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Search for optimal compromise compositions of fibrous polymer-cement dry mixes with computational materials science methods

When developing multi-component materials of specified or optimal properties methods of computational materials science can be validly used. This scientific line forming in recent decade encompasses (judging by international journal «Computational Materials Science» founded in 1992) all fundamental and applied aspects related to knowledge of materials. In studies of building materials the need for these methods arises when a solution can not be found directly in physical experiment or off the computer technology, without too much time and financial resources. The elements of computational building materials science are regularly analysed at annual International seminar "Modelling and Optimisation of Composites" (MOC, Odessa, Ukraine, http://www.moc-odessa.boom.ru). The methods are constantly upgraded and cover new areas in building materials.

The means of computational materials science based on experimental-statistical (ES) models [1-3] have substantially helped in solving the problems of concrete with complex additives and of foamed concrete, of polymer, polymer-cement, and other composites [2, 3]. In 1999-2005 they allowed the number of solutions to be obtained that proved to be useful in production and applications of dry building mixes, including the mixes with polymer and cellulose fibre [4].

Leaning against such experience this computer-aided research technology was used when developing dry mixes for industrial floor coverings, specifically, to find optimal compositions of dry mixes for fibrous polymer-cement composites.

Conditions of experiment

The experiment was carried out in the laboratory of Henkel Bautechnik (Ukraine) company, according to optimal three-level four-factor design. Varied in 18 mixes were two factors of mineral framework and two modification factors. The variables of the first group have defined the grain size distribution of mix mineral part, its total content being constant (56.5 mass parts in 100 m.p. of dry mix). Mass ratio of dolomite filler to sand X_1 at levels of 0.25, 0.5, and 1.0 was normalized to $|x_1| \le 1$ as $x_1 = \log_2 X_1 + 1$. Mass ratio of granite screenings (up to 1.2 mm) and sand X_2, at levels 5, 7.5, and 10, was conventionally normalized to $|x_2 = (X_2 - 7.5)/2.5| \le 1$.

The conditions of modification purposed to increase the crack resistance of floors were specified by the content (m.p.) of polyamide fibre (mark "6, 7/3") equal to 0.05, 0.1, 0.2 – factor X_3 normalised as $x_3 = \log_2 X_3 + 3.322$, and of redispersible polymer additive (Vinnapas LL 222) – X_4 equal to 1.5, 2.75, 4 m.p., $x_4 = (X_4 - 2.75)/1.25$).

The dosages of other components (mineral binder, anti-shrinkage additive, superplasticizer, etc.) did not vary. In 10 minutes after mixing with water all 18 mixes had the same fluidity – spread of 21cm, water-cement ratio W/C being from 0.68 to 0.91 (with C equal to amount of Portland and aluminous cements).

Among various properties (quality criteria) determined for 18 mixes at various terms of hardening was linear shrinkage ε (mm/m), measured with digital electronic micrometer on 100 cm long specimens hardening in special ruler-form with moving end.

ES-models and property fields in coordinates of mix proportions

The data obtained in the experiment have allowed non-linear ES-models describing the fields of material properties in coordinates of composition factors to be built, the influence of these factors on the properties to be analysed, the variety of optimisation problem to be formulated and solved.

In particular, the models (1) and (2) were built for two quality criteria of composite of 5-day age – the properties that should be optimised. The tensile strength when bending R_5 (MPa) should be maximised while linear shrinkage ε_5 – to be minimised, in order to increase the crack resistance of the early age floor. The models of this kind were also built for two other quality criteria of the material after 28 days of hardening the values of which in this optimisation problem were restricted by specifications: compression strength $R_{28} \geq 35$ MPa and linear shrinkage $\varepsilon_{28} \leq 1.5$ mm/m.

