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Optimisation of nano-silica modified self-compacting high-Volume fly ash mortar

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Abstract. Evaluation of the effects of nano-silica amount and superplasticizer (SP) dosage on the compressive strength, porosity and slump flow on high-volume fly ash self-consolidating mortar was investigated. Multiobjective optimisation technique using Design-Expert software was applied to obtain solution based on desirability function that simultaneously optimises the variables and the responses. A desirability function of 0.811 gives the optimised solution. The experimental and predicted results showed minimal errors in all the measured responses.

1. Introduction

The need to develop a durable and cost-effective high-performance concrete has led to the utilisation of industrial by-products termed cement replacement materials' or 'supplementary cementitious materials' such as ground granulated blast furnace slag (GGBS), silica fume (SF), fly ash (FA) or limestone powder. Numerous researches have been reported utilizing by-products or waste for cement replacement in high performance concretes [1], and [2].

Nano-silica has been widely accepted by many researchers due to its contribution to improving performance in a cementitious matrix. It has been reported that nano-silica densify matrix in cementitious composites; it reacts faster than other supplementary cementitious materials such as FA, GGBS, etc. it utilizes calcium hydroxide released during cement hydration to produce more calcium silicate hydrates (C-S-H) gel for further increase in strength of concrete [3] and [4]. This is due to nano size effect, which contributes to pore filling and subsequent packing density of cementitious matrix [5], and [6]; high surface area and nucleation effect, which aid faster reaction. Silica units from cement particles during cement hydration [7] and C-S-H gel produced during the first stage are attracted to nano-silica particles [5] and form nano-silica particles as a nucleus. The products of hydration on both the surface of cement grain and newly formed nucleation sites are well dispersed in the pore solution, creating a more dense matrix [6]. The resultant effect of accelerated hydration of cement by nano-silica by its nucleation effect leads to the production of more calcium hydroxide. The two processes, nucleation effect and hydration effect, through consumption of calcium hydroxide, leads to boosting a pozzolanic value of nano-silica [3]. In effect, the presences of nano-silica shorten the dormant period of cement hydration as nano-silica consumption of calcium hydroxide stimulates the further release of hydration products in order for the hydration reaction to attain a balanced reaction [8]. Nanosilica has demonstrated faster pozzolanic reactivity compared to other active powders, particularly at initial phases [9].

Despite the numerous benefits associated with the use of nano-silica in cementitious composites some drawbacks have been highlighted: The range of optimum quantity of nano-silica recommended for improvement of mechanical properties in the literature have not been consistent. Two sources have been identified in relation to this effect. Firstly, the source of the nano-silica is of different origins, which shows differences in types, particle size, specific surface or methods of production. Secondly, the difficulty in uniform dispersion of nano-silica in the fresh cement paste, grout, mortar or concrete [10], is an issue that cannot be exposed completely on a mechanistic basis. Even in their original form, slurry or hydrosol, if well dispersed, they have the potential of aggregating once in pore solution of a cement-based material due to the availability of ions such as Ca^{2+}, Na^{+} and K^{+} released during cement reaction with water [3]. These ions have a bridging effect, agglomerating silica units in the form: (SiO^{-})-(Ca^{2+})-(SiO^{-}) [8], which leads to disrupt the dispersion of nano-silica particles in the pore fluid and restraint their uniform dispersion. Similarly, the utilization of nano-silica restraint compatibility between the cement, superplasticizer and any other chemical admixture or mineral additions [10] and [11]. This results in a negative effect on the rheology/flow of cement paste and mortar. Consequently, increasing nano-silica content lead to significantly increasing yield stress values [11]. Identifying the threshold for both nano-silica and superplasticizer balances the threat pause by their incorporation in cement paste and mortar. The varied optimum content of nano-silica reported by numerous researchers ranges between 5% to 2% weight of cement [10] and [12]. The wide disparity in the optimum content reported indicates difficulty claiming maximum nano-silica content required to improve properties of cement composites.

