CHEMISTRY JOURNAL OF MOLDOVA. General, Industrial and Ecological Chemistry. 2021, 16(1), 79-87
ISSN (p) 1857-1727 ISSN (e) 2345-1688
http://cjm.asm.md
http://dx.doi.org/10.19261/cjm.2021.779

A MODIFIED SCHEFFE'S SIMPLEX LATTICE DESIGN METHOD IN DEVELOPMENT OF CERAMIC CARRIERS FOR CATALYTIC NEUTRALIZERS OF GAS EMISSIONS

Valery Ved, Hanna Ponomarenko *, Yevhenia Ponomarenko, Konstantin Gorbunov

Department of Chemical Engineering, National Technical University "Kharkiv Polytechnic Institute"
2, Kirpichova str., Kharkiv 61002, Ukraine
**e-mail: khpannap@gmail.com; phone :(+380 67) 91 02 917*

Abstract. A modified Scheffe's simplex lattice design method is proposed to study the properties of multicomponent materials. Usually, the Scheffe's simplex lattice method is used to describe three-component systems in design of chemical experiments. This modified Scheffe's method allowed determining the optimal compositions of cordierite and corundum based ceramic materials that are used as catalyst carrier for gas purification equipment. The obtained material (0.63-1.25 mm weight fraction of cordierite of 0.35 mass% fraction; <0.63 mm weight fraction of cordierite of 0.35 mass% fraction; <0.06 mm weight fraction of corundum of 0.2 mass% fraction; 1.25-2.5 mm weight fraction of cordierite of 0.2 mass% fraction) was used successfully for the manufacturing of catalytic neutralizers of gas emissions. This method was essential for the designing and manufacturing of a catalytic neutralizer for waste recycling complex at the Kharkiv - Passenger railway station in Ukraine.

Keywords: Scheffe's simplex lattice design method, ceramic carrier, catalytic converter, gas emission.

Received: 18 January 2021/ Revised final: 01 March 2021/ Accepted: 06 March 2021

Introduction

Environment protection against harmful gas emissions of industrial enterprises and transport is interconnected with related to a number of environmental problems requiring solutions. Worldwide experience in gas emission treatment proved that catalytic gas purification is the most effective method. The components of the catalytic converter unit are the body, the catalyst carrier and the catalytic compound. The efficiency of the converter depends on the catalytically active elements covering the carrier surface. The composition of the catalyst determines the kinetics of the process of gas emissions catalytic destruction [1]. The process in the diffusion area is determined by the structure and properties of the carrier [2].

Suitable materials for ceramic-based carrier must have high heat resistance and mechanical strength. Given hydrodynamic conditions must be provided in the catalytic converter. Due to a wide range of variations of the studied mixtures compositions, it is desirable to use the design of experiments. This procedure helps to select the number and conditions of experiments which are necessary and sufficient to solve the problem with the required accuracy. Experimental design procedures make possible the simultaneous variation of a significant number of factors, obtaining a quantitative estimation of the main parameters and their interaction, which ultimately lead to the increase of experimental efficiency [4]. The methods of optimization of experimental design allow the use of mathematical calculations not only at the stage of processing the measurement results but also at the stage of the experiment preparation and during carrying out. The application of the experimental design method based on simplex lattices is the most suitable to study the properties of mixtures with the simultaneous use of several components in the composition of the mixture.

Scheffe's simplex lattice design method was developed for experiments involving mixtures, with the purpose of predicting empirically the response to any mixture of the components, when the response depends only on the proportion of the components and not on the total amount [5]. It is the most optimal method of experiments design with simultaneous use of several components that can be used to study the properties of mixtures. In the selected method, the response function – property (*y*) is represented by projections of equidistant dependencies (equal value curves on the triangle plane). The simplex lattice for experiments with three variable components is shown in Figure 1.

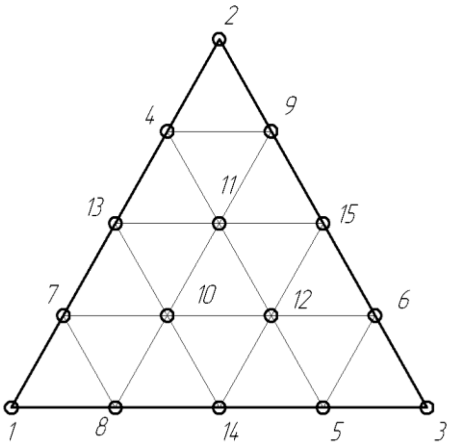

Figure 1. Spatial pattern of a simplex lattice.

The Scheffe's simplex lattice design method has been applied successfully by many scientists in the fields of biology [6], fuel energy [7], food chemistry [8], industrial chemistry [9], material science [10] and many others. Such experimental designs as the McLean - Anderson plan [11], *D*-optimal plans of Kiefer [12], Draper and Lawrence plans [13] are developed on the basis of the simplex lattices. Despite the wide use of the simplex lattice method in mathematical design, it is quite difficult to describe the simultaneous interaction of multicomponent systems and to use them for a graphical representation of dependencies of the obtained properties.

