COMPROMISE OPTIMISATION OF SLAG ALKALINE BINDERS WITH COMPUTATIONAL MATERIALS SCIENCE METHODS

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Abstract. The influence of eight composition and thermal treatment factors on strength, frost resistance, and chemical resistance of slag alkaline binders is evaluated by non-linear experimental-statistical models. The models are used to solve two optimisation problems. With the help of iterative random scanning of the fields of material properties in eight composition and process coordinates found have been the compromise solutions when maximising quality criteria and the conditions for saving resources.

Key words. *Binder system "slag – zeolite – natrium sulfate – portland cement", strength, durability, property field, Monte Carlo, compromise*

1 Introduction

The composites on alkaline binders are multi-component materials, with many parameters defining the processes of their structure formation and destruction. When studying, designing, and manufacturing these materials numerous criteria of their quality condition the choice of rational composition and process parameters.

Effective solutions can be obtained through combined realization of reasonable (by theoretical prerequisites and by fulfilment) natural and computational experiments. To carry out these latter certain models are necessary.

In the extensive and in-depth studies of alkaline composites conducted in V.D. Glukhovsky Scientific Research Institute for Binders and Materials [1-4] used during more than three decades have been experimental-statistical (ES) models [5]. Experience in ES-modelling when investigating materials of this kind was analysed at two International conferences [6-7] and generalised in the specialised manual [8].

New possibilities and advanced tools have been put forward [9] on incorporation of ES-models into computational building materials science, as descriptions of the fields Y(**x**) of material properties (any characteristics or quality criteria Y) in a region Ω_x of composition and

process coordinates (vector **x**). These means can be especially helpful in the development of complex alkali activated systems.

Specifically, to find optimal technological solutions (composition and process parameters) for the binder system “granulated blast furnace slag – zeolite – natrium sulfate – portland cement”, developed by P. Krivenko and J. Skurchinskaya [10], iterative random scanning [9] of the fields of material quality criteria has been used.

2 Conditions of experiment and modelling

The influence of eight composition and process factors on ultimate strength of the binder system after 28 days of hardening (R, MPa) and on two durability criteria, frost resistance (F, cycles) and coefficient of resistance after keeping in Na_2SO_4 solution (K), has been investigated at a certain stage of the study. Three groups of the factors X_i have been varied in experiment as $X_{i.0} \pm \Delta X_i$:

- “composition” – contents (% by binder mass) of zeolite ($Z = X_1 = 10 \pm 10$), alkali salt ($A = X_2 = 5 \pm 5$), and cement ($C = X_3 = 2.5 \pm 2.5$),
- “preparing conditions” – dosage of plasticizer ($P = X_4 = 0.4 \pm 0.4\%$) and fineness of grinding ($S = X_5 = 320 \pm 30$, m^2/kg),
- “thermal treatment conditions” – precuring ($\tau_0 = X_6 = 8 \pm 4$, h), isothermal heating temperature ($T = X_7 = 75 \pm 15°C$), heating time ($\tau_T = X_8 = 6 \pm 2$, h).

The factors are normalized to $-1 \le x_i \le +1$ as $x_i = (X_i - X_{i.0})/\Delta X_i$ [11].

Under non-linearity hypothesis the influence of k = 8 factors on binder quality criteria can be described by second degree polynomials Y(**x**), with $L = (k+1)(k+2)/2 = 45$ coefficients to be estimated on experimental results. To make this possible special design of experiment has been synthesised [10]. The design reduces the number of runs to 54 combinations of the levels of composition-process parameters, i.e., 54 slag alkaline composites (whereas full factor experiment of the 2^{nd} order would require $3^8 = 6561$ combinations).

Scatter diagram in fig. 1 represents the results of the experiment. The correlation between quality criteria either is not revealed (correlation coefficients $r\{F, R\} = -0.08$, $r\{F, K\} = 0.25$) or can be admitted with high risk ($r\{KR\} = -0.35$).

