Scientific Journal Impact Factor (SJIF): 5.278
International Journal of Modern Trends in Engineering and Research
ISSN (ONLINE) : 2349 - 9745
ISSN (PRINT) : 2393 - 8161

UTILIZING NANO- TO MICRO-SCALE PARTICLES BASED ADDITIVES TO ENHANCE CEMENT-DUNE SAND COMPOSITES

A.M. Alshehy[1], S.A. Ahmed[2], M.S. El-Feky[3] and Y.H. Helal[4]
[1,2] Faculty of Engineering, Zagazig University, Zagazig 44519, Egypt.
[3]National Research Centre, Cairo, Egypt.
[4]Higher Technological Institute, 10th of Ramadan City, Egypt.

Abstract-This study investigated the effect of using particle on nano- and micro- scale on the properties of cement mortar. Three types of fine powders, namely: nano-silica, silica fume and fly ash, were added to the mortar mix as a partial replacement of cement content by weight. A total of 64 mixes were screened using JMP program to obtain 12optimum combinations of mixes in these mixes, the compressive strength, heat of hydration, atomic force microscopy, and XRD were measured in order to evaluate the effect of the above mentioned powders on the behavior of cement mortar. The outcome of this study showed that use of nano-silica with silica fume and fly ash significantly improves the compressive strength of cement mortar, as a result of their high reactivity with the residual CH from the hydration of cement and thus the formation of high content of C-S-H. This C-S-H was the responsible for the compressive strength improvement.

Keywords-Nano-silica, Silica fume, Fly ash, Particle size, Cement and Compressive strength

I. INTRODUCTION

The concrete deteriorates either by micro-cracking within the cement matrix or by corrosion of the reinforcing bars. Most of the reasons behind concrete deterioration are related to the cement mortar rather than the fine and coarse aggregate. As the cement composite got highly strengthened, well compacted and denser, the penetration of the aggressive substances reduces and the life time increases.

In many works, properties of the cement mortar were highly improved through the use of fly ash (FA), silica fume (SF) and nano-silica (NS) as a partial substitution of cement [1, 2].It was found that replacing the cement content with 25% FA or 5-8% SF improved the concrete resistance against the combined freezing-thawing and sulfate attack, which in turn leading to significant improvement in the concrete durability NS should not be added alone to the cement mortar, but it should be added together with micro silica to ensure successful filling of voids; thus increasing the packing density of the cementitious materials and densifying the microstructure of hardened cementitious mortar. This is the best way to utilize the good filling effect of micro silica and the high pozzolanic reactivity of NS so as to further increase the strength and durability of concrete for the production of the next generation high performance concrete [3-12].

There are limited studies available in the literature about the use of dune sands in concrete. Guettalla et al. [13] have compared strength properties of mortar mixes made with conventional sands and dune sand. Mixes made with dune sand only resulted in lower strengths. Kay et al [14] made a comparison among concrete mixtures made with beach sand, wadi sand, dune sand, screened dune sand, and combinations of dune sand or screened dune sand with crushed rock fines. The results indicated that dune sand may provide a readily available alternative material for use as fine aggregate in concrete.

The aim of the proposed study is to enhance the packing of cement mortars, based on sand of dunes as a replacement of natural sand, by utilizing silica based additives at both; micro and nano scales. In order to reach our goal, number of mixtures were selected through screening technique to cover a variety of mix constituents choices, and then running over them mechanical and micro-structural analysis.

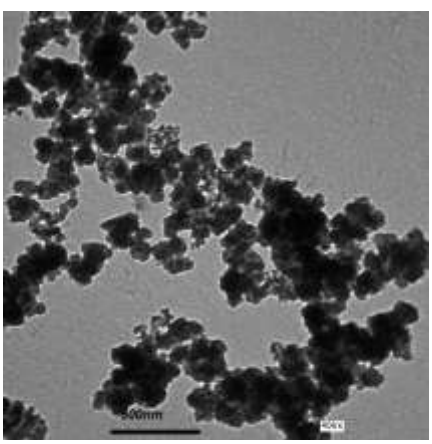
II. EXPERIMENTAL PROCEDURE

Ordinary Portland cement (OPC) with grade CEM I 52.5 N was used; its chemical composition is shown in Table (1).Commercial NS with average particle size of 20 to 80 nm was used. Its chemical composition, particle size distribution (PSD) and properties are shown in Table (1) and Figs. (1-3).As observed the peak centered at 2(Ɵ) =23°, which revealed the amorphous nature of NS particles Also, amorphous and agglomerated micro-silica (SF) with size ranging from 48 nm to 625 nm was used. Its chemical composition and properties are shown in Table (1) and Figs. (1, 2).As observed the peak centered at 2(Ɵ) =21°, which revealed the amorphous nature of SF particles.

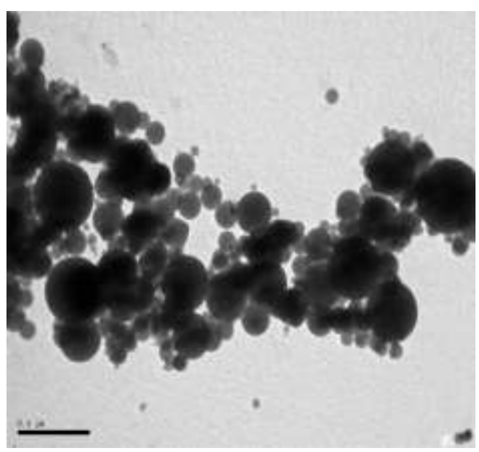
Fly ash, the fine particulate waste material produced by pulverized coal-based thermal power station, was used; its chemical composition and properties are shown in Table (1) and Fig. (1).Also, dune sand with sieve analysis shown in Fig. (4), portable water, and SikaViscoCrete-425 P super plasticizer were used in the experimental program.

Table (1): Chemical composition of OPC, NS, SF and FA (wt. %).

