was used. The used crushed dolomite is complying with the requirements of ASTM C33/C33M. The properties of fine and coarse aggregate are listed in table 3. The mix proportions are shown in table 3. Sika Viscorete 3425 was used as a polycarboxilic ether additive.

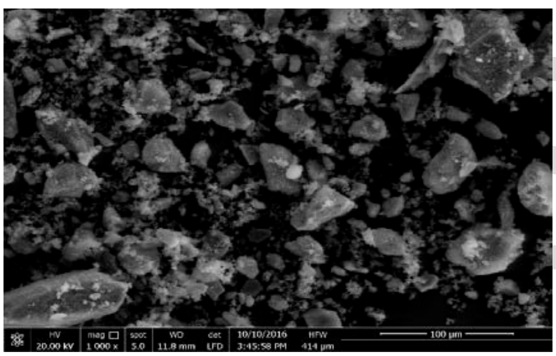


Figure 1: TEM Microscopy of Nano-Silica

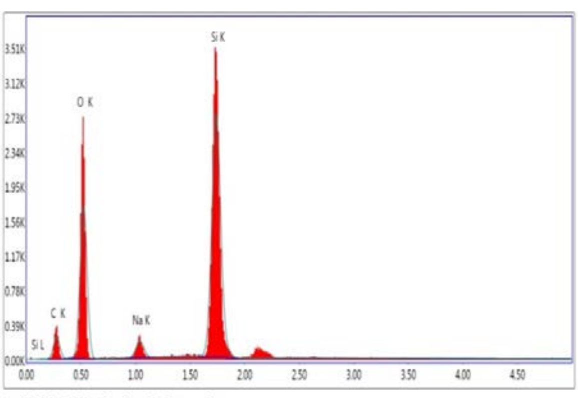


Figure 2: XRD Analysis of Nano-Silica

Table 1: Chemical Composition of Portland Cement, Silica fume, and Nano-Silica

Element	Cement	Silica Fume	Nano-Silica
SiO_2	20.13	92	99.1
Al_2O_3	5.32	1.1	0.13
Fe_2O_3	3.61	1.45	0.06
CaO	61.63	1.2	0.14
MgO	2.39	1.55	0.11
SO_3	2.87	0.25	-
Na_2O	0.37	0.45	0.4
K_2O	0.13	1.45	-
P_2O	-	-	0.01
L.O.I	1.96	-	-

Table 2: Properties of Sand and Crushed Dolomite

Property	Sand	Gravel
Specific Weight	2.89	2.76
Bulk Density	1.67	1.82
Fineness Modulus	2.75	
Water aAbsorption (%)	-	1.85
Crushing Value (%)	-	14.3
Clay and Fine Dust Content (%)	1.95	0.55

2.2 Experimental Program:

The experimental program is divided into two phases; the first phase aims to optimize the liquid to solid ratio of nano-silica using indirect sonication on different durations. Where a group of 4 different concentrations of nano-silica; 1:30, 1:15, 1:10, and 1:7.5, were sonicated for 4 different durations; 0, 5, 10, and 15 minutes. The optimum concentration was chosen based on the optimum particle size distribution, and specific surface area of the sonicated samples.

In the second phase the effect of nano-silica on the slump, the compressive strength, and on the rate of absorption was studied using the optimum concentration and with the corresponding indirect sonication time. Four mixtures were prepared in this study. The compressive strength of concrete was measured at ages of 7 and 28 days using concrete cubes of size 100x100x100 mm. Cubes were manually compacted in 3 layers according to ASTM C495. Then, the specimens were vibrated for 3 minutes using the vibrating table. Then the cubes were de-molded after 24 hours and cured in water at room temperature until the day of testing.

The mixture proportions are shown in table 3, with constant cement content 500 kg/m³ and with water to binder ratio 0.26. The silica fume was used with a fixed percentage of 20% of the cement content. The chemical admixture used with a ratio of 4% of the binder content. Nano-silica used with a percentage of 1, 2, and 3% replacement of cement content at the optimum solid to liquid ratio obtained from the first phase; 1:10.