Therefore, a modification of the Scheffe's simplex lattice design is proposed for compiling simplex for four-component systems. This simplex is a tetrahedron, with each vertex corresponding to components taken at 100% content. The edge of this tetrahedron is a two-component system, and the face is a three-component system. The points inside the tetrahedron correspond to four-component systems. It is quite convenient to represent such system graphically in a form of sections of a three-dimensional simplex with plane perpendicular to one of its axes. The composition of the four-component mixtures lying in the section plane is determined by the four-dimensional simplex. This approach allows representing the change in the properties of the system in the form of contour curves. Moreover, only three components can vary in one section. The combination of the faces of elementary concentration simplexes are matched taking into account the fact that the properties are a continuous function of the concentrations and do not have derivatives gaps in the regions adjacent to the faces.

This research aims to develop the Scheffe's simplex lattice design method that allows determining the optimal compositions of cordierite and corundum based ceramic materials that are used as catalyst carriers for gas purification equipment.

Experimental

Materials

Cordierite is used in the manufacturing of carrier for gas purification catalysts because of its high heat resistance and the possibility of variation of physical and mechanical properties [14]; whilst the α-Al_2O_3 is used for high compressive resistance of ceramic carrier [15]. Therefore, the experimental samples of ceramic materials for carriers of catalytically active compounds have been made from a mixture that consists of weight fraction of cordierite: <0.63 mm, 1.25-0.63 mm, 2.5-1.25 mm; and of corundum <0.06 mm; these were provided by the Ukrainian Research Institute of Refractories named after A.S. Berezhnoy (Kharkiv, Ukraine). The chemical composition of the corundum and cordierite is presented in Table 1.

Graphical representations of the obtained results were done using the TRIANGLE software.

The studies were conducted at the Chemical Engineering Department of the National Technical University Kharkiv Polytechnic Institute (Ukraine).

Ceramic experimental samples preparation (General procedure)

An aluminophosphate binder with molar ratio P_2O_5:Al_2O_3= 4.08 was introduced into all studied compositions in an amount of 10 mass% over 100 mass% dry matter. These samples were put into 20x20x20 mm block shapes that were vibrated for 170 sec. on a laboratory vibrotol VT-55. The obtained samples were heated at 400°C for two hours in a SNOL–8.2/1100 muffle furnace after being dried for one hour at 100°C in a SNOL–67/350 laboratory oven.

Table 1

Chemical composition of the ceramic materials, mass% fraction.

Material	*SiO_2*	*Al_2O_3*	*Fe_2O_3*	*CaO*	*MgO*	*Na_2O+K_2O*
Corundum	0.02	99.64	0.02	-	-	0.32
Cordierite	51.4	34.9	-	-	13.7	-

Description of the modified Scheffe's simplex lattice design method

A modified Scheffe's simplex lattice design method was developed, which allows determining the compositions of ceramic materials. The advantage of the modified method consists in the fact that it allows the significant reduction of the number of experimental points (from 60 to 34) describing the properties of the four-component simplex of concentrations. The simplex lattice for four variable components experiments is shown in Figure 2.

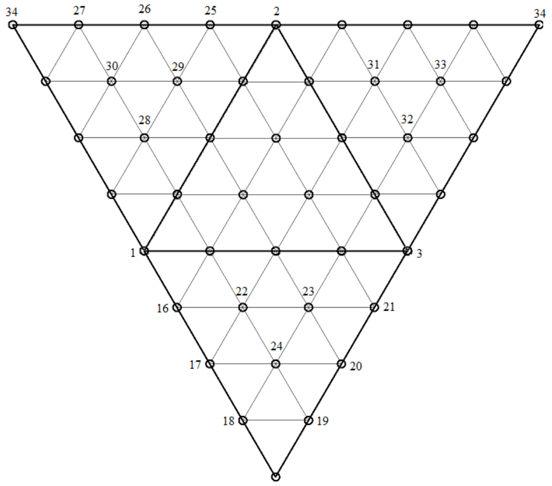

Figure 2. The simplex lattice for experiments with four variable components.

Procedure. Fractional compositions of materials in accordance with the Scheffe's simplex lattice design were expressed as independent variables: x_1 – 1.25-0.63 mm weight fraction of cordierite; x_2 – <0.63 mm weight fraction of cordierite; x_3 – 0.06 mm weight fraction of corundum; x_4 – 2.5-1.25 mm weight fraction of cordierite. The obtained variables were modified in the limits 0.2-0.6 (expressed in fractions). To represent the polyhedron with the given restrictions in the form of a regular simplex, the natural variables (x_i) were converted into coded (z_i) in Eq.(1). Meanwhile, the coded variables z_1, z_2, z_3, z_4, varied from 0 to 1.

$$\sum_{i=1}^{q} z_i = 1;\ z_i \geq 0;\ i = 1,2,3...q \tag{1}$$

The regular simplex of concentrations is constructed with respect to these coded variables. Table 2 shows the coordinates of the experiment plan for the simplex of vertex concentrations at points z_1, z_2 and z_3.

