CORELAȚII COMPOZIȚIE-PROCESARE-PROPRIETĂȚI LA OBȚINEREA CERAMICII DE MENAJ. PARTEA I: INFLUENȚA MATERIILOR PRIME ASUPRA PROPRIETĂȚILOR CORPULUI CERAMIC

CORRELATIONS COMPOSITION-PROCESSING-PROPERTIES FOR TABLEWARE CERAMICS. PART I: INFLUENCE OF THE RAW MATERIALS ON THE CERAMIC BODY'S PROPERTIES

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A technological issue was addressed in the part I of the paper, i.e. recipes of different proportions of materials were used to obtain ceramic bodies (seven raw mixtures). Shaped materials were fired in industrial tunnel kilns for biscuit/glost firing. Resulting ceramic bodies were tested for industry-relevant properties. Correlations composition-properties for the ceramic body were, initially, empirically identified, by observation. Regression analysis was used, afterwards, to scientifically quantify these correlations. Results confirm the existence of fair to up to strong correlations composition – properties; therefore, properties can be easily obtained by using the composition of the raw mix. In industry, these simple calculations can serve as a preliminary assessment of the properties, to reduce the amount of experimental efforts, given a set of raw materials. Multiple-Criteria Analysis was used to show that it is possible to select, on a scientifically basis, the best available alternative of the raw materials' proportions used i.e. the alternative that fulfills most requirements, as concerning properties. In a future, second part of the paper, the correlation processing – properties will be explored to demonstrate the influence of grinding on the glaze properties.

In acest prim articol a fost tratată o problemă tehnologică. Au fost obținute o serie de corpuri ceramice folosind șapte rețete – fiecare rețetă conținând proporții diferite din aceleași materii prime. Materialele fasonate au fost arse în cuptoare tunel industriale atât la arderea I (arderea de biscuit) cât și la arderea II (arderea de glazură). Corpurile ceramice au fost testate pentru proprietăți relevante în industrie. Au fost inițial, identificate în mod empiric, prin observație, corelații tip compoziție-proprietăți. Analiza de regresie a fost utilizată ulterior pentru a cuantifica pe baze științifice aceste corelații. Rezultatele confirmă existența unor corelații bune până la puternice între compoziție și proprietăți, ceea ce oferă posibilitatea de a calcula cu ușurință proprietățile corpului ceramic pe baza compoziției amestecului de materii prime. În industrie, aceste calcule simple pot folosi pentru estimarea preliminară a proprietăților, în scopul reducerii volumului de experimente. Analiza multi-criterială a arătat că este posibil să se selecteze, pe criterii științifice, cea mai bună variantă de amestec de materii prime (care satisface cele mai multe cerințe). În articolul următor se va investiga dacă măcinarea influențează proprietățile glazurilor (corelații tip procesare-proprietăți).

***Keywords:** tableware ceramics, correlations, Multiple-Criteria Analysis*

1. Introduction

Tableware ceramics industry production capacities are rapidly increasing worldwide, due to both, an inner characteristic of ceramics, i.e. their brittleness, and to the fast growing of the world population and, therefore, consumption. Although tableware fabrication has a long history, the continuous search for new object designs, colors and improved properties of the products require constant research and development. It is worthwhile to mention that raw materials differ, from region to region, in oxide/mineralogic composition. Therefore, when using one or more raw materials from another source (or by changing their proportions in the raw mix), some properties may be different; consequently, further experimental work is mandatory. The logical correlation chain that starts from the raw materials composition and ends with products' properties (referred in [1]) can be thus identified and it will be investigated herein.

The design of a tableware product starts from the desired properties that provide product with required quality indices. Properties are dependent on phase and mineralogical compositions; mineralogical composition depends on oxide composition of the raw materials and their proportions, on one hand, and on processing conditions on the other, thus completing the correlation chain: composition-processing-properties.

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This research will be organized in two parts: in the first paper (part I), correlations composition-properties for the ceramic mass (support) will be addressed, meaning raw materials' proportions will be modified and influences on biscuit's properties will be recorded and explained.

Processing conditions require grinding of the materials for ceramic support and glaze, followed by two stages of firing, one for the biscuit (1^{th}) and the second for glost (2^{nd}) firing. It is well known that, proper firing cycles should be selected according to the desired mineralogical composition, product's size and shape and on the green body composition and moisture content. Cooling should be adjusted according to the required properties for both, biscuit and glost firing. In both parts of the paper, thermal processing conditions will be maintained constant, so the other influences can be isolated. Note that firing cycles should be adjusted, based on thermal analysis results. In the part II of this paper (to follow) it will be tested if/how the mechanical processing (grinding) will influence thermal processing efficiency when obtaining glazes.