All three Y vary in the wide intervals: $4.0 \le R \le 41.3$, $2 \le F \le 200$, $0.25 \le K \le 3.85$. Though adequately describing such experimental results, ES-models for these Y (with only significant coefficients at 10% risk and generated errors of experiment [8, 9] $s_R = 3.48$ MPa, $s_F = 18.6$ cycles, $s_K = 0.26$) could produce unreal large or small estimates for

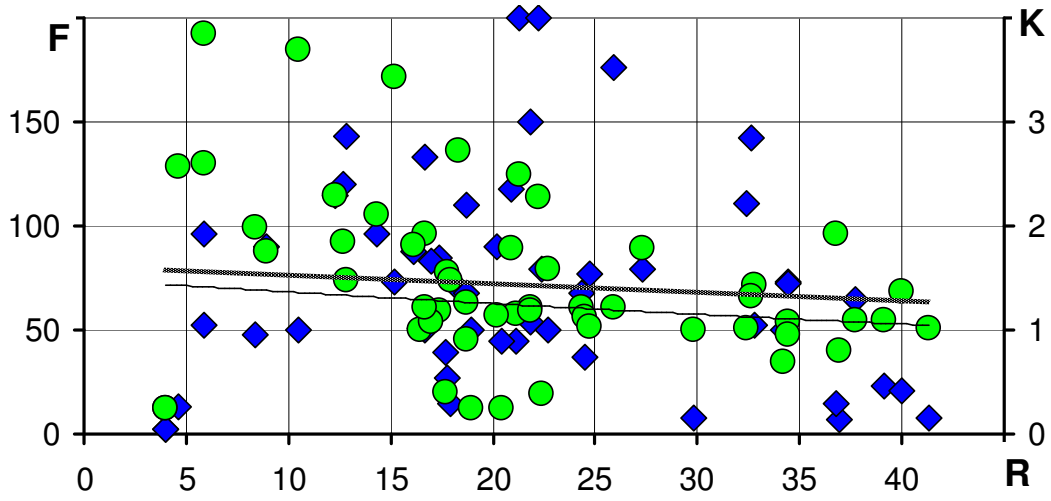

Fig. 1. Experimental values of strength R, frost resistance F (♦), and chemical resistance K (●) for 54 variants of binder system

extreme values of the properties under study. That is why the models should be built for transformed criteria κY [12], basing on logarithmic transformation ($\kappa Y = \ln [p\{Y\}/(1 - p\{Y\})]$ [5] put forward to provide the value of probability, calculated by any model, within existence limits, $0 \le p\{Y\} \le 1$.

So, in particular, put in correspondence to $p\{R\} = 0$ (i.e., $\kappa R \to -\infty$) and to $p\{R\} = 1$ (i.e., $\kappa R \to -\infty$) have been the very smallest value of strength $R^*_{min} = 3$ (instead of $R_{min} = 4$ MPa) and the greatest possible value $R^*_{max} = 43$ (instead of 41.3 MPa) respectively – to obtain, firstly, $p\{R\} = (R - R^*_{min}) / (R^*_{max} - R^*_{min})$ and then κR.

On the transformed data, specifically for strength, the model (1) has been built, with extreme values $\kappa R = -5.30$ and $\kappa R = +3.57$ corresponding to the lower and the highest levels of the field $R(\mathbf{x})$, $R_{min} = 3.2$ and $R_{max} = 41.9$ MPa.

$$\kappa R = -0.19 + \underbrace{0.20x_1 \pm 0\,x_1^2 + 0.48x_1x_2 + 0.34x_1x_3 + 0.86x_2 \pm 0\,x_2^2 - 0.37x_2x_3 + 0.25x_3 \pm 0\,x_3^2}_{(a)} + \underbrace{\pm 0\,x_4 + 0.55x_4^2 - 0.24x_4x_5 + 0.18x_5 + 0.72x_5^2}_{(b)} + \underbrace{0.27x_6 - 0.97x_6^2 - 0.15x_6x_7 \pm 0\,x_6x_8 + 0.33x_7 + 0.89x_7^2 \pm 0\,x_7x_8 + 0.24x_8 - 0.36x_8^2}_{(c)} + \underbrace{\pm 0\,x_1x_4 - 0.22x_1x_5 \pm 0\,x_2x_4 \pm 0\,x_2x_5 + 0.27x_3x_4 + 0.28x_3x_5}_{(d)} + \underbrace{\pm 0\,x_4x_6 \pm 0\,x_4x_7 + 0.14x_4x_8 \pm 0\,x_5x_6 + 0.14x_5x_7 - 0.23x_5x_8}_{(e)} + \underbrace{0.20x_1x_6 + 0.24x_1x_7 \pm 0\,x_1x_8 + 0.38x_2x_6 \pm 0\,x_2x_7 \pm 0\,x_2x_8 + 0.18x_3x_6 \pm 0\,x_3x_7 + 0.20x_3x_8}_{(f)} \quad (1)$$