In the indirect sonication the power was fixed at 135 Watt and the frequency was at 40 KHz. The mixing procedures were as follow: (a) Weight the mix components, (b) add the nano-silica to the needed water for sonication according to the required concentration, (c) place silica fume in the mixer with the concentration of water and nano-silica to the dry mix and rotate the mixer for 2 mins, (d) add cement to the mixer, and let the mixer for 2 mins, (e) add the remaining of mixing water with the superplasticizer to the mix and mixing for 3 mins, (f) add fine aggregate (sand) to the mix and mixing for 3 mins, (g) add dolomite coarse aggregate to the mixer, and mixing for 3 mins.

Table 3: Mix Proportions by Weight

Mix	C	NS1	NS2	NS3
Cement (kg/m³)	500	495	490	485
Water (kg/m³)	156	156	156	156
Silica fume (kg/m³)	100	100	100	100
Fine Agg. (kg/m³)	580	580	580	580
Coarse Agg. (kg/m³)	1078	1078	1078	1078
Nano-Silica (kg/m³)	0	5	10	15
Superpalsti-zier (Lit./m³)	20	20	20	20
Concentartion	as re-ceive	1:10	1:10	1:10
Sonication time	Control	5 mins	5 mins	5 mins

3. Results and Discussion:

3.1 Optimum Sonication time for each Concentration:

Figure (3) and table (4) show the relation between particle size distribution and volume density percentage for different indirect sonication time and for different solid to liquid ratio. As the indirect sonication time increased the nano- particle content to reach an optimum value, differs in every concentration, while by increasing the indirect sonication time above the optimum duration the nano-particle decreased. At solid to liquid ratio 1:30, at zero time sonication the D50 was 1230 nm and a specific surface area was 8829 m²/kg and after 5 minutes of indirect sonication the D50 was decreased to 57 nm and a specific surface area was increased to 136800 m²/kg. However, at solid to liquid ratio 1:10 the D50 was 57.2 nm and with a specific surface area was 137200 m²/kg. Where by increasing the indirect sonication time to 5 minutes the optimum results was obtained at a D50 equals to 45.3 nm and with a specific surface area 164800 m²/kg.

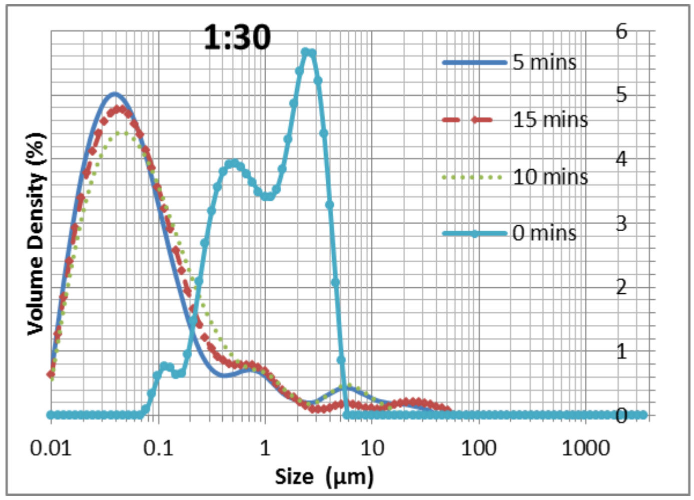


Figure 3: Particle size distribution of nano-silica samples concentration at 1:30

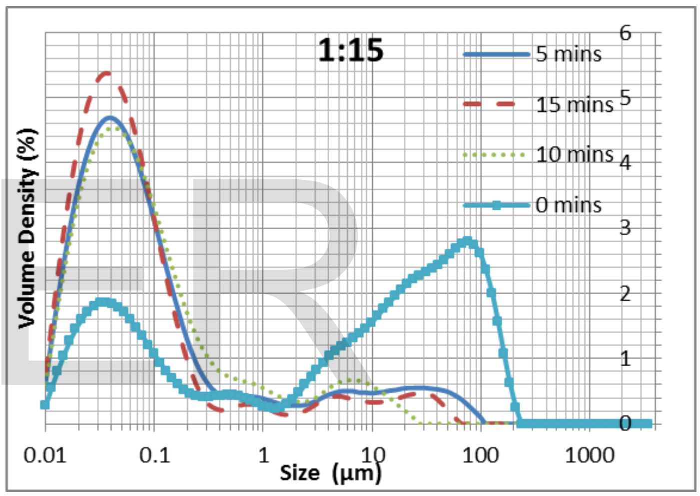


Figure 4: Particle size distribution of nano-silica samples concentration at 1:15

