INTERNATIONAL DEVELOPMENT PLANNING REVIEW
ISSN:1474-6743 | E-ISSN:1478-3401

A STATE OF CRITICAL REVIEW ON EFFECT OF NANOMATERIALS ON HIGH VOLUME FLY ASH CONCRETE COMPOSITES

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ABSTRACT :

The increasing demand for rigid pavements has necessitated the availability of cement concrete in which cement is the principal material. But production of cement is energy entringic. Using high volume fly ash, greater than 50% as a cement alternative has encouraged researchers as it reduces CO_2 emissions by minimizing cement production. But low strength of high volume fly ash concrete (HVFAC) at early edges are the main barrier to replace the cement with significant amount of fly ash. Recently various nano materials have been extensively used to achieve sustainability goals in the HVFA concrete. This paper presence state-of-the-art review involving the uses of different nano materials for enhancing the property of high volume fly ash concrete. The effects of nano material on the green concrete mechanical as well as durability properties are analyze. It is demonstrated that nano materials including Nano –SiO_2, Nano-Fe_2O_3, Nano Al_2O_3 Nano TiO_2, CNTs, Nano Clay, Nano ZnO, GO, PCE etc. can be used effectively to enhance the micro structures, mechanical property such as compressive strength, flexural strength and split tensile strength of the modified concrete. It also improves their durability properties such as water permeability, sorptivity, antichloride penetration, anti errosion etc. this paper may provide deep insight into the role of different nano materials inclusion in the concrete composite to enhance their overall properties.

Keywords: Energy intringic, HVFAC, Strength, Durability, Nanomaterials, C-S-H, Optimum.

1. INTRODUCTION

It is estimated that 10 billion tons of cement concrete is produced worldwide. The amount of consumption of these are bound to increase with increasing demand, especially from the large developing countries like India, China, Brazil, South Africa etc. wherein China is already the largest producer of cement followed by India (Schneider et al., 2011).

Cement production utilizes lot of energy at various stages. About 0.8 tons of CO_2 is estimated to be produced for each ton of production of Portland cement clinkers (Skinner et al., 2010). This in turn leads to chemical decomposition of limestone thereby again emitting large volume of CO_2 (Shanks et al. 2019, Salas et al. 2016). The emission of CO_2 by cement industry is 5 -7 % of total CO_2 emission in the world (Mathieu, 2006; Shanks et al. 2019). CO_2 is one of the significant greenhouse gases leading to disastrous global warming. This emission continues to increase with time resulting in environmental degradation (Salas, 2016). Lately, the growing concern for environment, has encouraged researchers to work for the development of green concrete. Green Concrete uses sustainable waste cementitious materials which results in reduced emissions and air pollutants (Xu & Shi, 2018).

One of the sustainable waste cementitious material which has seen global utility is fly ash obtained from coal based thermal power plants. Use of fly ash promotes energy conservation and eco-friendly construction. The utilization of fly ash in India and China are about 38% and 45% respectively (Dwivedi et al. 2014). The utilization of fly ash still remains limited and fly ash in concrete rarely exceed 30%. As per the definition given by Mehta et al. (2014); a concrete having a minimum cement replacement level of 50% by fly ash is termed as high volume fly ash concrete (HVFAC). Even though the HVFAC have lower compressive strength, the long term compressive strength is comparable with regular concrete after 90 days because of late hydration of fly ash (Mehta et al. 2002 Shivasundaram et al. 1989). Kumar et al. (2007) have reported that HVFAC mixture containing 50 to 60% fly ash could be designed to fulfil the requirement of strength and durability suitable for pavement quality concrete.

In spite of sustainable nature of high volume fly ash concrete (HVFAC), it has not gained popularity in utilization for road construction because of its lower initial strength as it shall need longer time after casting to open for traffic. Results have shown that nanotechnology can be one of the ways to deal with such situation. Nanotechnology can be defined as the design, production, characterization and application of devices, structures and systems by controlling size and shape of any material at the nanoscale. Studies have shown that nanomaterials such as carbon nanotubes, nano titanium dioxide, nano silica, nano zinc oxide, nano aluminium oxide and nano iron oxide to have improved strength and durability characteristics in various types of concrete (Kumar et al., 2007; Gopalakrishnan et al., 2010).

Despite all these superior qualities HVFAC cannot be used effectively for concrete roads due to lesser early strength. hence modification is sine – qua – non for achievement of early strength and therefore, in the present study, research of different researchers are studied about the effect of nanomaterials in the concrete and high volume fly ash concrete (HVFAC) are outlined.

Nanotechnology is certainly a key innovation in the construction in the construction sector and has led to a step forward in high-performance building materials with endurance. The outstanding scientific breakthroughs in the nanotechnology field have enabled the effective utilization of diverse nanoscale materials with distinctive characteristics, improving the basic properties of traditional construction components. (Huseien G. F. 2023). Following nanomaterials are reviewed;

2. NANOSILICA

Nano Silica is one of the most widely used nano-materials in concrete due to the observed enhanced properties and is found in powder or slurry form. It has a variable size with spherical morphology (Quercia & Brouwers, 2010).

Nanosilica has found its way into research to overcome the problems encountered using silica fumes. Nanosilica usage have similar positive reactions and has higher pozzolanic activity as compared to silica-fumes, which can be judiciously utilized to prepare concrete with better properties.

2.1 Effect of Nanosilica on strength of cementitious material

Zhang et al. (2011) found that usage of nano silica in concrete with high volume fly ash increases strength as depicted in figure under. Increased heat of hydration and early stage pozzolanic reactions was observed. At the same time, inactive period was reduced.

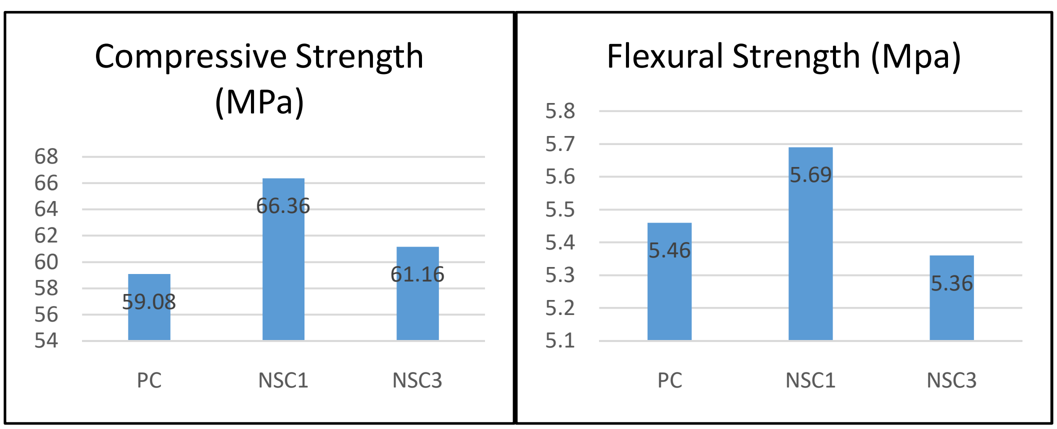

Figure 1.1 :- Variation of strength using different percentage of nanosilica for high volume fly-ash concrete (Zhang et al., 2011)

Use of nano silica on fly ash mortar compensates the negative effects associated to fly-ash incorporation in terms of setting time and initial strength (Lin et al., 2008). As per Sobolev et al. (2006) strength of HVFAC with addition of nano silica increased by 15–20%.

Rong et al., (2015) experimentally concluded that 3% addition of nano-silica resulted in enhancement of mechanical and microstructural properties of ultra-high performance cementitious composite. It was found that 1%, 3%, 5% addition of nano-silica resulted in increased compressive strength by 13.33%, 26.66%, 20% for 28 days and 15.58%, 29.87%, 20.77% for 90 days respectively with reference to normal concrete. Similarly, flexural strength was increased by 5.88%, 11.76% and 8.82% respectively at 28 days age for the above mentioned proportions. Similar trend was also observed for 90 days.