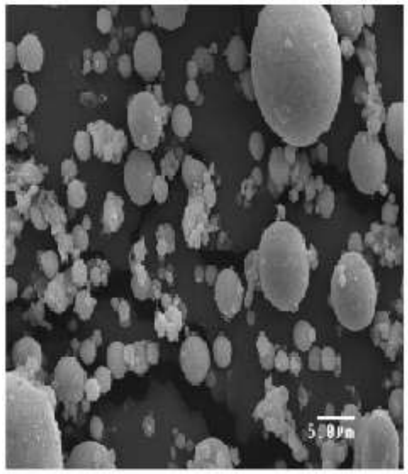
Element	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	L.O.I	P_2O_5
OPC	20.13	5.32	3.61	61.63	2.39	2.87	0.37	0.13	1.96	-
NS	99.17	0.13	0.06	0.14	0.11	-	0.4	-	-	0.01
SF	94.7	0.26	0.25	1.13	-	0.6	0.36	2.45	-	-
FA	49.9	19.2	10.1	8.21	2.84	0.71	1.01	0.72	5.21	-



NS

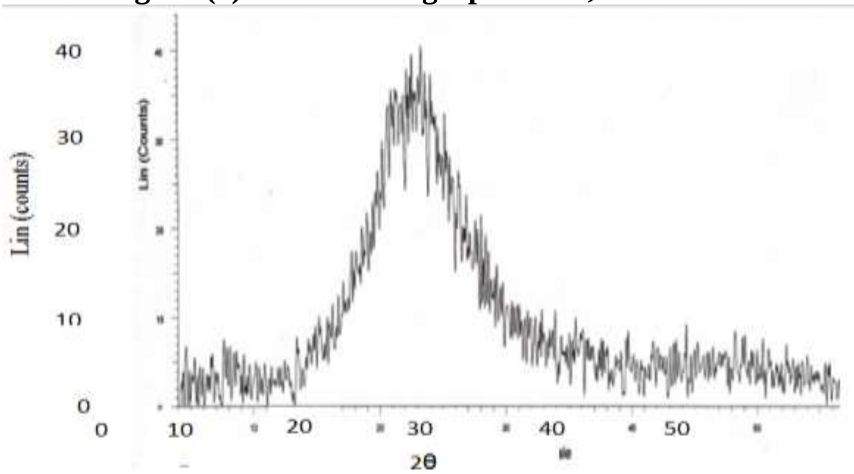


SF

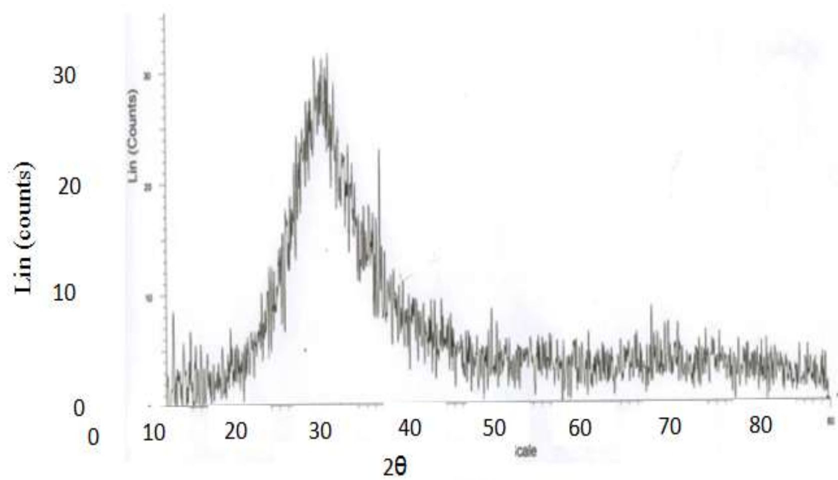


FA

Figure (1): TEM micrograph of NS, SF and FA.



NS



SF

Figure 2.XRD analysis of NS and SF

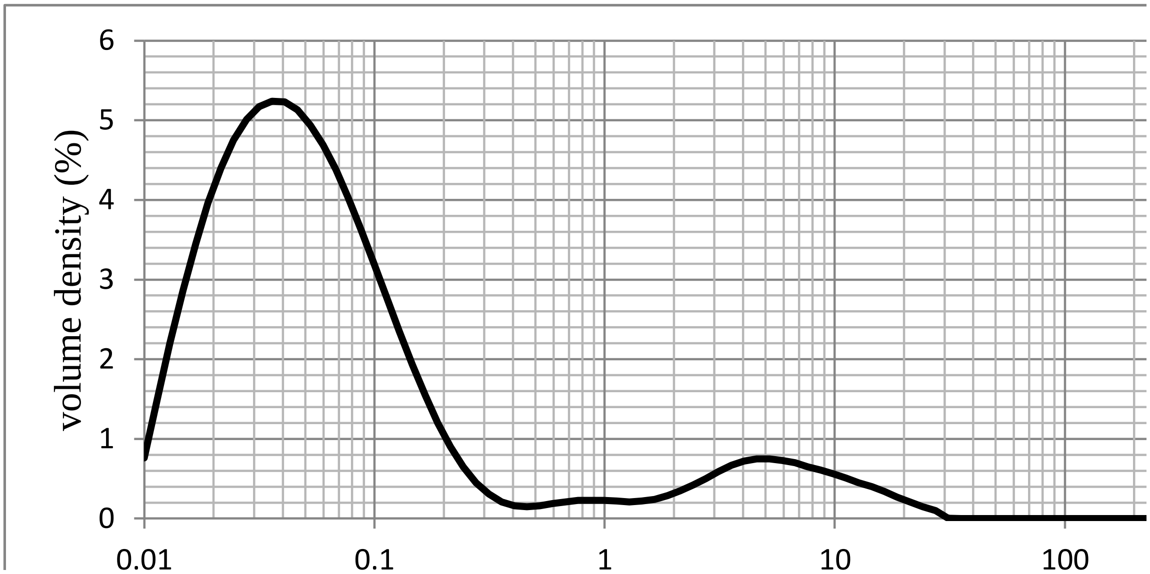


Figure. (3): Particle size distribution of NS.

