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ISOPARAMETRIC ANALYSIS WHEN STUDYING COMPOSITE MATERIALS

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Abstract. Presented is the method to analyse the variations in characteristics of composite materials under effects of composition-process factors in conditions when the factors must provide the unvarying level of one of the characteristics. The abilities of the isoparametric analysis, which is related to methodology of composition-process fields, are demonstrated. Experimental-statistical models describing the fields of the properties of filled carbamide binder in coordinates of degree of filling and filler mix proportions are used. The models have enabled to carry out the statistical trials in computational experiment. On its results the variations in strength and abrasion resistance of isoviscous compositions, with equal viscosity provided by various quantities of silicon carbide and coarse grains of andesite in the filler, are analysed.

Keywords: constant level of property, viscosity, composition field, experimental-statistical model, computational experiment, filled carbamide binder, strength, abrasion resistance.

ІЗОПАРАМЕТРИЧНИЙ АНАЛІЗ ПРИ ДОСЛІДЖЕННІ КОМПОЗИЦІЙНИХ МАТЕРІАЛІВ

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Анотація. Представлено метод аналізу змін характеристик композиційних матеріалів під впливом рецептурно-технологічних факторів в умовах, коли фактори повинні забезпечити незмінний рівень однієї із характеристик. Можливості ізопараметричного аналізу, пов'язаного з методологією рецептурно-технологічних полів, демонструються на даних про властивості наповненого карбамідного сполучного. За результатами обчислювального експерименту, який використовує експериментально-статистичні моделі, аналізуються зміни міцності та зносостійкості ізов'язких композицій, при забезпеченні рівної в'язкості різною кількістю в наповнювачі карбіду кремнію та крупних зерен андезиту.

Ключові слова: постійний рівень властивості, в'язкість, рецептурне поле, експериментально-статистична модель, обчислювальний експеримент, наповнене карбамідне сполучне, міцність, зносостійкість.

ИЗОПАРАМЕТРИЧЕСКИЙ АНАЛИЗ ПРИ ИССЛЕДОВАНИИ КОМПОЗИЦИОННЫХ МАТЕРИАЛОВ

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Аннотация. Представлен метод анализа изменений характеристик композиционных материалов под влиянием рецептурно-технологических факторов в условиях, когда факторы должны обеспечить неизменный уровень одной из характеристик. Возможности изопараметрического анализа, связанного с методологией рецептурно-технологических полей, демонстрируются на данных о свойствах наполненного карбамидного связующего. По результатам вычислительного эксперимента, использующего экспериментально-статистические модели, анализируются изменения прочности и износостойкости изовязких композиций, при обеспечении равной вязкости разным количеством в наполнителе карбида кремния и крупных зерен андезита.

Ключевые слова: постоянный уровень свойства, вязкость, рецептурное поле, экспериментально-статистическая модель, вычислительный эксперимент, наполненное карбамидное связующее, прочность, износостойкость.

Introduction. In many tasks of materials science there is a need to analyse the effects of composition-process (CP) factors, $\boldsymbol{x} = (x_1, x_2, \dots, x_i, \dots, x_k)$, on structural, technological, operational characteristics and other criteria related to materials (*properties*, *Y*) under condition of one of the properties (*base*, *B*, for instance, some technological criterion) being at constant level. It could be necessary to compare the properties of specific concretes [1-6] made from equally workable mixes or thermophysical and other properties of equal in density materials. The particular *isoparametric* (IP) conditions – isoviscous, of equal strength, of the same resources, etc., are defined by the purpose of a study. Experimental determination of the values of CP-parameters that would provide the same fixed level of the base property is quite laborious or practically unrealisable in some cases, in particular, when there is a need to study "iso-resistant" composites or those with equal portion of the pores in certain size range.

Put forward for such studies was [7] isoparametric analysis (IPA), on the base of experimental-statistical (ES) models $Y(\boldsymbol{x})$. Described by the models dependences of material properties on CP-factors could be analysed [8-10] under condition (1) of fixed level of one of the properties.

$$B(\boldsymbol{x}) = const = B_{is} \quad (1)$$

The complexes of ES-models, with $B(\boldsymbol{x})$ among them, enabled to reduce the volume of the experimental works many-fold and to obtain new information in studies of polymer binders [8, 11-13], of cellular and fine grained concrete [9, 14]. However graphic-analytical realisation [8-10] of this useful method was not sufficiently simple and convenient.