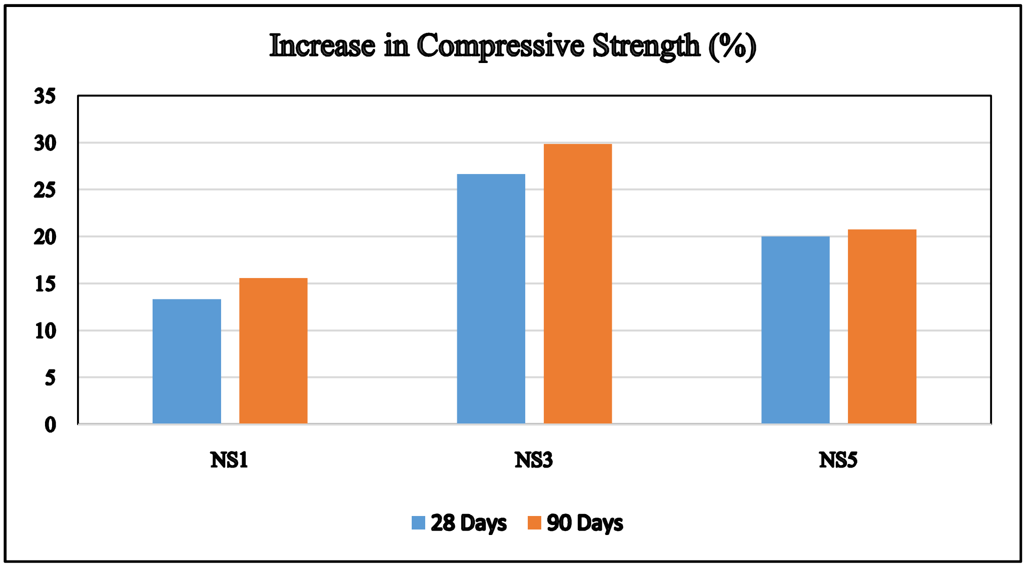

Figure 1.2:- Increase in compressive strength for ultra high performance cementitious composites using different percentage of nano-silica (Rong et al., 2015)

Madandoust et al. (2015) studied the effect of nano-silica and nano iron oxide on self-compacting concrete (SCC). The amount of fly ash was 25% of the cement in all mixtures and the compressive strength found for 3rd, 7th, 28th and 90th day are given under:

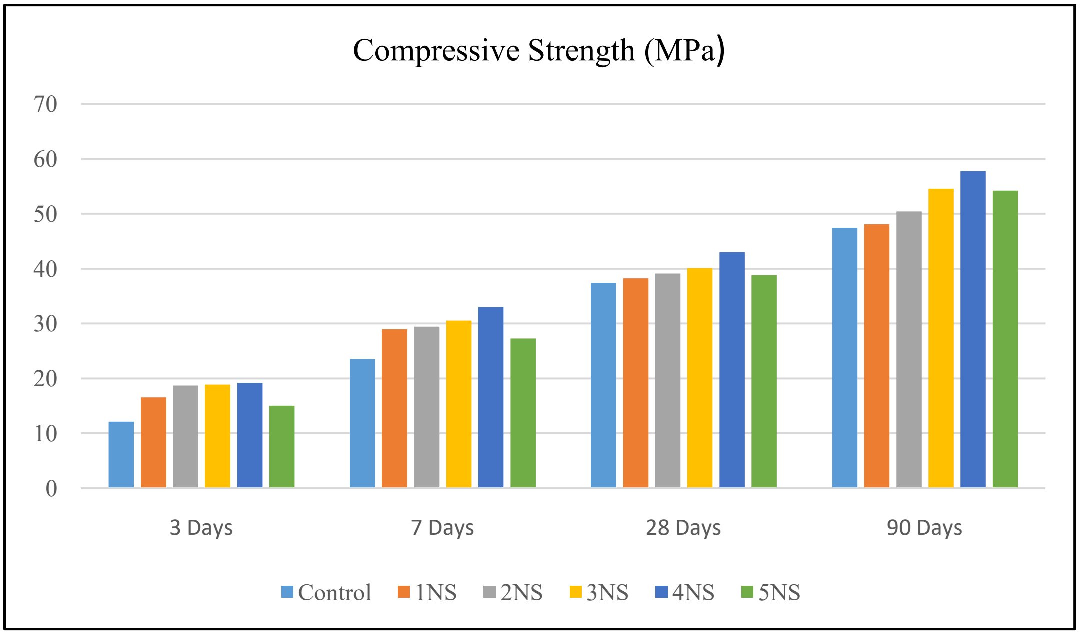

Figure 1.3 : - Variation of compressive strength using different percentage of nanosilica for self-compacting concrete (Madandoust et al., 2015)

Cement paste and concrete mixtures with 15% to 25% of the cement content replaced with fly ash and four different percentages (1.5%,3%,5% and 7.5%) of nano silica were prepared and studied. Nano-silica accelerated the reactivity of early- age concrete with fly ash . The optimal ratio of nano silica was 5% with 15% fly ash (Ehsani et al., 2017).

Labaj et al., (2016) conducted experiments with 0%, 40% and 60% fly ash replacement of Portland cement and with and without 0.5% nano-silica. Even with small dosage of nano-silica, results of compressive strength test, static and dynamics modulus of elasticity and resistivity against water pressure were found significantly improved.

Roychand, De Silva, & Setunge, (2018) conducted experiments to find alternatives for total replacement of ordinary portland cement. Varying percentages of fly-ash, nano-silica, granulated blast furnace slag (GBFS) and hydrated lime as cementitious additives was considered as raw materials for the experiment. It was suggested that 5 % nano-silica was the optimal value for usage as increasing the nano-silica content further resulted in micro-cracks, thereby preventing strength gain. Observations were made that silicious hydrogarnets increases with increase amorphous nano-silica, but it decreased with increase in hydrated lime content.

It was found that concrete produced by mixing 58% OPC, 40% fly-ash and 2% nano-silica had high early compressive strength (Sun, Shen, Tan, & Tanner, 2019). This was attributed to the fact that addition of nano-silica resulted in early stage C-S-H gel formation and also reduced calcium hydroxide thereby resulting in increase in compressive strength.

Liu et al. (2019) studied strength and microstructures of steam cured fly-ash cement mortar with 50% replacement of cement by fly-ash with varying dosage of nano-silica. Flexural and compressive strength revealed that nano-silica significantly increases the early strength of the fly-ash based cement mortar due to high degree of hydration as well as pore refinement.

Bimal et al. (2019) studied effect of strength of high volume fly ash pavement quality concrete due to addition of nano silica. PQC were made by replacing 55% cement with F-type fly ash. Flexural strength was found to be maximum for HVFAC modified with 2% nano silica. Similar trends were found for compressive strength tests and split tensile strength and for the nano silica modified HVFAC, 2% value was found to be the maximum one. As compared to Normal Concrete (M45), the flexural strength, which is the most important guiding parameter for the strength of rigid pavements, for 2% NS+HVFAC was found to be even more than that of Normal Concrete. Variation is shown below;

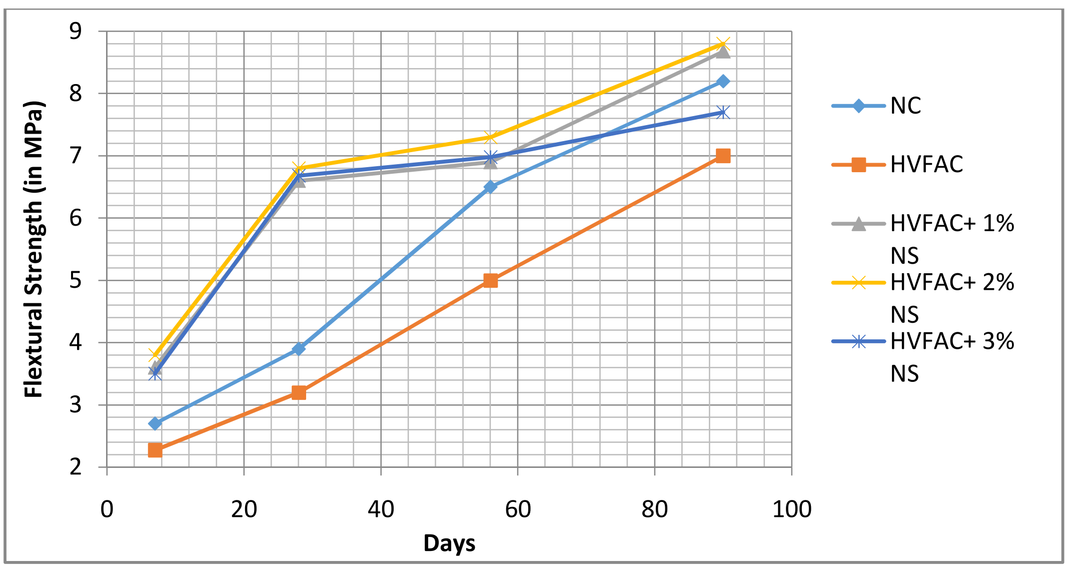

Fig 1.4: Variation of flexural strength for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at different ages.

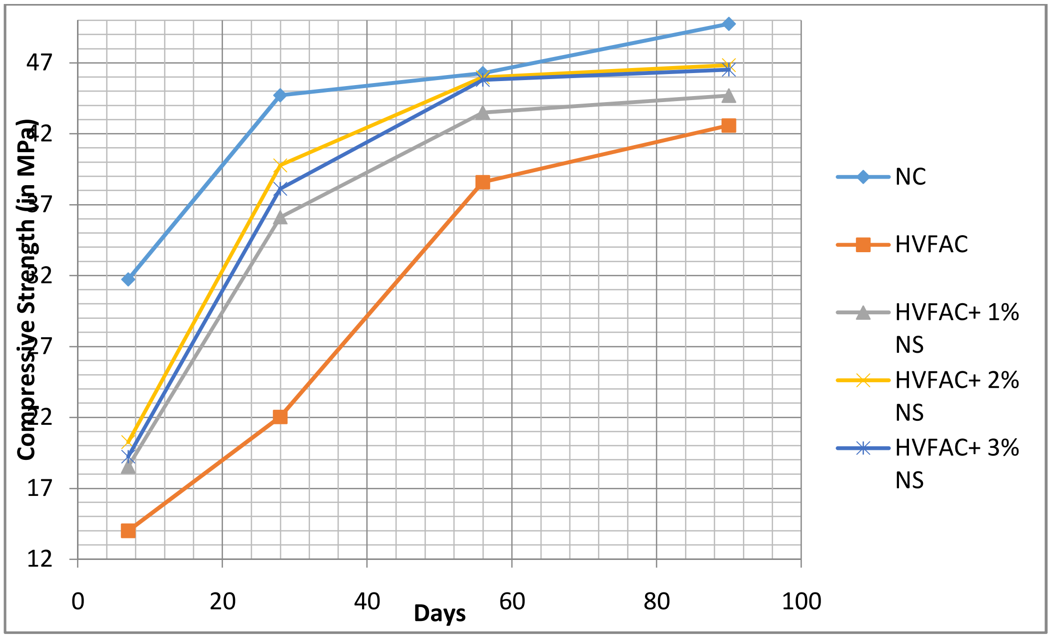

Fig 1.5: Variation of compressive strength for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at different ages.