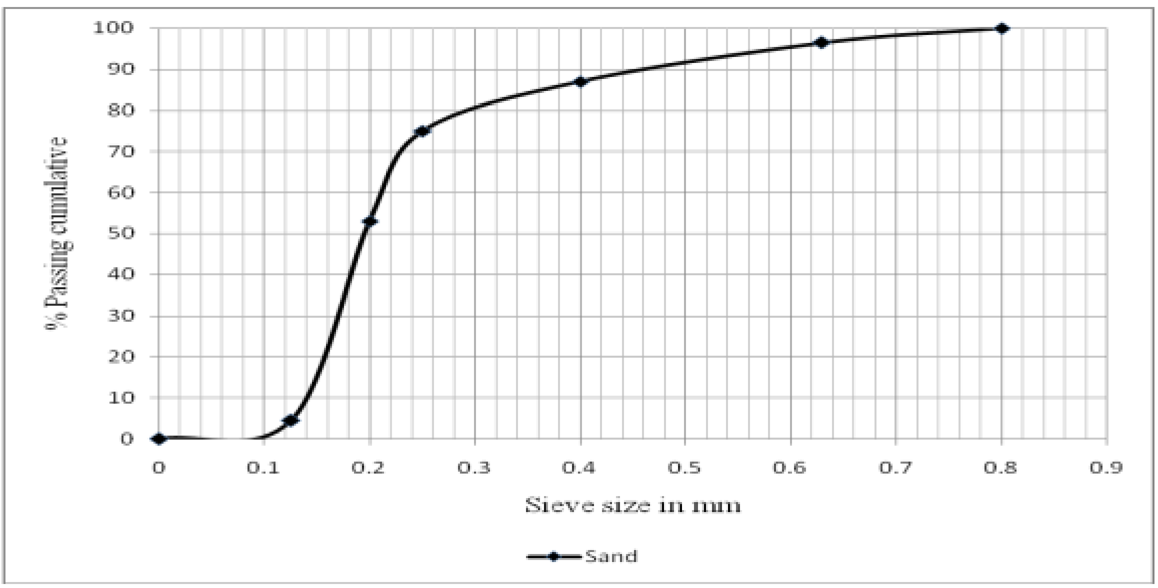


Figure. 4: Sieve analysis of sand.

The experimental program was divided into three main steps; the first step investigated the effect of using different percentages of NS (0, 1, 1.5 and 2%), SF (0, 4, 8 and 12%) and FA (0, 12, 16 and 20%) as a partial replacement of cement content on the compressive strength of cement mortar. The sixty four mixes obtained from these variations in mix variables were screened using program JMP (JMP is a business unit of The Statistical Analysis System SAS) to obtain the twelve optimum combinations, shown in Table (2). These optimum combinations were selected based on most screening experiments use designs at two-levels, with the addition of one centre points to provide a portmanteau test for curvature. An economic class of three level screening designs have been proposed, called" Definitive Screening Designs", to investigate d variables, generally in as few as n = 2d + 1 runs. In all mixes the content of cementitious material to dune sand (by weight) was kept constant at 1:2.

In the second step the heat of hydration of the twelve mixes was measured. In the third step a comparison was made between the twelve optimum combinations and the control mix using two tests. The first test was the atomic force microscopy (AFM), which was used to provide insight into the interpretation of the mechanical properties of particle size distribution. The second test was the X-ray diffraction technique (XRD).After performing the compressive strength test; the crushed cubes of each mix were finely ground and thoroughly mixed. A representative sample from each mix was undertaken and ground to a very fine powder that passes (75 µm) sieve and was tested immediately after that.

Table (2): The twelve optimum combinations of mixes

Mix No.	1 (Control)	2	3	4	5	6	7	8	9	10	11	12
OPC	100	84	72	83	75	79	78.5	90.5	74.5	94	74	66
NS	0	0	0	1	1	1	1.5	1.5	1.5	2	2	2
SF	0	4	12	0	4	8	0	8	12	4	8	12
FA	0	12	16	16	20	12	20	0	12	0	16	20

III. RESULTS AND DISCUSSION

3.1 Compressive Strength

The effect of NS, SF and FA on the compressive strength of cement mortar is shown in Fig. (5). Generally, an improvement in the compressive strength with varying degree was obtained with the use of NS, SF and FA in the mortar mix; compared with the compressive strength of the control mix. At 7 days age, the highest compressive strength reached about 68MPa for mix # 2; with gain in strength of about 31%.However, the lowest compressive strength reached about 45 MPa for mix # 8; with loss in strength of about 13%.At 28 days age, the highest compressive strength reached about 80MPa for mix # 7; with gain in strength of about 52%.However, the lowest compressive strength reached about 58MPafor mix # 8; with gain in strength of about 11%. Moreover, mix #7 where the nano silica was added with the fly ash only, showed clearly the significant effect of the nano silica through; 1- the filling effect where the nano particles fill the nano to micro pores of the cement matrix, as well as 2- the nucleation site effect of the nano silica where its particles act as nuclei for the hydration products resulting in much denser matrix, and finally through the higher pozzolanic reactivity as compared to both the silica fume, or the fly ash particles. In addition by comparing the mix #7 with # 8, we could find out the effect of fly ash particles in breaking down the agglomeration of the nano silica rather than the silica fume that most commonly included agglomeration itself.