$$\begin{aligned} R_5 = {} & 2.73 + 0.16x_1 \pm 0\, x_1^2 + 0.16x_1x_2 - 0.12x_1x_3 \pm 0\, x_1x_4 \\ & \pm 0\, x_2 - 0.28x_2^2 \pm 0\, x_2x_3 \pm 0\, x_2x_4 \\ & + 0.14x_3 - 0.28x_3^2 - 0.08x_3x_4 \\ & + 0.29x_4 + 0.44x_4^2 \end{aligned} \quad (1)$$

$$\begin{aligned} \varepsilon_5 = {} & 0.38 \pm 0\, x_1 - 0.09x_1^2 - 0.05x_1x_2 + 0.06x_1x_3 + 0.03x_1x_4 \\ & \pm 0\, x_2 + 0.19x_2^2 + 0.03x_2x_3 + 0.03x_2x_4 \\ & - 0.08x_3 + 0.08x_3^2 - 0.03x_3x_4 \\ & - 0.11x_4 + 0.04x_4^2 \end{aligned} \quad (2)$$

The models describe the full fields [5] of material properties in normalized composition coordinates (only with significant effects at risk 0.1 and the errors s_{R28} =1.28, $s_{\varepsilon 28}$ =0.154, s_{R5} =0.16 и $s_{\varepsilon 5}$ =0.018). The most important for the optimization problem under consideration generalizing indices of the fields (1) and (2) are: $R_{5.max}$= 3.64 MPa (at $x_1 = x_4 = +1$, $x_2 = +0.28$, $x_3 = -0.12$), $\varepsilon_{5.min}$ = 0.10 mm/m (at $x_1 = -1$, $x_2 = -0.26$, $x_3 = x_4 = +1$), relative increases $\delta\{\ \} = 2.4$ and $\delta\{\varepsilon_5\} = 9.0$.

The comparison of these indices indicates the impossibility to reach both maximal R_5 and minimal ε_5 concurrently (coordinates of two optima do not coincide, however evident is the tendency to favourable influence of increased content of Vinnapas on both properties).

Represented in Fig. 1 is the local field of 5-day strength in coordinates of mineral framework, at medium content of fibre and dosage of redispersible powder at upper level ($x_3 = 0$, $x_4 = 1$). Vector of averaged gradient points to zone of the best for R_5 filler-sand proportions at indicated conditions of modification.

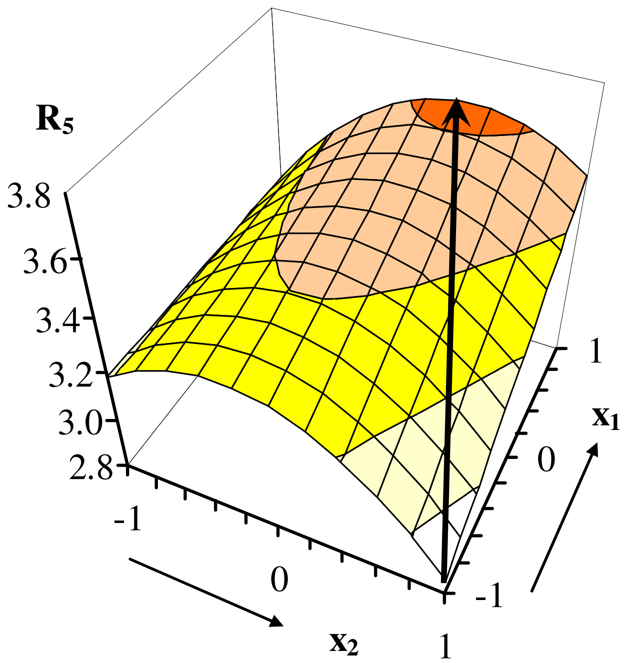

Fig. 1

The field of bending strength of the composite after 5 day of hardening, in coordinates of mineral framework at medium content of polymer fibre and highest dosage of polymer additive

With rather high negative correlation (up to r = –0.7), estimated in computational experiments [6] on local fields $R_5(x_1, x_2)$ and ε_5 (x_1, x_2) in certain zones of (x_3, x_4)-region, it is not sufficiently high in some other zones, specifically, at $x_3 = 0$, $x_4 = 1$ (conditions of Fig. 1 and Fig. 2), the estimate of correlation coefficient between R_5 and