The plan points 15_1, 15_2 and 15_3 in Table 3 are selected as test points to determine the adequacy of the design models for experimental data. Two experiments have been carried out for each point.

The introduction of the fourth variable made it necessary to make plan simplexes with vertices {z_1, z_3, z_4}, {z_1, z_2, z_4} and {z_2, z_3, z_4} according to Figure 2 (Table 3).

Table 2

The coordinates of the experiment plan for the three variable simplex {z_1, z_2, z_3}..

Points on the Figure 2	*Natural variables, weight fraction*				*Coded variables, unit fraction*			
	x_1	x_2	x_3	x_4	z_1	z_2	z_3	z_4
1	0.6	0.2	0.2	0	1	0	0	0
2	0.2	0.6	0.2	0	0	1	0	0
3	0.2	0.2	0.6	0	0	0	1	0
4	0.3	0.5	0.2	0	0.25	0.75	0	0
5	0.3	0.2	0.5	0	0.25	0	0.75	0
6	0.2	0.3	0.5	0	0	0.25	0.75	0
7	0.5	0.3	0.2	0	0.75	0.25	0	0
8	0.5	0.2	0.3	0	0.75	0	0.25	0
9	0.2	0.5	0.3	0	0	0.75	0.25	0
10	0.4	0.3	0.3	0	0.5	0.25	0.25	0
11	0.3	0.4	0.3	0	0.25	0.5	0.25	0
12	0.3	0.3	0.4	0	0.25	0.25	0.5	0
13	0.4	0.4	0.2	0	0.5	0.5	0	0
14	0.4	0.2	0.4	0	0.5	0	0.5	0
15	0.2	0.4	0.4	0	0	0.5	0.5	0
15_1	0.333	0.333	0.333	0	0.333	0.333	0.333	0
15_2	0.26	0.48	0.26	0	0.15	0.70	0.15	0
15_3	0.48	0.28	0.24	0	0.7	0.2	0.1	0

Table 3

The coordinates of the experiment plan for the simplexes with four variables {z_1, z_2 , z_3, z_4}.

Points on the Figure 2	*Natural variables, weight fraction*				*Coded variables, unit fraction*			
	x_1	x_2	x_3	x_4	z_1	z_2	z_3	z_4
16	0.5	0	0.2	0.3	0.75	0	0	0.25
17	0.4	0	0.2	0.4	0.5	0	0	0.5
18	0.3	0	0.2	0.5	0.25	0	0	0.75
19	0.2	0	0.5	0.3	0	0	0.75	0.25
20	0.2	0	0.4	0.4	0	0	0.5	0.5
21	0.2	0	0.3	0.5	0	0	0.25	0.75
22	0.4	0	0.3	0.3	0.5	0	0.25	0.25
23	0.3	0	0.4	0.3	0.25	0	0.5	0.25
24	0.3	0	0.3	0.4	0.25	0	0.25	0.5
25	0.2	0.5	0	0.3	0	0.75	0	0.25
26	0.2	0.4	0	0.4	0	0.5	0	0.5
27	0.2	0.3	0	0.5	0	0.25	0	0.75
28	0.4	0.3	0	0.3	0.5	0.25	0	0.25
29	0.3	0.4	0	0.3	0.25	0.5	0	0.25
30	0.3	0.3	0	0.4	0.25	0.25	0	0.5
31	0	0.4	0.3	0.3	0	0.5	0.25	0.25
32	0	0.3	0.4	0.3	0	0.25	0.5	0.25
33	0	0.3	0.3	0.4	0	0.25	0.25	0.5
34	0	0	0	1	0	0	0	1

Table 4

Compression strength of ceramic experimental samples.

Points on the Figure 2	*The compressive strength, MPa*	*Points on the Figure 2*	*The compressive strength, MPa*	*Points on the Figure 2*	*The compressive strength, MPa*
1	57.7	13	240.4	25	74.1
2	91.5	14	96.6	26	37.8
3	95.7	15	76.0	27	56.6
4	62.7	16	98.9	28	54.9
5	71.2	17	100.4	29	75.2
6	57.5	18	62.5	30	131.2
7	92.4	19	91.5	31	149.6
8	78.0	20	220.7	32	90.6
9	128.1	21	34.1	33	96.7
10	63.0	22	173.2	34	108.7
11	98.9	23	100.6		
12	90.9	24	114.2		

The 34 mixtures with corresponding compositions were made according to the obtained experimental properties values of compression strength (Tables 2 and 3).