In this first part, raw mixtures of different proportions of materials will be used to obtain the ceramic body (7 raw mixtures). Representative properties for the ceramic body will be experimentally measured or computed. The constructed database will be used, firstly, to identify if there is any correlation, by performing Multiple Linear Regression (MLR). Multiple-Criteria Analysis (MCA) will be used to select the best combination of raw materials/materials by considering all biscuit properties, on a scientific basis.

It is common sense to prepare biscuits (ceramic bodies) in a way that allow for a desired shape, proper mechanical strength and a good match with the glaze. In that purpose, in order, pyroplastic deformation of the fired body (referred in [2-4]), flexural strength [5], absorption and color [6, 7] will be investigated. Note that absorption influences the amount of glaze entering biscuit's pores, thus bonding ceramic body and glaze; too much or too small absorption will be detrimental.

2. Experimental

2.1. Materials

Five common raw materials (sand, feldspar, two kaolins with different provenience – named Kaolin 1, Kaolin 2, clay) and indigenous recycled biscuit in different proportions (see Table 1) were used to create 7 raw mixtures named R1-R7. Note that the recycled material and the clay have been used in only one combination, each. Kaolins, which have different proveniences and compositions, were used mostly 25/25% while for two raw mixtures, R1 and R2, only one kaolin has been used, at a time, as 50%. In combination R6, 5% of Kaolin 2 has been replaced by the same amount of clay. The aim of these changes is to identify any possible influence of the individual materials by reducing, as possible, the amount of experimental.

Table 1

Recipes of raw materials/recycled materials (wt%)
Rețetele de materii prime/material reciclat (%grav)

%	R1	R2	R3	R4	R5	R6	R7
Sand	25	25	25	20	30	25	25
Feldspar	25	25	25	30	20	25	20
Kaolin 1	0	50	25	25	25	25	25
Kaolin 2	50	0	25	25	25	20	25
Clay	0	0	0	0	0	5	0
Recycled biscuit	0	0	0	0	0	0	5

Raw materials and recycled material compositions are given in Table 2 along with the Loss on ignition (LOI). All oxide compositions were provided by the suppliers.

2.2. Preparation

Proportioned materials according to Table 1 have been wet-grinded and homogenized in a laboratory ball mill for 7000 rotations. Resulting slurry has been dried for 2 h in air, then oven-dried at 110°C for 24 h.

Grain size distribution of the powder has been analyzed by SediGraph III Plus. Particle size distribution was measured using the sedimentation method, by measuring particle mass via X-ray absorption. The equivalent spherical diameter of particles is determined by measuring the rate at which particles fall under gravity through a liquid having known properties and by using Stokes' Law [8]. Particle redispersion was made by sonication. An example of the size distribution of raw mix is given in Fig. 1 for R7 combination.

Table 2

Oxide compositions of raw materials and recycled biscuits
Compoziția oxidică a materiilor prime și a biscuitelui reciclat

	%SiO_2	%Al_2O_3	%Fe_2O_3	%TiO_2	%CaO	%MgO	%Na_2O	%K_2O	LOI
Sand	97.40	1.43	0.12	0.26	0.07	0.03	0.03	0.03	0.54
Feldspar	70.00	18.00	0.15	0.32	0.50	0.20	9.50	0.28	0.50
Kaolin 1	51.02	34.32	0.81	0.22	0.16	0.24	0.10	1.09	12.04
Recycled biscuit	69.30	24.00	0.57	0.40	1.02	0.53	1.47	1.83	0.00
Clay	61.90	23.80	2.28	1.19	0.28	0.48	0.08	2.22	7.46
Kaolin 2	49.63	34.12	1.76	0.59	0.49	0.37	0.07	0.89	12.05