Another approach to IP-analysis has been formed, based on the concept of CP-fields of material properties (presenting the levels of *Y* at each point $\boldsymbol{x}$ of CP-factor domain Ω_x). The IP-conditions are achieved over the equipotential surface (1) of the one of them. In the case of 2-dimensional Ω_x the changes of $\boldsymbol{x}$ that would provide IP-conditions correspond to the movement along the isoline (1). Then the levels of other *Y* are estimated (by ES-models) along the trajectory in Ω_x defined by this isoline. The use of Monte Carlo method (MCM) in tandem with ES-models for simulating CP-coordinates and the levels of CP-fields frees IPA of complicated procedures [8-10] of moving by equal steps directly in the isoline and makes the results of computational experiments closer to inaccessible real data. Besides, this could be carried out in factor region of any reasonable dimensionality.

Such computational approach has been already applied in the studies of some composites [15-16], but has not been adequately described. Its abilities are demonstrated in this paper. As in other papers on CP-fields methodology [17-18], the data obtained when developing the compositions for industrial floors [19] are used.

Conditions of natural experiment and the models. The following CP-factors (normalised to $-1 \le x_i \le +1$) were varied in the designed experiment when studying carbamide binder filled with mixture of andesite and silicon carbide grains (abrasive production waste):

- degree of filling *F*, filler-polymer mass ratio (from 2 to 2.5) → x_1;
- *SC* – mass part of silicon carbide (with specific surface 320±10 m^2/kg) in fine fraction of the filler (from 0 to 60%) → x_2;

♦ *CA* – mass part of andesite coarse grains (specific surface 70±5 m^2/kg) in total amount of the filler (40±20%) → x_3.

The data on the properties of 15 compositions, according to design of the experiment, were obtained. Specifically, effective viscosity η (Pa·s, at shear rate 1 s^{-1}) of 15 mixes, compression strength *R* (MPa) and abrasion resistance *A* (h/g) of the hardened composites were determined. These data enabled to build ES-models (2-4) describing the whole fields of η, *R*, and *A*, in coordinates of all 3 normalised formulation factors (with 1%-risk, at experimental errors s_e equal to 3.5, 1.3, and 0.41 respectively) .

$$\begin{aligned} \ln\eta = \; & 3.27 + 0.61x_1 + 0.33x_1^2 + 0.09x_1x_2 - 0.16x_1x_3 \\ & + 0.36x_2 + 0.36x_2^2 - 0.14x_2x_3 \\ & - 0.36x_3 + 0.46x_3^2 \end{aligned} \quad (2)$$

$$\begin{aligned} R = \; & 75.3 \pm 0\; x_1 - 1.7x_1^2 - 1.1x_1x_2 + 2.4x_1x_3 \\ & + 9.4x_2 - 4.7x_2^2 + 4.6x_2x_3 \\ & - 11.0x_3 - 4.8x_3^2 \end{aligned} \quad (3)$$

$$\begin{aligned} A = \; & 6.69 + 0.22x_1 - 0.47x_1^2 - 0.52x_1x_2 + 0.85x_1x_3 \\ & + 1.45x_2 \pm 0\; x_2^2 \pm 0\; x_2x_3 \\ & \pm 0\; x_3 - 1.12x_3^2 \end{aligned} \quad (4)$$

Shown in Fig.1 is the field η(***x***), in normalized coordinates of degree of feeling (x_1) and quantities of silicon carbide and coarse grains of andesite (x_2, x_3). Minimal level of the field (at low filling, about 20% of SiC, and 44% of coarse grains), η_{min} = 18.5, maximum η_{max} = 464 Pa·s (the most filled mixes, with maximal content of silicon carbide and minimum of coarse grains, x_1= x_2 = +1, x_3= −1), the difference equals 445.5 Pa·s, i.e. 25 times. Clearly such great change of viscosity and direction of growth vector are conditioned by decrease of quantities of polymer and coarse grains, resulting in thinner layers of liquid phase.

To demonstrate the abilities of IPA the "plane task" should be considered, so it is appropriate to turn to the local fields of the properties in coordinates of filler composition at medium quantity of the filler, *Y*(x_2, x_3 / x_1= 0). Such fields of η, *R*, and *A* (fig. 2) are described by models (2-4) with x_1= 0. The isoviscous compositions are considered, with η as unvarying parameter. The chosen level η_{is} = 45 Pa·s corresponds to the middle of the range 30 ≤ η ≤ 60 Pa·s (required by technology [19]).

