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OPTIMIZATION OF PARTICLE SIZE DISTRIBUTION IN CERAMIC TILE PRODUCTION

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A mathematical model was developed by correlating the experimental values of tile characteristics with the mass fractions of raw mixture components, whose particle size distribution was known and supplemented with the general constraints (such as the mass balance equation and the conditions of non-negativity). A typical problem was solved by a combined analytical-numerical procedure and by the geometric programming method.

The results of the two methods were compared and those of the latter were found to be preferable.

INTRODUCTION

The quality of both green and fired floor tiles depends considerably on particle size distribution in the raw material mix, which is usually determined by the characteristics of the mill. Consequently, an efficient procedure for optimum composing of the raw mix fractions may be of interest for improving the properties of floor tiles.

In order to solve the problem mentioned above, the present authors applied the well-known mixture design method to the components having equal chemical, but different granulometric composition.

Factorial granulometric design was first introduced into the field of ceramics by P. R. Jones and collaborators [1—4]. However, the method has not found wider use in material science and industry. As far as the mix design is concerned, few practical studies in ceramics have appeared lately [6—12], apart from the very good theoretical literature [5].

To correlate some product characteristics with particular grain size fractions of the raw mix, Sheefe's Simplex Latex design [13] was selected in this work. As the experiments were symmetrically distributed, the coefficient of regression functions could be determined by means of explicit relations. The equations obtained were used as objective functions, the extremes of which were found by two different techniques selected from the many possible methods and their variants [14—16], namely

— the combined analytical-numerical method (based on a non-linear mathematical model, developed from partial derivatives and solved by an original numerical procedure), and

— the geometric programming method.

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Table I

Mean values of green and fired tile properties

Experiment No.	Raw mix composition			Green tile			Fired tile		
	x_1	x_2	x_3	Bending strength MPa	Density kg/m³	Residual moisture content %	Bending strength MPa	Density kg/m³	Water absorptivity %
1	1	0	0	1.03	1934.67	6.51	40.96	2364.33	1.30
2	0	1	0	0.96	1952.00	5.98	46.24	2357.00	1.14
3	0	0	1	1.16	1936.33	5.37	51.01	2378.00	1.14
4	0.5	0.5	0	1.07	1981.67	6.29	45.36	2325.00	1.17
5	0.5	0	0.5	0.79	2020.33	5.99	50.42	2340.33	1.29
6	0	0.5	0.5	0.80	2002.00	5.50	44.40	2324.67	1.08
7	0.75	0.25	0	1.14	2040.00	6.37	48.33	2348.00	1.14
8	0.25	0.75	0	1.21	2023.33	6.32	47.80	2361.67	1.04
9	0.75	0	0.25	1.47	2013.33	6.34	46.16	2377.33	1.37
10	0.25	0	0.75	0.94	1980.00	5.34	43.40	2379.67	1.00
11	0	0.75	0.25	1.69	1967.67	5.61	50.61	2341.67	1.47
12	0	0.25	0.75	1.92	1967.33	5.23	44.51	2364.67	1.29
13	0.5	0.25	0.25	1.32	2018.67	5.76	45.24	2399.67	1.20
14	0.25	0.5	0.25	1.20	2019.33	5.77	42.27	2338.33	1.31
15	0.25	0.25	0.5	1.24	1992.67	5.55	38.58	2377.33	1.18

CORRELATION BETWEEN PARTICLE SIZE DISTRIBUTION OF THE RAW MIX AND THE PROPERTIES OF TILES

In the production of floor tiles pursued in the tile factory where the experimental work has been done, the particle size distribution was determined by means of seven fractions (i.e. the oversizes on sieves, 0.5 mm, 0.4 mm, 0.315 mm, 0.2 mm,

0.16 mm and 0.08 mm). In order to investigate the effect of particle size distribution on the quality of tiles, the whole size range was simulated by means of three fractions: fine (particles smaller than 0.16 mm), medium (particles between 0.16 mm and 0.4 mm) and coarse (particles larger than 0.4 mm).

In accordance with the design matrix for the (3,4) lattice [13] (Table I), fifteen different combinations of particle size distribution in the raw mix were compared and used for making floor tile samples and measuring their properties, i.e. bending strength, density and residual moisture content of green tiles, as well as bending strength, density and water absorptivity of fired tiles. The mean values of experimentally obtained results are listed in Table I.