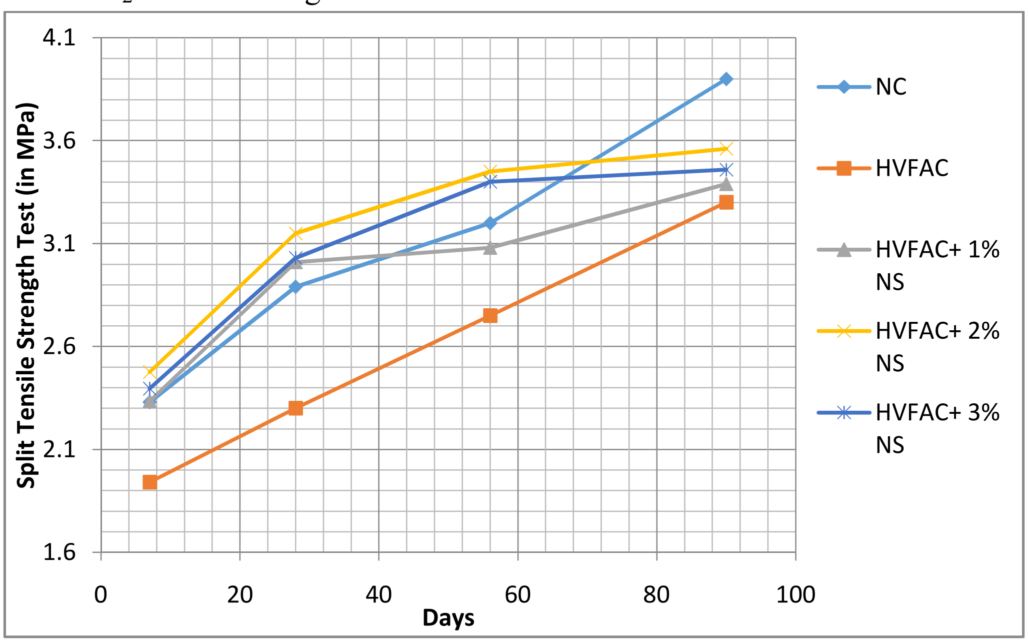

Fig. 1.6: Variation of split tensile strength for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at different ages.

2.3 Effect of Nanosilica on durability of cementitious material

Concrete with nano silica and fly ash has earlier pozzolanic reaction resulting in formation of C-S-H gel as compared to only fly ash replacement (Ji, 2004). It had a dense structure and its water permeability was also found to be less.

Cement pastes and concrete mixtures with incremental replacement of fly ash and four different percentages of nano-silica were prepared (Ehsani et al., 2017). Nano-silica accelerated the reactivity with passage of time. Depending upon interfacial transition zone (ITZ), water absorption and sorptivity coefficient of concrete, the optimal ratio of 5% nano silica with 15% fly ash was determined to be the best combination.

Madandoust et al., (2015) worked on durability of SCC containing nano-silica and Nano Iron oxide. The amount of fly ash was 25% of the cement in all mixtures. For nano silica optimum dosage was found to be 4%. Water absorption was found to reduce.

Supit and Shaikh (2015) studied the durability properties of HVFAC using nano silica. Four different sections of samples were prepared. These include normal concrete, concrete with cement replacement of 2 and 4% by nano-silica, concrete with 40 and 60% replacement of cement by fly ash. The fourth section was prepared with nano silica as tertiary replacement of fly ash. The sorptivity value obtained is furnished under

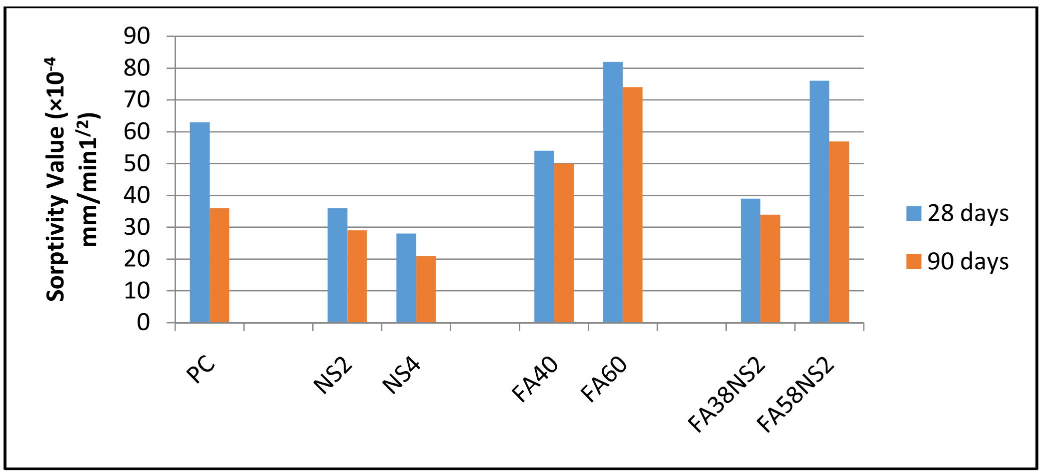

Figure ; 1.7 Variation of sorptivity value for different percentage of nanosilica for HVFAC (Supit and Shaikh, 2015)

Pitroda and Umrigar (2013) determined the water absorption and sorptivity coefficients for 9 samples of M25 (A1) and M40 (A2) with 10% (B1, B5), 20% (B2, B6), 30%(B3, B7) and 40%(B4, B8) replacement of cement by fly-ash. The results of durability obtained are depicted under:

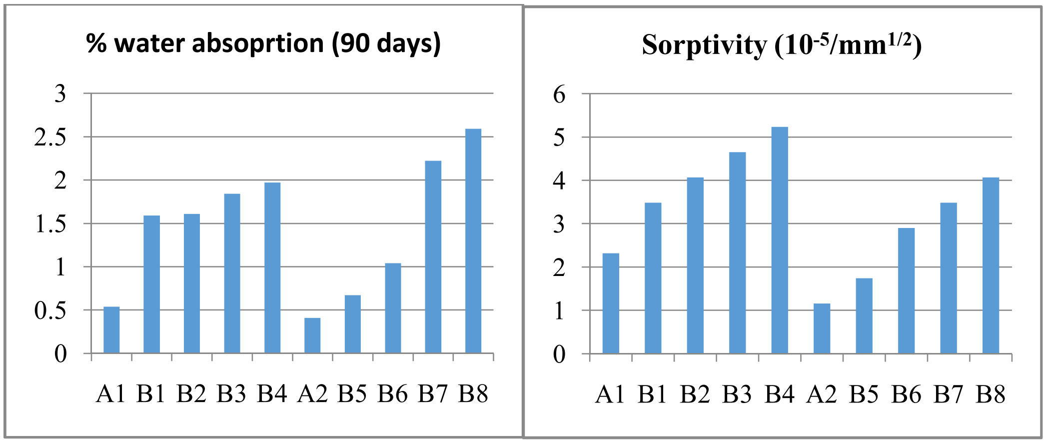

Figure 1.8: Water absorption and sorptivity coefficients for concrete having different percentages of fly-ash (Pitroda and Umrigar, 2013)

Behfarnia and Salemi (2013) determined the durability aspects of nano-silica modified concrete (3%, 5% and 7%) against frost resistance. ASTM C666A was used to stimulate frost cycle. Strength loss, mass loss and increase in water absorption after 300 cycles along with water absorptions were measured and plots are given below ;

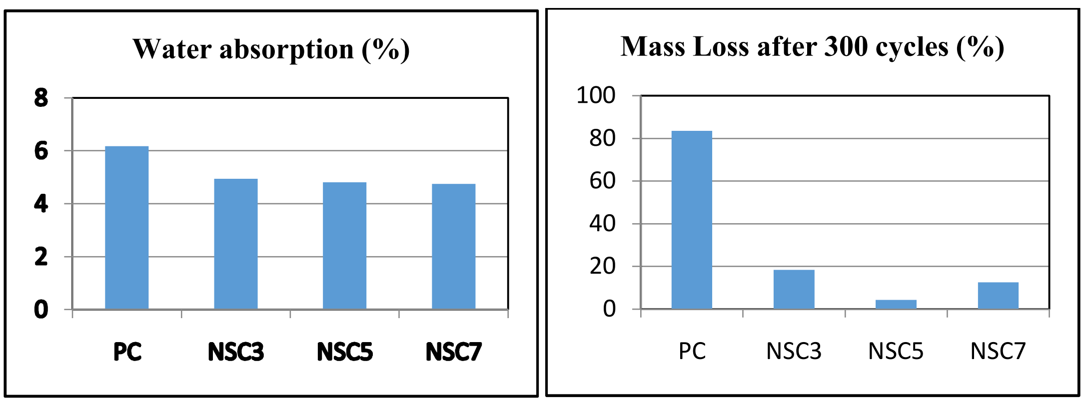

Figure 1.9: Water absorption and mass loss after 300 thaw cycles for samples containing different percentages of nanosilica (Behfarnia and Salemi, 2013)