2. Research Significance

The study evaluates the influence of nano-silica and superplasticizer quantities on compressive strength and mercury intrusion porosimetry (MIP) of self-compacting high-volume fly ash cement mortar. Response surface and multiobjective optimization are used to develop a solution of most effective nano-silica and SP amount based on the set objectives. This is essential to harmonise differences in the literature regarding the optimum nano-silica and superplasticizer amounts in nano-silica modified mortars.

3. Constituent Materials

Washed river sand of about 600 μm average diameter, ordinary Portland cement type 1, tap water, Class F fly ash with low Calcium less than 15% (ASTM C618), commercially available nano-silica particles ordered from Dongshen Petrochemical, China, with silica purity 99.8% and average diameter 20nm and polycarboxylate-based superplasticizer sika viscocrete R 5-700® were used in all the mortar mixes.

3.1. Mixture proportion

Some of the ingredients used in the mortar preparation are fixed to minimize over-fitting in the responses. The constituents that have been fixed are presented as shown in Table 1.

Table 1. Fixed constituents of mortar mixes

W/C	Water (Kg/m^3)	Cement (Kg/m^3)	Sand (Kg/M^3)	Fly ash (Kg/m^3)
0.32	187	583	467	700

3.2. Sample preparation

A mortar mixer of 4.5 L capacity was used to prepare all mortars at an average of 8 min. Water and SP were mixed together and half the quantity is poured into the mixer containing measured dry quantities of sand, cement, and FA for about 2 min, the remaining half quantity of mixed water and SP was further added to the mixture after another 2 min until the mixture attained homogeneity, which took

about 4 min and the mixer stopped. The mortars are tested for workability on a mini mortar flow. The mortars are cast in 50 mm cube moulds for compressive strength testing and MIP assessment of porosity. The compressive strength of mortar cubes was tested at 28th day after curing at lab temperature of 22 ± 2°C in accordance to ASTM C39/C39M-04 A. The samples for porosity assessment was cut from the mid-center of cube samples and tested in a MIP equipment in accordance with the requirements of ASTM C1202.

4. Method and Experimental Design

Design-Expert software was used for the response surface and optimization. The user-defined option was utilised for the experimental design to accommodate the range of values of trial mixes. The values of the responses were inputted and the model for each response was established after verification with the statistical requirement. Afterwards, a multiobjective optimisation was conducted utilising the models to obtain solutions of responses and variables that give a fair treatment to all the variables based on the rating in terms of weights/ranking inputs. Usually, model overfitting occurs when it is made excessively complex, such as having too many parameters relative to the number of observations [13]. Three parameters are selected in the present investigation for optimisation of the response variables of the mortars and other parameters are fixed. Trial mixes were also conducted to obtain a realistic range of variables and ensure further avoidance of overfitting. The range of variables of nano-silica (0%, 1%, 2%, 3%, 4%), and SP (0.65%, 0.7%, 0.8%, 1.0%, 1.2%).

5. Results and Discussion

5.1. *Development of Compressive Strength.* The experimental design and results obtained by a user-defined option in RSM are shown in Table 2. The model expression for compressive strength, f_c, of mortars at the age of 28 days is shown in equation (1). The relationship between the variables A, nano-silica and B, superplasticizer has R-squared = 0.788:

$$f_c = +8.97 - 0.22A + 0.11B + 0.35AB - 1.28A^2 \quad (1)$$

Response surface plot in fig.1 shows a quadratic trend depicted by equation (1). It illustrates the effects of nano-silica and SP on the compressive strength of mortars. From the plot, nano-silica maximise the compressive strength of mortars at about 2%. Nanosilica is more significant than SP, though, the interaction of the two showed significant effect as well.

Table 2. User-defined experimental design and responses

Run	Factor1 A: NS (%)	Factor2 B: SP (%)	Response1 Comp. strength (MPa)	Response2 Porosity (%)	Response3 Slump flow (mm)
1	3	0.65	50.5	8.1	175
2	4	1.2	60.4	10.95	215
3	4	0.8	52	12.5	195
4	1	1	72.15	6.8	230
5	4	0.65	50.55	13.5	170
6	2	0.8	89.5	5.45	210
7	1	0.8	74.5	5.9	220
8	2	1.2	81.2	7.1	228
9	0	0.7	64.09	7	219
10	2	1	87.15	6.1	220
11	1	0.65	71.5	6.1	208
12	2	0.65	80.15	7.4	192
13	4	1	56	11.55	205