The regressive equations for each single quality parameter Y (strength, density etc.) as a function of fractions x_1, x_2 and x_3 (coarse, medium and fine respectively) have been used in the form of fourth-degree polynomials:

$$Y = b_1x_1 + b_2x_2 + b_3x_3 + b_4x_1x_2 + b_5x_1x_3 + b_6x_2x_3 + b_7x_1x_2(x_1 - x_2) + \\ + b_8x_1x_3(x_1 - x_3) + b_9x_2x_3(x_2 - x_3) + b_{10}x_1x_2(x_1 - x_2)^2 + b_{11}x_1x_3(x_1 - x_2)^2 + \\ + b_{12}x_2x_3(x_2 - x_3)^2 + b_{13}x_1^2x_2x_3 + b_{14}x_1x_2^2x_3 + b_{15}x_1x_2x_3^2, \qquad (1)$$

where $b_i (i = 1,15)$ represents the coefficients to be determined from the explicit equations [13]. The results of calculating the b coefficients for the relevant Y-functions are given in Table II. Using adequacy testing it was proved that the second-degree equation (consisting of six terms) could be used for the subsequent bending strength determination. However, due to the relatively high replication variance as well as the t-value for point 15, equation (1) was adopted in its complete form. Regressive equations were expected to be the basis of a mathematical model of the optimization problem.

Table II

Coefficients be in regressive equations (1) for investigated quality parameters Y

Coef. No.	Green tile			Fired tile		
	Bending strength Y_1	Density Y_2	Residual water content Y_3	Bending strength Y_4	Density Y_5	Water absorptivity Y_6
1	1.03	1934.67	6.51	40.96	2364.33	1.30
2	0.96	1952.00	5.98	46.24	2357.00	1.14
3	1.16	1936.33	5.37	51.01	2378.00	1.14
4	0.28	153.33	0.17	7.02	—142.66	—0.20
5	—1.22	339.33	0.20	17.72	—123.33	0.26
6	—1.04	231.33	—0.68	—16.83	—171.33	—0.23
7	—0.55	135.11	—1.13	16.88	—92.44	0.10
8	3.14	182.22	2.26	—11.84	23.99	1.57
9	4.11	—40.00	0.36	45.27	—66.66	0.93
10	2.78	1271.11	1.47	67.11	446.22	—2.00
11	7.21	—52.44	—2.88	10.06	649.77	—1.82
12	10.46	—427.55	—2.72	44.82	379.55	6.08
13	9.20	—1087.12	—26.24	225.84	4976.91	—8.56
14	—16.50	1652.45	—2.26	—173.62	—2361.78	9.87
15	36.37	—1555.56	9.08	—579.73	1332.43	6.02

CALCULATION OF EXTREME VALUES

To obtain the optimum values of fractions x_1, x_2 and x_3, one has to calculate the extremes of function (1) for each measured characteristic, i.e. the maxima of the functions Y_1 (green tile strength), Y_2 (green tile density), Y_4 (fired tile strength), Y_5 (fired tile density) as well as the minima of the functions Y_3 (residual moisture content in green tile) and Y_6 (water absorptivity of fired tile).

A review of a number of optimization methods provided the opportunity for selecting two of them. The first involves an analytical approach based on partial differentiation of function (1), and the other is one of the modern numerical methods. The authors decided to compare the two quite different techniques and to assess the efficiency of their application in ceramic materials science and practice.

The analytical-numerical method

The first method of interest is based on the non-linear algebraic system, obtained by partial differentiation of the modified polynomial (1), respected to feasible variables $x_1 - x_3$ and artificial variables $x_4 - x_{10}$. Namely, before starting the optimizing procedure it is necessary to complement the objective function (1) with the respective constraints, namely the mass balance equation

$$\sum_{i=1}^{3} x_i - 1 = 0 \tag{2}$$

and the condition of non-negativity:

$$x_i + x_{i+3}^2 = 0 \qquad (\text{for } i = 1, \ldots, 3) \tag{3}$$

in the way proposed by Lagrange [14].
After rearrangement one obtains the modified optimization criterion:

$$F = Y + x_7\Big(\sum_{i=1}^{3} x_i - 1\Big) + \sum_{i=1}^{3} x_{i+7}(x_i + x_{i+3}^2) \tag{4}$$

which can always be readily differentiated because of its polynomial type.

