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Application of Silica Fume and Nanosilica in Cement and Concrete – A Review

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Abstract: This paper reviews the recent developments and present state of the application of silica fume (micro-silica) and nano-silica for sustainable development of concrete industry. This would save not only the natural resources and energy but also protect the environment with the reduction of waste material. Limited work is done on use of nano-silica and micro-silica in paste, mortar and concrete and whatever work is available is highly contradictory about their influence on mechanical strength development and durability properties. Various literatures have been reviewed to understand the influence of micro and nano-silica on fresh, hardened and microstructural properties of paste, cement mortar and concrete. Taking advantage of nanostructure and microstructure characterization tools and materials, the simultaneous and also separate optimal use of micro-silica and nano-silica will create a new concrete mixture that will result in long lasting concrete structures in the future.

Keywords: Micro-silica, Nano-silica, mortar, concrete, compressive strength.

1. INTRODUCTION

In the most customary sense, cement is a binder that sets and hardens independently as well as binds other materials together. Cement mortar is a building compound created by mixing fine aggregate and a selection of cementing material with a specified amount of water. Mortar has been used for centuries as a means of adhering bricks or concrete blocks to one another. Cement mortar continues to be used in many different types of construction such as the binder between bricks in walls, fences, and walkways, to make quick repairs in patio slabs and reset loosened stones or bricks in a walkway or retaining wall. Unfortunately, construction industry is not only one of the largest consumers of natural resources and energy, but is also responsible for large emissions of green house gases (GHGs) such as carbon dioxide responsible

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for global warming. It is estimated that one ton of Portland cement clinker production yields one ton of GHGs. In addition, due to the accumulation of natural aggregate extraction from quarries; it poses an immediate concern for sustainable construction development.

1.1 Concrete and Sustainability

Concrete is probably unique in construction, it is the only material exclusive to the business and therefore is the beneficiary of a fair proportion of the research and development money from industry. Concrete is a composite construction material composed primarily of aggregate, cement, and water, which is a nano-structured, complex, multi-phase material that ages over time.

Sustainability is defined by the World Commission on Environment and Development as the development that meets the needs of the present, without compromising the ability of the future generations to meet their own needs. It is basically an idea for concern for the well being of planet Earth with continued growth and human development. The current construction practices are based on the consumption of enormous quantities of building materials and drinking water, resulting in the scarcity of these resources after a long turn.

The sustainable development of the cement mortar would save not only the natural resources and energy but also protect the environment with the reduction of waste material. The mortar properties in fresh state such as workability are governed by the particle size distribution and the properties in hardened state, such as strength and durability, are affected by the mix grading and resulting particle packing. Rheological properties of a fresh cement paste play an important role in determining the workability of concrete. The water requirement for flow, hydration behavior, and properties of the hardened state largely depends upon the degree of dispersion of cement in water. Factors such as water content, early hydration, water reducing admixtures and mineral admixtures like silica fume determine the degree of flocculation in a cement paste (Sanchez and Sobolev, 2010).

1.2 Nanotechnology in Concrete

Nanotechnology is rapidly becoming the Industrial Revolution of 21st century (Siegel et al., 1999). It will affect almost every aspect of one's life (IWGN, 1999). In comparison to other technologies, nanotechnology is much less well-defined and well-structured. It is known that 'Nano' is a Greek word and means 'dwarf'. It does not mean dealing with dwarfs but it became a common word for everything which is smaller than 1 Micron or 1 million of a millimeter. 1 Micron is 1000 Nanometer. The nanoscience and nano-engineering (nano-modification) of concrete are terms that have come into common usage and describe two main approaches of applications of nanotechnology in concrete (Scrivener and Kirkpatrick, 2008; Scrivener, 2009).

Until today, concrete has primarily been seen as a structural material. Nanotechnology is helping to make it a multipurpose ‘‘smart’’ functional material. Concrete can be nano-engineered by the incorporation of nano-sized building blocks or objects e.g., nanoparticles, nano admixtures and nanotubes) to control material behavior and add trailblazing properties, or by the grafting of molecules onto the cement particles, cement phases, aggregates, and additives (including nano-sized additives) to provide the surface functionality adjusted to promote the specific interfacial interactions of the molecules. Recently, nanotechnology is being used in many applications and it has received increasing attention also in building materials, with potential advantages and drawbacks being underlined (Campillo et al., 2003; Pacheco-Torgal and Jalali, 2011).