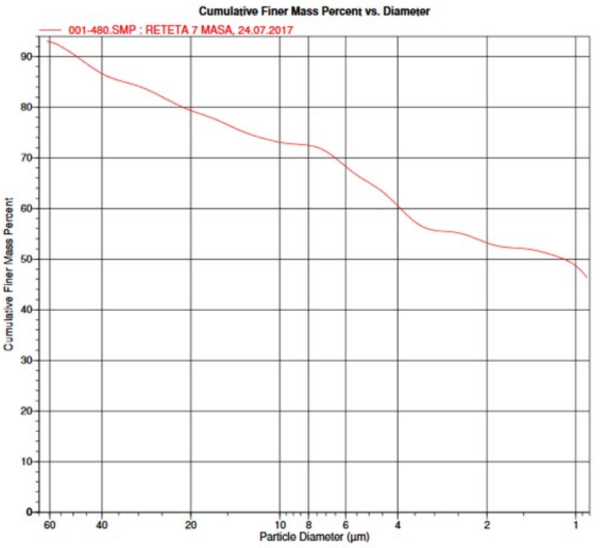

Fig. 1. - Grain size distribution analysis of the R7 recipe . *Distribuția granulometrică a rețetei R7.*

On all 7 cumulative distributions, D80 values were identified (note: D80 is the diameter at which 80% of the sample's mass is made of particles with a diameter less than the measured value). Almost all the D80 values are less than 30 µm, as it can be seen in Table 3. Differences between them can be attributed (mostly) to local agglomeration phenomena but, also, to the variations in composition and/or grain size distribution of the raw materials used. It is possible that the lowest D80, recorded for R4 recipe, may be due to the lowest amount of sand included. Sand - basically quartz - has the highest hardness of all materials that were used; as a consequence, the less quartz within the raw mix – the higher the grindability will be and, therefore, the smaller the value of the D80.

Table 3

D80 values of all seven raw mixtures, grinded in a laboratory mill for 7000 rotations

Valorile D80 ale celor șapte amestecuri de materii prime măcinate într-o moară de laborator la 7000 rotații

Recipe	D80 (µm)
R1	17
R2	32
R3	28
R4	8
R5	30
R6	9
R7	22

Powder has been shaped in samples of 100x50x10 mm by uniaxial pressing at 3 different, successive pressures (14, 28 and 46 MPa). According to the property measured, some of the samples went to biscuit firing (950°C), others to glost firing (1200°C).

Flexural strength is a major attribute of the tableware; it's value depends on several factors, one of them being the feldspar content. Basically, the higher the feldspar content, the higher the flexural strength. Specimens used to determine flexural strength were thermally treated in an industrial tunnel kiln. On the fired body, flexural strength was measured by 3-points testing; for each recipe, 3 tests were carried, and the average value of the flexural strength was retained.

Absorption has been measured in two steps: *i)* after the biscuit firing; *ii)* after glost firing, both by weighing the specimen before and after firing. Before firing, specimens were completely immersed in water at ambient temperature for 4 h for *i)* and 2h in boiled water for *II)*.

Fired shrinkage describes how bodies shrink: with temperature increase, in solid state particles pack closer and closer; by further increasing temperature, some of the particles melt, bringing the rest of the particles even closer; also, some of the particles, such as kaolin, shrink themselves due to dehydroxylation. Shrinkage can be obtained by measuring dried and after glost firing bodies.

Note that all dimensional measurements for all properties/features were made by using a precision caliper.

Loss on ignition (LOI) is primarily related to the amount of chemically bound water that is removed from the kaolin/clay structure with temperature; it was measured for both biscuit (LOI 1) and glost firing (LOI 2) by weighing the specimen.

The pyroplastic deformation has been recorded for glost firing, on the ceramic body. Pyroplasticity, which is the gravity-driven, viscous deformation of a ceramic material during firing, is attributed to the volume and nature of the liquid phase and to the porosity of the structure in which this phase is contained. The firing zone temperature, the heating rate and the amount of time in which the specimens remain at the maximum temperature, are processing variables that can affect the pyroplastic deformation. Some chemical components, such as feldspars, have a high fusibility and ability to form eutectics with other components, even at low temperatures.

The procedure used to determine the pyroplasticity index consists in measuring the curvature of a specimen during its firing over two refractory supports.

One of the most important attributes of the tableware is color. Color has been measured with a BYK Colorimeter in CIE L*a*b* color space (where L* stands for the lightness, a* and b* for the green–red and blue–yellow color components, ranging between 0 and 100). Each specimen was measured in three distinct points and the result was averaged.