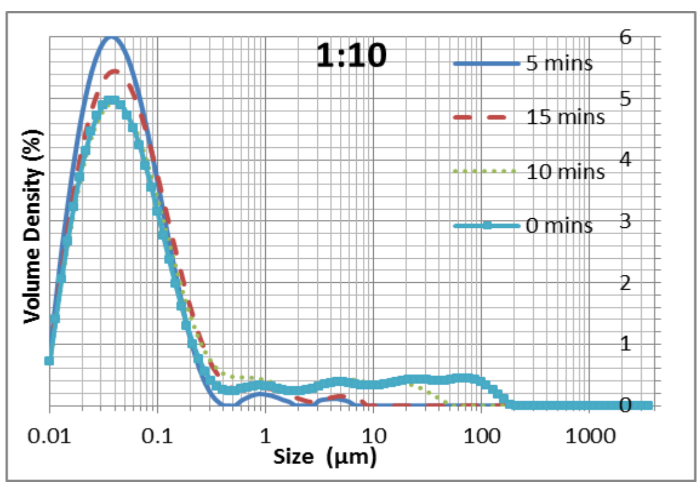


Figure 5: particle size distribution of nano-silica samples concentration at 1:10

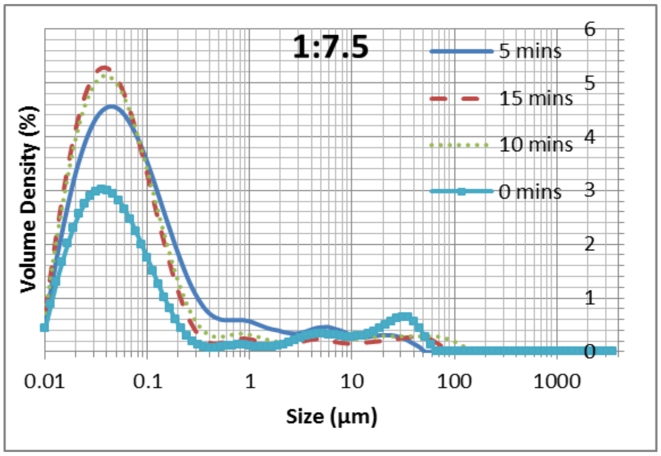


Figure 6: Particle size distribution of nano-silica samples concentration at 1:7.5

Table 4: Specific Surface Area and mean partcile size distribution for each sonication time and concentration by the mastersizer 3000

Concentration		Time (min.) 0	5	10	15
1:30	surface area (m^2/kg)	8829	138600	121500	131100
	D50 (nm)	1230	57	71.4	62.6
1:15	surface area (m^2/kg)	52650	129500	125400	148400
	D50 (nm)	1090	62.6	66.7	51
1:10	surface area (m^2/kg)	137200	164800	136100	14700
	D50 (nm)	57.2	45.3	58.5	51.8
1:7.5	surface area (m^2/kg)	83210	124400	141100	145400
	D50 (nm)	1450	67.6	55.3	52.7

3.2 Fresh and Hardened Properties of Concrete:

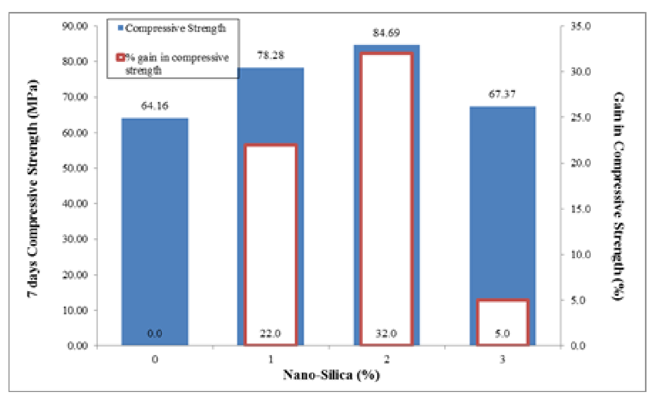
Four mixes of nano-silica concrete were prepared to study the effect of the optimum solid to liquid ratio and the corresponding indirect sonication time on the fresh (workability) and hardened properties (compressive strength, permeability) of concrete. The first three groups were NS1, NS2, and NS3 were prepared using sonicated nano-silica using solid to liquid ratio 1:10 with an indirect sonication time 5 minutes. Nano-silica used with a replacement 1, 2, and 3% of the cement weight. The fourth mix was the control mix with 0% nano-silica.

