

# The Effect of Indirect Sonication on the Reactivity of Nano Silica Concrete

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**Abstract**—The paper studied the effect of changing the duration time of indirect sonication on the reactivity, and dispersion of nano silica as cement substitution for concrete mixes, the optimum duration will then be used to produce nano silica concrete with different substitutions and their compressive strength is to be compared with the -as received- nano silica concrete. The results revealed that optimum nano silica dispersion was found to be reached by applying 5 min of indirect sonication to the nano silica before being added to concrete mix with a gain in compressive strength reached 33% and an increase in the workability as well. Increasing sonication time beyond 7 min resulted in an increase in the re-agglomeration which subsequently affected the compressive strength gain and a significant loss in strength was reported to reach more than -22% for 30 min sonication as compared with the un-sonicated nano silica concrete mix.

**Index Terms**— Indirect Sonication, Nano Silica, Zeta Potential, Mechanical Properties, Concrete.

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## 1 INTRODUCTION

Pozzolan materials have long been used for partial substitution of Portland cement in concrete in order to achieve economical or technical benefits. Among these materials, silica fume has found wide spread application in improving the durability and mechanical properties of cement composites.

Recently, nanotechnology has attracted considerable scientific interest due to the new potential uses of particles in nanometer scale. Thus industries may be able to re-engineer many existing products that function at unprecedented levels. Due to these developments in nano science and technology, various forms of nano-sized amorphous silica have become available. As these materials have higher specific surface area compared to silica fume, a considerable research effort has been attracted to investigate the influence of nano silica on the properties of cement based materials.

Although numerous papers have studied the influence of nano silica on the properties of cement composites, their effects have not been adequately characterized yet, and some discrepancies and inconsistencies in compressive strength, and workability results are witnessed. [1-5].

Since the major problem in utilizing Nano-particles is that they are highly agglomerated and if used directly in a bulk composite, they often lose their high-surface area due to grain growth. Effective means of de-agglomerating and dispersing are needed to overcome the bonding forces after wetting the powder; the ultrasonic breakup of the agglomerate structures in aqueous and non-aqueous suspensions allows utilizing the full potential of nano-sized materials, besides the addition of proper chemical dispersing admixtures.

Ultrasound is used in a wide range of physical, chemical and biological processes. Homogenizing, emulsifying, and dispersing are examples for physical processes. Most of the applications of high-intensity ultrasound

are based on cavitation effects. The physical effects of cavitations are being used in a top-down generation of Nano-particles. Here, particles are reduced in size by the forces of cavitations. This includes the breaking of agglomerates and aggregates. In recent years, the application of high frequency ultrasound treatments to disperse densified silica fume (DSF) has been explored. Sonication enhances the effectiveness of silica fume as an substitution cement material SCM, associated with higher degree of consumption of portlandite over time of curing and higher mechanical strengths. [6,7]. Sonicated aqueous suspensions of silica fume (SSF) show improved dispersion of sub-micrometer particles [6-8].

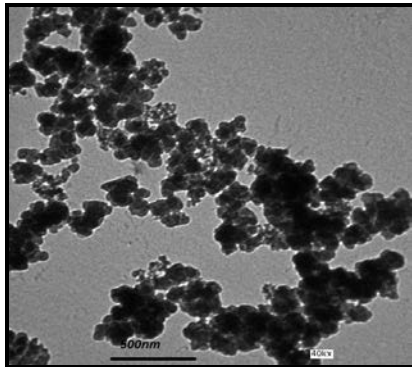
The aim of the proposed plan of work is to study the effect of changing the indirect sonication duration on the reactivity, and dispersion of nano silica as cement substitution for concrete mixes, the optimum duration will then be used to produce nano silica concrete with different substitutions and their compressive strength is to be compared with the -as received- nano silica concrete.

## 2 EXPERIMENTAL WORK:

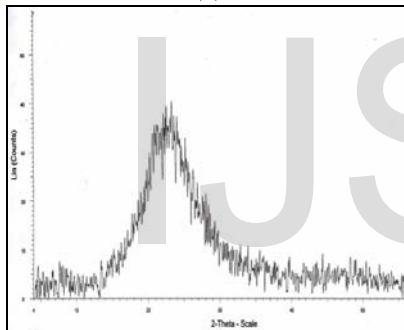
### 2.1. Materials:

Ordinary Portland Cement (OPC) conforming to ASTM C150 standard was used as received. Chemical and physical properties of used cement are given in Table 1. SiO<sub>2</sub> nano particles with average particle size of 30 nm and 45 m<sup>2</sup>/g Blaine fineness produced from WINLAB laboratory chemicals, UK was used as received. The properties of SiO<sub>2</sub> nano particles are shown in Table 2. Transmission electron micrographs (TEM) and powder X-ray diffraction (XRD) diagrams of SiO<sub>2</sub> nano particles are shown in Figs. 1(a, and b). Crushed limestone aggregates, as well as sand free of alkali-reactive materials were used to insure producing durable Concretes; the aggregates were mixed by percentages of 65%

for coarse aggregate, and 35% for fines by volume. A polycarboxylate with a polyethylene condensate de-foamed based admixture (Glenium C315 SCC) was used. Table 3 shows some of the physical and chemical properties of polycarboxylate admixture used in this study.



(a)



(b)

Figure 1: (a) TEM micrograph of SiO<sub>2</sub> Nano particles, (b) XRD analysis of SiO<sub>2</sub> Nano particles.

Table1. Properties of Portland cement (wt%).

Element	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	S	Na <sub>2</sub> O	K <sub>2</sub> O	L
Cement	20.13	5.32	3.61	61.63	2.39	2.87	0.37	0.13	1.96

Table2. Chemical composition of Nano SiO<sub>2</sub> (wt %).

Element	SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	MgO	CaO	Na <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>
NS	99.17	0.06	0.13	0.11	0.14	0.40	0.01

Table3. Physical and chemical characteristics of the polycarboxylate admixture.

Appearance	Off white opaque liquid
Specific gravity @ 20°C	1.095 ± 0.02 g/cm <sup>3</sup>
PH-value	6.5 ± 1
Alkali content (%)	Less than or equal to 2.00
Chloride content (%)	Less than or equal to 0.10

## 2.2. Experimental program:

The experimental program is to be divided into two main routes; the first investigates the effect of changing indirect sonication time on the nano silica concrete behavior; eight samples with seven different sonication durations; 0, 3, 5, 7, 10, 15, and 30 min as well as a control mix without nano silica addition are to be conducted, constituents as well as the suggested duration times are mentioned in table 4. , the second compares the effect of adding different percentages of -as received-nano silica on compressive strength of concrete with the optimally sonicated nano silica, the constituents of those mixes are mentioned in table 5.

A total of 16 mixtures were prepared as shown in table 4. Sets of 6 cubes (15\*15\*15 cm<sup>3</sup>) were cast to perform compression strength tests after 7, and 28 days of water curing. Cubes were consolidated in accordance to ASTM C 192 in three layers on a vibrating table, where each layer was vibrated for 10 seconds, and then the specimens were de-molded after 24 hours and cured in normal free water at room temperature until the day of testing.

Mixes prepared for this plan with constant binder/aggregate weight ratio (B/A) of 1:3.57, and water/binder (W/B) ratio of 0.4. Mixtures COD, A1, A2, and A3 were produced with 0%, 1.0%, 1.5%, and 2.0% nano silica, used as received, by weight replacing cement. The amount of Superplasticizer (SP) was set to 0.42 wt% of the binder (cement plus nano silica). The mixtures compositions are shown in Table 4. Preparation of mixtures was performed in the following sequence:

- (a) Weighing components, (b) mixing the solid components inside a turn tilt mixer for 1 min, (c) adding nano silica and superplasticizer into water for helping dispersing the nano silica, (d) adding the solution to the mixer and (e) finally mechanical mixing for 3 min.

While for the sonicated nano silica, the preparation of mixtures was performed in the following sequence:

- (a) Weighing components, (b) mixing the solid components inside a turn tilt mixer for 1 min, (c) adding sonicated nano silica with a portion of water and mixing for 1 min, (d) adding superplasticizer into the rest of water for helping dispersing the nano silica, and (e) finally mechanical mixing for 2.5 min.

Table 4: mix constituents by weight (kg) per 1m3.

Mix	Cement	Aggregate		Water	S.p.	N.s.	Sonication time
		Coarse	Fine				
Cod	480	1109	597	192	2.11	0	Control
Sd100	475.2	1109	597	192	2.11	4.8	0 min sonication
Sd103	475.2	1109	597	192	2.11	4.8	3 min sonication
Sd105	475.2	1109	597	192	2.11	4.8	5 min sonication
Sd107	475.2	1109	597	192	2.11	4.8	7 min sonication
Sd110	475.2	1109	597	192	2.11	4.8	10 min sonication
Sd115	475.2	1109	597	192	2.11	4.8	15 min sonication
Sd130	475.2	1109	597	192	2.11	4.8	30 min sonication

Table 5: mix constituents by weight (kg) per 1m3.