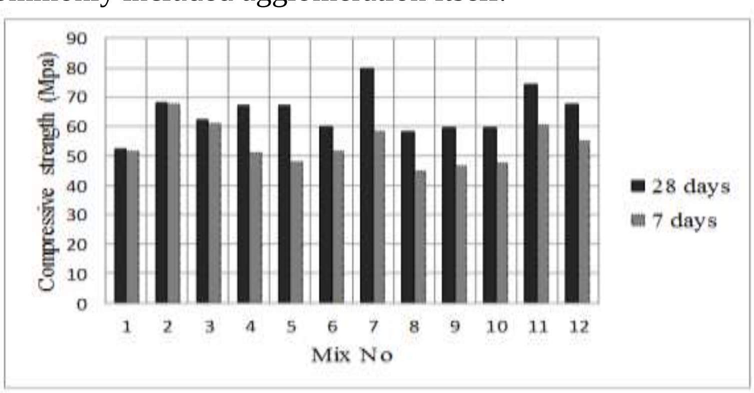


Figure 5: Compressive strength of different mixes at 7- and 28-day ages.

3.2 Heat of Hydration

Figures (6 and 7) present the results of normalized heat flow and normalized heat up to 40 hours for fresh mortars of all mixes tested. As shown in Fig. (6), the normalized heat flow was 0.0021 for mix # 11 rather than being 0.0019for the control mix and 0.00135 for mix # 7. The increase of normalized heat flow for mix # 11 was 10.5% as compared to the control mix, and55.5% as compared to mix #7.The first peak for the control mix and mixes # 9 and # 11 taken place at 31, 24 and 15 hours of hydration, respectively. Also, the maximum height of the first peak of the different mixes can be arranged as follows: 8, 6, 12,11,5, 9, 5, 3, 4, 7, 2 and then the control mix. This indicated that the cement substitution by NS, SF and FA accelerating the cement hydration and then the reaction with CH released from the hydration of cement. Thus producing an extra C-S-H gel at the early ages, which in turn resulting in the enhancement of mechanical properties of cements mortar at the early ages.

Figure (7) shows that the normalized heat signal from the hydration of mortars with NS, SF and FA was more than that from the cement mortar. The temperature rise in mortars with NS, SF and FA began at early time compared to the cement mortar; with the same trend of heat flow. This can be attributed to the high reactivity of NS particles, which accelerated the hydration of cement and rapid its pozzolanic reaction with the residual CH from the hydration of cement.

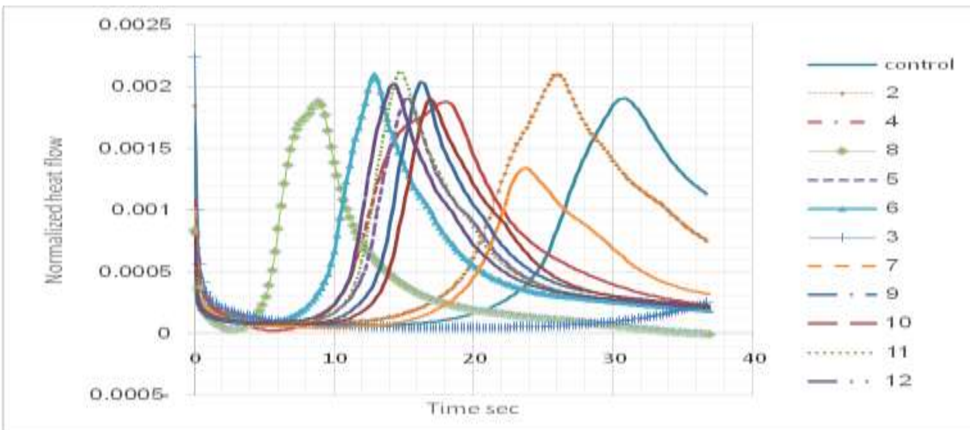


Figure. 6: Normalized heat flow for all mixes.

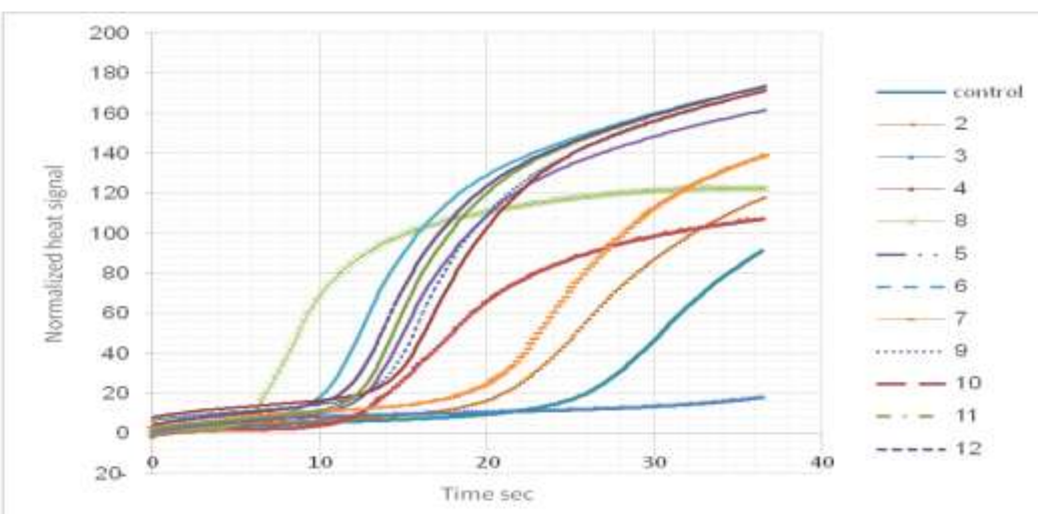


Figure. 7: Normalized heat of all mixes

3.3 Atomic Force Microscope

Figures (8 and 9) show the topographical images from AFM of the control mix and mix #11. Although the AFM images do not have great quantitative value other than for determination of morphology, but in this case AFM images can be useful for providing insight into the interpretation of the mechanical properties of using very fine particle in nano- and micro-scale (NS, SF and FA).Figure (9) shows that AFM topography images of polished cement mortar incorporating NS, SF and FA particles indicated that rounded particles grouped into clumps representing the CSH aggregated particles, which supports the colloidal spheres model of C–S–H, since the spherical features observed in Fig. (8).

