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The use of mathematical modelling in the composition of a composite material

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Abstract

The paper treats the use of modelling of the mixture proportioning of a new composite material (CM) on the basis of combined utilization of secondary industrial by-products [V. Petkova, *Composition of Heavy Strength Concrete*, Patent Ser. B No. 60100, 03.10.19094, Bulgaria]. A three-parameter polynomial model is used for the determination of the amount of the composite components cement, active additive and granulated blast-furnace slag. It is shown that using this approach, the component's influence on the changes of compressive strength in the course of up to one-year hardening of the composite can be evaluated.

Keywords: Mixture proportioning; Compressive strength; Granulated blast-furnace slag; Composite; Modelling

1. Introduction

The existence of sustainable and ecologically clean environment depends on the rational utilization of the secondary mineral by-products from different industrial branches, as well as on a number of other factors. The practice of cement production uses a wide range of industrial subproducts: anorhit from copper–nickel and raw taconite tailings [1], fly ash [2–4], granulated blast-furnace slag [2,4–7], etc. The utilization of the industrial waste products in concrete came to the fore during the energy crisis in the 1970s and was also the result from the rise of environmental consciousness. Many of the properties of concrete can be improved by including in its composition various types of industrial waste [8]. It is shown [9] that rubberized concrete (concrete with addition of rubber chips) demonstrates high resistance towards impact loading and is especially prospective for structures of crash barriers, bridges and roads. Silica fume finds still wider application in concrete [2,10–14]. Granulated slag is one of the most widely applied by-products in concrete [2,15]. Until recently, however, its application in high-strength concrete was restricted by its low activity. This shortcoming is partially overcome by the development and use of differing in composition and quality-active admixtures (AA) [6–8,16–18]. As a result of the program for the development and application of high-performance concrete launched in Japan in 1982, it was suggested to include a mineral additive promoting ettringite formation to reduce or refine concrete pore structure in addition to the utilization of the conventional mineral additives, including silica fume, as well as ground granulated blast-furnace slag, to produce very high strength concrete [19]. However, except for the ettringite-based mineral materials, it is recommended to use special mixers [20]. The problem of increasing the activity of the slag-containing binder substances is solved in the developed new high-strength composite material (CM) [18]. The silicate cement matrix is filled with medium-grained artificial sand and an AA. The obtained CMs possess increased density, high mechanical characteristics, good water impermeability, corrosion resistance, operation durability and low exothermic properties [21,22]. It is known that there are two

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methods for the faster determination of cement and concrete strength: by express strength testing and by applying suitable mathematical models [23–25]. Models for describing the hydration processes [26,27], the microstructure [28] and the electrochemical extraction of chlorides from concrete [29], as well as the cement setting when using $CaCl_2$ as an accelerator [30], can be found in reference literature. However, publications on the methods of mathematical statistics for modelling the formulation composition and strength properties of composite concrete with additives of mineral by-products are rather scarce in reference literature.

The present paper shows the results from the investigated quantitative influence of important formulation parameters (including new additive) on the mechanical properties of a cement-based CM in the course of one-year hardening. The method of the planned experiment has been applied, and regression equations have been obtained for these effects on the strength of the composite for different ages of hardening.

2. Materials and methods of investigation

Composite mortars were prepared with cement/aggregate ratio 1:3 and water/cement ratio of 0.50. Bulgarian cement brand PC35 (Zl. Panega) was used, which meets the requirements of the Bulgarian State Standard BDS En 196-1. The artificial medium-grained sand is a good balanced combination of granulated slag (CaO=13.0, SiO_2=26.4, Al_2O_3=5.0, Mg=1.5%, FeO=36.0, CuO=0.53, Na_2O=1.0 и K_2O=1.0%) and sterile material-inexpensive secondary mineral wastes from nonferrous metallurgy. This artificial medium-grained sand is acidic in character. It replaces entirely the natural sand in the mortars. The active additive is a waste product from the lime-producing industry, with high CaO contents 66% [18].

The most important feature of the formulation is the application of a two-stage technology in the production of the new CM [18]. The first stage includes the preparation of a water suspension with active admixture. The second stage includes 30 min of mechanical mixing of the granulated acidic slag with the prepared suspension. It allows for the preliminary mechanical–chemical activation of the granulated acidic slag. The result of this activation is a rapid surface decomposition of the melilite glass, which accelerates the hydrolysis and hydration of the slag component. In this way, the slag actively participates in the structure formation of the slag–cement stone at an early age, even under ordinary curing conditions. The two technological stages contribute to the successful overcoming of the low hydraulic activity of the slag. The advantages of the technology consist in the possibility to use the artificial sand and the alkaline additive in their industrial granulometry without additional grinding or dispersing. In this way, there is conformity with the requirements of the standard method for determining the activity of active additives [31].

