A Study on the Synergistic Effect of Silica Fume and Fly Ash Inclusion in High Performance Concrete

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ABSTRACT

The use of Supplementary Cementitious Materials (SCMs) in the construction industry to reduce the carbon footprint associated with cement manufacturing is increasing worldwide. This study examined the synergistic behavior of two SCMs, Fly Ash (FA) and Silica Fume (SF), in the production of High-Performance Concrete (HPC). Using various amounts of SF and FA to replace cement, several ternary mixes were created and their strength, microstructure, durability and fresh state properties were evaluated. A Rapid Chloride Permeability test (RCPT) was performed to determine the resistance of the mixture to chloride ion penetration. Microstructural analysis was carried out utilizing Scanning Electron Microscopy (SEM) images to study the morphological characteristics of the mixes. The results revealed that the combined use of SF and FA in HPC significantly increased its durability and compressive strength owing to the pozzolanic reaction and filler effect. Microstructural investigation exhibited improved particle packing, refined pore structure, and the creation of more Calcium Silicate Hydrate (CSH) phases. Thus, SF and FA, when used in conjugation, optimize HPC performance and promise to be a sustainable and viable solution for reducing cement requirements.

Keywords-pozzolanic reaction; supplementary cementitious materials; silica fume; fly ash; microstructure

I. INTRODUCTION

By mass, cement is the largest manufactured product on earth [1]. However, large amounts of CO_2 are released into the atmosphere during the manufacturing of Ordinary Portland Cement (OPC) 43 and OPC 53 grades [2]. The manufacturing of cement accounts for approximately 6–8% of the global anthropogenic CO_2 emissions [3]. The most significant and pressing issue the cement industry faces, is the decrease in the CO_2 emissions related to cement production. Numerous studies have shown that various pozzolanic additives, such as Fly Ash (FA), Silica Fume (SF), and Blast Furnace Slag (BFS), can increase the density and durability of concrete [4-7]. Consequently, the use of pozzolanic materials as Supplementary Cementitious Materials (SCMs) has emerged as a practical solution to several durability issues and as an environmentally friendly option to produce low-carbon sustainable cement. Natural pozzolans, activated minerals, and industrial waste products are the most commonly used types of SCMs in the cement industry. Most SCMs do not exhibit notable cementitious hydraulic responses, either by themselves or when in contact with water. However, they undergo a chemical reaction, known as the pozzolanic reaction, when exposed to calcium hydroxide or alkaline aqueous solutions, forming hydration products similar to those observed in OPC systems [8-10]. The rheology of the OPC 43 cement slurry was also improved by adding SCMs [11].

SCMs can affect the mixing and hydration of composite cement through two pathways. The first is by physical means and the filler effect, which is applied to inert fine powders or fillers [12, 13]. Secondly, chemical interactions result in the formation of hydration products [14-16]. The reaction proceeds via a dissolution-precipitation mechanism. There are two primary categories of the chemical behavior of SCM acting as solid reactants. When the SCM reaction consumes $Ca(OH)_2$, it is called the pozzolanic reaction. Reactions that do not require $Ca(OH)_2$ or in which $Ca(OH)_2$ primarily functions to activate pH are referred to as hydraulic or latent hydraulic reactions, respectively [6]. Because SF consists of very fine particles and contains a large amount of silica, it is mostly amorphous and exhibits pozzolanic behavior [17].

Depending on the type of coal from which the FA is produced, FAs can be either calcareous or siliceous. According to ASTM, FAs used in concrete are divided into two classes: C and F [18]. Class F is siliceous, has a higher silica content, and is a by-product of the burning of bituminous coal. FA resulting from lignite combustion and sub-bituminous coal is categorized as class C. Compared with class F, FA has a higher calcium content [19-21]. FA is a glassy material that must be activated with cement or lime. FA, when used in combination with cement, enhances flowability [22]. The sources of coal and combustion methods can have a substantial impact on the chemical and physical characteristics of FA. The performance and consistency of concrete can be affected by variations in FA characteristics.

This study investigates how the mechanical and durability qualities of concrete are affected by the concurrent application of SF and class F FA.