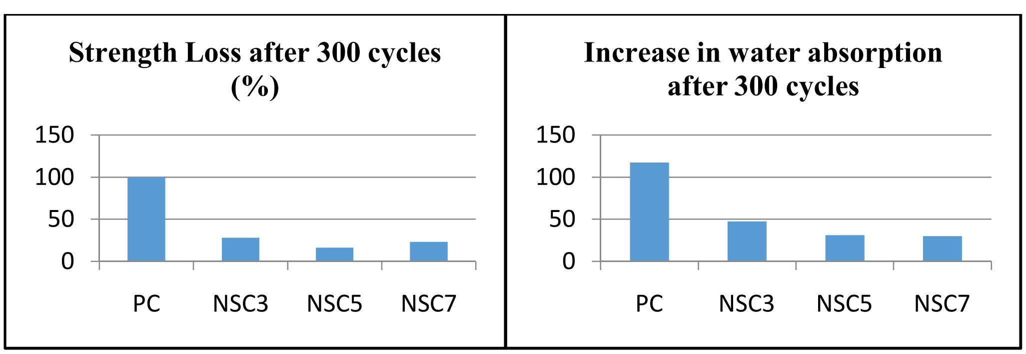

Figure 1.10: Strength loss and increase in water absorption after 300 thaw cycles for samples containing different percentages of nanosilica (Behfarnia and Salemi, 2013)

Bimal et al. (2019) Studied variations of durability parameters such as water permeability, sorptivity, Rapid Chloride Penetration by adding 1%, 2% and 3% nano silica in high volume pavement quality concrete. Results were shown in the figures. It has been observed through a series of experiments that HVFAC+2% NS samples gives optimum results

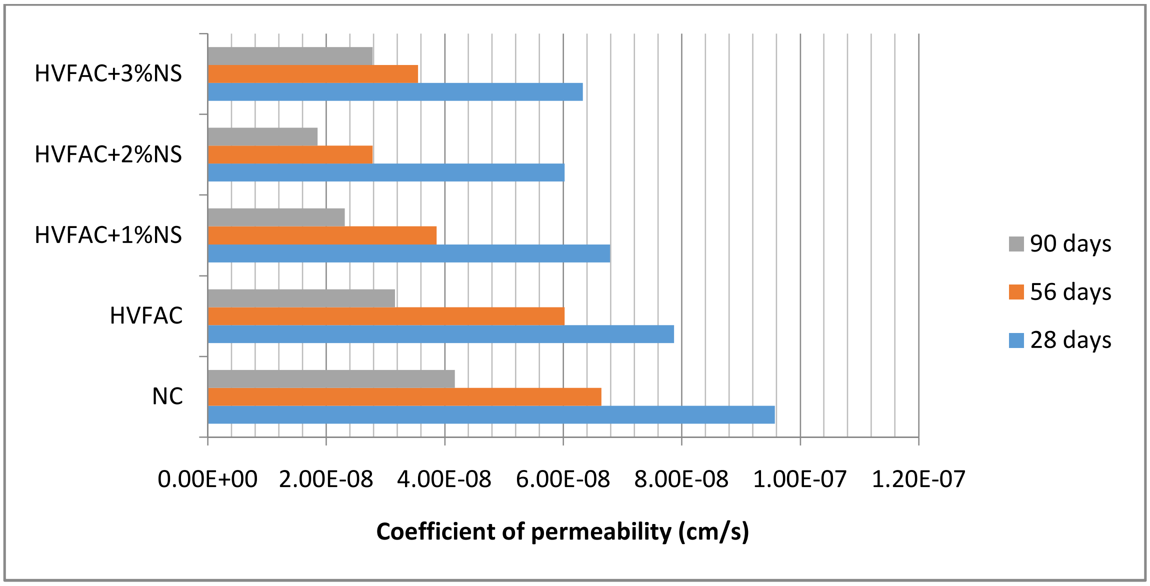

Fig 1.11: Variation of permeability for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at different ages.

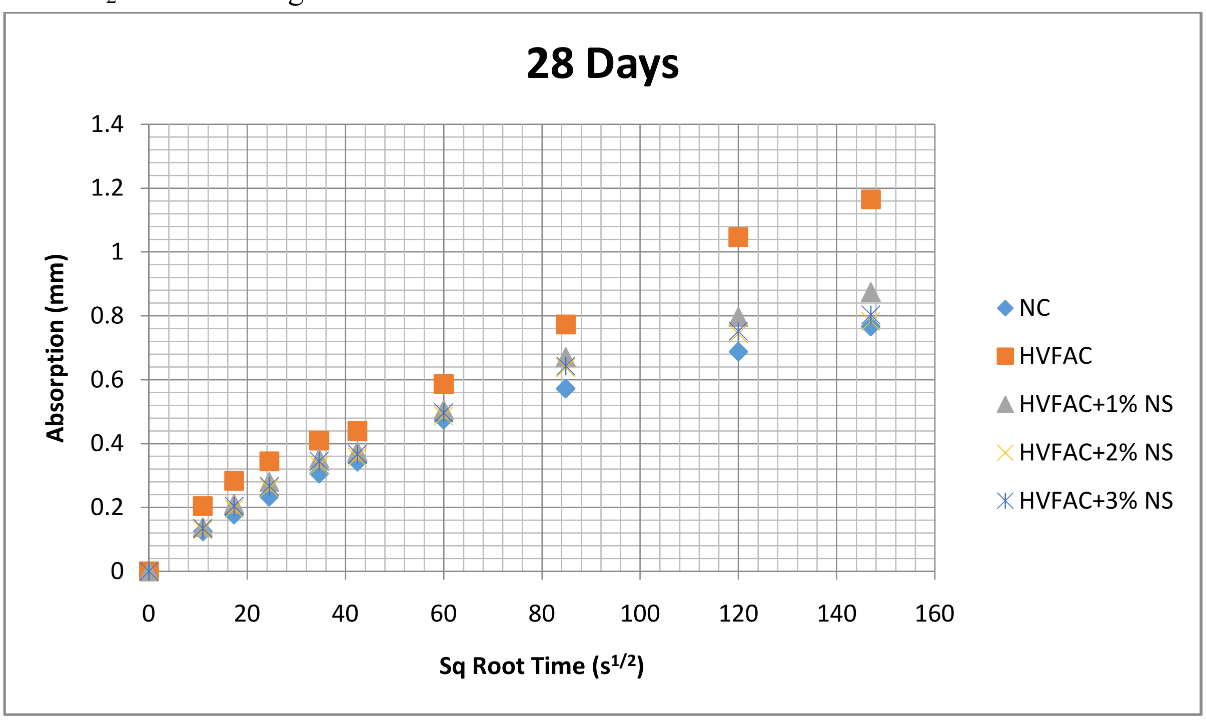

Fig 1.12: Absorption for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at 28 days.

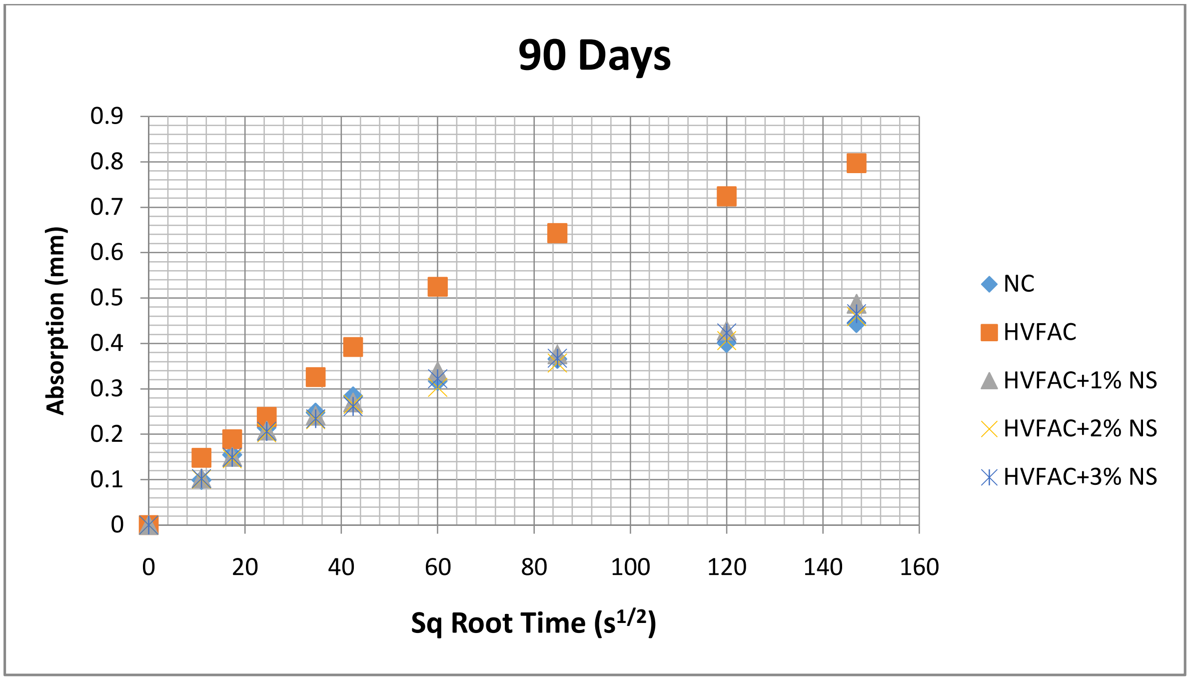

Fig. 1.13: Absorption for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at 90 days.