In order to obtain catalytic carriers with a sufficiently long service life, the effect of the material composition on the compressive strength of experimental samples was studied, under a press, using a hydraulic hand-held PGR–20 WTC press (Table 4). The load onto the sample was increased with steady speed, leading to samples destruction in 20-60 seconds.

Results and discussion

The proposed modification of Scheffe's simplex lattice design method

The proposed modified Scheffe's simplex lattice design method made it possible to obtain fourth-order polynomial dependencies for each of the four-component mixtures which describe the experimental values of the compressive strength property. The obtained dependence is presented in Eq.(2), where, a_1, a_2 ...a_{28} are the polynomial coefficients obtained with the least-square method Eq.(3).

$$\begin{aligned}y = {} & a_1x_1 + a_2x_2 + a_3x_3 + a_4x_4 + a_5x_1x_2 + a_6x_1x_3 + a_7x_1x_4 + a_8x_2x_3 + a_9x_2x_4 + a_{10}x_3x_4 + a_{11}x_1x_2\left(x_1 - x_2\right) + \\ & + a_{12}x_1x_3\left(x_1 - x_3\right) + a_{13}x_1x_4\left(x_1 - x_4\right) + a_{14}x_2x_3\left(x_2 - x_3\right) + a_{15}x_2x_4\left(x_2 - x_4\right) + a_{16}x_3x_4\left(x_3 - x_4\right) + \\ & + a_{17}x_1x_2\left(x_1 - x_2\right)^2 + a_{18}x_1x_3\left(x_1 - x_3\right)^2 + a_{19}x_1x_4\left(x_1 - x_4\right)^2 + a_{20}x_2x_3\left(x_2 - x_3\right)^2 + a_{21}x_2x_4\left(x_2 - x_4\right)^2 + \\ & + a_{22}x_3x_4\left(x_3 - x_4\right)^2 + a_{23}x_1^2x_2x_3 + a_{24}x_1^2x_2x_4 + a_{25}x_1^2x_3x_4 + a_{26}x_1x_2^2x_3 + a_{27}x_1x_2^2x_4 + a_{28}x_3x_2^2x_4 + \\ & + a_{29}x_1x_2x_3^2 + a_{30}x_1x_4x_3^2 + a_{31}x_2x_4x_3^2 + a_{32}x_1x_2x_4^2 + a_{33}x_1x_3x_4^2 + a_{34}x_2x_3x_4^2.\end{aligned} \tag{2}$$

$$\begin{aligned}y = {} & 57.7x_1 + 91.5x_2 + 95.7x_3 + 108.7x_4 + 663.2x_1x_2 + 79.6x_1x_3 + 68.8x_1x_4 - 70.4x_2x_3 - \\ & - 249.2x_2x_4 + 474x_3x_4 + 248.533x_1x_2\left(x_1 - x_2\right) + 137.6x_1x_3\left(x_1 - x_3\right) + 330.133x_1x_4\left(x_1 - x_4\right) + \\ & + 387.733x_2x_3\left(x_2 - x_3\right) + 139.2x_2x_4\left(x_2 - x_4\right) + 340.8x_3x_4\left(x_3 - x_4\right) - 2590x_1x_2\left(x_1 - x_2\right)^2 - \\ & - 363.2x_1x_3\left(x_1 - x_3\right)^2 - 328.533x_1x_4\left(x_1 - x_4\right)^2 + 264.533x_2x_3\left(x_2 - x_3\right)^2 + 255.467x_2x_4\left(x_2 - x_4\right)^2 - \\ & - 2737x_3x_4\left(x_3 - x_4\right)^2 - 6405x_1^2x_2x_3 - 8799x_1^2x_2x_4 + 14670x_1^2x_3x_4 - 1332x_1x_2^2x_3 - 2570x_1x_2^2x_4 + \\ & + 4570x_3x_2^2x_4 + 2610x_1x_2x_3^2 - 8413x_1x_4x_3^2 - 3615x_2x_4x_3^2 + 7981x_1x_2x_4^2 - 3540x_1x_3x_4^2 - 78.4x_2x_3x_4^2.\end{aligned} \tag{3}$$

Table 5

The results of adequacy with Student's *t*-distribution.

Control point	x_1	x_2	x_3	x_4	$\overline{y}$	y	s_y	φ	t
15_1	0.333	0.333	0.333	0	95.15	93.071	0.919	0.547	2.572
15_2	0.15	0.7	0.15	0	63.3	61.627	4.525	3.566	0.245
15_3	0.7	0.2	0.1	0	29.4	26.632	3.566	5.366	0.844

The adequacy of the obtained equation has been assessed in each control points (15_1, 15_2, 15_3) with Student's *t*-distribution [3], the obtained results are presented in Table 5.