The gels are defined as the dispersions in which the attractive interactions between the elements of the disperse phase are so strong that the whole system develops a rigid network structure and, under small stresses, behaves elastically. C-S-H in hydrated tricalcium silicate paste exactly fits this definition. AFM imaging of a flat surface of hydrated cement mortar showed that the elements of the disperse phase occurred in the form of identical aggregated nano particles of C-S-H. Images of C-S-H in hardened cement mortar showed nearly spherical particles of different sizes in different areas. Typical sizes of these spherical particles ranged from 100 to 406nm for the control mix, while reached 482.65nm for the mix #11. AFM images of C-S-H gel with brighter regions being higher than darker ones. Maximum height difference between two different points in the image of the as-received control mix was 406 nm and 482.65nmfor mix #11.

When compared to the control mix, the presence of CH plates could be easily identified as with small clumps of C-S-H particles. The surface of C-S-H phase is showed rougher, bumpy and grainy, while the surface of CH phase is showed smoother. The surface topography as well as the particles morphology with the wide spread of C-S-H gels in mix #11 as compared to the control mix, and the complete absence of CH phases within AFM images of mix # 11 highly indicated the reactivity of NS, SF and FA particles that results in the previously observed tremendous mechanical properties.

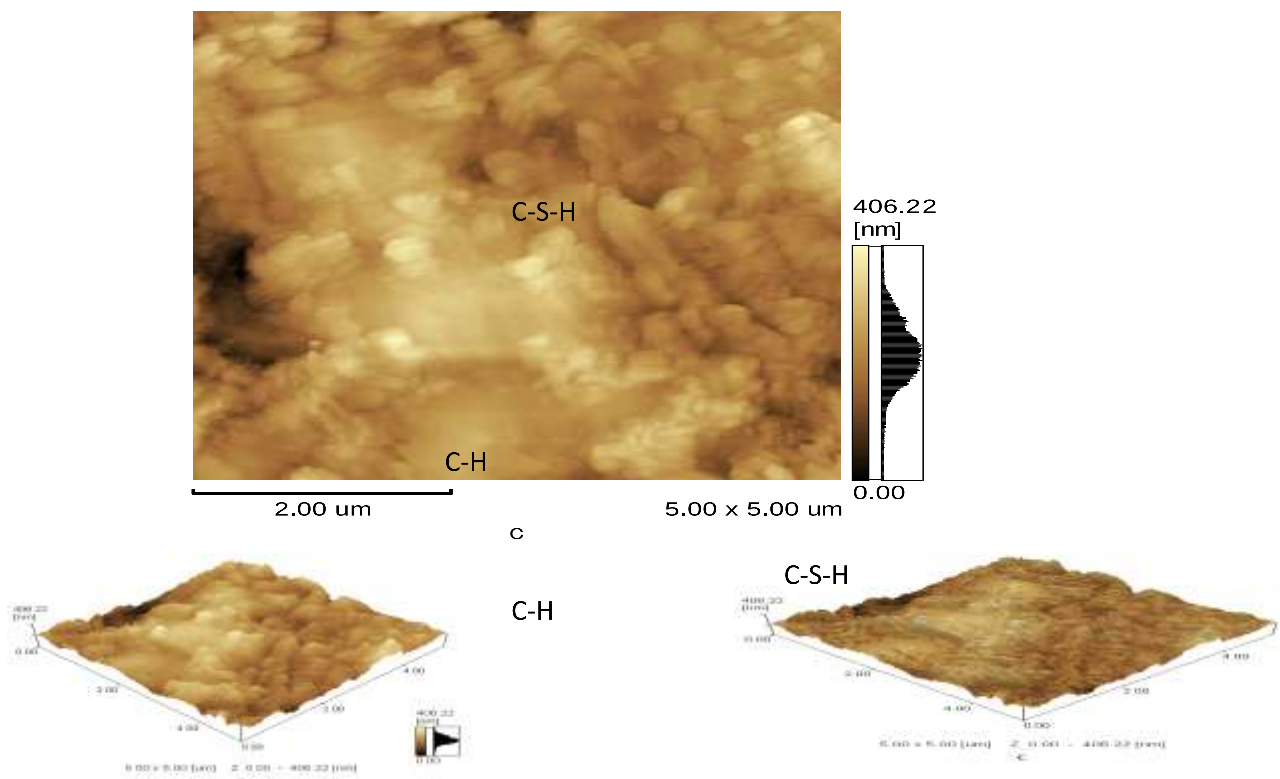


Figure. 8: AFM photographs of the control mix.