Computational experiment. The trial in computational experiment for IPA is carried out preferably in 2-dimensional region of forming the local field gradient factors (***x*** = $\boldsymbol{x}_{gr}$ = (x_i, x_j), Ω_x = Ω_{gr}), at fixed values of the other factors (components of vector $\boldsymbol{x}_{ch}$, changing the local fields). The trials can be specially realised at certain points in the region Ω_{ch} of changing factors according to certain design of computational experiment. In each trial the information for IPA is "mined" not in the line *B*(***x***) = B_{is} (it is η_{is} = 45 in the example), but in the isoparametric corridor Ω_{is} separated out of Ω_x. The trial consists of the following.

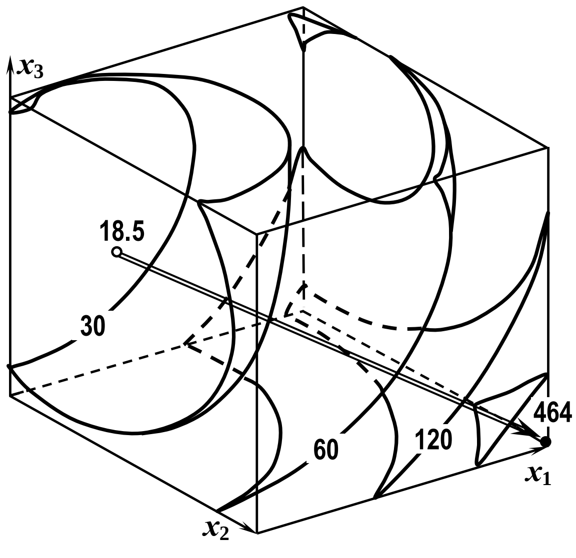

Fig. 1. The field of viscosity of filled carbamide resin

1. The *confidence corridor* (5) is determined, its boundaries accounting (with the risk α at which ES-model has been built) for "fuzziness" of pseudo determinate value of *B* calculated by statistical model. The width of the corridor (of isoparametric region of *B* values) is defined by

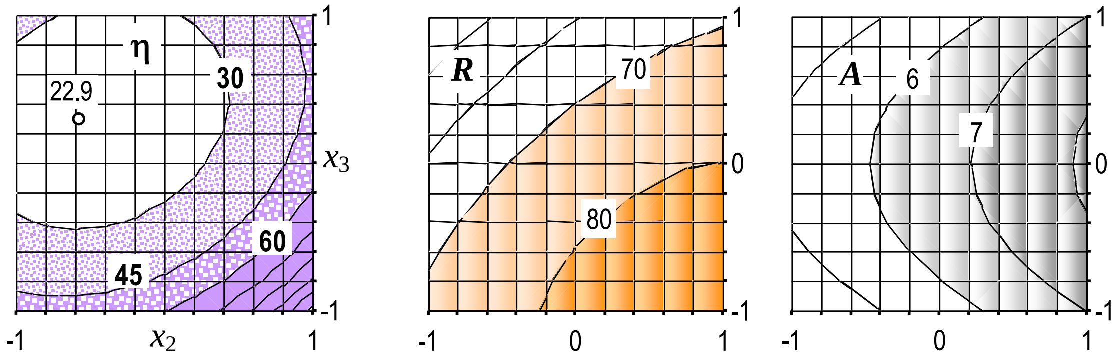

Fig. 2. The influence of filler composition ($SC \sim x_2$, CA $\sim x_3$) on mix viscosity (Pa·s), strength (MPa), and abrasion resistance (h/g) of the composite at medial filling (F = 2.25, x_1 = 0)

error s_e of valuating B and by the quantile of t-distribution and varies in dependence on $\boldsymbol{x}$ through prediction variance function d [8, 17-18]. Sometimes it is admissible to replace $d(\boldsymbol{x})$ with the average over Ω_x value $\bar{d}$ as well as to increase the risk; the confidence corridor can be also widened (with Δ multiplied by coefficient greater than 1).