The samples were prisms with sizes 4×4×16 cm. After stripping at the age of 24 h, the specimens were cured in water at a temperature 20±3 °C until gaining the corresponding age of testing.

The variable parameters are the following:

X_1—Portland cement, percent of the total dry mass.

X_2—active additive, percent of the total dry mass.

X_3—granulated slag, percent of the total mass of the artificial sand (granulometric mixture of slag and sterile material).

The boundaries of variation of these three factors in natural coordinates were determined on the basis of preliminary experimental examinations:

X_1—varies from 20% to 25%. The choice is made with the aim of achieving high compressive strength.

X_2—varies from 5% to 15%, considering the operation mechanism of the new additive and the realization of the preliminary mechanical–chemical activation of the acidic slag.

X_3—varies from 55% to 75%, to obtain medium-grained artificial sand.

The variation levels in natural and coded coordinates are shown in (Table 1). The selected values allow the accurate study of the relationship between the three formulation parameters and the compressive strength development for a long period of hardening.

The plan of the experiment contains 27 compositions (test points) for five ages of investigation (Table 2). Three prisms or six half-prisms have been tested at each point. The compressive strength is calculated as the mean value of the obtained test results.

The unknown target function $R_c=(X_1, X_2, X_3)$ is determined as a second order polynomial for the selected ages (1, 7, 28, 180 and 365 days):

$$R_c = b_0 + \Sigma b_i X_i + \Sigma b_{ii} X_i^2 + \Sigma b_{ij} X_i X_j \quad (1)$$

where i=(1..3); j=(1..3), b_0, b_i, b_{ii} and b_{ij} are the regression coefficients. They are calculated using multiple linear

Table 1
Levels of the variation of factors in natural and in coded (confidential) coordinates

Factors	Variation levels in coded coordinates			Variation levels in natural coordinates			Interval of variation
	$x_{i,\min}$	$x_{i,o}$	$x_{i,\max}$	$X_{i,\min}$	$X_{i,o}$	$X_{i,\max}$	ΔX_i
Cement (X_1), %	−1	0	+1	20	22.5	25	2.5
AA (X_2), %	−1	0	+1	5	10	15	5
Slag (X_3), % Art. S.	−1	0	+1	55	65	75	10

Table 2
Design of the experiment for the determination of compressive strength (R_c) as a function of studied factors

Point number	Plan of experiment			Compressive strength R_c (MPa) for different ages (days)				
	x_1	x_2	x_3	1	7	28	180	365
1	−1	−1	−1	4.0	11.1	16.5	18.9	20.9
2	−1	−1	0	4.4	12.9	17.9	20.3	22.3
3	−1	−1	+	3.3	9.9	1.9	17.0	19.4
4	−1	0	−1	5.7	15.6	23.2	26.8	29.8
5	−1	0	0	6.2	18.9	25.5	29.0	31.1
6	−1	0	+	4.8	14.0	21.4	24.2	27.4
7	−1	+	−1	5.1	14.9	20.7	23.9	26.0
8	−1	+	0	5.4	16.6	23.5	24.7	26.9
9	−1	+	+	4.1	12.5	18.9	19.7	24.2
10	0	−1	−1	6.1	17.0	25.1	28.9	31.6
11	0	−1	0	6.4	19.6	27.5	30.8	33.9
12	0	−1	+	5.1	15.1	22.8	26.1	29.3
13	0	0	−1	8.7	24.0	35.5	41.1	44.8
14	0	0	0	9.3	28.8	39.1	44.0	47.5
15	0	0	+	7.3	21.6	32.7	37.2	41.7
16	0	+	−1	7.8	22.6	31.6	36.7	40.1
17	0	+	0	8.2	25.4	36.0	38.4	41.2
18	0	+	+	6.3	18.7	29.1	33.2	37.3
19	+1	−1	−1	5.5	15.3	22.5	26.0	28.3
20	+1	−1	0	5.8	17.7	25.1	27.8	30.8
21	+1	−1	+	4.5	13.5	20.5	23.2	25.4
22	+1	0	−1	7.8	21.6	32.1	37.0	40.5
23	+1	0	0	8.5	26.1	35.3	39.7	42.9
24	+1	0	+	6.5	19.3	29.5	33.5	37.8
25	+1	+	−1	7.0	20.2	28.1	33.0	36.0
26	+1	+	0	7.4	22.9	32.5	34.9	37.0
27	+1	+1	+1	5.7	17.0	26.1	29.8	33.7

regression Linest in MS Excel. The latter gives the possibility of obtaining correct coefficients, regardless of whether the variables are standardized (with uniform distribution around the zero point).

3. Results and discussion

The polynomial models of the compressive strength of the CM for the abovementioned ages are given in (Table 3).