The desired value of Student's *t*-distribution has been calculated using the Eq.(4):

$$t = \frac{\Delta y\sqrt{r}}{s_y\sqrt{1+\varphi}} \tag{4}$$

where, *Δy* is calculated according to Eq.(5):

$$\Delta y = \left|\overline{y} - y\right| \tag{5}$$

where, $\overline{y_i}$ – arithmetic mean of experimental data in the control points;

y– calculated value of studied property in the control points;

r– number of parallel testing in one control point;

s_y– standard deviation of experimental points;

α– level of importance (α= 0.05);

φ– value depending on the mixture composition on the simplex surface and that has been calculated by Eq.(6) for fourth-order polynomial dependencies.

$$\begin{aligned}\varphi = {} & \sum_{1\le i\le q} d_i^2 + \sum_{1\le i<j\le q} d_{ij}^2 + \sum_{1\le i<j\le q} d_{iiij}^2 + \sum_{1\le i<j\le q} d_{ijjj}^2 + \\ & \sum_{1\le i<j<k\le q} d_{iijk}^2 + \sum_{1\le i<j<k\le q} d_{ijjk}^2 + \sum_{1\le i<j<k\le q} d_{ijkk}^2\end{aligned} \tag{6}$$

where, *q*– number of the mixture components.

The values d_i, d_{ij}, d_{iiij}, d_{ijjj}, d_{iijk}, d_{ijjk}, d_{ijkk} have been calculated by Eqs.(7-13).

$$d_i = \frac{1}{6}x_i(4x_i - 1)(4x_i - 2)(4x_i - 3) \tag{7}$$

$$d_{ij} = 4x_ix_j(4x_i - 1)(4x_j - 1) \tag{8}$$

$$d_{iiij} = \frac{8}{3}x_ix_j(4x_i - 1)(4x_i - 2) \tag{9}$$

$$d_{ijjj} = \frac{8}{3}x_ix_j(4x_j - 1)(4x_j - 2) \tag{10}$$

$$d_{iijk} = 32x_ix_jx_k(4x_i - 1) \tag{11}$$

$$d_{ijjk} = 32x_ix_jx_k(4x_j - 1) \tag{12}$$

$$d_{ijkk} = 32x_ix_jx_k(4x_k - 1) \tag{13}$$

The sampling variances have been calculated by Eq.(14).

$$s_y^2 = \frac{\sum_{i=1}^{r}(\overline{y} - y)^2}{r - 1} \tag{14}$$

The number of degrees of freedom is *f*= 3; the obtained values of *t*-criteria have been compared to table value $t_{tab}(\alpha = 0.05, f = 3) = 3.182$. The condition $t < t_{tab}$ is valid in all control points. That fact confirms the adequacy of the obtained fourth-order polynomial dependencies for each of the four-component mixtures.

Optimization of the composition of ceramic materials based on compressive strength evaluation using Scheffe's method

The response functions of the studied property of materials in simplex lattices of the given components (formed by the variables z_1, z_2, z_3 and z_4) are represented graphically as projections of equal values curves as well as using 3D-figures that illustrate the surfaces of the changing values of the studied properties in Figures S1-S4 (Supplementary material). The increasing of the compressive strength index value in the mass fractions simplex lattice at the coded variables z_1, z_2 and z_3 (Figure S1 in Supplementary material) is indicated towards the centre of the $\{z_1, z_2\}$ axis while reaching 220 MPa. The compressive strength index reaches 220 MPa in the centre of the $\{z_1, z_2\}$ axis on the mass fractions simplex lattice at the encoded variables z_1, z_2 and z_4 (Figure S2 in Supplementary material). According to Figure S3, the values of compressive strength increase from the simplex apex formed with the coordinates z_2, z_3 and z_4 up to 160 MPa in the centre of the simplex and up to 190 MPa on the $\{z_3, z_4\}$ axis. The highest values of the compressive strength on the mass fractions simplex lattice of the three-component system at the coordinates z_1, z_3 and z_4 (190 MPa) are indicated in the centre of the $\{z_3, z_4\}$ axis (Figure S4 in Supplementary material).

The joint of the elementary three-component simplexes along the faces made it possible to obtain the simplex lattice of compressive strength for four parameters (Figure 3).

Figure 3 clearly shows that the index of the compressive strength varies within significant limits 40-220 MPa. The areas with maximum index values are of great interest because these determine the resistance of the ceramic carrier to external physical loads in operation. There are two such areas of maximum values on a four-dimensional simplex lattice. The first area with the compressive strength of 220 MPa is within the coordinates of the coded variables $\{z_1, z_2, z_3\}$ and $\{z_1, z_2, z_4\}$. This area is given for the following mixture proportion: 0.63-1.25 mm weight fraction of cordierite with 0.35-0.5 with the coded variables; <0.63 mm weight fraction of cordierite with 0.3-0.45 with the coded variables; corundum with 0.2-0.3 with the coded variables; 1.25-2.5 mm weight fraction of cordierite with 0.2-0.25 with the coded variables.