3.2.1 Workability:

The value of the slump obtained was the same at all nano-silica concrete mixes 210±25 mm and the same in the control mix (without nano-silica). This can be attributed to by using the optimum solid to liquid ratio; 1:10, the nano-particles will be dispersed properly and show higher workability as a consequence of the free water among the particles that causes the rolling effect. Also, nano silica acts as filler that occupy the void space between the cement particles releasing the water that was fill the voids and increase workability.

3.2.2 Compressive Strength:

Adding nano-silica increased both the 7 and 28 days compressive strength of nano-silica concrete. The gain in 7 days compressive strength reached 22%, 32%, and 5% by the addition of 1%, 2%, and 3% nano-silica respectively. While for the 28 days the improvement was 18.4%, 24.0%, and 16.0% by the addition of 1%, 2%, and 3% nano-silica respectively, as shown in figures (7 and 8). This significant improvement in strength indicates that nano-silica acts as nuclei for cement phases to promote cement hydration due to its pozzolanic reaction with CH creasing the production of C-S-H gel. Although the gaining in strength after 7 days is significantly higher than the gaining after 28 days, these can be explained that the larger and/or agglomerated particles in the mix that had not completely dissolved in solution will reduce the porosity in the capillaries of the CSH gel packing into some of these voids. In doing so, the density of the CSH will slightly increase in the plastic form and convert into denser compressive bearing structure after hydration completes. From the results shown below, the mum nano-silica percentage is 2% as a partial substation of the cement weight which gives the optimum compressive strength



after 7 and 28 days, with a solid to liquid ratio 1:10.

Figure 7: 7 days compressive strength of nano-silica concrete and the gain in compressive strength as compared to the control mix

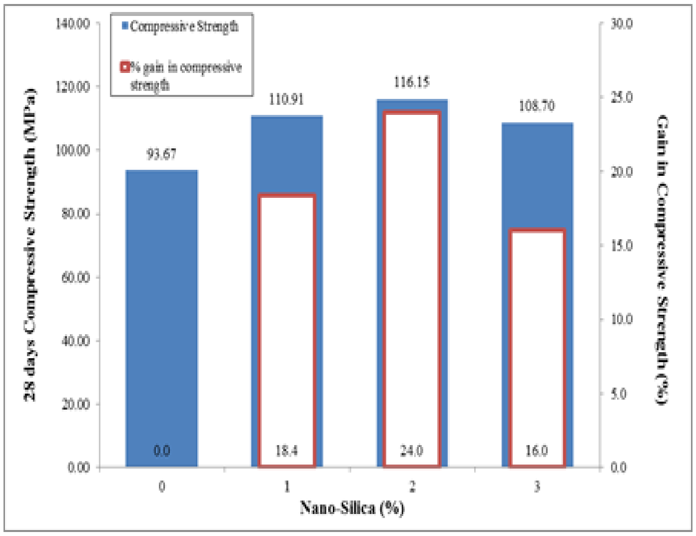


Figure 8: 28 days compressive strength of nano-silica concrete and the gain in compressive as compared to the control mix

3.2.3 Absorption of Nano-Silica Concrete:

Increasing nano-silica percentage generally decreased the absorption of concrete. By using 1%, 2%, and 3% nano-silica, the absorption reduced to 2.36, 2.22, and 2.07% compared to the control mix (containing 0% nano-silica), as shown in figure (9). This can be attributed to the filling effect of nano-silica due to its nano particles as compared with the cement particles and micro size of silica fume even if nano-silica particles were agglomerated. In addition to the above, nano-silica reacts with CH to form more C-S-H gel which makes the matrix more dense and homogenous and decrease the permeability of concrete. Nano-silica can absorb the $Ca(OH)_2$ crystals and reduce the size and the amount of the $Ca(OH)_2$ crystals, thus making the interfacial transition zone (ITZ) of aggregates and binding paste matrix denser.