The analysis shows that the value of the coefficients b_{ij} is negligible and could be neglected. This does not affect significantly the standard error and the determination coefficient. The coefficient values found determine the direction and degree of influence of the studied factors on the compressive strength. The negative values of the coefficients b_{ii} show that all the three factors exert positive effect on the strength value and they have maximums. When the b_{ij} coefficients are zero, the extremum coordinates (for the optimal formulation of the composite) are easily calculated from the formula $X_{i,\mathrm{extr}}=-b_i/(2b_{ii})$. The coefficient values for the normative age of 28 days show that the maximum strength is obtained for the composition with the following values of the investigated parameters: X_1=23.2%, X_2=12.1% and X_3=64.7%. The coordinates of the optimal composition for the rest four ages do not differ substantially from the above mentioned values. The maximum compressive strength of this composition varies from 9.3 MPa for the first day to 47.3 MPa for the age of 365 days.

The changes in the compressive strength depending on two factors simultaneously are illustrated in (Figs. 1, 2 and 3) by the isolines for the 28-day strength. The two-factor graphs are plotted by varying two of the factors within the accepted range and fixing the third one at the medium level.

The influence of the quantitative participation of the active additive and the granulated slag on the strength development is shown in (Fig. 1). The amount of Portland

Table 3
Models for the determination of R_c at different ages

Equation for the compressive strength R_c (MPa) for X_1 (%), X_2 (%), and X_3 (%)	Determined coefficient R^2	Standard error (MPa)
$R_c^1=-173.9+11.83X_1+1.354X_2+1.237X_3-0.255X_1^2-0.0611X_2^2-0.00994X_3^2$	.981	0.25
$R_c^7=-569.2+35.29X_1+3.848X_2+5.222X_3-0.761X_1^2-0.171X_2^2-0.0411X_3^2$	.972	0.91
$R_c^{28}=-753.5+51.38X_1+5.454X_2+5.208X_3-1.108\ X_1^2\ 0.243X_2^2-0.0409X_3^2$	.982	0.97
$R_c^{180}=-816.2\ +59.15X_1+6.468X_2+4.392X_3-1.275X_1^2-0.293X_2^2-0.0350X_3^2$	.984	1.05
$R_c^{365}=-846.1+63.64X_1+7.014X_2+3.706X_3-1.372X_1^2-0.317X_2^2-0.0294X_3^2$	.985	1.08

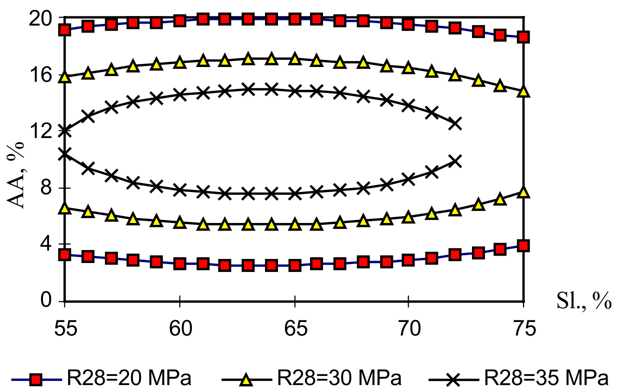

Fig. 1. Isolines of the changes in R_{28} depending on the amount of the formulation components AA and Sl for PC=22.5%.

cement is constant and equals to 22.5% of the total mass of the dry mixture. A broad range with high compressive strength values has been established, in which the changes in the granulated slag content exert negligible effect. For example, the strength of 35 MPa can be attained for two different mass combinations of the admixture—AA=8% or 15% for almost one and the same slag amount of 64%–65%. It is also seen in Fig. 1 that a sufficiently broad range of the AA variation exists, in which attaining the necessary design strength is guaranteed. At the same time, an optimal amount of the active additive exists, which ensures the high 28-day strength for fixed constant cement and slag amounts. Increasing or decreasing the active additive quantity with respect to this optimal amount leads to strength drop. The same relationship can be observed for changes in the components PC-AA and PC-Sl within the shown ranges of variation.

Figs. 2 and 3 show that the contribution of the alkaline additive in obtaining a compressive strength of 35 MPa is observed for the amount AA=10–11% and Sl=64–65%, while the participation of the Portland cement varies from 22.5% to 25%.

The data in Fig. 4 exhibit the same character of changes of the compressive strength due to the variation of the active additive and the cement quantity, but an increase of the strength value R_c to 40 MPa is observed.