1.3 Silica fume

Silica is the common name for materials composed of silicon dioxide (SiO_2) and occurs in crystalline and amorphous forms. Silica fume or micro-silica (SF) is a byproduct of the smelting process in the silicon and ferrosilicon industry. The American concrete institute defines silica fume as ‘Very fine non-crystalline silica produced in electric arc furnaces as a by-product of production of elemental silicon or alloys containing silicon’ (ACI Committee 226., 1987b). It is a grey colored powder, similar to Portland cement or fly ashes. It is an ultrafine powder collected as a by-product of the silicon and ferrosilicon alloy production and consists of spherical particles with an average particle size (diameter) of 150 nm. The main field of application is as pozzolanic material for high performance concrete (Prasad et al., 2003).

1.4 Nano-silica

Nanosilica is typically a highly effective pozzolanic material. It normally consists of very fine vitreous particles approximately 1000 times smaller than the average cement particles. It has proven to be an excellent admixture for cement to improve strength and durability and decrease permeability (Loland, 1981; Aitcin et al., 1981). NS reduces the setting time and increases the strength (compressive, tensile) of resulting cement in relation with other silica components that were tested (Roddy et al., 2008). Nano-silica is obtained by direct synthesis of silica sol or by crystallization of nano-sized crystals of quartz.

2. EFFECT OF ADDITION OF SILICA FUME AND NANO-SILICA

Silica fume has been recognized as a pozzolanic and cementitious admixture which is effective in enhancing the mechanical properties to a great extent. The pozzolanic reaction results in a reduction of the amount of calcium hydroxide in concrete, and silica fume reduces porosity and improves durability. It accelerates the dissolution of C-S and formation of C-S-H with its activity being inversely proportional to the size, and also provides nucleation sites for C-S-H. It is responsible for an additional increase in strength and chemical resistance and decrease in water absorption (Diab et al., 2012). The addition of micro and nano silica particles to cement paste could effectively reduce the degradation rate as well as its negative consequences. Even small additions (0.5 wt. % binder) of these particles are very efficient in terms of improvement in mechanical properties of cement based materials. This is especially pronounced at early ages and for concretes with regular strength grade. Therefore, application of SF and nS could be a successful method for improvement of low strengths of cement based materials. In addition, when low water content is used, economical advantages and higher durability are expected.

However, when mortars with nanosilica (nS) and silica fume (SF) are produced using low water content, the resulting material has inadequate workability for most applications. In this case, adding extra amount of water has to be done, but the benefits of mineral additions on the hardened state properties would be minimized. The use of plasticizers and superplasticizers (SP) is always desirable to improve the rheological properties without the need for addition of extra water (Qing et al., 2007).

3. LITERATURES REVIEWED

The fundamental processes that govern the most pertinent issues to the study of concrete technology (strength, ductility, early age rheology, creep, shrinkage, durability, fracture behavior, etc) are affected (dominatingly or not), by the performance of the material at the nanoscale. The use of supplementary cementing materials have become an essential part of the Portland cement concrete production, and the research on new materials with supplementary cementing potential is receiving considerable attention from the scientific point of view.

3.1 Influence on Fresh and Mechanical properties

Experiments using nanosilica and silica fume were conducted and the results showed that with 5% replacement of cement by nS (mean size 15±5 nm), 7

& 28-days compressive strength of mortars were increased by 20% and 17%, respectively, whereas 15% silica fume replacement increased mortar strengths by 7% and 10% compared with those of control Portland cement mortar. With the experimental analysis, it was proved that the compressive and flexural strengths of the cement mortars with nano-silica and with nano-Fe_2O_3 were both higher than that of the plain cement mortar with the same water to binder ratio (Li et al., 2004).

In a study to evaluate the effect of silica fume on the compressive strength, split tensile strength and modulus of elasticity of low quality coarse aggregate concrete was conducted whose results indicated that the type of coarse aggregate influenced the compressive strength, split tensile strength and modulus of elasticity of both plain and silica fume cement concretes. Incorporation of silica fume enhanced the compressive strength and split tensile strength of all concretes especially that of the low quality limestone aggregates (Abdullah et al., 2004).