A general form of the mathematical model can be expressed as follows:

$$\frac{\partial F}{\partial x_1} = b_1 + b_4x_2 + b_5x_3 + + b_7x_2(2x_1 - x_2) + b_8x_3(2x_1 - x_3) + \\ + b_{10}x_2(x_2^2 - x_1^2) + b_{11}x_3(x_3^2 - x_1^2) + 2b_{13}x_1x_2x_3 + b_{14}x_2^2x_3 + b_{15}x_2x_3^2 + \\ + x_7 + x_8 = 0. \tag{5}$$

$$\frac{\partial F}{\partial x_2} = b_2 + b_4x_1 + b_6x_3 + b_7x_1(x_1 - 2x_2) + b_9x_3(x_2 - 2x_3) + \\ + b_{10}x_1(x_1 - x_3)(x_1 - 3x_3) + b_{12}x_2(x_2 - x_3)(x_2 - 3x_3) + \\ + b_3x_1^2x_2 + b_{14}x_1x_2^2 + 2b_{15}x_1x_2x_3 + x_7 + x_{10}. \tag{6}$$

$$\frac{\partial F}{\partial x_3} = b_3 + b_5x_1 + b_6x_2 + b_8x_1(x_1 - 2x_3) + b_9x_2(x_2 - 2x_3) + \\ + b_{11}x_1(x_1 - x_3)(x_1 - 3x_3) + b_{12}x_2(x_2 - x_3)(x_2 - 3x_3) + b_{13}x_1^2x_2 + \\ + b_{14}x_1x_2^2 + 2b_{15}x_1x_2x_3 + x_7 + x_{10} = 0. \tag{7}$$

$$\frac{\partial F}{\partial x_4} = 2x_4x_8 = 0, \quad \frac{\partial F}{\partial x_5} = 2x_5x_9 = 0, \quad \frac{\partial F}{\partial x_6} = 2x_6x_{10} = 0. \qquad (8)\text{-}(10)$$

$$\frac{\partial F}{\partial x_7} = x_1x_2 + x_3 - 1 = 0, \quad \frac{\partial F}{\partial x_8} = x_1 + x_4^2. \qquad (11)\text{-}(12)$$

$$\frac{\partial F}{\partial x_9} = x_2 + x_5^2 = 0, \quad \frac{\partial F}{\partial x_{10}} = x_3 + x_6^2. \qquad (13)\text{-}(14)$$

In this way the problem of optimum granulometric design of the raw mixture for both green and fired floor tiles is oriented at determining the $x_1 - x_{10}$ values by solving the (5)-(14) system. This appeared to be a difficult task. The authors decided to resolve it by applying the original numerical method called "Ridgepath". It is discussed in more detail elsewhere [17].

The geometric programming method

The second method is known as the geometric programming method. Although it was published twenty years ago, it has been continuously corrected and improved ever since and is generally regarded to be very efficient. The general principle of using Legendre's transformation, which constitutes the basis of geometric programming, can be found in many books and papers [18, 19]. In our investigation we used the most advanced standardized version of the numerical procedure, considering only the positive values of the variables.

The mathematical model contains two equations; one defines the property being measured as a function of mass fractions, polynomial (1), and the other represents the mass balance equation (2). From the previous comments it is obvious that the polynomial takes the place of an objective function, while the mass balance constrains the variable values.

The results of both methods

Analysis of the calculated values of fractions reveals some interesting facts.

One of the partial solutions provided by the analytical-numerical production:

$$x_1^c = 0.48,\ x_2^c = 0.34,\ x_3^c = 0.18 \text{ (calculated values)}$$

$$x_1^p = 0.46,\ x_2^p = 0.40,\ x_3^p = 0.14 \text{ (current production)}$$

Unfortunately, this combination of components gives low values of the bending strength of fired tile. The observation is in agreement with the experimental data.

According to the values of mass fractions conforming to the theory of packing, the local maximum of the bending strength (49 MPa) was:

$$x_1^c = 0.705 \cong 0.7, \quad x_2^c = 0.199 \approx 0.2, \quad x_3^c = 0.095 \approx 0.1$$

This case is represented by point 7 in Table I.

