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Studies on optimization of silica nanoparticles dosage in cementitious system

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ABSTRACT

Optimization of silica nanoparticles (SNPs) dosage in cementitious system was carried out analytically as well as experimentally by understanding the early stage hydration reaction of tricalcium silicate (C_3S). XRD and TGA results show the maximum nucleation effect of SNPs at 8 h, when the rate of product formation was higher than the control (~66% additional C–S–H and ~61% more CH with 10% SNPs addition). While at 24 h of hydration, ~25% additional C–S–H was formed and CH content reduced by ~32% with 10% addition showing the pozzolanic effect of SNPs. Further, FTIR results reveal that SNPs accelerate the polymerization in silicate chain and with 10% SNPs addition more crystalline (probably tobermorite like) structure is formed. This is responsible for the formation of highly compact and dense microstructure at 24 h as observed in electron micrographs, which may be responsible for the slow hydration rate at later age. XRD, FTIR and TGA studies on C_3S revealed that up to 5% addition of SNPs is beneficial, whereas higher dosages do not contribute significantly. Based on these investigations, studies were performed on cement paste, mortar and concrete samples, which revealed that 2–3% addition of SNPs is the optimum quantity for significant contribution in strength properties.

1. Introduction

Increasing use of Nanotechnology in cementitious materials is gaining widespread attention as remarkable improvement in mechanical properties has been reported [1–5]. Among the various nanoparticles viz. silicon oxide, titanium oxide, zirconium oxide, aluminum oxide, carbon nanotube, etc. explored, silica nanoparticles (SNPs) have gained extensive attention as they provide improved mineralogical and morphological characteristics [6–9]. During the last one decade, numerous research papers have reported varying amount of SNPs dosage ranging from 0.5 to 10% in different cementitious systems such as paste, mortar and concrete. The physical state of these nanoparticles varied from colloidal to nano-powder and the size of nanoparticles also ranges from 5 to 10 nm to few microns. Also, it has been reported that the mechanical properties such as compressive strength decreases beyond the optimised dosage apart from the problems of agglomeration of nanoparticles, improper mixing, etc. [10–12]. Only a little is available to explain the role of SNPs at early stage of hydration. It is reported that SNPs accelerate the hydration reaction rapidly than the silica fume as SNPs serve as a site for the precipitation of calcium-silicate-hydrate (C–S–H) due to their high surface area, thus C–S–H is first formed on the surface of silica particles rather than the most reactive phases; C_3S and C_3A [13–16]. Singh et al. [17] have reported that beside the physical effect of SNPs, there may be two possible reaction mechanisms, nucleation and well known pozzolanic reaction (Fig. 1). They have reported that in presence of SNPs, additional C–S–H is formed during early stage of hydration, which is responsible for the shift in the super-saturation stage of calcium and acceleration period [18–20].

Jo et al. [21] have suggested that 6% of SNPs addition with a w/c ratio of 0.23 brings maximum positive results in cement mortar, while Singh et al. [22] mentioned that with 5% addition of SNPs, the calcium hydroxide (CH) content is reduced by 86% at 1 day and up to 62% at 28 days of hydration than the control. Riahi and Nazari [23] have reported that the incorporation of SNPs improves the abrasive resistance as well as compressive strength of concrete. Ltifi et al. [24] described that compressive strength increases with the increasing dosage of SNPs, whereas Li [25] has testified that SNPs increases both short term and long term strengths in high volume fly ash based system. He has reported that 8% replacement of SNPs improves compressive strength by 81% at 3 days and 7% at 2 years.

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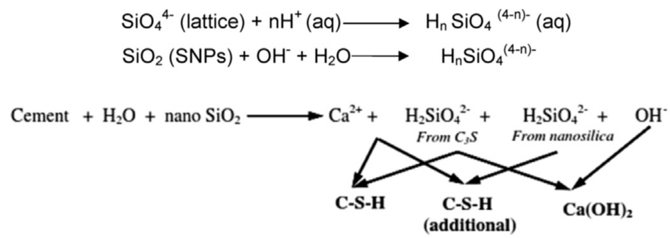

Fig. 1. Possible hydration reaction in the presence of SNPs.