14	1	1.2	70.4	7.05	240
15	2	0.7	87.6	5.85	205
16	0	0.8	62.5	7.85	223
17	0	1	60.75	8.2	227
18	3	0.8	65	7.2	195
19	0	0.65	65.77	6.88	215
20	3	1.2	68.08	8.85	227
21	3	1	70.3	6.97	210
22	1	0.7	73.65	5.85	217
23	3	0.7	60	7.5	186
24	4	0.7	51.75	12.2	184
25	0	1.2	56.25	9.15	240

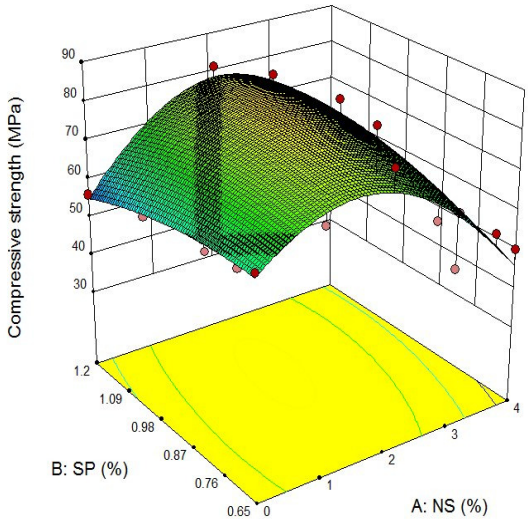

Figure 1. Response surface plot compressive strength vs nano-silica amount and superplasticizer dose.

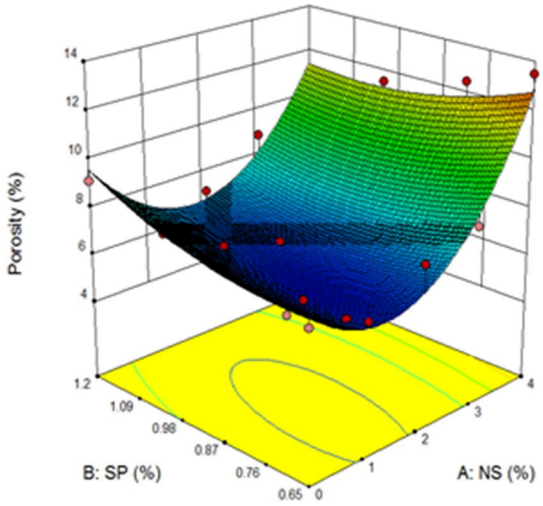

Figure 2. Response surface of porosity vs NS and SP.

5.2. *Porosity.* The experimental results of porosity based on experimental design are shown in Table 2 while the *response* surface plot based on equation (2) is shown in fig.2. The effect of nano-silica on porosity is more significant (R-squared = 0.95) while SP insignificant. However, the interaction effect of nano-silica amount and SP dose showed a significant effect. The nano-silica amount minimises porosity at about 2%, the same value as required to maximise compressive strength.

$$Porosity = +5.85 + 1.82A + 0.24B - 0.92AB + 3.74A^2 + 0.69B^2 \quad (2)$$

5.3. *Slump flow*. Equation (3) has been used to plot response surface of slump flow as shown in fig.3. The SP dosage is statistically significant with regard the slump flow (Sf) (R-squared =0.98). Contrary to this, the slump flow decreases with increasing nano-silica amount. The SP dosage is in percentage while the slump flow is in mm. looking at equation (3) and figure 3, it can be observed that the effect of SP was not truly linear, and the interaction between the two variables, the nano-silica and SP also affected the slump flow. Consequently, variation in slump flow due to SP dose was dependent on the nano-silica amount.

$$Sf = 216.83 - 23.26A + 10.24B + 5.44AB - 2.28A^2 - 4.16B^2 - 2.23A^2B - 5.12AB^2 + 11.87A^3 + 9.41B^3 \quad (3)$$