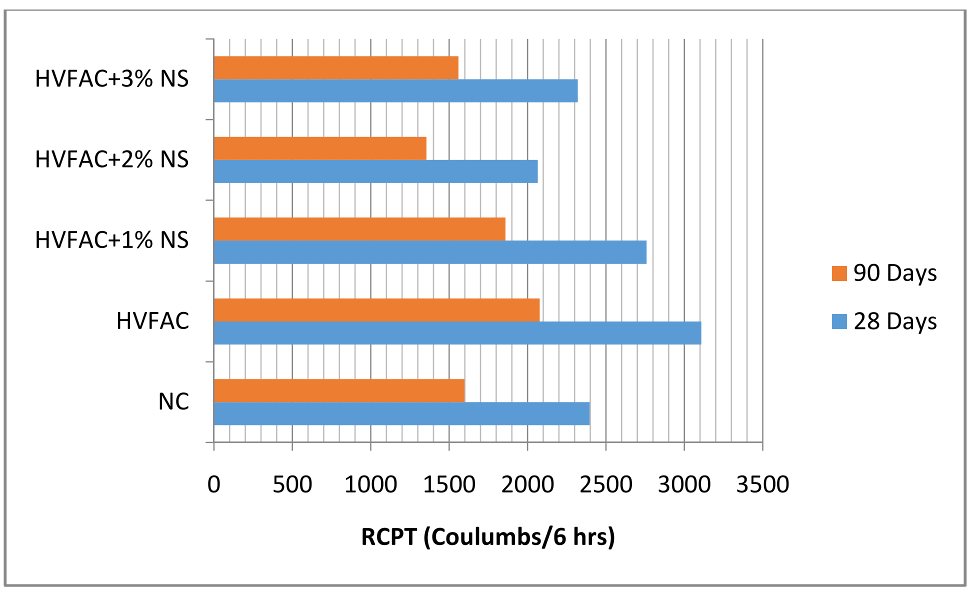

Fig. 1.14: Variation of RCPT for NC, HVFAC and HVFAC modified with 1%, 2% and 3% Nano SiO_2 at 28 days and 90 days.

3. Nano Ironoxide

Nazari et al., 2010 investigated compressive strength and workability of concrete prepared by partial replacement of cement using nano iron oxide particles of 0.5%, 1.0%, 1.5% and 2% by weight. The size of nanoparticles were determined to be 15 nm. It was observed that the strength of concrete increased with respect to control samples with increased replacement. However, the ultimate strength was attained at 1.0% replacement. However, the workability decreased continuously with increase in replacement. In another study, Nazari et al., 2010 investigated split tensile strength, flexural strength and setting time of concrete with same replacement as above. The split tensile strength and flexural strength showed an increasing trend with increase in replacement, however, the setting time decreased. It was thereafter suggested that the split tensile strength could improve by using needle type reinforcement.

Yazdi et al., 2011 conducted research to determine the compressive and tensile strength of cement mortar containing Fe_2O_3 nanoparticles of 1, 3 and 5% by weight of cement. An increase in mechanical properties was observed for 1 & 3% use of nano iron oxide which decreased for 5%. SEM images confirmed that the nanoparticles fill up the pores. Reduction in large crystals of $Ca(OH)_2$ nanoparticles were observed, and thus, hydrated products were observed to be denser and compact by filling C-S-H gel nanostructures. The decrease for 5% addition of nano-Fe_2O_3 were subjected to the fact that the nanoparticles at higher concentration starts to agglomerate.

Mechanical strength properties such as compressive, flexure and split tensile strength as well as co-efficient of water absorption on high performance self-compacting concrete was measured. 4% addition of nano-Fe_2O_3 was found to provide best results for strength and water permeability. This was explained by the hypothesis that nano-Fe_2O_3 acts as foreign nucleation sites thereby accelerating C-S-H gel formation due to increase in $Ca(OH)_2$ crystals at initial stage of hydration. Moreover, the nanoparticles act as filler and improve the pore structure of specimen by containing the pores harmful as per water permeability is concerned. This is supported by the accelerated peak in conduction calorimetry tests, significant increased weight loss during thermo-gravimetric analysis and more rapid hydrated product related peaks in XRD (Ali Khoshakhlagh, 2011)

Oltulu et al., 2013 studied the compressive strength and capillary water absorption of cement mortars containing fly-ash using Nano-Fe_2O_3 (NF), Nano-Al_2O_3 (NA) and Nano-SiO_2 (NS) powders and their ternary and binary combinations at ratios corresponding to 0.5%, 1.25% and 2.5% of binder. An increase in 7-32% of compressive strength and 14% decrease in capillary absorption were determined to be the best result.

In another study, the compressive strength and flexure strength of plain cement mortar was measured at 7th and 28th day by mixing nano-Fe_2O_3 and nano-SiO_2. The result indicated a gain in strength when compared to control samples. SEM images were obtained to find that nano-Fe_2O_3 and nano-SiO_2 filled up the pores and reduced $Ca(OH)_2$ among hydrates proving that nano-Fe_2O_3 not only acts as fillers but activators to promote hydration. (Li et al., 2014)

Madandoust et al. (2015) worked on durability of SCC containing Nano-silica and Nano iron oxide. The amount of fly ash was 25% of the cement in all mixtures. For Nano silica optimum dosage was found to be 4% and 2% was found to be the optimum dosage for Nano iron oxide. Water absorption was found to reduce.

Bimal et al. (2019) observed that by the addition of small quantities of nano iron oxide, both the strength and durability characteristics of modified HVFAC get significantly affected. The flexural strength of modified concrete with 0.75 percent nano iron oxide had identical strength as that of normal concrete (NC) after 28 days and had even more strength than NC after 56 days. The rate of gain of flexural strength was highest in modified HVFAC with 0.75 % nano iron oxide after 56 days. It was about 20% higher than that of HVFAC. It might have occurred due to higher quantity of fly ash present in HVFAC, as the pozzolanic reactivity associated with fly ash increases with the time. Increase in flexural strength was also observed with modified HVFAC with 0.5% and 1% nano iron oxide but it was lesser than 0.75% nano iron oxide. The compressive strength of NC was observed to be highest followed by modified HVFAC with 0.75 % nano iron oxide. The rate of gain of compressive strength was highest in HVFAC after 28 days. The results of modified HVFAC with 0.75 percent nano iron oxide, was the highest and was even higher than that of NC. The tensile strength of concrete was found to increase with the addition of 0.5 percent and 1 percent nano iron oxide as shown in fig. 1.15.

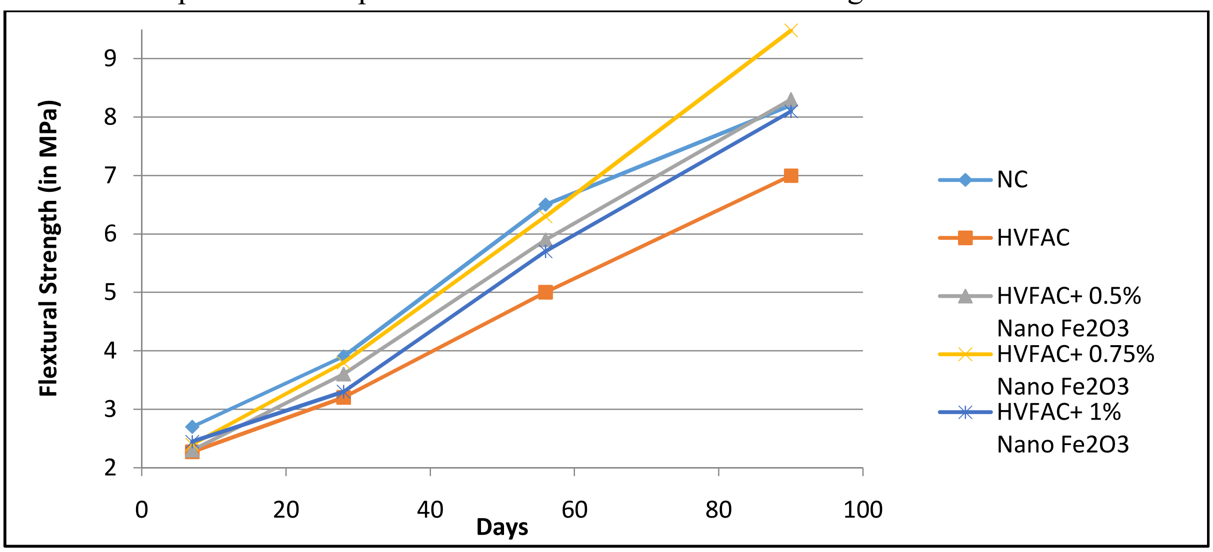

Figure 1.15: Variation of flexural strength of NC, HVFAC and HVFAC modified with nano-Fe_2O_3

Bimal et al. (2019) olso observed about the variation of durability by addition of nano iron oxide. They find HVFAC have more permeability than that of Normal concrete. Whereas the addition of nano iron oxide to the HVFAC causes decrease in permeability. The absorption of water was highest for HVFAC in the sorptivity test whereas that was least for Normal concrete . A dosage of 0.75 percent nano iron oxide decreased the absorption up to the level of normal concrete. The chloride penetration obtained from RCPT results showed that NC performed the best with least chloride penetration, whereas the HVFAC had highest penetration. Amongst all the modified HVFAC with nano iron oxide, 0.75% addition of nano iron oxide gave the best results. The figures show that the improvement starts declining with further addition of nano iron oxide. This may be due to agglomeration of nano iron oxide particles in concrete specimens.