Mix	Cement	Aggregate		Water	S.p.	N.s.	Sonication time
		Coarse	Fine				
A1	475.2	1109	597	192	2.11	4.8	As received
A2	470.4	1109	597	192	2.11	9.6	As received
A3	465.6	1109	597	192	2.11	14.4	As received
S1	475.2	1109	597	192	2.11	4.8	Sonicated 5 min
S2	470.4	1109	597	192	2.11	9.6	Sonicated 5 min
S3	465.6	1109	597	192	2.11	14.4	Sonicated 5 min

### 3 RESULTS AND DISCUSSION:

#### 3.1. The effect of sonication duration time on compressive strength

As it can be seen from figure 2, the compressive strength increased by increasing sonication time till 5 min to reach 368 kg/cm<sup>2</sup> at 7 days of curing, and 492 kg/cm<sup>2</sup> after 28 days of curing with an increase of 26% than the un-sonicated nano silica mix, and then the strength gain starts to decrease, this can be attributed to the re-agglomeration of nano silica particles by increasing the sonication time, the same trend was reported by (Erich D. Rodriguez et. al 2012) [9], while from figure 3 we can conclude that increasing sonication time beyond 7 min resulted in an increase in the re-agglomeration which subsequently affected the compressive strength gain and a significant loss in strength was reported to reach more than -22% for 30 min sonication as compared to the un-sonicated nano silica control mix, the TEM results reported in figure 4 proved the re-agglomeration behavior. This can be as a result of the entrapment of some of mix water in the aggregates of nano silica formed in cement paste environment, making less water available for the progress of cement hydration.

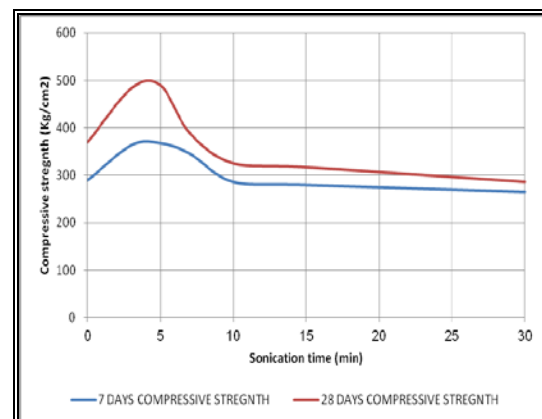


Figure 2: the effect of sonication time on the early and late compressive strength results of nano silica concrete.

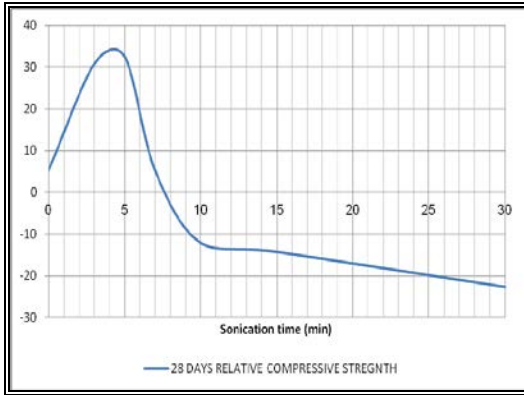
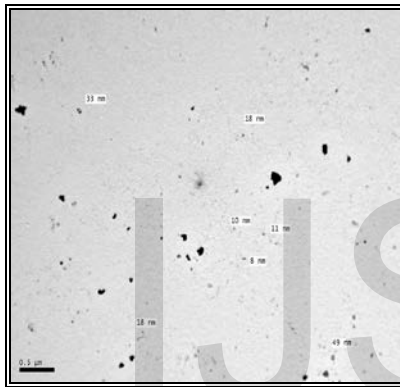
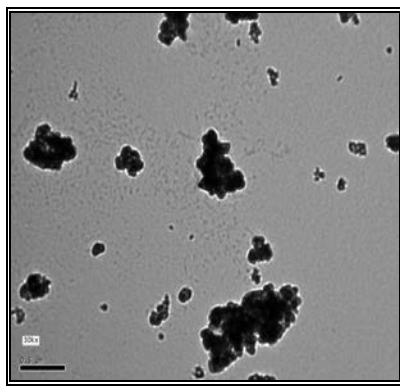


Figure 3: effect of sonication time on the 28 days compressive strength gain of nano silica concrete.



a) NS sonicated for 5 min



b) sonicated for 30 min

figure 4: TEM micrographs for a) nano silica sonicated for 5 min, and b) sonicated for 30 min

A linear relation between the early and late strengths of sonicated nano silica concrete can be seen from figure 5, which indicates that de-agglomeration as well as re-agglomeration affects the reactivity of nano silica particles at both; the early, and late strengths, the proposed equation accounts for 90% of the results, and helps in predicting the late strength from the 7 days strength result.