II. MATERIALS USED

Ternary mixtures were prepared deploying the three cementitious materials OPC53, FA, and SF. The FA was provided by NTPC Kahalgaon Thermal Power Plant. The SF was provided by Elkem South Asia Pvt. Ltd. Tables I and II provide an overview of the physical and chemical characteristics of the cementitious materials used in this study, respectively. Sand was collected from the Sone River bed. According to IS 383:2016, sand can be classified as of grade zone 3 [23]. CHRYSO India Private Limited supplied the chemical admixture CHRYSO Premia S6123PM utilized in this study. The amount used during mixing was meticulously regulated because the active solid content (i.e., solids that are employed to disperse cement particles) was more than 35%. CHRYSO Premia S6123PM is a superplasticizer based on polycarboxylic materials that can be applied in situations requiring high initial and final compressive strengths. For a given concrete mix, a considerable reduction in the mixing water of up to 32% can be achieved while keeping the cement consumption and workability unchanged. After the initial dispersion caused by electrostatic forces, the carboxylic ether polymer with extended lateral chains can offer the required steric hindrance to prevent further coalescence of the cement particles.

TABLE I. PHYSICAL PROPERTIES OF CEMENTITIOUS MATERIALS

Physical Properties	OPC53	FA	SF
Specific Gravity	3.15	2.2	2.1
Consistency (%)	30	35	--
Soundness (%)	0.8	0.06	--
Specific surface (m^2/kg)	322	340	20,400

TABLE II. CHEMICAL PROPERTIES OF CEMENTITIOUS MATERIALS

Chemical Properties	OPC 53	FA	SF
CaO (%)	64.48	2.42	--
Al_2O_3 (%)	6.1	28.7	--
Fe_2O (%)	2.78	8.92	--
SiO_2 (%)	20.54	59.5	88
MgO (%)	1.73	1.98	--
SO_3 (%)	3.03	2.07	--
Loss of Ignition (%)	0.65	2.53	--
Chloride content (%)	--	0.33	--
Moisture content (%)	--	--	0.58

III. EXPERIMENTAL METHOLOGY

Trial mixes of OPC53 with SF and FA were prepared to achieve a characteristic compressive strength corresponding to the M65 grade. The techniques outlined in ACI 211.4R-08 [24] were applied in the proposed methodology and adjusted according to the Indian standard code IS 10262(2019) [25]. Utilizing significant quantities of a single type of SCM (binary mixes) can result in unfavorable side effects, such as longer setting times and delayed strength [26, 27]. Therefore, different trial mixes of ternary blends were prepared to determine the optimal amounts of FA and SF in an attempt to obtain the High-Performance Concrete (HPC) formation. The Indian Standard for mix proportioning, IS 10262(2019), recommends an FA dose between 15% and 30% for high-strength mixes [25]. Therefore, two mixtures in which cement was replaced with 20% and 15% FA were used in the experiments. Two groups of mixtures were prepared. In mix group 1, the FA content was 15%, whereas in mix group 2, the FA content was 20%. In both mixes, the SF varied from 2.5% to 10% in 2.5% increments. The designation of the mix is given by the notation SF(TM)FA, where TM denotes the ternary mix, while SF and FA represent the contents of SF and FA in the mix, respectively.

According to the Indian Standard Code for mix proportioning, the concrete must exhibit a minimum compressive strength M65 in order to be classified in the High Strength Concrete (HSC) category [25]. HPC is different from HSC in that the desirable compressive strength constitutes only one of the required parameters. However, HPC must also possess a desirable fresh state, durability, and dense microstructure as additional attributes. Therefore, in addition to the compressive strength test, a slump test, Rapid Chloride Permeability Test (RCPT), and morphological SEM analysis were performed to determine the synergistic effect of SF and FA on the concrete properties.

IV. RESULTS AND DISCUSSION

The results of the tests conducted in this study, including the compressive strength test, the RCPT, and the slump test, are reported in this section. The morphology of the HPC mixes was assessed by SEM images.