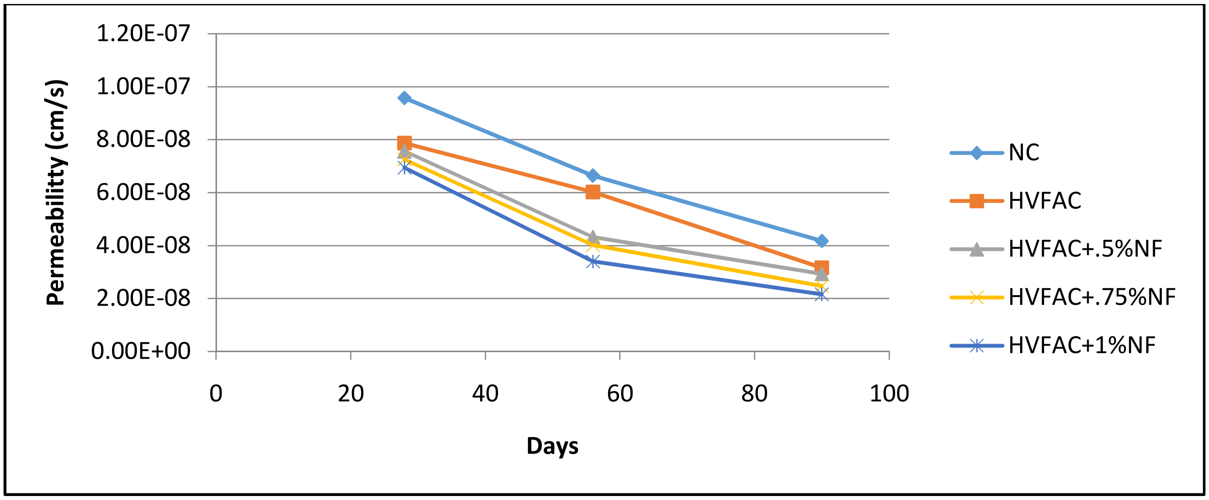

Figure 1.16 : Variation of permeability of NC, HVFAC and HVFAC modified with nano-Fe_2O_3

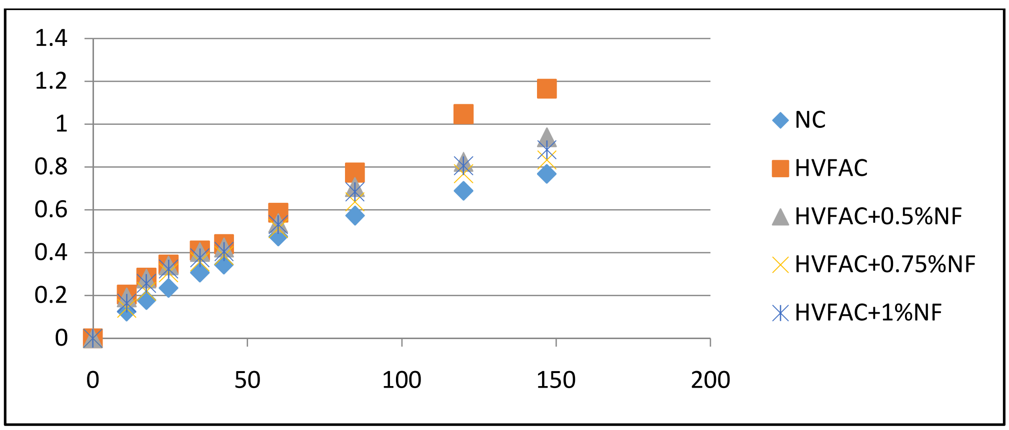

Figure 1.17. Variation of sorptivity of NC, HVFAC and HVFAC modified with nano-Fe_2O_3 (28 days)

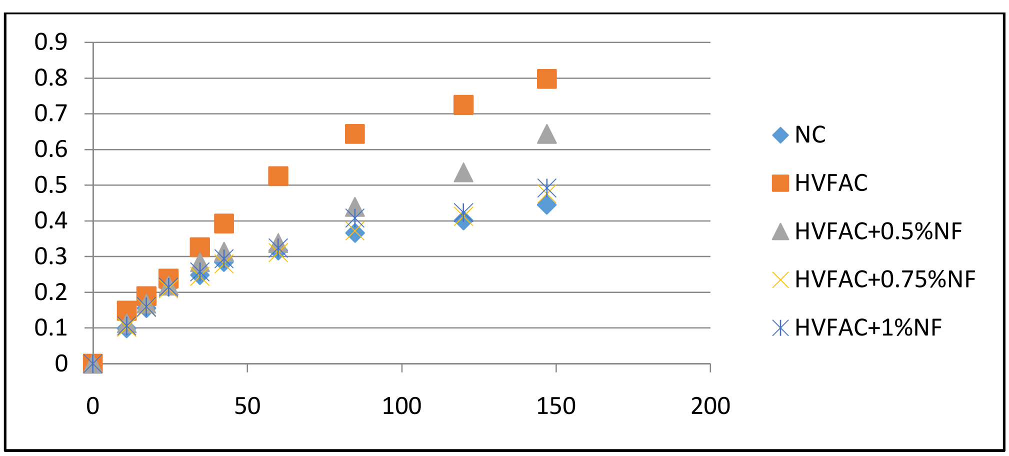

Figure 1.18 : Variation of sorptivity of NC, HVFAC and HVFAC modified with nano-Fe_2O_3 (90 days)

4. Nano alumina (Nano Al_2O_3)

Nano alumina is formed from alumina itself. Silica and alumina are major chemicals involves in cement hydration. The function of silica is to change strength properties where alumina controls the setting time of cement. Due to this property, nano alumina is specially used in UHPC (ultra high power concrete). The function of nano alumina is to speed up the initial setting time for UHPC. This will reduce segregation and flocculation. (Norhasri et al.,2017).

As per Nazari et al., (2011) physical, mechanical and thermal properties of concrete containing nano Al_2O_3 of average particle size 15 nm also depends upon curing medium. Concrete with 2% replacement of Portland cement with nano-Al_2O_3 and cured in saturated lime water had improved compressive strength and setting time. When it is cured in water, the optimal level of cement replacement was found to be 1% by cement. Without nanoparticles, strength of concrete cured in limewater is reduced relative to the specimen cured in water. TGA and XRD results shows mechanical and physical properties of the concrete using nano-Al_2O_3 have been found improved.

Effect of different types of hybrid system of nano-particles in concrete were also found differently. The optimum content of aminosilane for improving 28 days strength was about 0.75%. The increment in 28 day compressive strength of cement mortar with hybrid nano-SiO_2 / amino-silane and nanoclay / amino-silane were about 19% and 20% respectively. Whereas lower efficacy on mechanical performance were observed with application of nanosilane with nano-$CaCO_3$ and nano-Al_2O_3. At early age (7 days) compressive strength of cement mortar were reduced using aminosilane but it is increased at longer time (28 days). Different percentage of (<= 1%) aminosilane also reduced the electric resistivity of the cement mortars at different curing times. Nanoclay had no effect on setting time but nano-Al2O3 decreased the final setting time whereas nano-SiO_2 and nano-$CaCO_3$ exhibited accelerating effect on both initial and final setting time. Nano-$CaCO_3$ and nano-Al_2O_3 had higher impact on viscosity and yield stress than the other two. Nanoparticles were more effective on flexural strength than on compressive strength. (Hosseinia et al., 2014).

Due to fine size of nano-particles, it increases the quantity of C-S-H gel responsible for improved strength and water permeability of the concrete. nano-Al_2O_3 also acts as a nanofillers and therefore recover the pore structure of the concrete specimens. Accelerated peak appearance in calorimetry test, more weight-loss in TGA indicated that nano-Al_2O_3 could improve the properties of specimen. (Nazari et al., 2011)

Concrete having water-binder ratio of 0.4 with 2% and 4% replacement of OPC by nano-Al_2O_3 were found with no changes for early age compressive strength. Although much denser microstructure were formed as observed by SEM. (Barbhuiya et al., 2014).