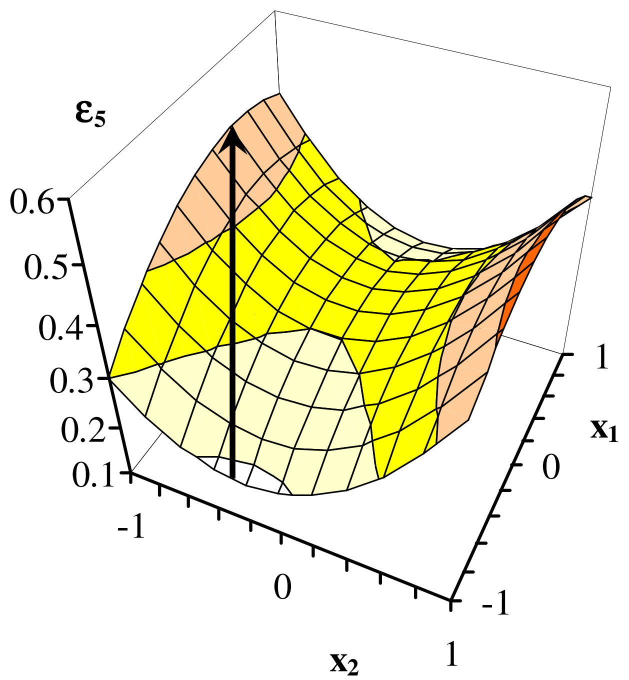

Fig. 2

The field of linear shrinkage of the composite after 5-day hardening, in coordinates of mineral framework at medium content of the fibre and highest dosage of Vinnapas

ε_5 being $|r| < 0.5$. The direction of averaged gradient of ε_5-field shown in Fig. 2 does not coincide with that of R_5-field (Fig. 1) at the same values of modification parameters. So it has been necessary to search for compromise compositions. The rational compositions should be found by four criteria. Bending strength R_5 and linear shrinkage ε_5 after 5 day of hardening have been the optimality criteria, compression strength R_{28} and linear shrinkage ε_{28} of composite of 28-day age have presented restricting criteria.

Conditions of search for compromise compositions within guaranteeing limits of restriction criteria

To find the compositions that would provide, with certain risk, the required levels of restriction criteria the requirements must be made more stringent during the search, guaranteeing levels [3, 7] must be introduced. To assign the guaranteeing requirement of compression strength R_{28} taken into account are experimental error s_{R28}=1.28 MPa, average value of prediction variance function $\overline{d}$ = 0.70 for the model obtained on results of experiment realised by above-mentioned design, and corresponding to 10% risk quantile of Student distribution t_{10}= 1.284. Thus, the level of R_{28} that would guarantee the minimal required level R_{28} = 35 MPa was determined as $R_{28.10} = R+\Delta R = 35 + 1.28\cdot 0.7^{0.5}\cdot 1.284 = 36.38$ MPa. To tighten the requirement for 28-day shrinkage the greater risk could be accepted (α=0.25), subtracted from specified level ε_{28} = 1.5 mm/m was $\Delta\varepsilon_{28} = 0.154\cdot 0.7^{0.5}\cdot 0.674$, giving the guaranteeing level $\varepsilon_{28.25}$ = 1.413 mm/m.

As initial values to start the search for compromise between possible maximal R_5 and minimal ε_5 the median levels of the fields (1-2) of optimality criteria are taken: $R_{5.M} = (R_{5.max}+ R_{5.min})/2 = (3.64+1.51)/2 = 2.57$ MPa and $\varepsilon_{5.M} = (0.90+0.10)/2 = 0.50$ mm/m.

Computational experiment

The search is based on results of computational experiment on the fields Y(**x**) of M properties (here M=4) in composition coordinates, described by ES-models, with iterative use of Monte Carlo method. Iteration procedure of the search allows for the dialogue between researcher and computer and makes it possible the guaranteeing composition-process parameters (acceptable, optimal, compromise) to be found.