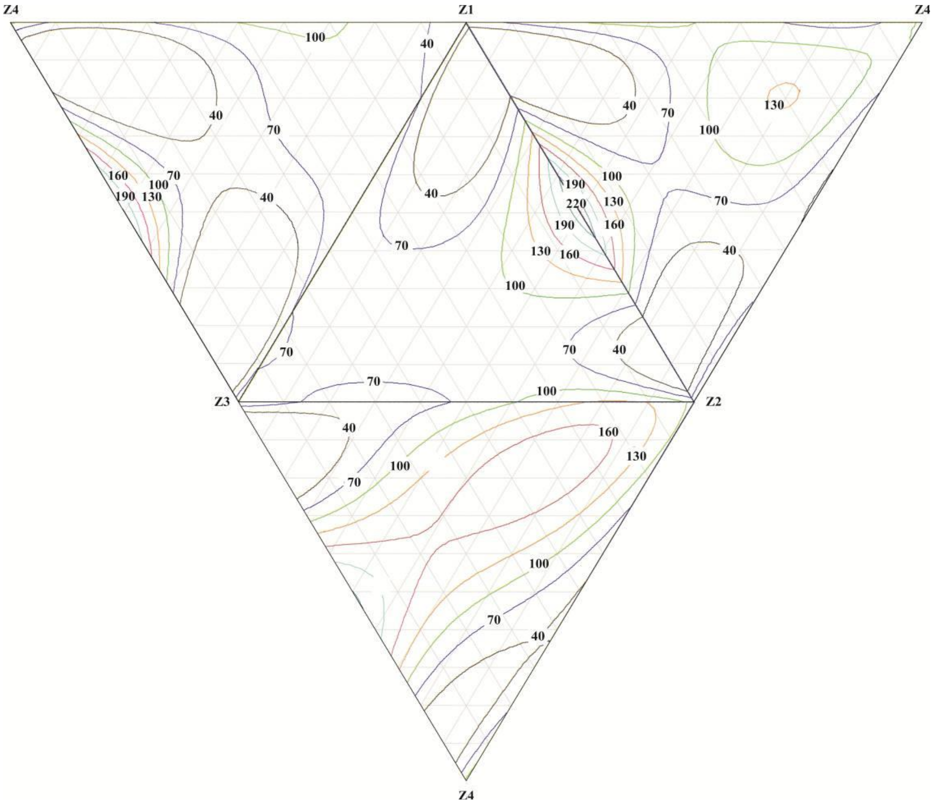

Figure 3. The projection of equal values curves of compressive strength (MPa) in mass fractions simplex lattice of the four-component system at the coded variables z_1, z_2, z_3 and z_4.

The second area of maximum values (190 MPa) is located inside the simplex formed by the coordinates z_1, z_3 and z_4 with the transition to the coordinates $\{z_2, z_3, z_4\}$ and corresponds to the following mixture proportion: 0.63-1.25 mm weight fraction of cordierite with 0.2-0.25 with the coded variables; <0.63 mm weight fraction of cordierite with 0.2-0.25 with the coded variables; corundum with 0.3-0.45 with the coded variables; 1.25-2.5 mm weight fraction of cordierite with 0.3-0.4 with the coded variables.

Thus, the proposed modified Scheffe's method of experimental design has made it possible to describe the change in the four-component system with compressive strength as parameter. The analysis of data regarding the response surface allows selecting an optimal mixture composition. This mixture consists of (mass% fraction): 0.63-1.25 mm weight fraction of cordierite with 0.35-0.5; <0.63 mm weight fraction of cordierite with 0.3-0.45; corundum with 0.2-0.3; 1.25-2.5 mm weight fraction of cordierite with 0.2-0.25.

Scheffe's method is widely used in chemical engineering but it was limited by graphical description of the simultaneous interactions of multicomponent systems [16,17]. The proposed method extends significantly the limits of the usage of simplex lattices in the study of multicomponent systems in all branches of chemistry.

Application of the modified Scheffe's method in the design and manufacturing of a neutraliser of gas emissions

The compressive strength of ceramic mixtures was determined using the proposed method. Analysis of results made it possible to select the optimal composition of the ceramic mixture to produce the item with the maximum compressive strength. This mixture consists of 0.63-1.25 mm weight fraction of cordierite with 0.35 mass% fraction; <0.63 mm weight fraction of cordierite with 0.35 mass% fraction; <0.06 mm weight fraction of corundum with 0.2 mass% fraction; 1.25-2.5 mm weight fraction of cordierite with 0.2 mass% fraction.