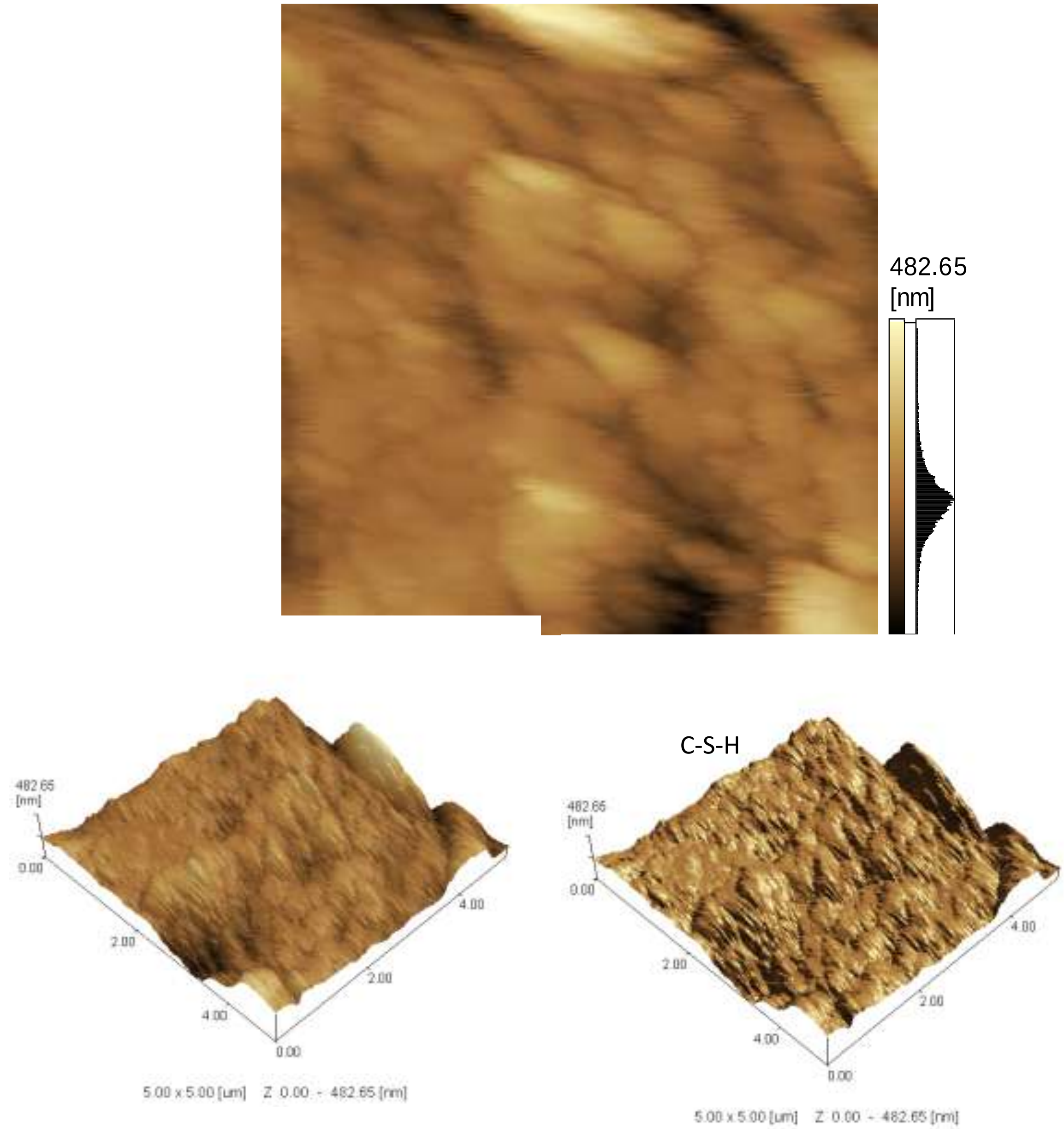


Figure. 9: AFM photographs of mix #11.

3.4 X-ray Diffraction

XRD was performed to detect changes in the hydration products due to the presence of different ratios of NS, SF and FA in the mortar. Figures (10-13) present XRD peaks of the control, # 9 and # 11 mixes. Due to their crystalline nature, calcium hydroxide and calcium carbonate peaks appear clearly in XRD diagrams, while amorphous materials such as calcium silicate hydrate cannot be directly detected by this technique. The CH peaks could be clearly found in the control mix, while it significantly decreased in mixes # 9 and11.The decrease of CH peaks indicated the reactivity of fine particle in micro-scale (SF and FA) and nano-scale (NS), thus the formation of higher C-S-H content in mixes #9 and 11.This higher content of C-S-H gel enhanced ITZ between the aggregate and the cement mortar, and therefore, the compressive strength of these mixes. Mix #11 with 2% NS, 8%SF and 16%FAshowed much lower peaks of CH than mix #9 with 1.5% NS, 12%SF and 12%FA.The calcite (CC) peaks

Volume: 5, Issue: 05, [May– 2018] **ISSN (Online):2349–9745 ; ISSN (Print):2393-8161**

followed the trend of the calcium hydroxide peaks, which also indicated much lesser content of CH in mixes #9 and 11, as compared to the control mix. The presence of CH in large amounts ease the reaction with CO_2 in air producing more calcite ($CaCO_3$) as a result of the carbonation process, see equation (1).

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \qquad (1)$$

Generally, the mixes could be arranged in reference to CH and CC contents as follows: control mix, mix # 9 and finally with the lowest content; mix #11.