At the first stage of the fist iteration (denoted as "1-1") N uniformly distributed random vectors **x** are generated in the region Ω_x of fields Y(**x**). In this particular problem Ω_x presents 4-dimensinal cube corresponding to the variety of compositions. Generated inside the cube have been N=10000 points, with 16 vertices (±1, ±1, ±1, ±1) added. This is as if multi-dimensional net was thrown on factor region. With values of each factor varying from −1 to +1, its range Δx_i = 2 and average coordinate step between knots equals $\Delta x_i / N^{0.25} = 2 / 10 = 0.2$. Levels of all M properties in N points (here M=4, N=10016) are calculated by ES-models.

At stage "1-2" N_Ω points, fallen within admissible region, are picked out by levels of restriction criterion fields. The other $N - N_\Omega$ points are eliminated. In the problem to be solved 13 compositions (the number displayed in Fig. 3) have remained in Ω (offering $R_{28} \geq 35$ MPa and $\varepsilon_{28} \leq 1.5$ mm/m and levels of the criteria to be optimised not worse than median), with 10003 unacceptable compositions removed. The region of search has contracted – its volume [1, 3] in relation to initial region is almost two orders less (K_Ω in Fig. 3).

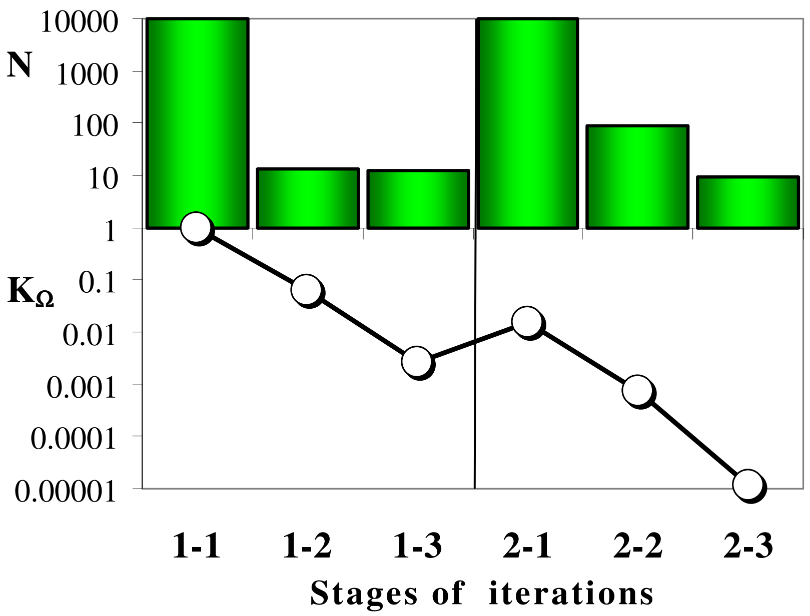

Fig. 3.

The changes of number of competing compositions (N) in search region and of its relative volume (K_Ω) in 4-factor cube during the course of computational experiment

At stage "1-3" step-by step approach to individual optima ($R_{5.max}$= 3.64 MPa and $\varepsilon_{5.min}$= 0.09 mm/m) is performed. Search region shrinks to the region of compromise Ω_{comp} (K_Ω about 0.001, fig. 3). The particular results of optimisation at this stage are shown in Fig. 4., the values of optimality criteria being better than median levels ($R_5 = 2.9 > R_{5.M} = 2.57$, $\varepsilon_5 = 0.22 < \varepsilon_{5.M} = 0.50$). The further progress to the optima at this iteration is impossible.

The process should be continued at the next iteration, with new 10000 points being generated in extended (accounting for step Δx_i, fig. 5) region of compromise, plus 12 points ("good" compositions) obtained at previous iteration (fig. 2a). The primary emphasis at the second iteration has been on the search for compositions providing the reduced early age shrinkage. Stage "2-3" has brought the boundaries of Ω_{comp} much closer together (with factor intervals from 0.06 to 0.029 in units of normalised factor space, Fig. 5). The upper and lower levels of optimality criteria over compromise region have practically closed up (Fig. 4). So the search process could be stopped.