In an experiment it was showed that the compressive and tensile strengths increased with silica fume incorporation, and the results indicated that the optimum replacement percentage is not constant but depends on the water-cementitious material ratio of the mix. They also found that compared with split tensile strengths, flexural strengths have exhibited greater improvements (Bhanja and Sengupta, 2005) while in another, it was showed experimentally that the compressive strengths of mortars with nano-SiO_2 particles were all higher than those of mortars containing silica fume at 7 and 28 days (Jo

Table 1: Compressive strength (MPa) after 7 and 28 days comparing the mortars containing nano-silica and silica fume (Jo et al.,2007).

	7 Days	28 days
OPC	18.3	25.6
SF5	22.5	35.1
SF10	24.7	37.4
SF15	26.1	38.0
NS3	39.5	54.3
NS6	46.1	61.9
NS10	49.3	68.2
NS12	50.7	68.8

et al., 2007). It was demonstrated that the nano-particles are more valuable in enhancing strength than silica fume. The addition of nano-silica and silica fume enhances mechanical properties of cement-based materials. Various conclusions were made regarding the effect of nano-silica that made cement paste thicker and accelerated the cement hydration process.

Compressive strengths of hardened cement paste increased with increasing the nano-SiO_2 content, especially at early ages. The pozzolanic activity of nano-SiO_2 is much greater than that of silica fume (Qing et al., 2007). The effect of silica fume on compressive and split tensile strength of lightweight concrete after high temperature was studied in which the level of importance of percentage of silica fume and heating degree on compressive and splitting tensile strength was determined by using analysis of variance (ANOVA) method (Tanyildizi and Coskun, 2008).

Researchers carried out an experimental investigation to study the effect of nano-silica on rheology and fresh properties of cement pastes and mortars. It was seen that nano-SiO_2 modified the characteristics of fresh mortars. The mortar with nanosilica showed the higher torque along all the testing period due to the plastic viscosity and yield stress increase (Senff et al., 2009). The addition of nano-silica reduced the spread diameter on the flow table of mortars,

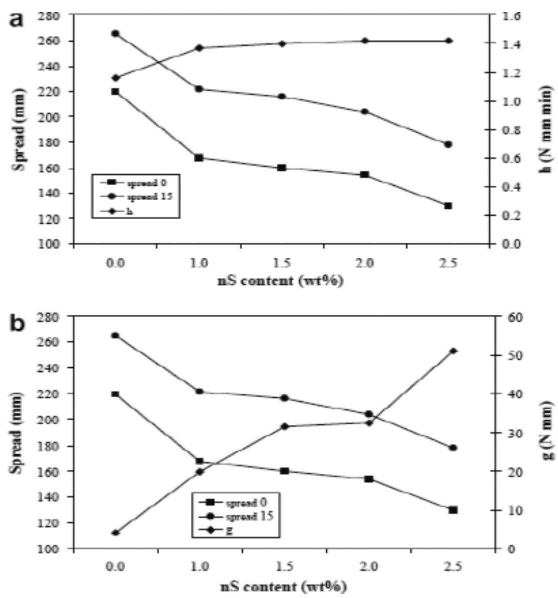

Figure 1: Influence of nS content on spread (after 0 and 15 strokes) and rheological parameters estimated after mixing (Senff et al., 2009).

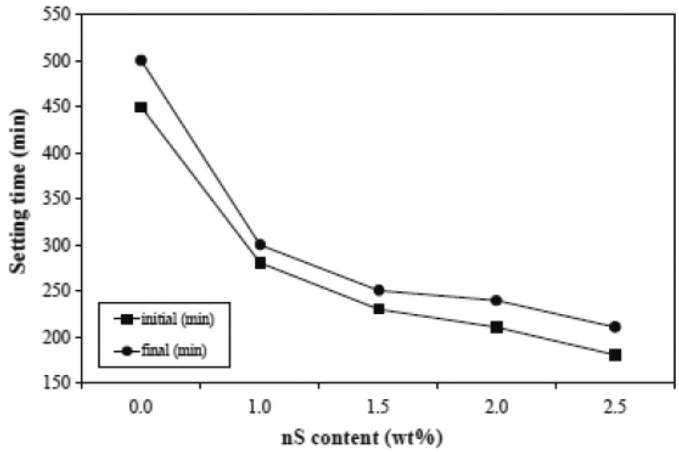

Figure 2: Variation of setting time (initial and final) on the mortar with the nS content (Senff et al., 2009).