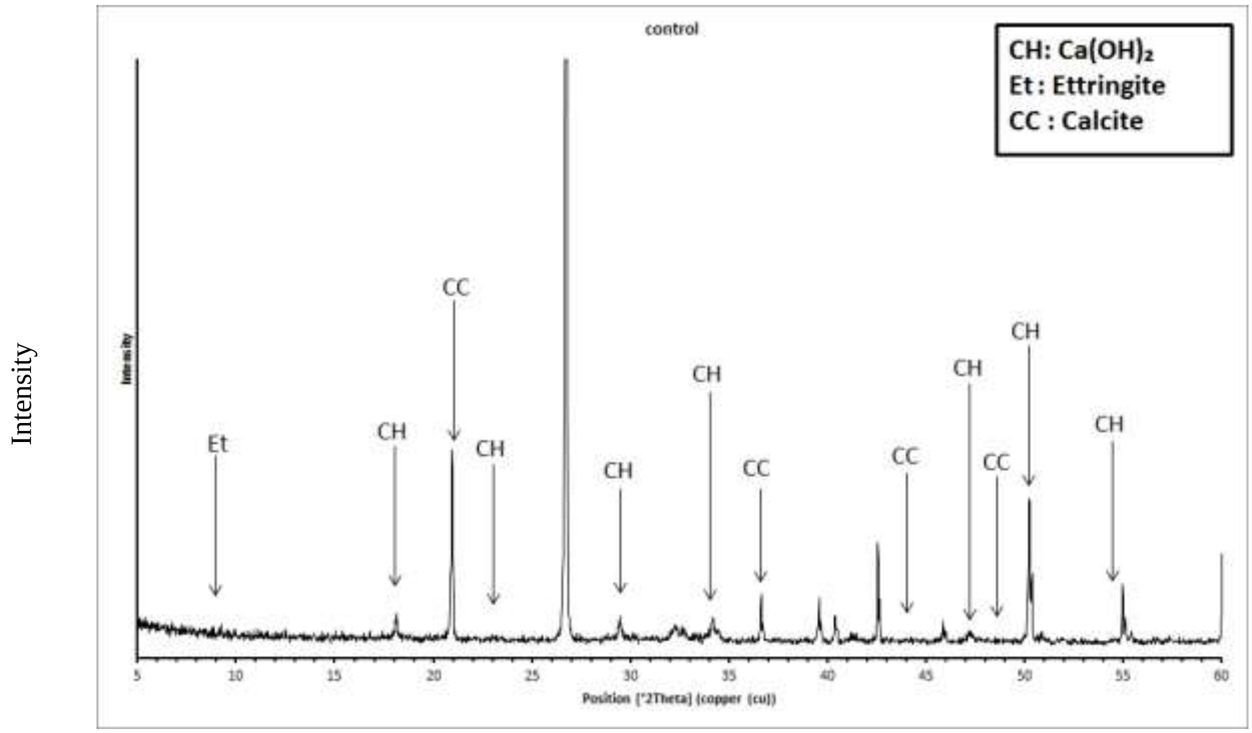


Figure. 10: XRD of control mix.

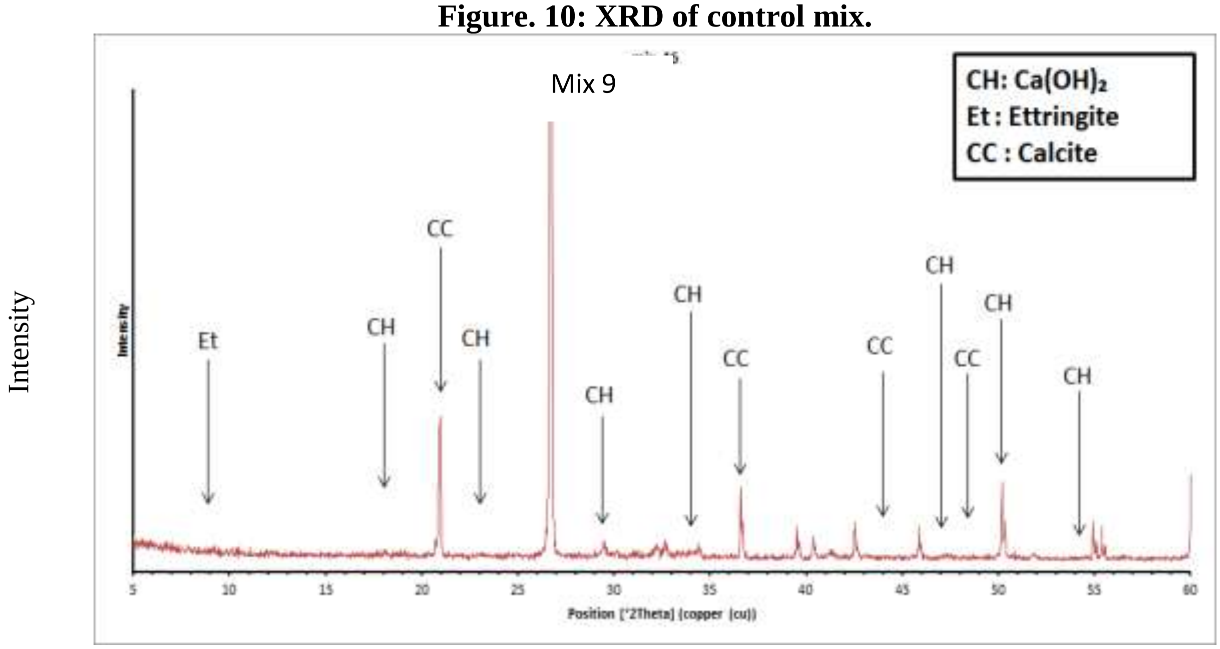


Figure. 11: XRD of mix # 9.