The composition providing the global maximum (53.5 MPa) was obtained by geometric programming. It can be expressed as

$$x_1^d = 0.213, \quad x_2^c = 4.92\ 10^{-7}, \quad x_3^c = 0.786$$

In fact, this solution represents the two-fraction (binary) system and can be related to the composition at point 10 (Table I). There is an explanation for the calculated quantities, especially for the high amount of the third component. Namely, an analysis of the effect the "pure" fractions have on bending strength (points 1, 2 and 3 in Table I) shows a positive influence on both final products. This can be explained by the fact that particles smaller than 0.16 mm are not uniform in size.

The optimization procedures were also employed in determining the other qualitative parameters. For example, water absorptivity Y_6 achieved its minimum at the point

$$x_1^c = 0.79, \quad x_2^c = 0.18, \quad x_3^c = 0.02$$

which also gives the optimum bending strength.

An analysis of selected numerical results allowed some general remarks about the efficiency of the methods to be made. Experience gained in solving a number of diverse problems shows that the geometric programming method exhibits a very high level of reliability, efficiency and accuracy. Using geometric programming, it is possible to carry out multicriterial optimization (for example, determination of maximum bending strength while keeping the water absorptivity at a required level).

In the present case its application was not necessary because all the experimental values for water absorptivity were well below the allowable limit.

CONCLUSION

The purpose of this paper was to clarify how the particle distribution affects the quality of the final product and to suggest an efficient method for determining the optimum composition of the raw mix. However, in spite of correlations of experimental results, it has to be pointed out that the variations in fifteen sample properties occur not only because of the differences in particle size distribution, but also as a result of other factors. The "noise" of the experiment cannot be regarded as insignificant.

Recent investigation results [20] have indicated an interesting aspect. They showed that the particular fractions had different mineralogical compositions, which is of essential significance for the properties of the fired product. The procedure described may be useful for analysing the most complex relationships between particle size distribution and the quality of floor tiles. It can be applied either to examining the relations between the coarse, medium and fine fractions, or to investigating the mixtures of all seven fractions from every-day tile production. Finally, on the basis of the acquired experience it is possible to design the experiments in local phase-diagram regions [5].

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OPTIMALIZACE ZRNITOSTI PŘI VÝROBĚ KERAMICKÝCH DLAŽDIC

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Práce se zabývá matematickou optimalizací zrnitostní křivky surovinové směsi, zaručující dosažení požadovaných kvalitativních ukazatelů vyráběných keramických dlaždic. Byl vypracován matematický model korelace mezi experimentálně zjištěnými kvalitativními charakteristikami výrobku a hmotnostními podíly jednotlivých frakcí složek surovinové směsi a doplněn obecnými omezujícími podmínkami, jako rovnicí hmotnostní bilance a podmínkami nezápornosti. Je řešen typický problém kombinovaným analyticko-numerickým postupem a metodou geometrického programování. Ke konečnému řešení se dospělo porovnáním výsledků obou metod, které vyšlo ve prospěch metody druhé.

ОПТИМИЗАЦИЯ ГРАНУЛОМЕТРИЧЕСКОГО СОСТАВА ПРИ ПРОИЗВОДСТВЕ КЕРАМИЧЕСКИХ ПЛИТОК

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В предлагаемой работе рассматривается оптимизация кривой гранулометрического состава сырьевой смеси, обеспечивающей получение требуемых качественных показателей производимых керамических плиток. Была разработана математическая модель корреляции между установленным экспериментальным путем качественными характери-

стиками изделия и весовыми долями отдельных фракций компонентов сырьевой смеси к которой добавили общие лимитирующие условия в виде уравнения весового баланса, и условия неотрицательности. Решается типичная проблема при помощи комбинированого аналитико-нумерического приема и метода геометрического программироавния. Окончательное решение было получено сопоставлением обоих методов и второй метод оказался более пригодным.

ALEXANDER MUCK: SYMETRIE KRYSTALŮ A VIBRAČNÍ SPEKTRA. 266 str., 26 obr., 60 tab., SNTL, Praha 1987

Kniha sa zaoberá teóriou grúp vo vzťahu k fyzikálnym vlastnostiam molekúl a predovšetkým kryštálov. Je rozčlenená do 4 častí. V úvode je veľmi elegantne a originálne popísaný výskyt a význam symetrie v prírode a história náuky o symetrii vrcholiaca v ucelenej teórii grúp. Samotná teória grúp tvorí náplň druhej kapitoly. Po abstraktnej teórii grúp, včítane teórie reprezentácií, autor popisuje detailne bodové grupy symetrie a korelácie medzi nimi. Oceňujem, že autor sa zmieňuje i o permutačne — inverzných grupách, ktoré hrajú významnú úlohu v molekulovej spektroskopii nerigidných molekúl. Ďalej sa popisujú priestorové grupy symetrie.