2% by weight replacement of OPC with 15 nm average size of nano-Al_2O_3 produces concrete with improved strength, but ultimate strength of concrete was obtained at 1% weight of OPC replacement. When nano-Al_2O_3 is increased, workability of fresh concrete were found to decrease suggesting essential use of superplasticizers. (Nazari et al., 2010).

Mechanical properties of cement mortar containing 1 and 3% nano-Al_2O_3 are desirable but 5% replacement reduces the mechanical properties significantly. As per SEM study, nano-Al_2O_3 reduces the $Ca(OH)_2$ crystals and fills the pores and hence, increases the density of concrete. (Arefi et al., 2011).

Voids between cement particles in a concrete can be filled by the addition of ultra-fine particles to prepare dense, compact concrete. Concrete specimen containing 3% colloidal nano-Al_2O_3 having better mechanical properties than normal concrete when doses of nano-Al_2O_3 is taken more than 4% by weight of cement, the mechanical properties of concrete is reduced. (Hase et al., 2015).

Addition of nano-Al_2O_3 to the concrete mixtures can increase both shear and bending strength but 3%, 4.5% interaction of nanoparticles with the steel fibers can have a negative effect on the beam strength. Whereas addition of nano-SiO_2 with low dosages of cement has a negative impact on both shear and flexural strength of RC beam. When beam in produced with higher dosages of cement, addition of nano-SiO_2 or nano-Al_2O_3 leads to an increase in the shear strength of 9%, 7% respectively. (Silva et al., 2016).

Effect of electrokinetic nano-Al_2O_3 treatment on strength of rebar concrete interface were evaluated through pull out tests. It was found that bond strength of rebar concrete interface were increased by increasing current density (Hangtong et al., 2016).

Concrete exposure to elevated temperatures has detrimental effects. Nanoparticles has been introduced thus, to decrease thermal degradation. These particles not only has been seen to protect strength loss but also protect from disruptive cracking. Nano-SiO_2 has been found to be most effective. However, CNT, GO, Nano-clays, nano-Al_2O_3 or nano-Fe_2O_3 can be used to produce heat-resistant concrete. (Powalsikora et al., 2018).

5. Nano titanium dioxide (Nano-TiO_2)

It is also called titania. It is naturally occurring oxide of titatium. It can be obtained from ilmenite, rutile and anatase. Because of various advantages such as low toxicity, high chemical stability, semi-conductivity, low industrial cost and high availability, TiO_2 has been used as cosmetics, paints and drugs (Hamdy and Ion 2011). Nano-TiO_2 can be obtained from TiO_2. This nanomaterial has extraordinary photo-catalyst activity and is therefore widely used as a photo-catalyst (Chen et al 2012). It has also reported to be used in cement concrete (Diamanti et al., 2008; Chen et al., 2012; Jalal et al., 2012).

Murata et al. (1999) pointed out nano-TiO_2 as a zero-dimensional material which got attention from industry as well as research community. It was indicated that NT could modify material structures, thereby developing high performance, durable, multifunctional and environment friendly cementitious composites.

Jalal et al., (2013) investigated the characteristics of high strength self-compacting concrete containing nano-TiO_2 and fly-ash. Here, Ordinary Portland Cement was replaced by 15% fly-ash by weight and 5% nano-TiO_2 was added. Rheological, mechanical and durability properties of concrete at higher age improved. Nano-TiO_2 improved the C-S-H gel formation and hence, improved the durability properties. Increase in FA decreased the flexural strength at 7 and 28th day but improved for 90days. Workability and rheological properties were enhanced due to increased percentages of FA. Water absorption, capillary absorption and chloride penetration are decreased at lower volume of FA replacement. NanoTiO_2 improved the consistency and reduce the probability of bleeding and segregation of concrete. The optimum level of addition of 4% for nano-TiO_2 was observed. nano-TiO_2 modified samples had much higher strength as compared to FA concrete. As nano-TiO_2 acts as nanofillers, hence water permeability, chloride penetration decreased.

Kalaitzakia et al., 2013 studied the effect of nano-titania replaced 4.5 to 6% of binder in the hydration and carbonation of the concrete as well as the mechanical properties and the adhesion capabilities of the designed mortars. As per DTA-TG, FTIR, SEM and XRD analysis, enhanced carbonation, hydration and modulus of elasticity of the mixtures with nano-TiO_2. The mechanical characterization indicated that the mortars with nano-TiO_2 showed increased modulus of elasticity enhanced carbonation compared to the specimens without nano-titania. nano-TiO_2 also improves the adhesive properties of the mortar applied to porous stones.

Zhang et al., 2015 studied the effect of nano-TiO_2 of average size 25 nm in cementitious materials. Compressive strength was found to increase due to accelerated cement hydration and pore refining effect. Also, decrease in contact angle was observed.

Liu et al., 2015 had evaluated the workability, mechanical and microstructure properties of mortar containing different types of nano-particles. Tests results indicated that increased amount of nano-SiO_2 and nano-Al_2O_3 led to the increased water demand whereas nano-TiO_2 and nano-ZnO had negligible effect on the mortar fluidity. The optimum content of nanoparticles was suggested to depend on the average particle size of nanoparticles. Nano TiO_2 and nano ZnO had negligible effect on the mortar fluidity due to their lower specific surface area. The compressive and flexural strength of the specimens slightly decreased for the mortar containing nano-Al_2O_3 and nano-TiO_2 but it was seen to be significant after 28 days age.

Senffa et al., 2013 compared the effects of nano-SiO_2, nano-TiO_2 prepared with 0-2 % NS,0-20% NT, 0.45 -7 % SP and 0.45 - 0-58 water binder ratio. NT influenced more significantly the results of rheological behaviour and hydration temperature but such differences were not found for compressive strength and water absorption. The maximum admissible amounts of nanoparticles are restricted when the dosage of SP and W/B ratio remained invariable. The effect of NT predominated over NS. NOx photocatalytic degradation up-to one hour testing ranged from 65% to 80% and the formulation 0.5. NS+5 NT+ 1.5 SP showed the best performance.

Lee et al., 2010 studied the effect of nano-TiO_2 powder on early age hydration kinetics of tricalcium silicate. 0%, 5%, 10% and 15% of nano-TiO_2 was added and increase in peak reaction for all mixes was observed. Degree of hydration at 12th hour and 24th hour increased. The rate of hydration of 10% and 15% nano-TiO_2 paste accelerated, while 5% delayed.

Baoguoma et al., 2015 studied the influence of this nanomaterial on micro-structures and mechanical properties of cement mortars. The result shows that 3% NT remarkably increased tensile strength by 65.6% and flexural strength by 61.9%. Also, the toughness is improved. Water absorption decreased by 40-65% and water vapour permeability by 43.9%. The total pore volume and the harmless pores volume reduced significantly by about 48.2% and 34.6% respectively. It was observed that when the dosage of NT was increased by more than 3%, the morphology of products synthesized due to hydration changed from short columnar types to polyhedral shape of complete or incomplete form and hence, the flexural and tensile strength decreased significantly. However, 3% NT could significantly improve characteristics such as the durability, toughness and structure compactness. It effectively avoids the high brittleness of cement-based materials. Main cause of improved toughness and compactness are nucleation effects, improvement in orientation of crystal of CH and control of grain size effect and micro aggregate filling effect of NT during the hydration process of cement.

Khataee et al., 2013 observed that increasing the amount of TiO_2 self-cleaning properties of cement increased. The addition of TiO_2 up-to maximum replacement of 1% improved the compressive, flexural strength and decreased its setting time.

As per Pacheco-Torgala et al., 2011, nanoparticles are used to improve the strength and durability of cementitious composites, the photocatalytic capacity and also nano-toxicity risks. TiO_2 modified building materials has been used as white pigments, self-cleaning and self-disinfecting and to reduce urban and indoor pollution levels have been confirmed by both laboratory research and field work.

As per Lee at al., 2009, nano-TiO_2 accelerates the rate of hydration and also increases the degree of hydration whenever used in the concrete.

As per Zhang et al., (2011), the flexural strengths as well as the compressive strengths of concretes containing nano-SiO_2 are lower than that of concrete containing the same amount of nano-TiO_2. This effectiveness of nano-TiO_2 observed for enhancement of compressive and flexural strengths of concretes varies in the order NTC5<NTC3<NTC1, i.e, with the decrease on nano-TiO_2 content.

The resistance to chloride penetration of concrete and the pore structures which contain nano-TiO_2 were observed to be superior to that of concrete having same quantity of nano-SiO_2.

6. Carbon Nanotubes (CNTs)

Morsy et al., 2011 used blended cement consisting of OPC, CNT and nano metakaolin (NMK) for their research. CNT was added by ratios 0.005%, 0.02%, 0.05% and 0.1% by weight of cement. Upto 0.02% addition, NMK cement mortar shows improved compressive strength by 11%.

Change in mechanical properties of mortars and corrosion of embedded steel bar were studied with the addition of CNT. No significant change in bending strength or compressive strength at 28 days were observed. However, a slight increase in porosity was observed. Samples were subjected to chloride attacks and carbonisation which revealed CNT modified samples to have higher steel corrosion intensities. (Camacho et al., 2014)

MWCNT was also used in Portland cement and microstructure and compressive strength was evaluated. MWCNT increased the mechanical properties such as compressive strength and flexural strength. SEM was conducted and it was found that CNT was well dispersed in cement hydration products. Gel pores were increased with CNT usage which was measured by pressure mercury testing thus, signifying positive influence on mechanical properties (Xu, 2015).