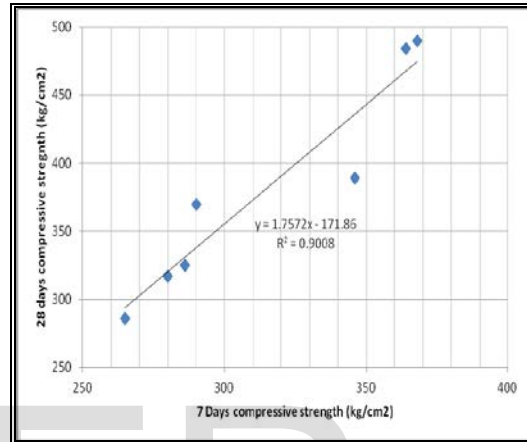


Figure 5: relation between 7, and 28 days compressive strength at different sonication time.

### 3.2. The effect of Sonication time on slump

The slump results are in very well complying with the compressive strength results as it can be seen from figure 6 as we can conclude that the slump is highly affected with the level of agglomeration of nano silica particles, as the nano particles get well dispersed the workability increases, this can be attributed to the fact that ultra fine particles have a higher workability as a consequence of high free water among the particles where the rolling effect can be observed. However, the use of densified silica particles has a negative effect on the workability of fresh mixes, where this rolling effect disappears due to the presence of large irregular agglomerates. The earlier mentioned rolling effect is similar to the effect of fly ash (E.B. Nelson et al. 2006) [10]. It improves or facilitates the flowing of the

cement paste, resulting in a lower water demand to obtain the same slump (G. Quercia et al. 2012) [11], this conclusion was also mentioned by (Erich D. Rodriguez et al. 2012) [9].

When nano-silica is incorporated it has a direct influence on water demand of the mixture. If the water content is kept constant, addition of nano-silica is believed to promote the packing of cement particles, decrease the volume between them and increase the free water contributing to fluidity in the paste. However, the agglomerates in nano silica possess a high water adsorption due to their high surface area and high nano-scale porosity.

Theoretically, addition of nano-silica will help improve fluidity of the paste if all agglomerates can act as fillers to occupy the void space among cement particles and release some free water in the space originally not contributing to fluidity, even though some free water in the space was adsorbed by the agglomerates, as illustrated in Figure 7a.

Otherwise, those agglomerates that are unable to act as fillers will consume some free water that originally contributes to fluidity. They may also push away the cement particles around them, causing an increase of the void space, see Figure 7b. Therefore, the influence of nano silica addition on rheological behavior of concrete is mainly dependent on whether the agglomerates can act as fillers or not.

The fact that addition of sonicated NS in concrete led to a slight increase in workability implied that not all the agglomerates in NS act as fillers though they are obviously smaller than most of the cement particles. For the mixes with sonication increased more 7 min, with the same NS addition, most agglomerates cannot function as fillers due to their much larger agglomeration size. Little free water in the void space was released by the filling effect of the small agglomerates but much water was

adsorbed by the large agglomerates, see Figure 7c.

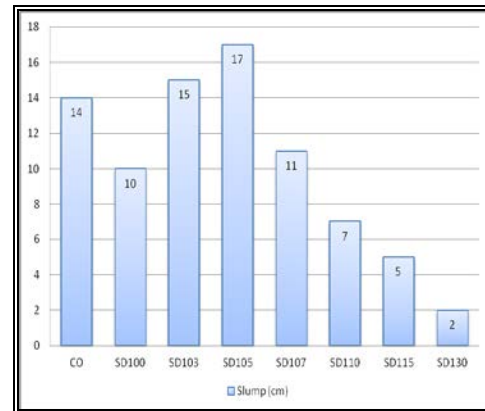


Figure 6: the effect of nano silica sonication time on slump as compared to control mix.