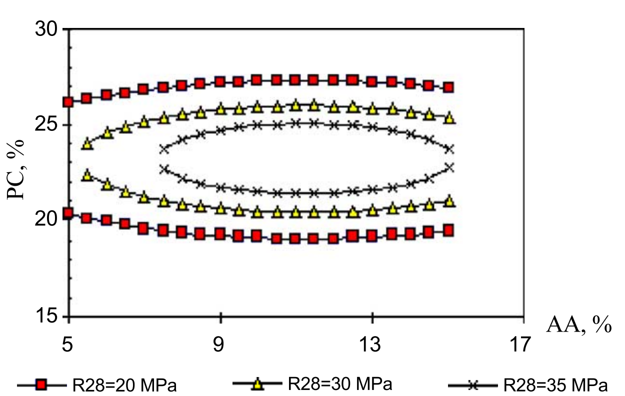

Fig. 2. Isolines of the changes in R_{28} depending on the amount of the formulation components PC and AA for Sl=65%.

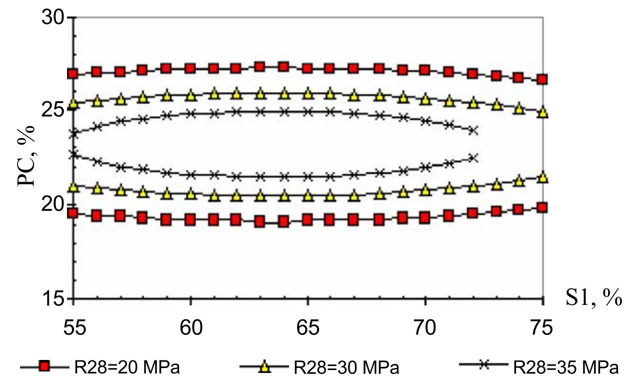

Fig. 3. Isolines of the changes in R_{28} depending on the amount of the formulation components PC and Sl for AA=10%.

The investigation carried out proves that depending on the preliminary set design of the compressive strength, a broad range of combinations exists to optimize the quantitative participation of the main formulation components of the silicate composite for all ages.

At the same time, the experimental and model data show that for each ratio of binder substance to granulated slag, there is a corresponding optimal amount of the active additive AA. Increasing or decreasing the AA amount above or below this optimal quantity leads to strength drop. This dependence obviously results from the interaction of $Ca(OH)_2$ obtained at the first stage of forming the suspension containing a high percent of glassy phase, as already mentioned above. It should be taken into account that during the mechanical processing of the slag and the suspension, surface decomposition of the melilite glass takes place, which contributes to increasing the participation of slag in the processes of composite hydration and hardening. Our investigations [32] and the studies of Mindess and Joung [33] show that blast-furnace slag with a high content of glassy C-S-H phase and tobermorite gel contributes to overcome the lower activity of slag in the beginning of the hardening process of alkali-activated slag cement. The optimal amount of active additive found is sufficient for obtaining products formed by a dissolution and reaction, as well as for the later stage, when this process continues according to a solid state mechanism. With these optimal quantities of AA, hydrate products of the

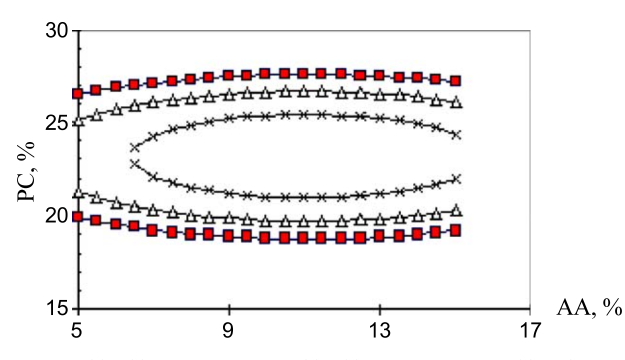

Fig. 4. Isolines of the changes in R_{365} depending on the amount of the formulation components PC and AA for Sl=16.5%.

$C_2SH(A)$ and C_3AH_6 type are formed [32], which are typical for slag Portland cement. If the amount of the active additive (AA) is less than the established maximal one, then the reactions mentioned above do not proceed in the entire volume, and hence, lower strength of the composite is recorded. The understanding of the character of the reactions and hydration products for quantities of the active additive (AA) higher than the optimal ones will obviously need further investigations.

4. Conclusions

Three-parameter models have been used for the description of the influence of the main components on the compressive strength of the composite hardening for up to 365 days. The influence of the studied parameters on the compressive strength is analyzed on the basis of these models. It is shown that an optimal quantity of the new active additive exists, depending on the composition of the other two components. For specific values of the Portland cement and granulated slag amounts, any increase or decrease of the active additive (AA) amount with respect to its optimal values is accompanied by the compressive strength drop for the two investigated ages—28 and 365 days.

The obtained results confirm the positive effect of the preliminary mechanical–chemical activation of the acidic granulated slag and the used new active additive on the strength properties of the silicate composite. It is shown that this positive effect is a result from the initiation of the alkali–slag reaction.

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