Table 4

Properties measured for all seven recipes / *Valorile măsurate ale proprietăților pentru cele șapte rețete*

	R1	R2	R3	R4	R5	R6	R7
Flexural strength [MPa]	2.50	4.34	4.06	5.95	3.46	3.00	3.28
Absorption_2 (%)	9.77	5.40	6.70	4.11	8.74	6.14	7.42
Absorption_1 (%)	17.39	16.85	16.09	16.47	15.88	15.85	16.24
Shrinkage (%)	4.26	5.91	4.85	6.51	3.80	4.96	4.68
LOI 1 - at 1st firing	5.82	6.03	5.88	5.90	5.57	5.68	6.09
LOI 2 - at 2nd firing	6.43	5.99	5.84	5.98	5.87	6.20	6.32
Pyroplastic deformation [mm]	25.03	15.63	17.83	20.45	15.40	16.67	14.87
Color L*	85.33	88.97	87.95	85.91	87.27	85.79	87.16
Color a*	2.63	0.97	1.58	1.61	1.51	1.61	1.57
Color b*	10.69	9.78	10.06	10.96	9.54	10.96	9.58

2.3. Results and discussion

All measured results are presented in Table 4.

Note that 1st firing refers to biscuit firing while 2nd firing to glost firing. Color parameters were recorded as the final color of the ceramic body.

Normal shrinking (less than 7%) characterizes all raw mixtures; the highest shrinkage was recorded for R4 combination, which contains the highest amount of feldspar (30%) and the lowest amount of sand (20%). This is consistent with the known fact that feldspars act like fluxes, thus promoting densification. The result correlate well with the lowest Absorption_2 recorded for the same R4 combination (this is explained by: higher density, lower porosity thus lower absorption), and with the highest flexural strength, both due to the feldspar increase. Deformation was also high, as expected.

Combination R5 contains the highest amount of sand (30%) and the lowest content in feldspar (20%), which is inversely to R4; consequently, shrinkage was quite the opposite, meaning that R5 combination exhibit the lowest value of the shrinkage of all.

For combination R1, made of equal amounts (25%) of sand and feldspar and 50% of only one type of kaolin, K2, the lowest flexural strength (2.5MPa) and the highest absorption were recorded, meaning that the use of only kaolin K2 should be avoided.

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3. Mathematical modeling

3.1. Multiple-Criteria Analysis of the ceramic body properties

Real-life issues can be expressed as Multiple-Criteria Analysis (MCA) problems, that consist in the simultaneous evaluation of a known, finite number of alternatives, by considering all criteria. A performance (consequences) matrix can be constructed, in which each row describes an alternative and each column describes the performance of the alternatives against each criterion. The problem may be defined as finding the best alternative while conflicting criteria can occur and/or various nature, measurement units and order of magnitude of the variables exist.

Several methods can be used to assess the performance (consequences) matrix [9]. The *Simple Additive Weighting* (weighted sum method) is the simplest MCA method and it was used here for the ceramic body.

The first step was to construct the consequences matrix; this was made by listing, on rows, the alternatives (R1-R7) and on columns the properties, as criterions. Let x_{ij} be the value given to alternative i^{th} with respect to criterion j^{th}, w_j being the weight of criteria j, n is the number of alternatives and m is the number of criteria.

Data were normalized such as their values range from 0 to 1, 0 being the *worst* and 1 being the *best*, with respect to their optimality criterion;

Table 5

Performance matrix in normalized values, weights and scores
Matricea consecințelor în valori normalizate, ponderile și funcțiile de utilitate

	FS	A_2	A_1	Sh	LOI 1	LOI 2	PD	L	a	b	Score
R1	0.00	0.00	0.00	0.83	0.52	0.00	0.00	0.00	1.00	0.81	0.12
R2	0.53	0.77	0.35	0.22	0.12	0.75	0.93	1.00	0.00	0.17	0.59
R3	0.45	0.54	0.84	0.61	0.40	1.00	0.71	0.72	0.37	0.37	0.60
R4	1.00	1.00	0.60	0.00	0.37	0.76	0.45	0.16	0.39	1.00	**0.70**
R5	0.28	0.18	0.98	1.00	1.00	0.95	0.95	0.53	0.33	0.00	0.60
R6	0.14	0.64	1.00	0.57	0.79	0.39	0.82	0.13	0.39	1.00	0.55
R7	0.23	0.42	0.75	0.68	0.00	0.19	1.00	0.50	0.36	0.03	0.50
w_j	0.300	0.180	0.100	0.100	0.045	0.045	0.200	0.010	0.010	0.010	

the results are given in Table 5. For example, Flexural Strength (FS) optimal value must be at its maximum, while Absorptions (A_2, A_1), Shrinkage Sh, Loss on ignition (LOI1 and LOI2) and Pyroclastic deformation (PD) should be at their minimum. Note that absorption A_1 may not be, in some cases, at its minimum, as it is required a certain value to create a strong bond of the biscuit with the glaze! Further work should be done here to properly consider this criterion.