The model is structured by three factor groups. Block (a) includes the

estimates of effects of three composition factors (at central levels of other factors $x_i = 0$). The content of alkali salt (x_2) has the greatest influence on strength, with extent of the effect depending on amounts of zeolite and cement (the greater is Z and the less is C, the higher is effect of A). Block (b) estimates the role of factors defining mix properties; it should be noted that increased levels of P and S could be useful for increasing binder strength. The effects of thermal treatment parameters evaluated in block (c) show that excessive increase of τ_0 and τ_T does not make sense.

Blocks (d, e, f) contain 11 estimates (commensurable with effects in main blocks) and evaluate the changes in the influence of factors from one group in dependence of the levels of factors from another group. So the positive influence of precuring (x_6) would grow for the systems with increased portions of zeolite (coefficient at x_1x_6 – $b_{16}>0$), alkali salt ($b_{26}>0$), and cement ($b_{36}>0$).

Similar ES-models have been built for the other criteria as well, the estimates of their maximal and minimal levels ($F_{max} = 207$, $F_{min} = 0$, $K_{max} = 4.0$, and $K_{min} = 0.20$) being close to extreme values in the experiment (fig. 1). The models can be used, in tandem with Monte Carlo, for simulating material properties in computational experiments, with various purposes [9], specifically to find optimal composition-process solutions.

3 Search for optimal composition-process parameters

3.1. Guaranteed levels of the quality and conditions of the search

The following requirements ($Y \geq Y_{norm}$) were specified for three quality criteria of binder system in a specific design task: strength $R \geq R_{norm} = 30$ MPa, frost resistance $F \geq 100$ cycles, coefficient of resistance $K \geq 1$. However, to increase the reliability of the new material it has been decided to orient the search for optimal composition and process parameters towards more stringent requirements, turning to probabilistic quality criteria Y_α, which would involve the risk of modelling error [5, 9].

So R as a quality criterion has been substituted by guaranteed strength R_α, with risk $\alpha = 0.05$ being accepted. In optimisation procedure considered below the level of R_{05} at any point $\mathbf{x} = (x_1, \ldots, x_8)$ of factor region is determined by model (1) as $R_{05}(\mathbf{x}) = R(\mathbf{x}) - \Delta R_{05}(\mathbf{x})$, where subtracted semi-interval $\Delta R_{05}(\mathbf{x}) = t_{05} \cdot d^{0.5}(\mathbf{x}) \cdot s_R$ is defined by risk level (through quantile of Student criterion, $t_{05} = 1.645$), by value of prediction variance function d [11] (in this particular case average value over the region is used, $d = 1$), and by experimental error ($s_R = 3.48$). Thus all calculated values of R are reduced by 5.7 MPa.

The probabilistic criteria for frost resistance and sulfate resistance have been taken at softer risk conditions, $\alpha = 0.10$ (quantile $t_{10}=1.282$). With inclusion of experimental errors ($s_F = 18.6$ cycles, $s_K = 0.26$) this give the semi-intervals $\Delta F = 23.9$ and $\Delta K = 0.33$.

Two series of computational experiments have been fulfilled to solve two optimisation problems. In the first series (A) compromisingly maximised should be $R_{05} > 30$ and $F_{10} > 100$, under restriction by chemical resistance $K_{10} \geq 1.0$.

In the second series (EA) the search for minimal expenditure of some "expensive" factors is carried out, with requirements $R_{05} \geq 30$, $F_{10} \geq 100$, and $K_{10} \geq 1.0$ to be fulfilled.

Iterative procedure of the search for the optima in the region Ω_x of the fields $R_{05}(\mathbf{x})$, $F_{10}(\mathbf{x})$, and $K_{10}(\mathbf{x})$, of guaranteed properties in composition and process coordinates, is arranged on random scanning of these fields, described by ES-models, with Monte Carlo method.