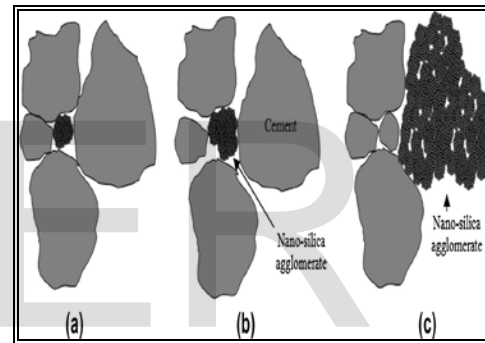


Figure 7. Illustration of the filling effect of the nano-silica agglomerates: (a) the smaller agglomerates act as fillers to release some free water in the gap to enhance the fluidity of the paste. (b) The larger agglomerates cannot act as fillers; they tend to push away the particles around them, resulting in an increase of the void space. (c) The very large agglomerates absorb some free water originally contributing to the fluidity of the paste [12].

### 3.3. The effect of sonication time on concrete density

As it can be seen from figure 8, as the nano silica get well de-agglomerated, the concrete density increased, this can be attributed to the fact that better dispersion helps in a better particle size distribution (PSD) and so better compaction of mix and as it was concluded by

(H.J.H.Brouwers and Radix. 2005) [13] in a previous work that it could be expected that if one would optimize the PSD down in the nanometer range, the workability and stability could be maintained while further reducing the necessary super plasticizer content, on the other hand agglomerated particles has less PSD and so less compaction and more voids that decreases the concrete mix density(G. Quercia et al. 2012) [11].

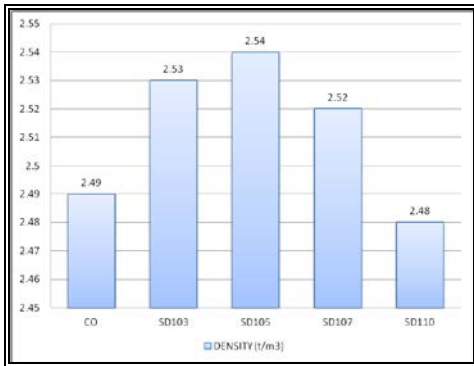
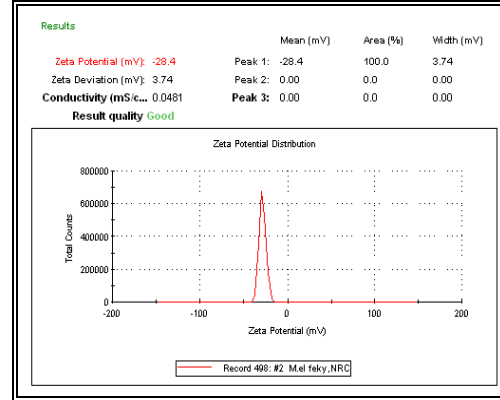


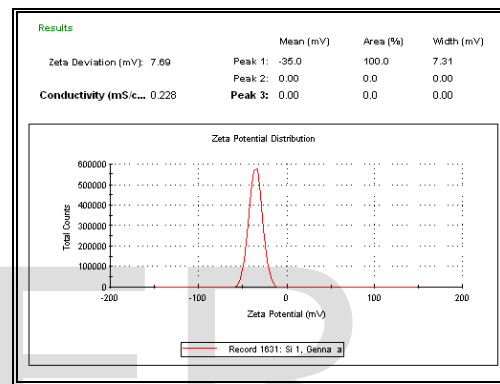
Figure 8: the effect of changing sonication time on the nano silica concrete density.

### 3.4. The effect of sonication time on nano silica zeta potential results

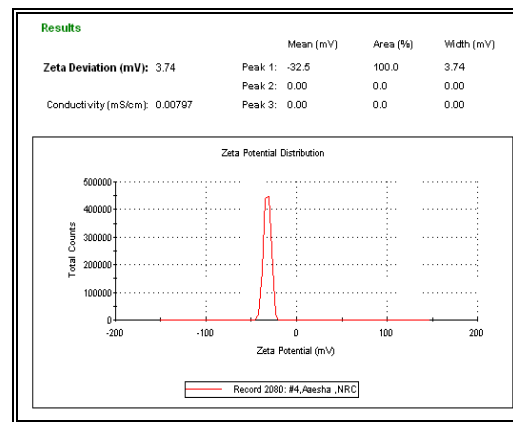
As we mentioned before zeta potential is an indicator of the stability of a colloidal system. If the suspension has a large negative or positive zeta potential, particles will tend to repel each other and there will be no tendency for the particles to come together or to agglomerate. From figure 9, we can conclude that increasing sonication to 5 min increased nano silica zeta potential to reach -35 mv as shown in figure 9b instead of -28 mv for the as received as shown in figure 9a, also we can see that increasing sonication time for 7 min decreased the zeta potential absolute value to reach -32 mv as in figure 9c, which explains the reported loss in compressive strength by increasing sonication time above 5 min.