due to the gain in cohesiveness of the paste. By adding nS, the beginning of setting was anticipated and the dormant period was reduced. Samples with nS (0–7 wt. %), SF (0–20 wt. %) and water/binder ratio (0.35–0.59), were investigated through factorial design experiments. Nanosilica with 7 wt. % showed a faster formation of structures during the rheological measurements.

It was investigated that there are effects of size of nS on compressive, flexural and tensile strength of binary blended concrete. It was found that the cement could be advantageously replaced by nS up to maximum limit of 2.0% with average particle sizes of 15 and 80 nm. Although the optimal replacement level of nano-Silica particles for 15 and 80 nm size were gained at 1.0% and 1.5%, respectively (Givi et al., 2010).

In another experiment, the properties of cement mortars with nano-SiO_2 were studied. Test data showed that nano-SiO_2 made cement paste thicker and accelerates the cement hydration process. Compressive strengths increased on increasing the nano-SiO_2 content (Ltifia et al., 2011). Researchers addressed the effect of nano-silica on the rheological behaviour and mechanical strength development of cementitious mixes. The addition of nano-silica to cementitious mixes produced a remarkable reduction of the mix workability (Berra et al., 2012). It was experimentally investigated about the influence of nano-SiO_2 on the Portland cement pastes. It was concluded that nano-SiO_2 appeared to affect the mechanical properties and the structure of high-strength cement pastes even in low concentration. The addition of nanosilica seemed to create two competing mechanisms in terms of the overall chemomechanical response of cement pastes. On one hand, the addition of extra water to the paste increased the water/cement ratio with all the well-established consequences, while the addition of nanoparticles tended to primarily increase the mechanical response. In that case, 0.5% up to 2% w/w of cement nanoparticles caused 20–25% strength increase despite the increased demand in water in the fresh state. In the second set of specimens the above mentioned problem was restricted (Stefanidou and Papayianni 2012).

It was reported that the use of nS and nano-TiO_2 in cement pastes and mortars have an effect on various properties. Rheological and flow table measurements were carried out. The values of torque, yield stress and plastic viscosity of mortars with nanoadditives increased significantly, reducing the open testing time in rheology tests. Meanwhile, the flow table values reduced. Mechanical properties were not significantly affected by nano particles in the range considered in the work (Senffa et al., 2012).

Mini-slump and rheometric tests were carried out on cement pastes made with three dose levels of nanosilica at different water/binder ratios. Cement paste workability resulted to be significantly lower than expected for the adopted water/binder ratios, as a consequence of instantaneous interactions between nanosilica sol and the liquid phase of cement pastes, which evidenced the formation of gels characterized by significant water retention capacity. The resulting reduction of the mix workability was avoided by suitable addition procedures of superplasticizers. No appreciable improvement in the compressive strength development of cementitious mixes by nanosilica addition was observed, in contrast with some results from literature. This confirmed conflicting experience on the problem, but some parameters (composition and content of mineral in mortars; water cement ratio and hydration degree of cement; size, number and distribution of capillary) affecting the strength development were identified and discussed (Berra et al., 2012). It was further studied about the effect of colloidal nano-silica on concrete incorporating single (ordinary cement) and binary (ordinary cement + Class F fly ash) binders. Significant improvement was observed in mixtures incorporating nano-silica in terms of reactivity and strength development (Said et al., 2012).