$$B_{is.\alpha}(\boldsymbol{x}) \;=\; B_{is} \pm t_\alpha \cdot s_e \cdot [d_B(\boldsymbol{x})]^{0.5} \;=\; B_{is} \pm \Delta B(\boldsymbol{x}) \tag{5}$$

2. In 2-factor region Ω_x (it is square $\{x_2, x_3\}$ in the example demonstrated below) n uniformly distributed points are generated (1600 compositions of the filler in the example, the first involvement of MCM). Those n_{is} points $\boldsymbol{x}_{is}$ out of n remains to take part in the analysis at which the values of B calculated by ES-model $B(\boldsymbol{x})$ fall into the corridor $B_{is} \pm \Delta B_{is}(\boldsymbol{x})$. Thus the upper and lower boundaries $B_{is.\alpha}(\boldsymbol{x})$ of the confidence corridor (5) define the *isoparametric corridor* Ω_{is}, $\boldsymbol{x}_{is} \in \Omega_{is} \subset \Omega_x$. In the example bellow 123 out of 1600 generated points have remained in IP-corridor.

3. The data on the levels of the fields of other criteria Y in IP-conditions, unavailable in real experiment, are simulated by estimates (6) of the level of random field [17-18] at any u-th of n_{is} points. Added to values of Y calculated by ES-models is normally distributed prediction error (the second involvement of MCM); generated for each point values of t distributed by standard normal law (i.e. normal $t \cdot s_e$) are used.

$$Y_u = Y(\boldsymbol{x}_{is.u}) \pm t_u \cdot s_e \cdot [d\,(\boldsymbol{x}_{is.u})]^{0.5} \tag{6}$$

If just the tendency in changes of Y at constant B is important, the estimates $Y(\boldsymbol{x}_{is})$ of average levels of Y at points $\boldsymbol{x}_{is}$ (merely calculated by the model) are acceptable.

These operations (par. 1-3) results in the "corridors" of 3 types (by parameter the values of which they contain):

"1" – with n_{is} levels of B in confidence limits (5);

"2" – IP-corridor Ω_{is} with values of factors that conditioned the levels of B in "1";

"3" – with n_{is} estimates of other Y by (6), at ($x_{i.is}$, $x_{j.is}$) from "2", forming the corridors of these properties under IP-conditions; in the example these are strength and abrasion resistance of 123 isoviscous compositions (with η in the range 45 ± Δη($\boldsymbol{x}_{is}$) Pa·s).

4. To visualise the results of above-described statistical trial the obtained corridors (Ω_{is} and those of the values of B and Y) are "unfolded". This may be realised in two ways.

The first one (focal) is along the line $B(\boldsymbol{x}) = B_{is}$, with the uniform angular displacement φ of the vector of both factors directed to this line from some focus. The choice of the focus must provide the best view of the whole corridor. Fig. 3 shows 2 examples of the focuses: the point of minimal level of the local viscosity field η (x_2, x_3 / x_1= 0) and the vertex (–1, +1) of the square

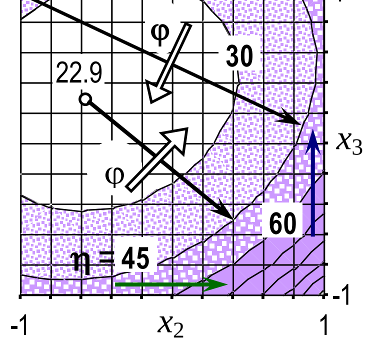

Fig. 3. Directions of viewing IP-corridor along the line η=45 and along factor axes

{x_2, x_3}; in latter case the angle of view (π / 2) is in one quadrant.

The second simpler way is to unfold the corridor along one of the factor axes (fig. 3). It could be that of the factor through changes of which both B and other Y would be controlled or the one with longer projection of the isoline $B(x_i, x_j) = B_{is}$.

Displayed in the unfolded corridor are the levels of the properties and providing them values of the factors.

Such "corridor" plots for medium-filled (x_1 = 0, F = 2.25) isoviscous (η_{is} = 45 Pa·s) binders when viewing from the point (x_2 = –1, x_3 = +1) are presented in fig. 4, the abscissa being clockwise angle (as shown in fig. 3).