The next step was to assign criteria weights. These weights, showing the relative importance of criteria in the multi-criteria problem were given here empirically. Therefore, weights

$w_j,\ 0 \le w_j \le 1,\ \sum_{j=1}^{m} w_j = 1,$

were assigned to each property as in Table 5. Some properties such as LOI 1 and 2 and color parameters were considered to be less important for biscuits, so they were assigned with very low weights. On the last column the score has been computed by the Eq. (1):

$$S_i = \sum_{j=1}^{m} w_j \cdot x_{ij},\ i = 1..n \qquad (1)$$

Finally, the best alternative was selected as a solution, i.e. the option with the highest score, $S_{max} = \max\{S_1, \ldots S_i, \ldots S_n\}$ fits the multiple-criteria analysis best.

The analysis of Table 5 indicates that the best alternative is given by the R4 combination with a distinctive score S_{max} = 0.70. Notice that, also, R4 combination recorded the smallest D80 value (see Table 3) at grinding i.e. it had the best grain size distribution. The worst score has been recorded for R1 combination (S=0.12). The rest of the raw mixtures obtained clustered scores, of 0.55±0.05.

It is important to reiterate that MCA does not provide an *optimal output* possible but helps to identify the *best solution* among the ones being analyzed.

3.2. Regression analysis of the ceramic body properties

Multiple Linear Regression (MLR) method was used to correlate ceramic body properties with raw materials percentages, based on their values given in Table 1. MLR method is well known as a simple, easy-to-use method to correlate an output (being the dependent variable) with various inputs (independent variables) and has applications in various areas [10, 11]; therefore, it will not be a subject for further discussion here.

All seven raw mixtures in Table 1 were used to obtain the regression equations for all properties. Results of the MLR are given in Table 6 showing fair to up to very good correlations between properties and the content in raw materials.

Figures 2 and 3 exemplify the extreme cases when the lowest *acceptable* (0.78) and the highest (0.98) R^2 coefficients were obtained.

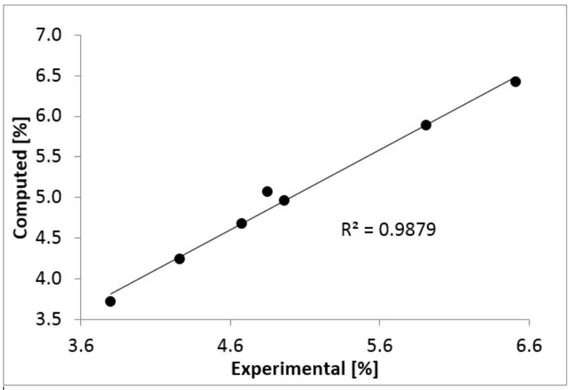

Fig. 2 - Computed versus experimental values of the shrinkage
Valorile calculate vs. experimentale ale contracției la ardere.

Table 6

MLR results for all properties / *Rezultatele analizei de regresie pentru toate proprietățile*