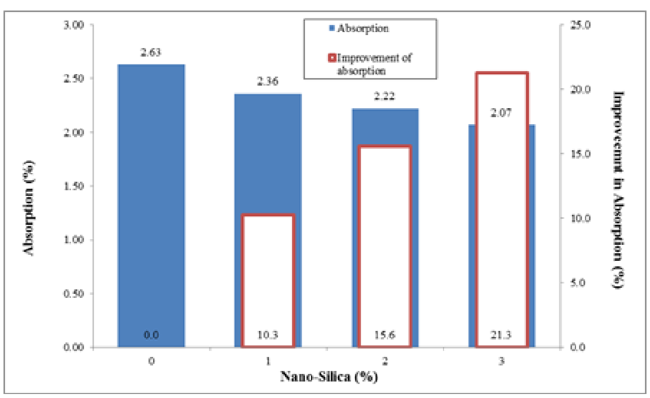


Figure 9: Absorption ratio of Nano-Silica Concrete

3.2.4 Microstructure Test:

Microstructure test was carried out using Scanning Electron Microscopy (SEM) as shown in figures (10, 11, and 12). This figure shows the SEM of mortar samples without nano-silica, with unsonicated nano-silica, with sonicated nano-silica, respectively with age 28 days. From the figures below, it is more obvious that the mortar samples with nano-silica are denser than samples without nano-silica, also, the effect of sonication on the microstructure on the mortar samples. Nano-silica caused a secondary reaction between the silica oxide located in nano-silica and $Ca(OH)_2$ crystals existing in the paste. Sonication makes the cement paste more homogenous than unsonicated nano-silica through the filler effect and the good dispersion of nano-particles in the matrix. XRD analysis was performed to detect changes in the hydration products due to the presence of nano-silica. Nano-silica addition resulted in a nificant decrease in the calcium hydroxide peaks especially in the sonicated nano-silica sample, figure (15), compared to the both unsonicated nano-silica sample, figure (14), and the control mix, figure (13).