Tretia kapitola tvorí spojovací článok medzi abstraktnou teóriou grúp a symetriou fyzikálnych vlastností. Všeobecne platné vzťahy medzi symetriou kryštálu a symetriou jeho fyzikálnych vlastností sú aplikované na sériu efektov (piezoelektrický, pyroelektrický, feroelektrický, optická aktivita). Autor sa nevyhýba ani komplikovanejším systémom (rotácia častíc v kryštáloch, reálne kryštály, OD štruktúry, kvázikryštály).

Posledná, štvrtá kapitola je venovaná vibračným spektrám. Popisujú sa princípy teórie vibračno-rotačných spektier, symetria normálnych vibrácií častice a kryštálu a výberové pravidlá, ako aj vplyv skupenských stavov na vibračné spektrá. Ďalšia časť tejto kapitoly popisuje site — grupovú, faktor — grupovú a korelačnú analýzu s konkrétnymi aplikáciami. Kniha je doplnená obsiahlymi dodatkami (spolu 54 strán).

Pri čítaní knihy, čitateľ ocení logičnosť a jasnosť výkladu, ale mňa osobne si táto kniha získala najmä neobvyklým informačným bohatstvom a úplnosťou. Napríklad autor uvádza kompletné informácie o všetkých bodových grupách a nielen o tých, ktoré sám považuje za „dôležité". U všetkých grúp autor dôsledne uvádza niekoľko príkladov zlúčenín, ktoré k nim patria. Myslím si, že mimoriadne užitočné sú i poučky a vety (napr. teorémy o produktoch operácií symetrie, na str. 40, 100, alebo explicitný tvar tenzorov polarizovateľnosti v Tab. 60, a mnoho iných), ktoré som si z nedostatku lepšieho názvu pomenoval „malé vety". Vo „veľkých knihách" teórie grúp sa tieto „malé vety" obvykle neuvádzajú, pretože sa dajú odvodiť zo základných viet. Odvodenie je však často časovo náročné a je preto príjemné mať k dispozícii hotové výsledky. Pozitívne hodnotím ten fakt, že na rozdiel od niektorých kníh s ktorými som sa stretol, autor neredukuje analýzu vibračných spektier kryštálov len na analýzu diamantu a NaCl.

Je len pochopiteľné, že v tak relatívne útlej knihe musí byť bohatstvo informácií zaplatené úsporným, telegrafickým štýlom výkladu, ktorý hlavne v časti o vibračných spektrách často predpokladá, že nejde o prvé stretnutie čitateľa s problematikou. Napr. definícia normálnych vibrácií na str. 163 je síce vecne úplne správna, ale čitateľ, ktorý sa s týmto pojmom stretne prvýkrát by si asi ťažko vytváral z popisu jasný obraz. Domnievam sa, že pri budúcom vydaní knihy by bolo k časti o vibračných spektrách užitočné pridať 10—20 strán venovaných popisu silových konštánt, dynamickej matice, súradniciam symetrie, a pod.

V kapitole venovanej abstraktnej teórii grúp, teóriu reprezentácií by bolo vhodné doplniť o veľmi užitočnú Schurovu lemmu a kapitolu venovanú symetrii o symetriu vzhľadom k obráteniu času.

V knihe sa vyskytli niektoré drobné nepresnosti: Zriedené plyny nemajú čiarové spektrá (str. 165), pretože konečná doba života a Dopplerov efekt spôsobujú rozšírenie línií. Vzhľadom k definíciám (2.54), (4.11) by asi malo byť v rov. (4—10) správne exp $[i(qt^h)/2\pi]$. Na strane 170 by bolo správnejšie hovoriť o symetrii vyššieho vibračného stavu než o symetrii frekvencie. Výberové pravidlo uvedené na str. 173 pod rovnicou (4—27) platí len vtedy, ak nižšia hladina je totálne symetrická.

Záver: kniha je mimoriadne užitočným a bohatým zdrojom informácií a môže slúžiť ako príručka pre všetkých, ktorí používajú teóriu grúp, hlavne však v oblasti spektroskopie, štruktúrnej analýzy a fyziky tuhých látok.

Sarka