A. Workability Test Results

The workability of the ternary mix was evaluated using a slump test, and the results are summarized in Figure 1. The cohesiveness of the mixture was evident because of the finer nature of the particles of the SCMs used. Segregation and bleeding were eliminated. With the application of a chemical admixture, a ternary mix of FA and SF generated workable concrete. However, as the SF content increased, the slump decreased. There are two opposing forces at play. SF is responsible for the reduction in flowability owing to its extremely fine size, which increases its water-absorbing tendency. On the other hand, the addition of FA slightly increased the workability owing to the spherical size of the FA, imparting a ball-bearing effect.

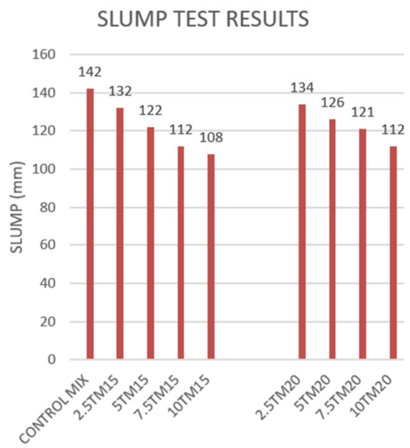

Fig. 1. Slump values obtained for different mixes.

B. *Compressive Strength Results*

The compressive strength test results for M65 grade concrete are graphically depicted in Figures 2 and 3. The compressive strength increased with increasing SF content up to 7% and remained stable at 10% SF. Typically, blending with FA alone results in a decrease in compressive strength [28-30].

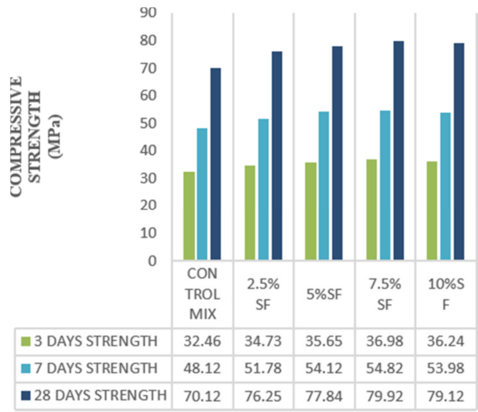

Fig. 2. The compressive strength values of mix group 1 samples, in which 15% cement was repleced with FA and 2.5%,5%,7.5%, and 10% with SF.

However, in the present study, the ternary blending composition contained SF in addition to FA, which compensated for the decrease in the compressive strength expected from FA. Consequently, compared to the control mix, the compressive strength increased as the percentage of SF in the mixture increased, as evidenced in Figures 2 and 3. This can be traced to the reactive silica provided by SF, which reacts with portlandite to produce a secondary C-S-H gel. Moreover, SF, owing to its extreme fineness, provides additional sites for the nucleation of hydration products, promoting the nucleation effect [31, 35]. From the comparison of Figures 2 and 3, it can be seen that as the FA content increased from 15% to 20%, the compressive strength values slightly decreased.

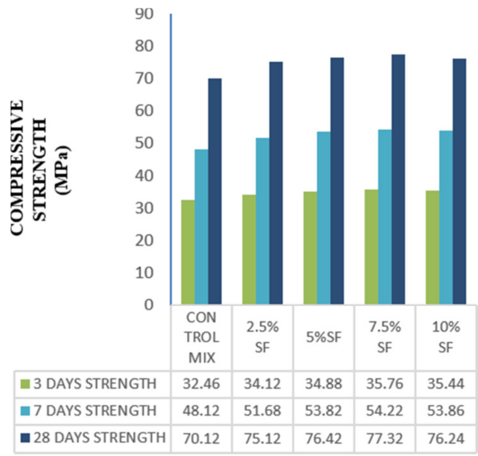

Fig. 3. The compressive strength values of mix group 1 samples, in which 20% cement was replaced with FA and 2.5%,5%,7.5%, and 10% with SF.

C. *RCPT Test Results*

To assess the durability of the mix, RCPTs were performed on the 28-days old HPC specimens following ASTM C 1202 [32]. The RCPT results are graphically presented in Figure 4. The control mix had an RCPT test value of 2246 coulombs per 6 h. From Figure 4, it is evident that the synergistic effect of SF and FA significantly reduced the permeability of all ternary blends, which is indicative of microstructural compactness. In addition, as the SF content increases, the penetration resistance to chloride ions decreases. This is because of the effective pozzolanic reaction and the micro sizes of the added pozzolana, which reduced the voids in the microstructure to a maximum extent.