One of the advantages of the procedure is the technologist-computer dialog mode available at any iteration. The results obtained at current iteration can be reviewed, in particular, from economic and technological positions. Discussed could be the rate $\partial Y/\partial x_i$ of changing (maximising or minimising) any criterion Y with some factor or group of them. Consecutive strategy of computational experiments is realised.

3.2. Search for compromise

To find composition-process variants that would provide compromise maximum for both guaranteed strength and guaranteed frost resistance, with guaranteed level K_{10} of chemical resistance coefficient not less than required level $K_{norm} = 1$, the series A of computational experiments has been conducted. For this particular problem general search procedure is presented and particular values of its parameters are indicated, with results shown in table 1 and fig. 2.

At initial stage (A11) of the first iteration N=10000 uniformly distributed random vectors **x** (of 8 normalised coordinates of composition and thermal treatment in the intervals from –1 to +1) are generated. Added to this points inside Ω_x are $N_{\pm 1}= 2^k = 2^8 = 256$ hypercube vertices ($\pm 1, \ldots, \pm 1$) – determinate points (that could not be generated). At each of $N_{11} = N + N_{\pm 1} = 10256$ points the levels of the fields of three criteria, R, F, and K, are estimated by respective model.

At the next stage (A12) after assorting (by criteria values) N_{11} points (composition-process variants) those of them are eliminated at which binders would have low guaranteed levels $R_{05}<30$ MPa, $F_{10}<100$ cycles, and $K_{10}<1$. It turns out to be 10253 such variants and only $N_{12} = 3$

Table 1. Intervals of factors at stages of the search for compromise (A series)

x_i	A11	A12	A13	A22	A23	A32	A33	A42	A43
x_1		–0.12...0.90	–0.12...0.90	–0.20...0.99	0.48...0.76	0.40...0.85	0.41...0.76	0.35...0.85	0.36...0.68
x_2		–0.28...0.70	–0.28...0.70	–0.35...0.79	0.45...0.76	0.35...0.84	0.35...0.62	0.25...0.70	0.25...0.53
x_3		0.20...0.99	0.20...0.99	0.11...1	0.91...0.98	0.85...1	0.92...0.99	0.92...1	1
x_4	–1...1	0.82...0.99	0.82...0.99	0.70...1	0.94...0.99	0.85...1	0.94...1	0.94...1	1
x_5		0.93...0.99 –0.98	0.93...0.99 –0.98	0.85...1 –1...–0.90	0.91...0.97	0.85...1	0.90...1	0.90...1	1
x_6		–0.36...0.91	–0.36...0.91	–0.45...1	0.19...0.63	0.10...0.75	0.11...0.38	0.00...0.45	0.01...0.27
x_7		–0.05...0.35	–0.05...0.35	–0.05...0.45	0.01...0.31	–0.10...0.40	–0.08...0.34	–0.15...0.45	0.08...0.39
x_8		–0.03...0.88	–0.03...0.88	–0.10...1	0.08...0.44	0.00...0.55	0.03...0.29	–0.10...0.35	–0.09...0.27

points (fig. 2) fall within the region of acceptable solutions Ω_{12}.

At final stage of the iteration (A13) the levels of R_{05} and F_{10} should be increased, the variants not satisfying new requirements eliminated, thus narrowing the addmissible region (to $\Omega_{13} < \Omega_{12}$). In this particular problem, however, the values of qualty criteria at all 3 point in Ω_{12} are already close to lower limits (strength between 30-31.3). So the result of A12 could not be improved at A13 and is taken as the result of the first iteration ($R_{05} \geq 30$, $F_{10} \geq 115$, $K_{10} \geq 1$, fig. 2).

The second and the following iterations Aj are arranged by the same principles, but with some special expedients at the stages.

At initial stages Aj1:

• The boundaries of search region are extended in relation to Ω_{j-1} achieved at previous iteration, by about 0.1 in both sides for each normalised variable x_i (i=1, …, 8). This can produce the values of criteria (R_{05}, F_{10}, K_{10}) below limiting levels (fig. 2). Thus in particular, widening the interval for dosage of alkali salt from $-0.28 \leq x_2 \leq 0.70$ at A13 (table 1) to $-0.35 \leq x_2 \leq 0.80$ at A21 reduces the lower level of R_{05} at this stage to only 13 MPa.

• Added to N generated points are all the admissible variants from preceding iteration. In particular, $N_{21}=N + N_{13} = 10003$ (fig. 2).