The effects of nano-silica on setting time and early strengths of high volume slag mortar and concrete was experimentally studied and results indicated that the incorporation of a small amount of nS reduced setting times, and increased 3-day and 7-day compressive strengths of high-volume slag concrete, significantly, in comparison to the reference slag concrete with no silica inclusion. The results also indicated that length of dormant period was shortened, and rate of cement and slag hydration was accelerated with the incorporation of 1% nS in the cement pastes with high volumes of fly ash or slag. The incorporation of 2% nS by mass of cementitious materials reduced initial and final setting times by 90 and 100 min, and increased 3- and 7-day compressive strengths of high-volume fly ash concrete by 30% and 25%, respectively, in comparison to the reference concrete with 50% fly ash (Zhang et al., 2012).

The effect of micro and nano-silica under various dosages of carboxylated-polyether-copolymer-type superplasticizer on the rheological properties of grouts in the fresh state was determined. Data mentioned that the maximum strength in nS-system was reached at 1.0 wt%, whereas in SF-systems, it was at a level of replacement in the order of 15 wt%. In addition, the highest compressive strength was obtained in SF-systems (Zapata et al., 2013). In another experiment, the addition of nano-silica (NS), nano-Al_2O_3 (NA) and nano-Fe_2O_3 (NF) powders and their binary and ternary combinations on the compressive strength of cement mortars containing flyash (FA) was determined and the results showed that addition of any single type of oxide powders at 1.25% increased compressive strength of the mortars much further than the other proportions (Oltulu and Sahin, 2013).

Thus, it was found that in most of the cases, addition of nano-silica and silica fume enhanced the compressive strength and flexural strength with optimized percentages.

3.2 Influence on Durability properties

The water absorption, capillary absorption and distribution of chloride ion tests indicated that the nano-silica concrete has better permeability resistance than the normal concretes. This was evident from the studies carried out that the water permeability resistant behaviour whose results showed that nS concrete is stickier than normal concrete due to the larger specific surface area (Ji, 2005). Through various experiments carried out, it was evident that for mixtures with 0.35W/B, the water absorption and apparent porosity reached the maximum values for mortars with 7% nS (Senff et al., 2010). The factorial design showed that the unrestrained shrinkage and weight loss of mortar did not follow a linear regression model and the mortars with nS showed higher values than SF. With 7 days the shrinkage increased 80%, while at 28 days it increased 54%. The chloride permeability of concrete containing nano-particles (TiO_2 and SiO_2) for pavement and compared with that of plain concrete, concrete containing polypropylene (PP) fibers and concrete containing both nano-TiO_2 and PP fibers (Zhang and Li, 2011). The test results indicated that the addition of nano-particles refines the pore structure of concrete and enhances the resistance to chloride penetration of concrete. The nS addition decreased the apparent density and increased the air content in the mortars. It was investigated that the addition of superplasticizers in 1% w/w of cement reduced the water demand and the strength increase varied from 30% to 35% (Stefanidou nad Papayianni 2012); Quercia et al., 2012) addressed the characterization of six different amorphous silica samples with respect to their application in cement paste. It was determined that the addition of 0.5 to 4.0% nano-silica to the cement paste reduced the water demand without the use of superplasticizers. A linear relationship between the deformation coefficient and the specific surface area of nS/mS particles was confirmed. Higher deformation coefficients (E_p) for amorphous silica with high content of nanoparticles were found which were bigger than that of cement. Guidelines in compressive strength assessment of concrete modified with silica fume due to magnesium sulfate attack were suggested. These guidelines could be used to check the safety of any structural element subjected to any concentration of magnesium sulfate attack after any service time knowing the mix proportions of the used concrete mix. Application of these guidelines shows the hazards of using Portland cement and silica fume in concrete subjected to magnesium sulfate attack.

The possibility of using waste ground ceramic powder and the combination of ground ceramic powder with nano-silica as a replacement for cement was studied and the results showed that concrete with ceramic waste powder ultimately demonstrated only minor strength loss, and ceramic waste powder exhibits very good pozzolanic reactivity and could be used as a cement replacement. Water absorption capacity of concrete was decreased by using pozzolan. The greatest decrease was observed in the sample containing 20% pozzolan (Heidari and Tavakoli 2013). Kawashima (2013) summarized the current work being done at ACBM-NU on nano-modification of cement-based materials. Shear rheology results indicated that nano clays have an immediate stiffening effect, governed by flocculation not water adsorption, but with little influence over time.