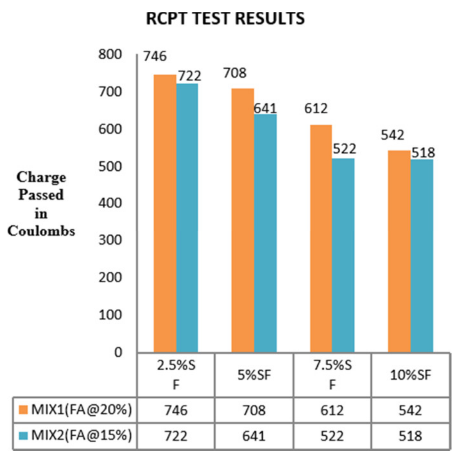

Fig. 4. The RCPT results for the mix groups 1 (20% FA) and 2 (15% FA) and SF content of 2.5%,5%,7.5%, and 10%.

D. Microstructure Study

A Carl Zeiss Field Emission SEM 500 system was used for the microstructural images. The type 2 Secondary Electrons (SE2) were recorded to obtain an SEM image of the sample. High spatial resolution is possible with SE imaging because secondary electrons are produced near the surface of the sample. The topographical variations of the sample surface are the main cause of the contrast in SE-based images. The hydration reaction of cement produces lime as an undesirable product that tends to leach. The essence of the pozzolanic reaction of SCMs lies in how well they are able to utilize lime by performing the reaction and converting it into a product of cementitious value, which is called additional CSH or secondary CSH. A few notable features observed in the SEM images of the ternary blends are discussed below.

The hexagonal shape of the lime crystals is evident in Figure 5, along with CSH aggregation, which appears fibrous and poorly crystalline. Similar results were observed in [33]. There is a place in the microstructure, marked in Figure 5, where the pozzolan and portlandite crystals meet, in other words, the nucleation of the lime crystals around the SCM occurs there, resulting in the reduction in the size of the lime crystals and leading to the "grain size refinement" as it is known. Grain size refinement basically means a reduction in the size of large lime crystals as well as a reduction in the orientation of the lime crystals, making them smaller and less oriented. The greater the grain size refinement is, the stronger is the conclusion that the pozzolanic reaction utilized to form secondary CSH.

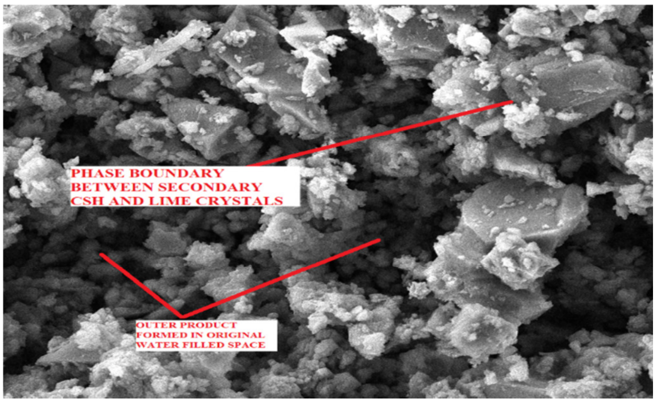

Fig. 5. The SEM image of the ternary mixture containing FA and SF illustrates the nucleation of lime crystals around the pozzolanic additions.

In addition, as illustrated in Figure 5, outer hydration products, marked as the Outer product (Op), were observed. There are two types of CSH hydration products, the inner product (Ip) and the outer product, depending on the hydration mechanism involved. One such mechanism is called the through-solution mechanism in which anhydrous compounds first dissolve in their constitutive ions, then the hydration products are formed in the solution, and finally these hydration products precipitate from the solution due to their low solubility [34]. Through this mechanism, an outer hydration product was formed in the pore space, as displayed in Figure 5. The other mechanism is solid-state hydration in which the reaction occurs on the surface of the hydration compounds. Both mechanisms can form hydration products.