Khanzadi et al. [26] have stated that compressive strength and tensile strength increases with SNPs especially at early stage and better permeability is also observed, whereas, Madani et al. [27] have reported that SNPs shows accelerated early hydration rate at 1 day and acceleration in hydration rate increases as the surface area of SNPs is increased. However, lower degree of hydration was observed at 7 days of hydration in SNPs incorporated samples. Table 1 representing the optimised dosage of SNPs reported by the various other researchers in different cementitious systems, clearly shows that there is a state of imperfection regarding the optimised dosage of SNPs [28–35]. The studies reported so far towards the optimisation of dosage have been carried out only on the basis of mixing of different amount of SNPs and evaluating the engineering properties (compressive strength, flexural strength, permeability, porosity, etc.) only. In the present paper, we have attempted to quantify the dosages of SNPs by instrumentation techniques as well as by systematically evaluating the engineering properties in cementitious system i.e. paste, mortar and concrete.

2. Materials and methods

2.1. Materials

The ordinary Portland cement (OPC) with Blaine fineness 390 m^2/kg, confirming to IS 8112:1989 was used for the present studies and its chemical composition is given in Table 2. The tricalcium silicate (C_3S) was prepared in the laboratory using an electric furnace at an elevated temperature of 1500 °C as reported elsewhere in detail [18]. Powdered SNPs were prepared in the laboratory through sol-gel method using water glass (sodium silicate solution) as a precursor. The powdered SNPs are amorphous and have a specific surface area (116 m^2/g) as reported earlier [18,36].

Two types of sand, the first being standard Ennore sand having the specific gravity 2.60 and fineness modulus of 2.77 was used for mortar studies, whereas, second one being river sand having specific gravity of 2.57 and fineness modulus of 2.98 was used for concrete samples. The angular crushed aggregates having maximum size of 10–20 mm were used as coarse aggregates, having specific gravity of 2.62 and fineness modulus being 7.34. The aggregates were found to be satisfying the specifications of IS 383:1970 (Table 3).

Table 2
Chemical properties of OPC.

Compounds	Content (%)
SiO_2	22.3
Al_2O_3	6.1
Fe_2O_3	2.2
CaO	61.1
MgO	3.1
SO_3	2.1
LOI	3.1

Table 3
Physical characterization of fine aggregates.

Property	Standard sand	River sand	Code followed
Water absorption (%)	0.42	0.57	IS 2386 Part 3
Specific gravity	2.60	2.57	IS 2386 Part 3
Fineness modulus	2.77	2.98	IS 2386 Part 1
Grading zone	Zone-I	Zone-II	IS 383 1970

2.2. Sample preparation for hydration studies

To understand the early hydration process in presence SNPs, experiments were carried out on C_3S phase. Different dosages of SNPs (1, 3 5 and 10%) were added by the weight of C_3S and total water binder ratio (w/b) was kept 0.4 for all the samples. The hydration process was stopped at different time intervals starting from 1 to 24 h, which completely covers all the major stages of early hydration process.

2.3. Sample preparation for cement, mortar and concrete mix

For the cement paste studies, the content of SNPs was varied from 1 to 5% by the weight of cement and the samples were prepared into a standard cube molds (25 × 25 × 25 mm) using w/b ratio 0.4. For mortar studies, a mix proportion (1:3) of cement and sand was used and the content of SNPs (1, 2 & 3%) was added by the weight of cement. Mortar cubes were prepared in 50 × 50 × 50 mm standard molds using 0.4 w/b ratio. For the concrete study, 2% SNPs by the weight of cement was used and mix proportioning was based on M-40 grade concrete (Table 4). Fuller and Thompson gradation curve was used for the aggregate grading. The cubes were de-moulded after curing of 24 h followed by immersion in water till testing time at room temperature.

2.4. Determination of CH and C–S–H content in hydrated C_3S paste

The quantification of C–S–H, in C_3S with varying amount of SNPs at different time intervals of hydration was carried out using

Table 1
Optimized dosages of SNPs in cementitious system.

Reference	Cementitious matrix	Type of SNPs used	Size of SNPs (nm)	SNPs dosages (%)
Berra et al. [28]	cement paste	colloidal	10	3.80
Qian et al. [29]	(cement+40%fly ash) mortar	colloidal	10	2.25
Qing et al. [30]	cement paste	powder	–	5.00
Sobolev et al. [31]	concrete	powder	5–50	2.00
Singh et al. [32]	cement paste	powder	40–50	5.00
Mukherjee et al. [33]	cement mortar	colloidal	8–20	3.00
Heikal et al. [34]	cement-ground granulate blast furnace slag paste & mortar	powder	15	2.00
Shaikh et al. [35]	mortar and concrete	powder	25	2.00