CNT was used in fly ash cement system (0.5% and 1% by weight). CNT resulted in higher strength of the mortar. Highest strength was obtained for 1% CNT, showing that 20% fly-ash replaced cement could result in 100% strength as compared to Portland cement (Aron Chaiparich, 2009).

MWCNT was added by 0.5% in cement matrix composites. It was found that defectiveness and chemical properties of MWCNT had a substantial influence on mechanical properties of cement composites. Lower amount of tobermorite gel was found for functionalized CNTs (f-CNTs) revealing a reduction in performance as compared to pristine cement (Musso, 2009).

Kousta-Gdoutos et al., 2010 found that lower amount of MWCNT (0.025-0.048%) provides effective reinforcement in comparison of short MWCNT (0.08% by weight). The strength also depended on effective dispersion and a ratio by weight of surfactant and MWCNT has identified to be close to 4.

0.1% and 0.5% by weight of CNT and 0.1% CNF was used to study flexural strength, Young's modulus and compressive strength of CNT & CNF reinforced mortar. The increase in flexural strength and Young's modulus was substantial whereas the compressive strength increased marginally. Corrosion potential, corrosion current density and electrochemical mass loss were also evaluated. It was confirmed that addition of 0.1% by weight of CNT and CNF resulted in corrosion resistive environment, whereas 0.5% does not produce such state. It was explained by the hypothesis that galvanic couples are formed for higher dosage of CNT (0.5%) (Kousta-Gdoutos et al., 2017)

In another study, usage of 0.08 % MWCNT was found to improve compressive strength and flexural strength. With further increase in MWCNT, mechanical properties showed a decreasing trend. X-ray tomography confirmed that with increase in CNT to 0.08%, 3D defects in volume fraction decreased and filled pore structure was observed. At higher %, the CNT agglomerated and lead to a poor structure (Liu, 2014).

Mechanical properties were further investigated with 0.1% and 0.2% addition of CNTs and CNFs. It was observed that the peak displacement increased by 150% showing application in high ductility cases could be devised using this modification (Tyson et al., 2011).

Fly ash mixes were modified with CNT and highest strength was achieved for CNT content of 1% by weight. CNT enhanced the flexural strength more than CNFs. Young's modulus was found to increase whereas porosity and total pores decreased by use of CNTs leading to denser structure (Siddique & Mehta, 2014).

Graphite nano-plates (GNP-C & GNP-M) and CNFs were used to evaluate rheological properties, hydration kinetics, autogenous shrinkage and pore structure of UHPC. GNP-C and GNP-M resulted in lower viscosity as compared to CNFs showing lubrication effects. However, no significant change in yield stress was observed. CNFs resulted increase in induction period whereas for GNPs it was porosity decreased significantly. Autogenous shrinkage also increased with respect to UHPC (Mang et al., 2018).

7. Nano fly ash

As per Paul et al. (2018), influence of nano fly ash (NFA) having average particles size of 44 nm on 7 days compressive strength of mortar showed a reduction in strength with replacement of 10.9% of cement with NFA. However, a 14.8% increase in 7days strength was observed when 13.1% of cement was replaced with nano-lime (1.2%) and NFA (11.9%) blend. At higher replacements, the compressive strength was lower, and a reduced strength was observed. The compressive strength increased with higher NFA content, and the increase was highest for the low grade concrete. The observed increase of strength for 10%, 20% and 30% replacement with NFA was 46.3%, 56.0% and 59.9% respectively, compared with plain cement concrete.

8. Nano Clay

As per Korayem et al. (2017) review paper, nano-clay used as an additive for mortar and cement paste had shown promising results from the addition of small percentage, usually up to 10% of cement weight. However, too much Nano clay can be fatal to both compressive and flexural strength, causing a considerable decrease. Nano metakaolin (NMK) and nano montmorillonite (NMMT) are the most widely used types of NC. NMK acts as a complementary element to the mixture, added mainly to strengthen the bonds between fibers and cementitious matrices.

As per Paul et al. (2018) review paper, the performance of Nano-clay mortar was compared with the reference mortar (without Nano-clay) for the same sand-cement ratio (S/C), having W/C of 0.4. In all mixes, significantly higher compressive and tensile strength were observed in Nano-clay mortar.

9. Nano Zinc Oxide

As per Vishwakarma et al. (2018) review paper, ZnO nanoparticles are having similar properties like TiO_2 nanoparticles. It has antibacterial, antifungal and anti- corrosive properties. Mohammad and Saeed (2012) observed that zinc oxide nanoparticles reduced the permeability of concrete structures and increased the mechanical and flexural strengths. The partial replacement cement by 4% ZnO nanoparticles could accelerate C-S-H gel formation at beginning of hydration. It has been also found that ZnO nanoparticles act as fillers and reduce the pores of the specimens by decreasing harmful pores.

As per Nazari and Riahi (2011), when the amount of ZnO nanoparticles is increased up to 3% by weight, the flexural strength of the specimens is increased.

10. Graphene Oxide (GO)

As per Paul et al (2018), graphene oxide has hydrophilic nature due to oxygen present in this compound make peaks with hydrogen and carbon as hydroxyl (OH), carbonyls (C=O) respectively. Hydrophilic nature contributing more in dispersion when used in the making cementitious mortar or concrete. Due to its high specific surface area being in the range of 160 m^2/gram, contact area with cementitious materials is highly improved hence reactivity increased as per other nanomaterials.

As per Long et al., (2017), the flexural strength of cement mortar were increased more than the value of increment in compressive strength after addition of 0.05%,0.1% and 0.2% respectively by weight of cementitious materials. The flexural strength was increased about 16%, 27% and 41% whereas compressive strength was increased about 7%, 10% and 10% respectively at 28 days age.

But Wang et al., (2017), found that increment in the strength was slightly more in the specimen replaced with GO by 0.02% relative to the specimen prepared without addition of GO.

11. Zeolites

Zeolites is a crystalline solid mineral. It mainly consists of silicon, aluminium and oxygen, hydrogen atoms. It is found in nature as volcanic roc and ash but commercial form is used in large scale. Due to porous nature, its commercial form is used as an absorbent, catalysts etc. It can be found in sodium or hydrogen ion (H^-) form. (S. C. Paul et al., 2018)

As per Perraki et al., (2010), even micro-size zeolite affected the strength of cement concrete.

As per Ahmadi and Shekarchi (2010) studied, the compressive strength of the concrete prepared by replacing the cement with zeolite, clinoptilolite by 5%,10%, and 20% by weight of total cementitious materials were found higher than the specimen containing no zeolites. The highest compressive strength was observed at 28 days in the specimen containing 20% higher dose of the zeolite.

But Caputo et al. (2008) reported the reverse of the above results. They found that the strength of the concrete containing zeolite decreased with the increased doses of zeolite. The compressive strength of the concrete specimen containing 5-25% of zeolite were decreased by 12-45% at 28 days.

Najimi et al., (2012) research, supported the findings of Caputo et al.,(2008). As per them, the value of compressive strength of the specimen in which cement was replaced by 15% natural zeolite were reduced by 10% and 7% respectively at 7days and 365days. The reduction was more at the higher doses of the zeolites. The reduction in compressive strength was found 38%,13.5% respectively at 7days and 365days on the 30% replacement of cement with natural zeolites.

12. Polycarboxylates (PCE)

This is basically a linear polymer compound. It is obtained from methoxy-polyethylene glycol co-polymer. In the past, it was also used as a nanomaterial in the concrete. Now a days, this compound is highly used in high strength concrete and high performance concrete to improve the workability. It acts as high range water reducer (HRWR) agent.

As per Huseien et al., (2019) studied, workability of a concrete can be effectively controlled at lower water-cement ratio by the addition of this group compounds.

The doses of PCE in a cement concrete affects the characteristics of the concrete. The higher amount may harm in the setting of the concrete. The higher doses may reduces the hydration process in the cement paste. To increase the workability of a self compacting concrete, the optimum amount of polycarboxylate compound is added. Due to addition of the optimum amount of polycarboxylate in the ultra-high performance concrete, voids can be remarkably reduced.

CONCLUSION:

This critical overview on effect of nanomaterials on concrete composites with fly ash have the following conclusions:

1. Inclusion of nanomaterials into the concrete mix can reduce its flowability, improve workability as well as reducing initial and final setting times.
2. Nanomaterials improving the rate of hydration of concrete mixes.
3. Negative effect due to addition of high doges of fly ash in concrete can be compensated by addition of optimum amount of nano-particles in the concrete composites.
4. Nanosilica and nanoironoxide are widely used in concrete and cement mortar. It also used in high volume fly ash pavement quality concrete to improve energy intringic problem. Optimum doses of nanosilica were found between 2 % to 4% whereas optimum doses for nanoironoxide was found less than 1% depending upon variation of amount of fly ash and types of cement.
5. The durability property such as water permeability, sorptivity and chloride penetration of concrete composites modified with nanomaterials can be inhanced and improved remarkably.
6. Access amount of nanomaterials content in the concrete composite more than the optimum percentage can negatively affect the mechanical as well as durability properties of concrete due to the agglomeration effect.

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