Compromise compositions

The compositions with following values of the parameters have been chosen on results of computational experiment on the full fields of four criteria in 4 composition coordinates. Mineral framework: mass ratio of dolomite filler to sand at the lower level X_1 = 0.25 (x_1= −1); mass ratio of granite screenings to sand in the range near medium level, 7.0 ≤ X_2 ≤ 7.4 (−0.20 ≤ x_2 ≤ −0.04). Modifiers: the content of polyamide fibre X_3 in the interval from 0.17 to 0.20 m.p. (0.77 ≤ x_3 ≤ 1); the dosage of polymer additive Vinnapas LL 222 at the upper level X_4 = 4 м.ч. (x_4=+1).

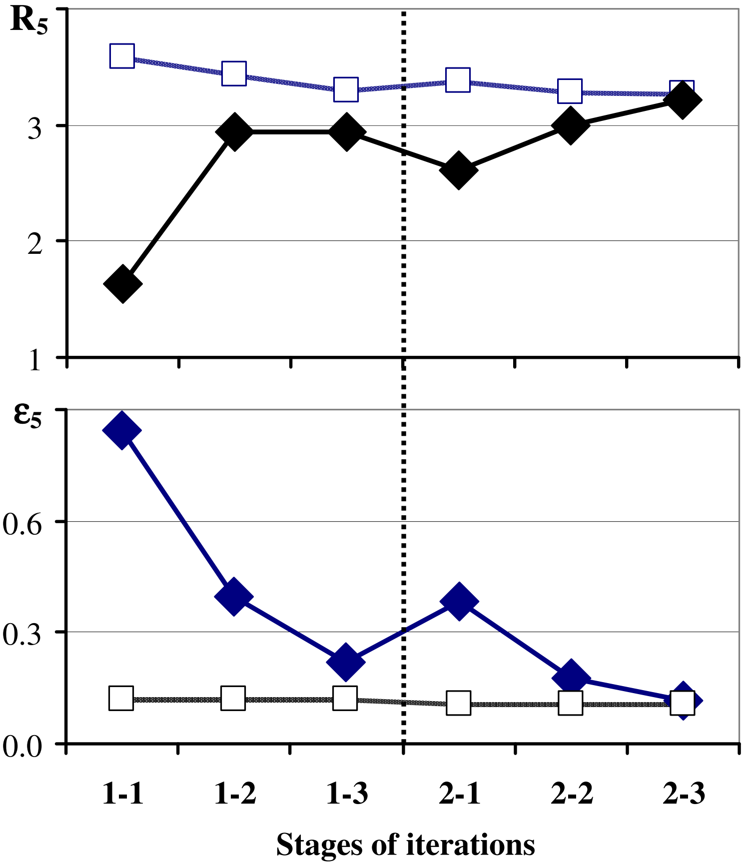

Fig. 4. Changes of lower and upper levels of optimality criteria (bending strength and linear shrinkage of composite after 5-day hardening) in computational experiment during the search for compromise solution

The indicated mix proportions would provide the following values of the composite quality criteria.

Specified:

- compression strength 37.5 ≥ R_{28} ≥ 36.5 ≥ $R_{28.10}$ = 36.4 MPa,
- linear shrinkage 1.32≤ ε_{28} ≤1.34 ≤ $\varepsilon_{28.25}$ = 1.41 mm/m.

Optimised:

- tensile strength of 5-day material when bending R_5 = 3.2-3.3 MPa (being about 25% greater than median level),
- linear shrinkage after 5 day of hardening ε_5 in the range 0.099-0.113 mm/m (this being almost 5 times less than median level).