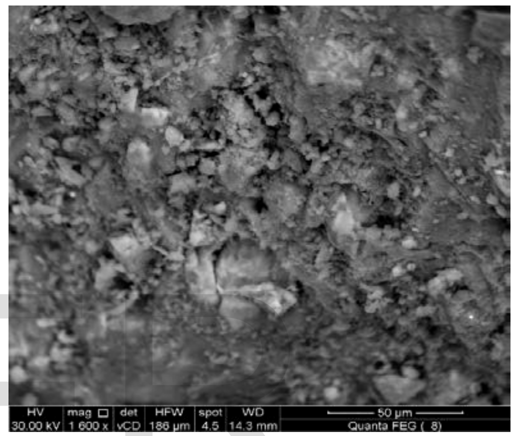


Figure 10: SEM pictures of cement matrix without nano-silica

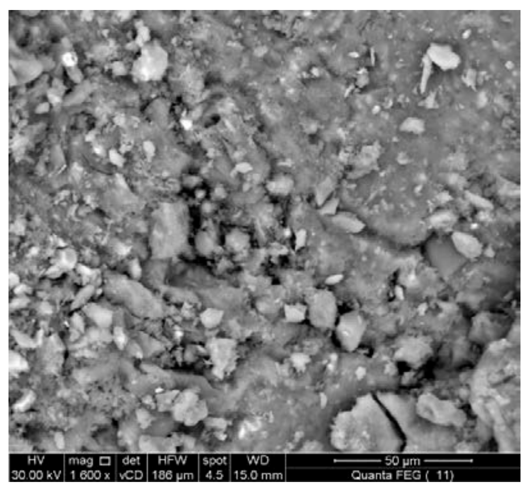


Figure 11: SEM pictures of cement matrix with unsonicated nano-silica

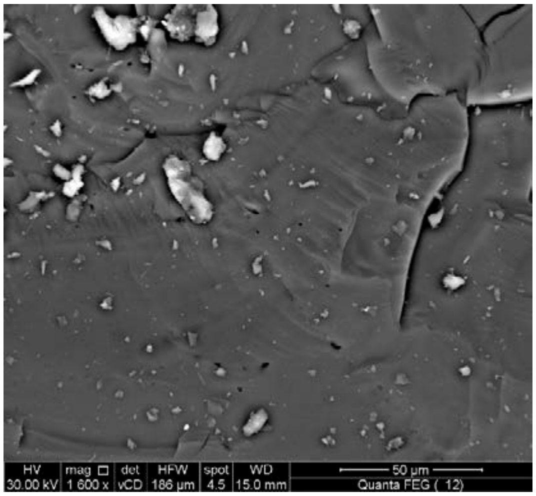


Figure 12: SEM pictures of cement matrix with sonicated nano-silica

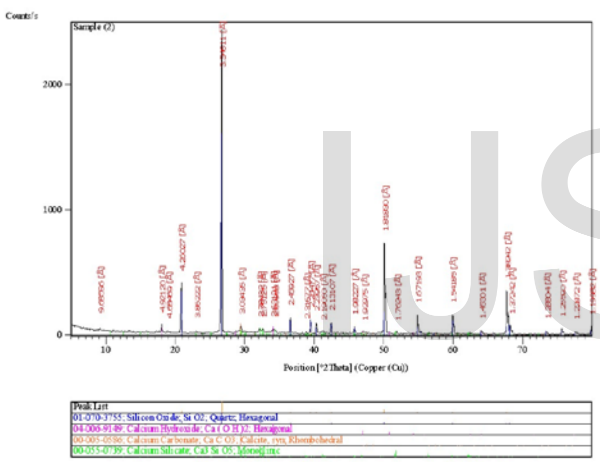


Figure 13: XRD analysis of cement mortar without nano-silica

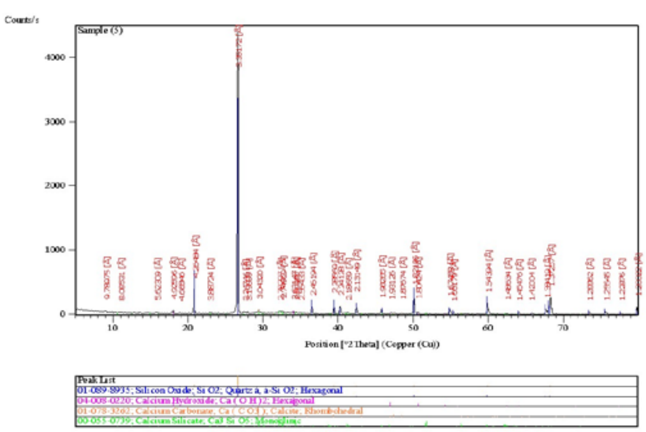


Figure 14: XRD analysis of cement mortar with unsonicated nano-silica

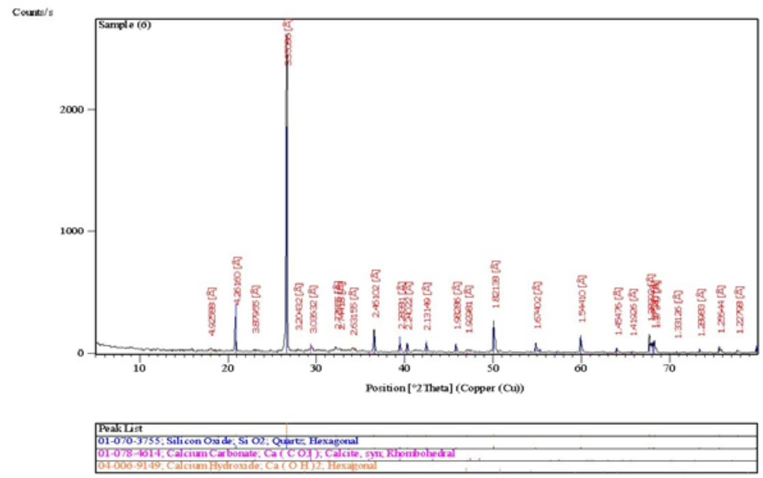


Figure 15: XRD analysis of cement mortar with sonicated nano-silica

4. Conclusion

From the experimental works the following conclusions can be drawn:

- The optimum solid to liquid ratio of nano-silica is 1:10 and with an indirect sonication time 5 minutes, which results in the optimum specific surface area and particle size distribution.
- Indirect sonication for 5 minutes and with a concentration 1:10 gives higher compressive strength improvement as compared to the control mix by 39% and 25% after 7 and 28 days, respectively.
- Using sonicated nano-silica improves the microstructure of the cement matrix and consumes calcium hydroxide crystals to produce more and denser C-S-H crystals.
- The optimum nano-silica replacement percentage is 2% of the cement weight.
- Within the range studied, changing the solid to liquid ratio of nano-silica more or less than 1:10 will results in a decrease in the strength gained, as an increase in the re-agglomeration occurred.
- Microstructure analysis reveals that nano-silica improve the microstructure of the cement paste and produce a denser and more homgenous matrix than without nano-silica.

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