At intermediate stages Aj2 the borders of admissible region Ω_{j2} are defined not by initial requirmenrs (Y_{norm}) to optimality criteria but by the worst levels of the criteria improved at previous iteration. Specifically, the low level of frost resistance at stage A32 (fig. 2) is 130 cycles, not 100 (initial, at A12).

At final stages of the iterations:

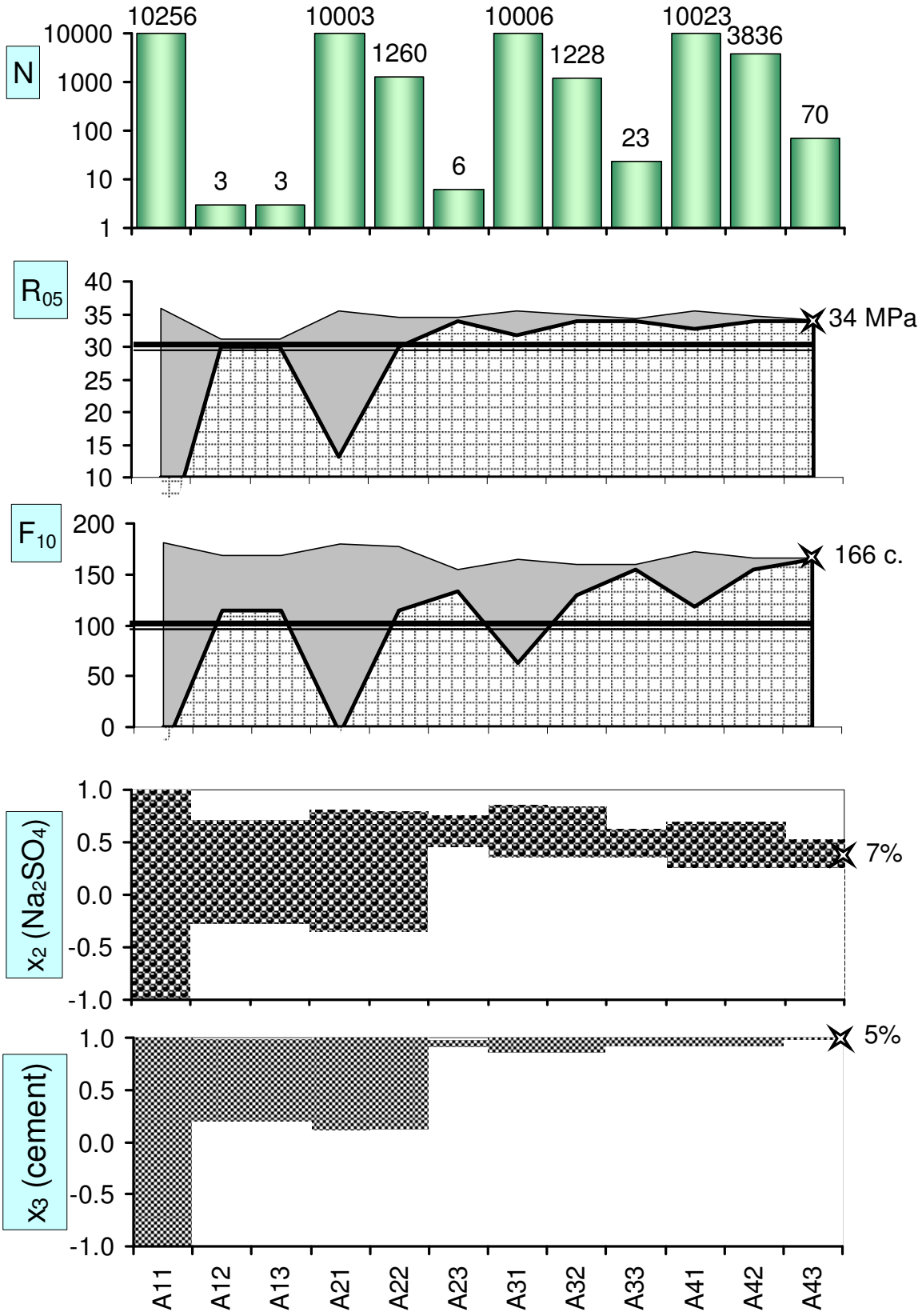

Fig. 2. Changes in the number (N) of solutions being tested, in ranges of optimality criteria, and in limits of two factors at stages of the search for compromise