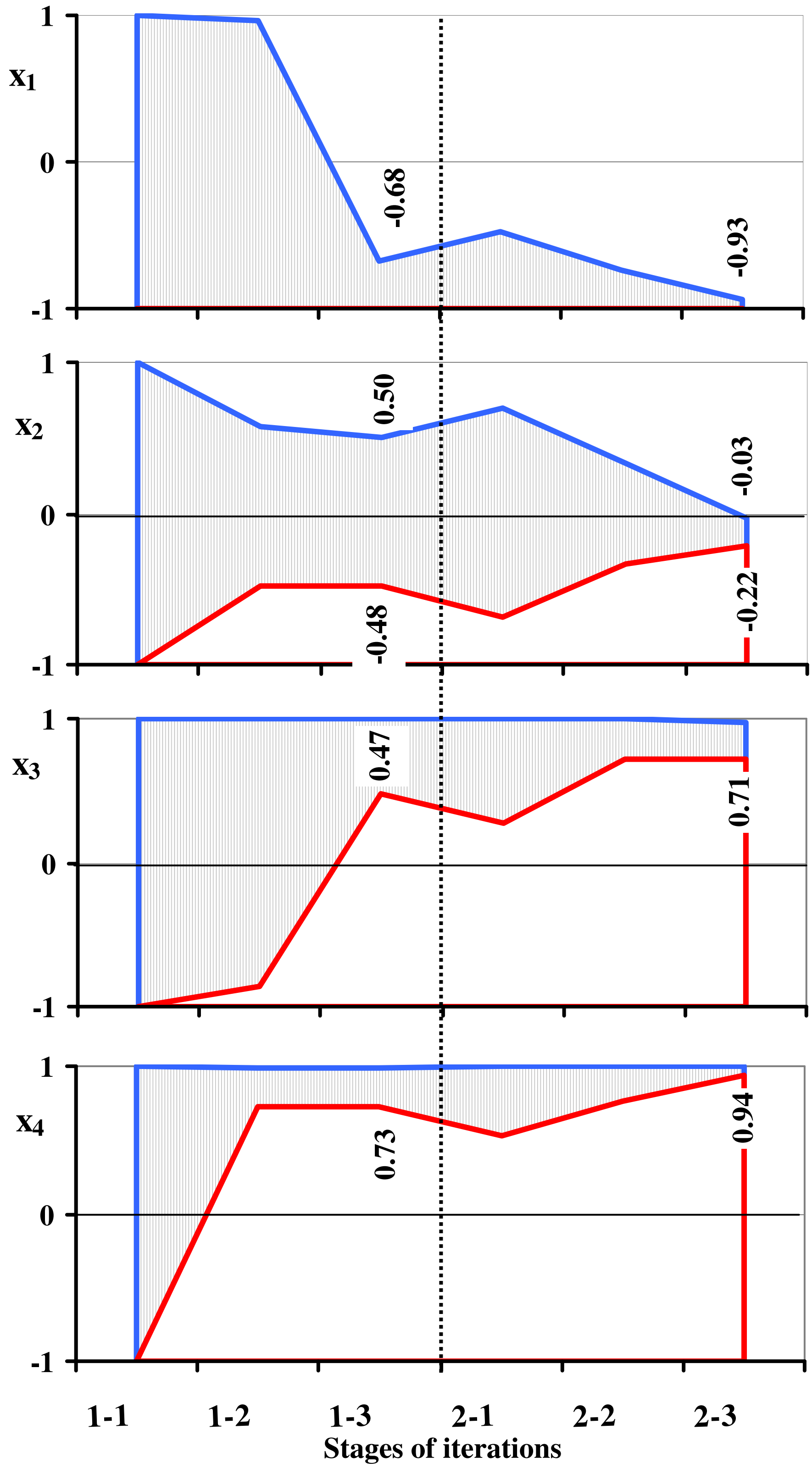

Fig. 5. Changes of intervals of 4 normalised factors at stages of the search for compromise compositions

Conclusion

Fibrous polymer-cement dry mixes are recommended to be used in high strength coverings for industrial floors. With optimal content of polyamide fibre “6, 7/3” and polymer powder Vinnapas LL 222 and optimal grain composition of mineral framework such mixes provide not only norm requirements to composite at 28-day age but increased bending strength and lessened shrinkage at early terms of hardening as well, serving to increase the crack resistance.

The methods of computational materials science allow rational technological solutions to be found when researching and developing new multicomponent materials for sustainable construction.

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