Figures 6 and 7 provide the microstructural images of the two samples containing 20% FA and 2.5% or 7.5% SF. The magnification was kept the same for both images for the differences arising as the SF content increased to be analyzed. In these two figures, the green rectangles show capillary voids of greater than 50 nm, which are macropores, and the red rectangles mark voids of less than 50 nm, which are micropores [34]. As the SF content increased, there was a shift in porosity towards a finer distribution. Thus, the addition of SF results in a denser microstructure. This improvement can be attributed to the strengthening effect of the pozzolanic reaction and the microfiller effect.

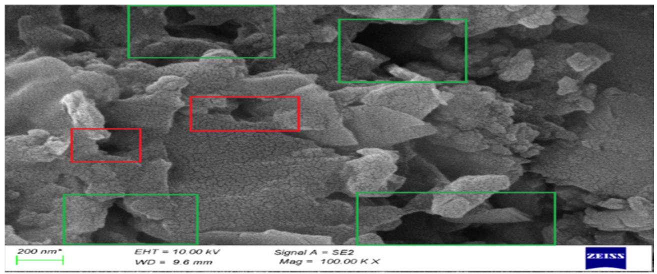

Fig. 6. SEM image of the sample with 20% FA and 2.5% SF.

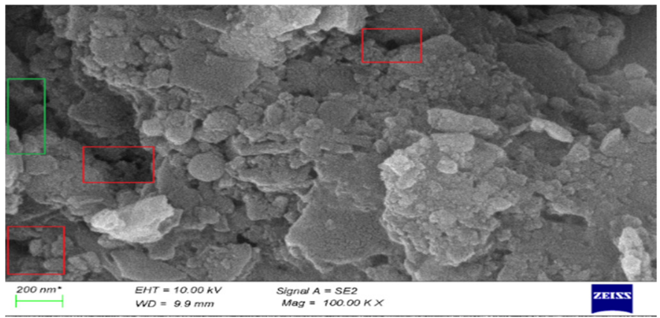

Fig. 7. SEM image of the sample with 20% FA and 7.5% SF.

V. CONCLUSIONS

The results of this study demonstrated that ternary blended mixtures of cement with Fly Ash (FA) and Silica Fume (SF) enhanced the workability, strength, and durability of concrete. Ternary mixing with 15% FA and 7.5% SF yielded good results and improved characteristics compared to the control. This mixture exhibited the highest compressive strength and RCPT values of less than 1000 coulombs per 6 h, indicating efficient microstructural compactness. FA and SF work in tandem and enhance each other's qualities, while addressing each other’s discrepancies to maximize strength and durability. The lower reactivity of FA was compensated for by the addition of SF. SF offers an early strength advantage, whereas FA possesses latent hydraulicity, which offers a compressive strength advantage on later days. Thus, their combination ensures sufficient supply of pozzolanic material for both the early and late days of hydration. Furthermore, the micropores of the FA particles and the spaces between the cement particles were filled with SF, making the concrete matrix denser and less permeable. FA improves workability because its spherical particles can slide past each other according to the ball bearing effect, making the mix workable, therefore compensating for the high water demand when adding SF because of the extremely finer size of SF. Microstructural examination using SEM verified that employing FA and SF together produced a denser, more uniform matrix. These improvements were attributed to the refined pore structure and reduced permeability achieved through the synergistic action of SF and FA.

SF is more expensive than FA; consequently, it is more cost-effective to use SF in conjunction with FA than utilizing it alone. Since both FA and SF are industrial byproducts, they contribute to sustainability and decrease the carbon footprint related to concrete production.

The glassy phase content, composition, and fineness of SCMs can significantly impact the reaction outcome. The use of silica-rich SCMs affects the type and quantity of hydrates formed, which, in turn, influences the porosity and durability of concrete. Although the application of FA and SF in a synergistic manner might improve the quality of concrete, it can also present some issues that must be handled properly. The inclusion of SF can significantly increase the viscosity of the concrete mixture, making it more difficult to be handled, be placed, and finish. Compared to regular Portland cement concrete, the addition of FA may reduce the strength at an early age. Although SF can increase compressive strength, it is important to handle this trade-off, particularly for applications that require early loading.

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