• Step-by-step approach to the individual maxima of guaranteed levels (R_{05} and F_{10}) is executed by moving the lower limits of these criteria upward and excluding the composition-process variants, which do not satisfy the new limits. Thus at stage A23 the strength has been raised to $R_{05} \geq 34.0$ and frost resistance to $F_{10} \geq 130$ (sulfate resistance coefficient K_{10} remaining ≥ 1); as rhe result only $N_{23} = 6$ points out of $N_{22} = 1434$ have remained in the region of addmissible solutions (fig. 2).

• The final stage of the last iteration (4^{th} in fig. 2) leaves, as a rule, rather many competing solutions ($N_{43} = 70$) with the criteria varying in narrow ranges ($34.0 \leq R_{05} \leq 34.1$, $165 \leq F_{10} \leq 166$). To determine the terminal normalised values of factors it is reasonable to average the coordinates of only the points providing the best levels of the criteria (3 points with $R_{05} = 34.1$ and 12 points with $F_{10} = 166$).

Thus the resulting compromise optimal values of the normalized factors have been obtained:

$x_1 = 0.45$, $x_2 = 0.41$, $x_3 = 1.0$;

$x_4 = 1.0$, $x_5 = 1.0$;

$x_6 = 0.15$; $x_7 = 0.22$; $x_8 = 0.04$.

On reverting to named values and rounding them to the levels feasible for technical realization the following results have been obtained:

• quantities of zeolite, alkali salt, and cement

Z = 14.5%, A = 7%, and C= 5%,

• dosage of plasticizer P = 0.8%, spesific surface S = 350 $м^2$/kg,

• precuring $\tau_0 = 8.5$ h, heating temperature T = 80°C, during $\tau_T = 6$ h.

This composition at these conditions of thermal treatment would provide the guaranteed strength $R_{05} = 34$ MPa, frost resistance $F_{10} = 166$ cycles, and resistance coefficient $K_{10} = 1.2$.

3.3. Search for resources-saving conditions

The problem of minimising the expenditure of resources, with requirements for material quality fulfilled (optimisation of the 2^{nd} type [11]), arises regularly in building materials technology. It becomes a real challenge as the number of factors (x) taken into consideration and of specified quality criteria ($Y \geq Y_{norm}$) increases.

Iterative random scanning of material property fields in composition and process coordinates proved to be efficient in search for solutions of reduced matter and power expenses that would guarantee the compliance with specifications for the slag alkaline binder quality criteria.

It has been required, in particular, the guaranteed levels of strength, frost resistance, and chemical resistance to be $R_{05} \geq 30$ MPa, $F_{10} \geq 100$, and $K_{10} \geq 1$. For a certain version of problem conditions (EA)

looked for have been the ways to reduce the quantities of Na_2SO_4 and portland cement (factors A, C) with concurrent reduction of treatment temperature and time (T, τ_T).

Factor costs have been assumed to be the same for all these factors, over the ranges $X_{i.0} \pm \Delta X_i$ (i = 2, 3, 7, 8). Expenditure criterion to be minimised has been formed as $E_{2378} = x_2 + x_3 + x_7 + x_8$, without weight (price) coefficients. It takes the values from –4 (alkali salt and cement are not introduced, T = 60° during 4 hours) to +4 (at upper level of 4 factors). All other resources (x_1, x_4-x_6) can be used without restrictions (within the limits of $|x_i| \leq 1$) to provide the required levels of material properties.

To find the solutions of the problem above EA-series of computational experiments has been carried out.

It has been appropriate to use (as initial approximation) the region of admissible variants already obtained at stage A12, while searching for compromise maxima of R_{05} and F_{10}. There are N_{12} = 3 points in the region (fig. 2), the criterion E_{2378} being between 0.93 and 2.51.

The subsequent procedure of minimising E_{2378} does not differ in principle from presented above (A13-A43). At each new iteration (EAj) the former region of search is extended along each factor by 0.1- 0.2 of unit semi-interval to each side (a little more than during the search for compromise) and new “10000” random points are generated (stages EAj1). The variants not complying with the requirements and having the level of E_{2378} higher than that at preceding iteration are eliminated (stages EAj2). The highest value of E_{2378} is decreased so that its range is not greater than 0.1-0.2 (stages EAj3).