	R^2	MLR Equation
Flexural strength [MPa]	0.78	$Y = 3.430-0.094 \cdot x_1-0.1554 \cdot x_2+0.000 \cdot x_3-0.036 \cdot x_4-0.2481 \cdot x_5+0.000 \cdot x_6$
Absorption_2 (%)	0.81	$Y = 8.720-0.49 \cdot x_1-0.061 \cdot x_2+0.000 \cdot x_3-0.106 \cdot x_4+0.211 \cdot x_5+0.000 \cdot x_6$
Absorption_1 (%)	0.96	$Y = -10.766+0.089 \cdot x_1-0.107 \cdot x_2+0.000 \cdot x_3-0.223 \cdot x_4-0.0378 \cdot x_5+0.000 \cdot x_6$
Shrinkage (%)	0.98	$Y = -12.830+0.000 \cdot x_1+0.270 \cdot x_2+0.220 \cdot x_3+x_4 \cdot 0.220+0.209 \cdot x_5+0.195 \cdot x_6$
LOI 1	0.81	$Y = -2.303+0.000 \cdot x_1+0.033 \cdot x_2+0.14 \cdot x_3+0.146 \cdot x_4+0.107 \cdot x_5+0.080 \cdot x_6$
LOI 2	0.60	$Y = 9.057-0.071 \cdot x_1-0.0596 \cdot x_2+0.000 \cdot x_3+0.008 \cdot x_4+0.044 \cdot x_5+0.000 \cdot x_6$
Pyroplastic deformation [mm]	0.91	$Y = -13.187+0.294 \cdot x_1+0.799 \cdot x_2+0.000 \cdot x_3+0.188 \cdot x_4-0.251 \cdot x_5+0.000 \cdot x_6$
Color L*	0.87	$Y = 86.246+0.1212 \cdot x_1-0.014 \cdot x_2+0.000 \cdot x_3-0.072 \cdot x_4-0.033 \cdot x_5+0.000 \cdot x_6$
Color a*	0.95	$Y = 0.180+0.008 \cdot x_1+0.018 \cdot x_2+0.000 \cdot x_3+0.033 \cdot x_4+0.232 \cdot x_5+0.000 \cdot x_6$
Color b*	0.98	$Y = 7.041-0.016 \cdot x_1+0.1252 \cdot x_2+0.000 \cdot x_3+0.018 \cdot x_4+0.169 \cdot x_5+0.000 \cdot x_6$

where *Y* means property, x_i, *i*=1..6 stands for percentages of the six materials used in raw mixtures, given in Table 1. In order, they are: sand, feldspar, Kaolin 1, Kaolin 2, Clay, Recycled biscuit. Only one property, LOI 2, exhibited low R^2 coefficient (R^2 = 0.60), probably due to improper specimen's manipulation, with loss of material due to accidental friction/damage.

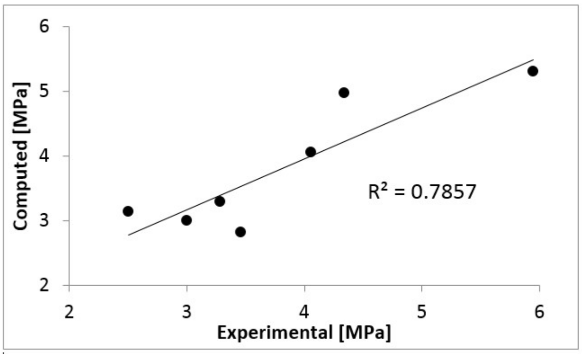

Fig. 3 - Computed versus experimental values of the flexural strength / *Valorile calculate vs. experimentale ale rezistenței la încovoiere.*

Although the intention was to identify correlations (and not to make predictions), preliminary tests on 5 datasets on raw materials were run. At the time the experimental for the preliminary tests were made, some of the raw materials provenience has changed. Even so, encouraging prediction errors on ceramic bodies' properties, averaging ~8% *for all properties,* were found. More experimental work is needed to accurately assess the average error and its standard deviation, but, again, MLR equations were here a byproduct while the main aim was to identify correlations and their strength.

4.Conclusions

Seven raw mixtures of different proportions of materials were used to obtain the ceramic body specimens. Raw materials and other materials were biscuit/glost fired in industrial tunnel kilns. Industry-relevant properties of the ceramic body (pyroplastic deformation of the fired body, flexural strength, absorption, shrinkage, loss on ignition and color) were experimentally measured.

Correlations composition-properties for the ceramic mass (support) were identified, firstly, empirical, by observation. Afterwards, Multiple Linear Regression was used to scientifically attest the existence and, if so, to assess the strength of these correlations. It was demonstrated that fair to very strong correlations occur between measured ceramic body's properties and composition of the raw materials. Although the reason behind employing MLR analysis was, basically, to evidence correlations, the resulting equations can be used for a preliminary calculation of the properties investigated here, within the given compositional range.

Multiple-Criteria Analysis has been used to demonstrate the possibility of selecting a combination of raw materials/materials on a scientific basis, by considering all biscuit properties. These properties are different in nature and in order of magnitude, measurement units etc. thus making, sometimes, difficult to compare and properly select the best option, empirically. MCA technique has been successfully employed here and the best combination has been identified.

A major factor of influence on biscuit/glaze properties has been deliberately omitted here (that could, also, explain in a certain extent the relative high value of the prediction errors on prediction preliminary tests). This influence is due to the mechanical processing of the raw materials mix (grinding); the reason of the omission is, this is the subject of the correlations processing-properties to be addressed in Part II of the article.

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