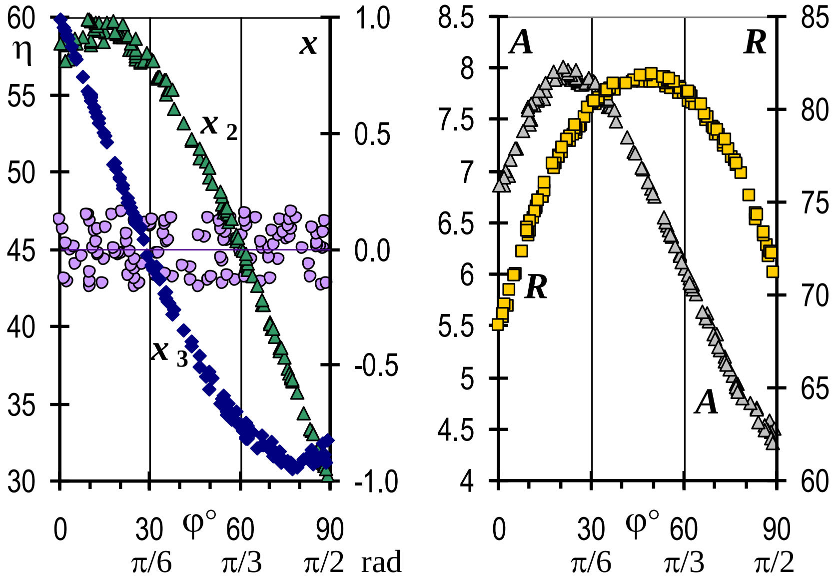

Fig. 4. Viscosity, filler composition (x_2, x_3), abrasion resistance and strength of isoviscous carbamide binders in IP-corridor (focal viewing)

The displays in fig. 5 are unfolded along the axes of the factors.

The values of Y in IP-corridor present the local field $Y(\mathbf{x} / B(\mathbf{x}) = B_{is} \pm \Delta B_{is}(\mathbf{x}))$, with criterion borders [17] defining its domain Ω_{is}. Such are the fields: $A(x_2, x_3 / x_1 = 0, \eta = 45 \pm \Delta\eta_{is}(x_2, x_3 / x_1 = 0))$ and $R(x_2, x_3 / x_1 = 0, \eta = 45 \pm \Delta\eta_{is}(x_2, x_3 / x_1 = 0))$. The "corridor plots" in fig. 4-5 representing them are obtained by "overlapping" Ω_{is} on the square domain of random fields $A(x_2, x_3 / x_1 = 0)$ and $R(x_2, x_3 / x_1 = 0)$.