The results of this search are represented in fig. 3. At stage EA43 the expenses makes up –0.01…+0.05 (fig. 3a). The variation of average level for each item in E_{2378} (fig. 3b) shows that the economy is achieved due the savings in power resources: decrease in temperature of heating to 70-71°C and reduction of its duration to 5 h. The reduction in costs of composition is not substantial: the decrease in salt content is nullified by increase in cement dosage. Two of the other four factors (P and S) have rather quickly come to upper level, the content of zeolite also nears its upper limit, precuring time τ_0 remains practically the same. The final solution has been averaged over 18 variants obtained at the last stage (EA43) and rounded with account for possibilities to fix the factors:

Z = 19%, A = 5.6%, C = 4.4%, P = 0.8%, S = 350 $м^2$/kg,

τ_0 = 9.5 h, T = 70°C, τ_T = 5 h.

At these values of composition and process parameters the level of the expenditure E_{2378} = 0.05, with the normative requirements for three criteria of binder system quality satisfied.

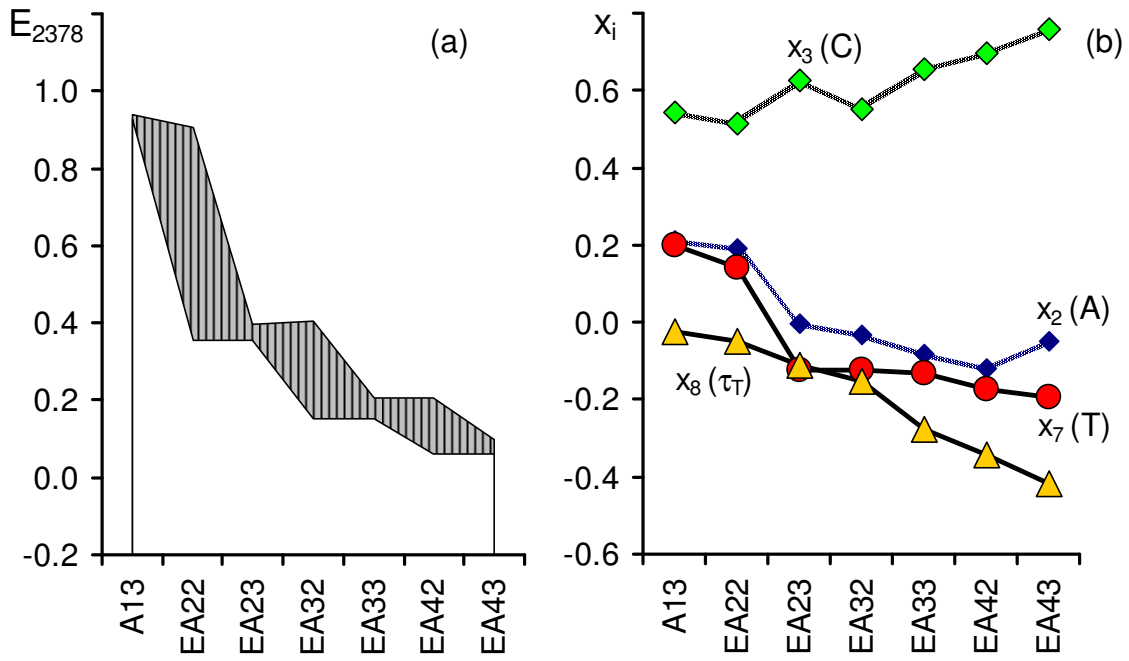

Fig. 3. Changes of expenses criterion (a) and its components (b) along the search for optimum

4 Conclusion

It is advantageous to use experimental-statistical models in research and development of materials on alkaline binders and other multi-component systems (especially with by-products among the components). ES-models enable new information about materials to be extracted from experimental data with methods of computational materials science.

Using Monte Carlo and ES-models for scanning the fields of three properties of alkaline geocement in eight composition and process coordinates found have been: 1) the compositions and process parameters that maximise the guaranteed level of the binder system quality, 2) minimal resources to guarantee the required quality level. The method allows the compromise solutions to be found in combined optimisation problems, with set of optimality criteria including both the properties and the factors defining them.

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