A) As-received nano silica zeta potential



B) Zeta potential of 5 min sonicated nano silica



C) Zeta potential of 7 min sonicated nano silica

Figure 9: the effect of sonication time on zeta potential results of commercial nano silica

### 3.5. Effect of adding (as received) NS as compared to optimally sonicated nano silica on the gain of compressive strength

The following conclusions can be drawn from figure 10:

The mechanical strength of tested mixes increased with increasing the amount of the silica nano particles up to 2%, and then slight reduction in compressive strength was reported. The results can be attributed to the clear agglomeration in the -as received - NS which is clear in TEM, and XRD figures shown previously (Fig 1).

For the contents higher than 2%, the nano particles cannot easily disperse within the cement matrix, and due to their high surface energy, they become more agglomerated, hence a weak area of empty spaces such as voids appeared. Consequently, the structure formed in such conditions cannot be homogenous and compacted (M.R. Arefi et al. 2011) [14].

As for the sonicated nano silica particles; the gain in strength increased significantly to reach five times higher than the as received nano silica.

The gain in strength increased by increasing sonicated nano silica to 2% then a significant drop appeared, this can be attributed to that the nano silica particles cannot easily disperse within the cement matrix, and due to their high surface energy, they started to be agglomerated, and so we can conclude that the sonication time should be adjusted with the percentage of nano silica addition.

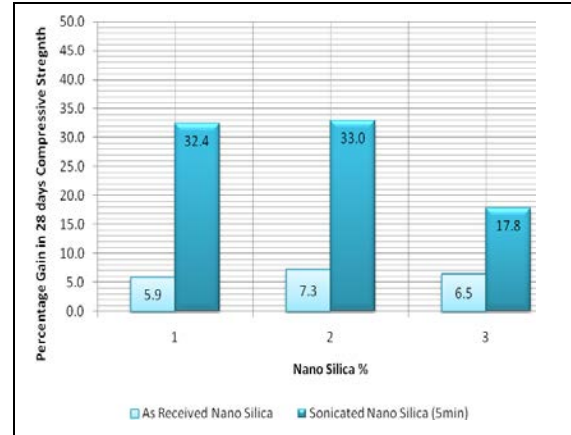


Figure 10: Gain in compressive strength of - as received- NS Concrete mixes as compared to (5 min) sonicated nano silica.

## 4 CONCLUSIONS

From the conducted plan we can conclude the following:

- Optimum nano silica dispersion was found by applying 5 min of indirect sonication to the nano silica before being added to concrete mix.
- Increasing sonication time beyond 7 min resulted in an increase in the re-agglomeration which subsequently affected the compressive strength gain and a significant loss in strength was reported to reach more than -22% for 30 min sonication as compared to the un-sonicated nano silica mix.
- A linear relation between the early and late strengths of sonicated nano silica concrete was found which indicates that de-agglomeration as well as re-agglomeration affects significantly the reactivity of nano silica particles at both, early, and late strengths, the proposed equation accounts for 90% of the results.
- The slump result is highly affected with the level of agglomeration of nano silica particles, as the nano



- particles get well dispersed the workability increases and vice versa.
- Re- agglomeration by increasing sonication time for more than 5 min was confirmed by comparing the TEM micrographs for nano silica sonicated for 5 min with the one sonicated for 30 min.
  - Zeta potential results also confirmed the re-agglomeration behavior of nano silica with increasing sonication time, as for 7 min sonication the zeta potential decreased (10%) as compared to 5 min sonication.
  - The use of nano silica particles as received from the manufacturer will result in unpredictable, and varying compressive strength values ranging from loss to gain in strength depending on its level of agglomeration.
  - The mechanical late strengths of tested mixes increased with increasing the amount of the as received silica nano particles up to 2%, and then a slight reduction in late compressive strength was reported.
  - The use of 5 min sonication showed a significant effect on de-agglomerating nano silica particles when compared with the as received nano silica addition.
  - The sonication time should be adjusted with the percentage of nano silica addition.

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