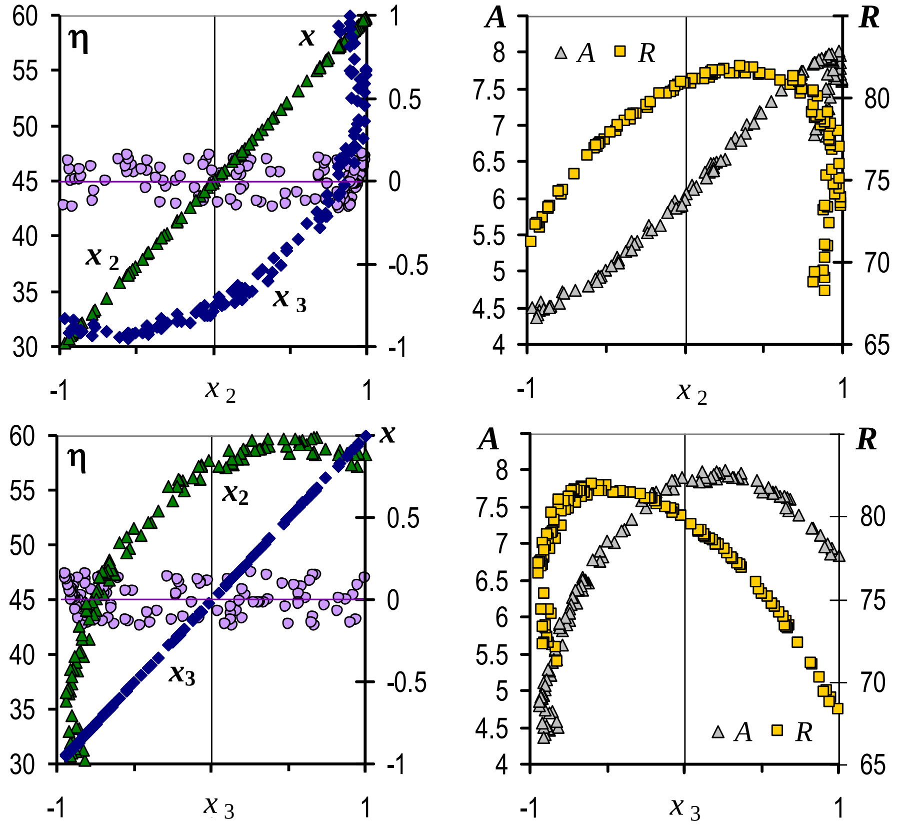

Fig. 5. Viscosity, filler composition (x_2, x_3), abrasion resistance and strength of isoviscous binders versus contents of silicon carbide (x_2) and coarse grains of andesite (x_3)

5. The specific features of CP-corridors, as of any CP-fields, can be expressed with generalising indices G [10, 17] – maximal and minimal levels of the property in Ω_{is}, the differences, and other numerical characteristics $G_Y\{\Omega_{is}\}$, allowing to evaluate the variation of Y in isoparametric conditions. The estimates of G (and their distributions) can be obtained as results of multiple trials or using "carrying" function, describing changes of Y along the axis of its corridor.

In particular, equation (7) describes (with determination coefficient 0.999, φ in radians) the trend line in the corridor of abrasion resistance of isoviscous compositions (fig. 4). The minimal and maximal levels by (7) are: $A_{max}\{\eta_{is} = 45\} = 8.0$, $A_{min}\{\eta_{is} = 45\} = 4.4$ h/g (the increment along the line of required viscosity is 1.8 times). The maximum is provided in zone of φ = 0.33-0.43 (x_2 in the range 0.86-0.93, about 57% of SiC in the filler, x_3 between 0.16 and 0.35, 46% of coarse andesite). The minimum is located in direction of φ = π/2 (from vertex «–1, +1» in fig. 3), i.e. at the absence of silicon carbide and 20% of coarse grains.

$$A\{\eta_{is} = 45\} = 6.73 + 6.61\varphi - 10.30\varphi^2 + 3.12\varphi^3 + 0.10\varphi^4 \qquad (7)$$

6. Isoparametric trials at various fixed values of factors x_{ch} (which would change CP-corridors of the properties), according to certain design of computational experiment in the region Ω_{ch}, produce the estimates of G for each fixed $\boldsymbol{x}_{ch}$. The secondary models built on these estimates would present the next level of generalisation in relation to primary natural data (the results of designed natural experiment), on which the primary ES-models, (2-4) for instance, are built.

The secondary model (8) describes the influence of degree of filling ($F \sim x_1$) on maximal level of abrasion resistance that could be achieved through composition of the filler ($SC \sim x_2$, $CA \sim x_3$) under condition of specified viscosity of the mix, η = 45 ± 2.5 Pa·s (fig. 6). This equation fits the results of the trials at x_1 = –1, –0.5, 0, +0.5. At x_1= +1 (F = 2.5, the most filled binders) there are no compositions of the filler that would provide the viscosity indicated; in this section of the cube in fig. 1 the acceptable region (by required η), the corridor Ω_{is}, does not exist.

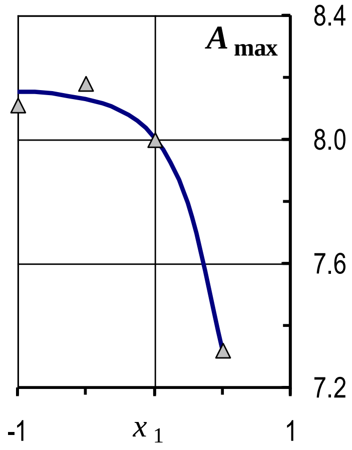

Fig. 6. Maximal abrasion resistance of isoviscous binders in dependence on degree of filling

$$A_{max}\{\eta(x_2, x_3) = 45\pm2.5\} = 0.1578\cdot(51.734 - \exp(3.3504x_1)) \qquad (8)$$

Conclusions. The information of this kind can be extracted from the data of natural experiment, which has been designed, only with the help of the virtual experiments. Used in these computational experiments are ES-models describing the composition-process fields of the properties of composite materials and Monte Carlo method. As a tool of computational materials science and a part of CP-fields methodology isoparametric analysis gives a possibility to obtain new knowledge that could be helpful for choosing reasonable technological solutions.

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