3.3 Influence on Microstructural properties

The Scanning Electron Microscope (SEM) observations revealed that the nano-particles were not only acting as filler, but also as an activator to promote hydration and to improve the microstructure of the cement paste if the nano-particles were uniformly dispersed (Li et al., 2004). The results of the experimental analysis indicated that nano-scale SiO_2 behaves not only as a filler to improve microstructure, but also as an activator to promote pozzolanic reaction (Qing et al., 2007); Jo et al., 2007).

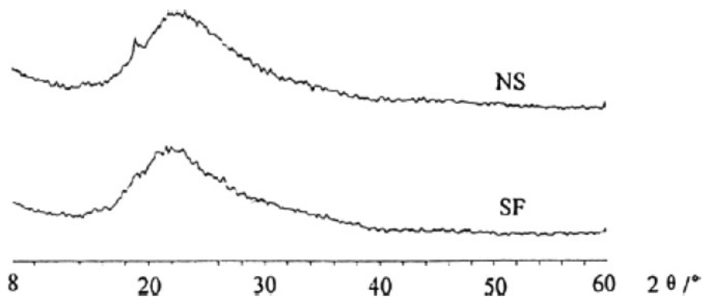

Figure 3: XRD powder pattern of nano-silica and silica fume (Qing et al., 2007).

The X-Ray Diffraction (XRD) showed the presence of CH, already after 9 hours, in samples with nanosilica addition. The nS addition contributed to an increased production of CH at early age compared with samples without nanosilica (Senff et al., 2010). Impressive changes were recorded in the structure of nanomodified samples as the calcium silicate crystal size was larger in samples with high nano-SiO_2 content (Stefanidou and Papayianni (2012). This was obvious in pastes with 5% nanoparticles where crystals were formed at 14 days, while at the same age, in pastes with 1% nano-SiO_2 the average crystal size was 600 nm. Microstructure observation also recorded a denser structure in nano-modified samples. The results showed that nS can reduce the size of CH crystals at the interface more effectively than SF (Qing et al., 2007).

It was showed that C–S–H gels from pozzolanic reaction of the agglomerates cannot function as binder. The nano-indentation test results revealed that the pozzolanic C–S–H gels from reacted agglomerates showed nearly the same properties as the C–S–H gels from cement hydration (Kong et al., 2012).

The effect of colloidal nano-silica on concrete and significant improvement was observed pertaining to refinement of pore structure and densification of interfacial transition zone. Micro-structural and thermal analyses indicated that the contribution of pozzolanic and filler effects to the pore structure refinement depended on the dosage of nano-silica (Said et al., 2012).

4. CONCLUSION

Nanotechnology has the potential to be the key to a brand new world in the field of construction and building materials. The role and application of the nano and micro silica particles with cementitious materials have been reviewed and discussed in details. It is evident from the literatures reviewed that none of the researchers have carried out extensive or comprehensive study of the properties of paste and mortar, with nano silica, micro silica and their simultaneous use. There is a limited knowledge about the mechanisms by which nano silica & micro silica affects the flow properties of cementitious mixes. In India, the research work on use of nano silica is still in elementary stage. Thus, a need arises to study extensively the various properties of paste, mortar, and concrete containing various percentages of nano silica, micro silica alone as partial replacement of cement and then studying their combined percentage effects. As the properties of nano-silica and micro-silica reported in literatures relate with those manufactured or exported from abroad, there is urgent need to study the effect of these materials (manufactured in India) on various properties of cement paste, mortar and concrete. Major parties in the construction materials industry should divert more funds to research work on incorporating nanotechnology in construction materials.

Thus, the main motive is to provide practical information, regarding the strength, sustainability & durability properties of nano silica, micro-silica and their simultaneous use in paste, mortar and concrete. Also, the aim is to carry out the extensive studies to conceive the general purpose of testing new sustainable building processes and modern production systems, aimed at saving natural raw materials and reducing energy consumption. Taking advantage of nanostructure and microstructure characterization tools and materials, the simultaneous and also separate optimal use of micro-silica and nano-silica will create a new concrete mixture that will result in long lasting concrete structures in the future. Thus, there is a gap or room available for further research towards the fruitful application of especially nano-silica for construction with different nano structure characterization tools, which will be enable to understand many mysteries of concrete.

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