These ceramic items with the maximum compressive strength (220 MPa) were used to make catalytic gas purification blocks for the waste processing complexes (WPC) with a capacity of 300 kg/hour ("WPC-300") at the Kharkiv – Passenger railway station (Ukraine) without the need to use burning (Figure 4) [18]. These ceramic blocks were hole-profiled and their operation surface was coated with catalytically active compounds (based on Co_3O_4) [18].

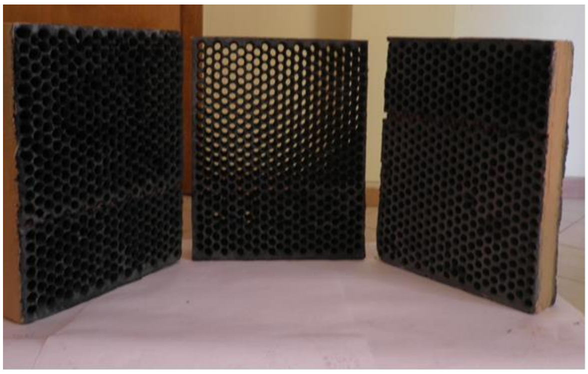

Figure 4. The hole profiled ceramic carrier with catalytically active centres.

The efficiency of the neutraliser of gas emissions of the waste processing complexes (WPC) was determined by sampling during its operation (Table 6). The combustion of solid household wastes releases a large amount of toxic substances into the atmosphere [19]. Even low concentrations of CO in ambient air can trigger physiological changes in humans. Polycyclic aromatic hydrocarbons, chlorinated hydrocarbons, alkanes C_3–C_{20} and benzapyrene are dangerous carcinogens [19]. The results presented in Table 6 prove that the developed catalytic gas purification equipment has high effectiveness; purification rate is ≥98% for all the observed indicators (Table 6).

Table 6

Comparative results of toxic substances emissions before and after the installation of catalytic gas purification equipment "WPC-300" [18].

Components	*Emission rate, kg/h*		*Purification rate, %*
	Before cleaning	*After cleaning*	
Carbon monoxide	15.2	0.12	99.2
Polycyclic aromatic hydrocarbons	0.402	0.008	98.0
Chlorinated hydrocarbons	0.101	0.002	98.0
Alkanes C_3–C_{20}	1.514	0.015	99.0
Benzapyrene	0.135×10^{-5}	0.4×10^{-8}	99.72

Conclusions

The modified Scheffe's simplex lattice design method was developed and used successfully to determine optimal compositions of multicomponent ceramic carriers of catalysts for gas emissions neutralizers.

The changes in the investigated property of compressive strength for four-component mixtures of ceramic materials were presented by the obtained fourth-order polynomial dependencies which are based on the modified Scheffe's simplex lattice method. This method gave an opportunity to select the optimal compositions of ceramic material (0.63-1.25 mm weight fraction of cordierite with 0.35 mass% fraction; <0.63 mm weight fraction of cordierite with 0.35 mass% fraction; <0.06 mm weight fraction of corundum with 0.2 mass% fraction; 1.25-2.5 mm weight fraction of cordierite with 0.2 mass% fraction) to produce a carrier for a gas emissions neutralizer with the compressive strength of 220 MPa. The gas treatment equipment was manufactured on the basis of this optimal selected composition and was installed on the waste processing complex "WPC-300" at the Kharkiv – Passenger railway station (Ukraine).

The efficiency of the manufactured catalytic gas purification equipment was determined during operation of the waste processing complex; the purification rate reached and exceeded 98% for all the indicators. Thus, the modified Scheffe's simplex lattice design method can be applied successfully in the study of the properties of mixtures in wide range.

Supplementary information

Supplementary data are available free of charge at http://cjm.asm.md as PDF file.