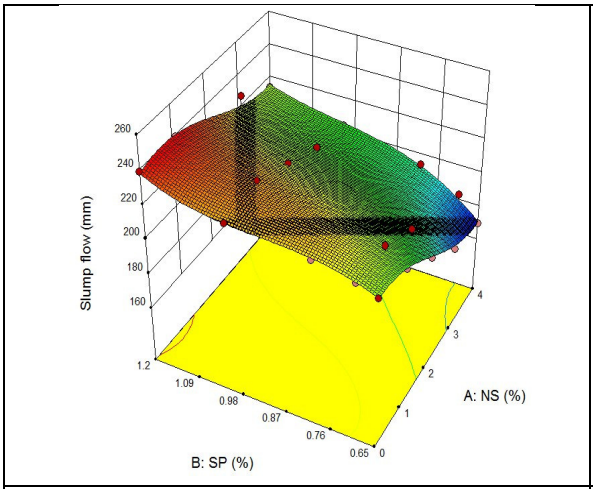

Figure 3. Response surface of slump flow vs NS and SP

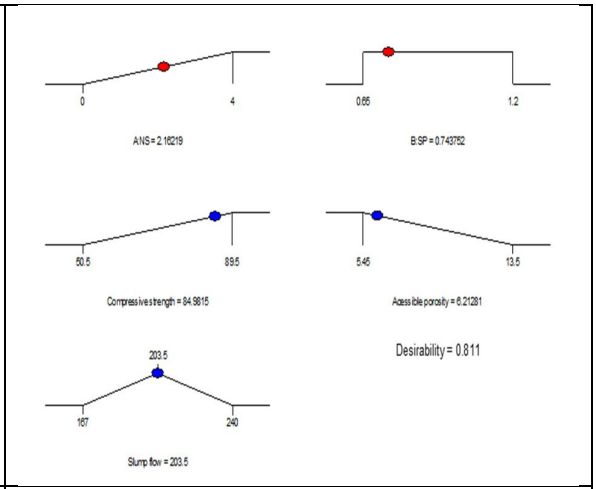

Figure 4. Rams of multiobjective optimisation of variables and responses

6. Multiobjective Optimization

The appropriate required amount of nano-silica and superplasticizer that is needed to maximise compressive strength may not necessarily be the same with the amount required to minimise porosity and maintain slump flow on target. Therefore, there is a need to determine the amount of nano-silica and dosage of superplasticizer that will satisfy all the responses. The multiobjective optimisation process is necessary to determine the most suitable solutions using desirability coefficient. Multiobjective optimisation utilises the model of all the responses and simultaneously develop solutions variables and responses values that will meet the requirements of all the responses. The rams in fig.4 illustrate the multiobjective solution based on desirability function. Using the solution variables and the fixed constituents in the mortar mix shown in Table 1, the new mixture was prepared with a slight adjustment of superplasticizer to attain the required range of flow. The experimental and

predicted results are as shown in Table 3 based on desirability value of 0.811. The error calculated is minimal which indicates the reliability of the optimisation technique.

Table 3. Experimental and prediction results for optimised solution

A-NS (%)	B-SP (%)	y1 (Mpa) Exp.	y1 (Mpa) pred.	Error (%)	y2 (%) Exp.	y2 (%) pred.	Error (%)	y3 (mm) Exp.	y3 (mm) pred.	Error (%)
2.2	0.75	81.5	84.98	4.27	6.05	6.21	2.64	209	203.5	2.63

7. Conclusion

To optimise the compressive strength and MIP at a set slump flow of 203.5 mm of nano-silica modified self-compacting high-volume fly ash mortar, response surface and multiobjective optimisation were applied. The optimised mortar is achieved at 2.2% nano-silica and 0.75% superplasticizer by weight of binders. Increasing nano-silica beyond this amount at the fixed SP dose is accompanied by decreasing slump flow and subsequent decrease in compressive strength and increase in porosity. Contrary to this, the compressive strength of the mortars increased with increasing SP within this range at the fixed nano-silica amount. The increasing slump flow aided proper packing density of mortar mixtures. Application of multiobjective optimisation is suitable for obtaining a solution of variables and objective functions that meet the requirements of all the variables involved in the mixture.

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