Intensity

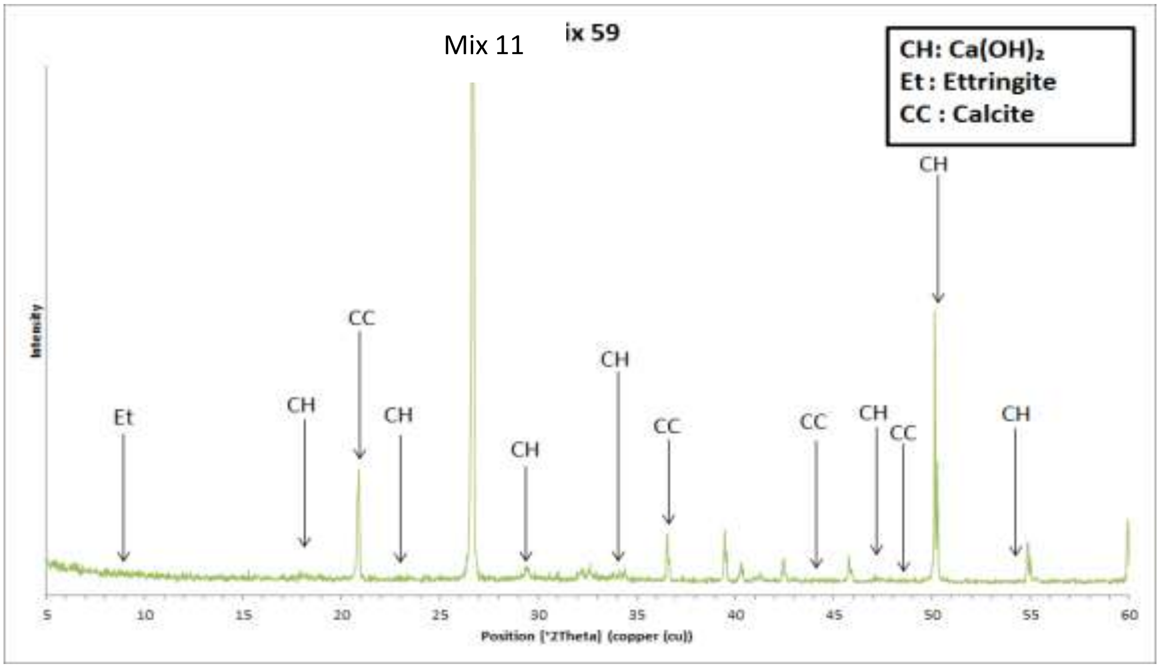


Figure. 12: XRD of mix 11.

Intensity

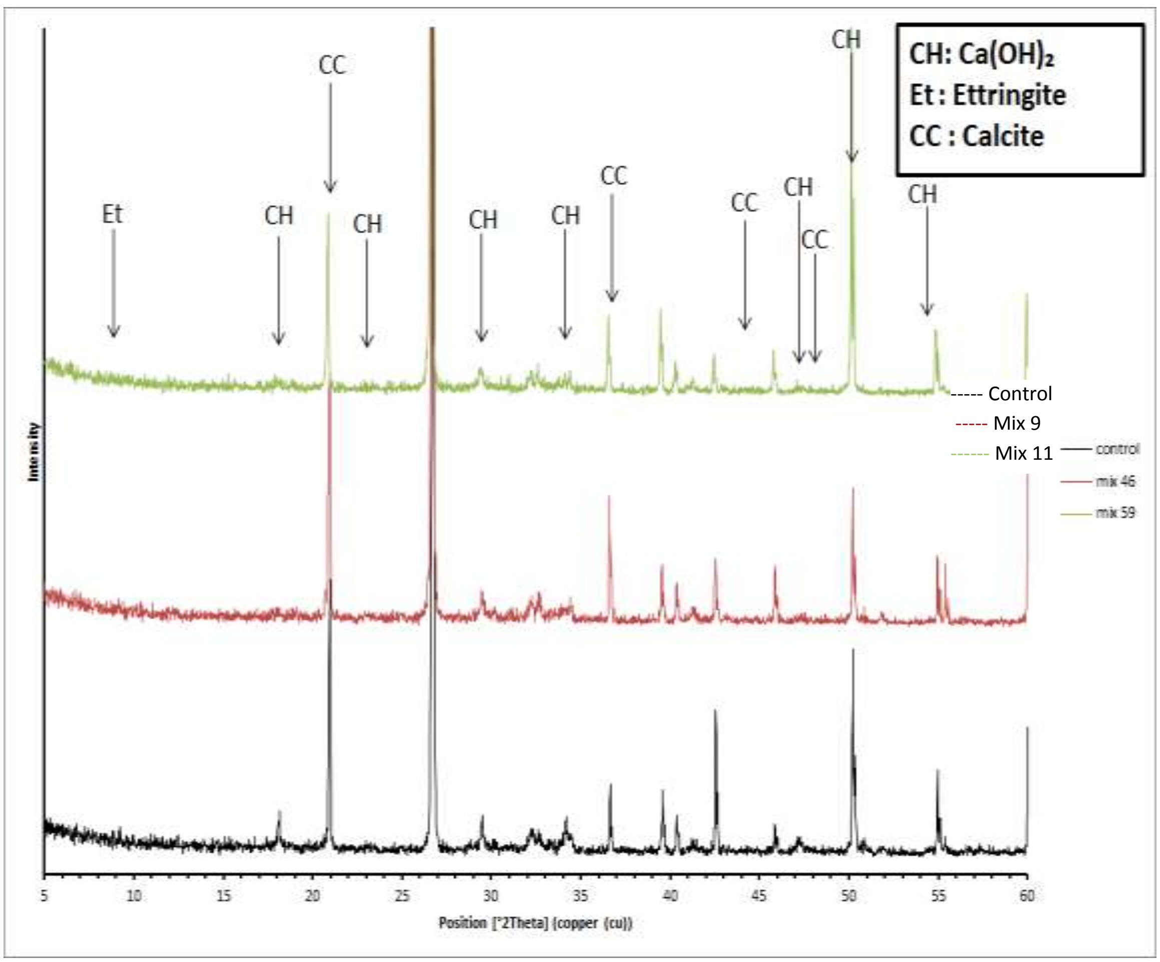


Figure. 13: XRD of control, # 9 and # 11 mixes.

IV. CONCLUSION

1- Replacing the cement content with 4% SF and 12% FA improved the compressive strength of cement mortar at 7 days age with 31%, while replacing 1.5% NS and 20% FA improved the compressive strength at 28 days age with 52%.
2- The temperature rise in mortars with NS, SF and FA began at early time compared to the cement mortar; with the same trend of heat flow. This can be attributed to the high reactivity of NS particles, which accelerated the hydration of cement and rapid its pozzolanic reaction with the residual CH from the hydration of cement.
3- The surface topography as well as the particles morphology with the wide spread of C-S-H gels in mix # 11 with 2% NS, 8% SF and 16% FA beside 74% OPC as compared to the control mix with 100% OPC, and the complete absence of CH phases within AFM images of mix # 11 highly indicated the reactivity of NS, SF and FA particles that results in the observed tremendous mechanical properties.
4- XRD showed high content of CH and CC in control mix, while less content was observed in mix # 9 with 1.5% NS, 12% SF and 12% FA beside 74.5% OPC and lowest content in mix # 11 that indicated the reactivity of fine particles and the formation of higher C-S-H content.

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