References

1. Ponomarenko, A.V.; Ved`, V.E.; Varbanov, P.; Sataev, M.I. Kinetic regularities of heterogeneous catalytic processes of gas emissions purification. Integrated Technologies and Energy Saving Conservation, 2015, 3, pp. 51-56. (in Russian). http://repository.kpi.kharkov.ua/handle/KhPI-Press/23803
2. Ponomarenko, A.V.; Ved`, V.E. Determination of the mass-transfer coefficient in designing heterogenous catalytic processes. Eastern-European Journal of Enterprise Technologies, 2014, 2(6), pp. 42-45. (in Russian). DOI: https://doi.org/10.15587/1729-4061.2014.23148
3. Patience, G.S. Experimental Methods and Instrumentation for Chemical Engineers. Elsevier: USA, 2013, 416 p. DOI: https://doi.org/10.1016/C2010-0-66690-9
4. Scheffe, H. Experiments with mixtures. Journal of the Royal Statistical Society: Series B (Methodological), 1958, 20(2), pp. 344-360. DOI: https://doi.org/10.1111/j.2517-6161.1958.tb00299.x
5. Fan, Y.; Cai, Y.; Li X.; Jiao, L.; Xia, J.; Deng, X. Effects of the cellulose, xylan and lignin constituents on biomass pyrolysis characteristics and bio-oil composition using the Simplex Lattice Mixture Design method. Energy Conversion and Management, 2017, 138, pp. 106-118. DOI: https://doi.org/10.1016/j.enconman.2017.01.075
6. Nouadjep, N.S.; Nso, E.; Gueguim Kana, E.B.; Kapseu, C. Simplex lattice mixture design application for biodiesel production: Formulation and characterization of hybrid oil as feedstock. Fuel, 2019, 252, pp. 135-142. DOI: https://doi.org/10.1016/j.fuel.2019.04.088
7. Crespo, Y.A.; Sánchez, L.R.B.; Quintana, Y.G.; Cabrera, A.S.T.; del Sol, A.B.; Mayancha, D.M.G. Evaluation of the synergistic effects of antioxidant activity on mixtures of the essential oil from *Apium graveolens* L., *Thymus vulgaris* L. and *Coriandrum sativum* L. using simplex-lattice design. Heliyon, 2019, 5(6), e01942. DOI: https://doi.org/10.1016/j.heliyon.2019.e01942
8. Grytsenko, O.; Pukach, P.Ya.; Suberlyak, O.V.; Moravskyi, V.S.; Kovalchuk, R.A.; Berezhnyy, B.V. The Scheffe's method in the study of mathematical model of the polymeric hydrogels composite structures optimization. Mathematical Modelling and Computing, 2019, 6(2), pp. 258-267. DOI: https://doi.org/10.23939/mmc2019.02.258
9. Onyelowe, K.; Alaneme, G.; Igboayaka, C.; Orji, F.; Ugwuanyi, H.; Van, D.B.; Van, M.N. Scheffe optimization of swelling, California bearing ratio, compressive strength, and durability potentials of quarry dust stabilized soft clay soil. Materials Science for Energy Technologies, 2019, 2(1), pp. 67-77. DOI: https://doi.org/10.1016/j.mset.2018.10.005
10. Lazic, Z.R. Design of Experiments in Chemical Engineering: A practical guide. John Wiley: Weinheim, 2004, 620 p. https://www.wiley.com/en-us/Design+of+Experiments+in+Chemical+Engineering%3A+A+Practical+Guide-p-9783527604593
11. Korir, B. Optimal Kiefer ordering of simplex designs for third-degree mixture Kronecker-Models with three ingredients. Mathematical Theory and Modeling, 2019, 9(1), pp. 64-85. DOI: https://doi.org/10.7176/MTM/9-1-06
12. Draper, N.R.; Lawrence, W. Mixture designs for three factors. Journal of the Royal Statistical Society: Series B (Methodological), 1965, 27(3), pp. 450-465. DOI: https://doi.org/10.1111/j.2517-6161.1965.tb00604.x
13. Frolova, L.; Khmelenko, O. Investigation of the magnetic properties of ferrites in the CoO-NiO-ZnO using simplex-lattice design. Journal of Nanomaterials, 2018, 23, pp. 1-8. DOI: https://doi.org/10.1155/2018/5686741

14. Ponomarenko, A.V.; Ved`, V.E. Ceramic bearer material development of catalytic converter gas emission. Integrated Technologies and Energy Conservation, 2014, 2, pp. 36-42. (in Russian). http://repository.kpi.kharkov.ua/handle/KhPI-Press/16050
15. Reference Document on Best Available Techniques in Ceramic Manufacturing Industry. The European IPPC Bureau, 2007. https://eippcb.jrc.ec.europa.eu/sites/default/files/2019-11/cer_bref_0807.pdf
16. Arimanwa, J.I.; Onwuka, D.O.; Arimanwa, M.C.; Ajoku, C.A. Simplex lattice design models for the determination of modulus of rupture of concretes. Nigerian Journal of Technological Development, 2019, 16(4), pp. 211-217. DOI: http://dx.doi.org/10.4314/njtd.v16i4.8
17. Claudius, K.; Elinwa, A.U.; Duna, S. Simplex lattice method: A predictive tool for concrete materials. American Journal of Engineering Research, 2017, 6(4), pp. 19-27. http://www.ajer.org/papers/v6(04)/C06041927.pdf
18. Ponomarenko, A.V.; Ved', V.E.; Krasnokutskij, E.V. Creation of a catalytic unit for the purification of gas emissions from a waste-processing plant with a capacity of 100 kg/h. ONAKhT, 2014, 45(3), pp. 180-185. (in Russian). http://repository.kpi.kharkov.ua/handle/KhPI-Press/16062
19. EMEP/EEA air pollutant emission inventory guidebook 2019. Technical guidance to prepare national emission inventories. EEA Report No 13/2019. https://www.eea.europa.eu/publications/emep-eea-guidebook-2019