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Dedicated to Professor Liviu Literat, at his 80th anniversary

SOFTWARE APPLICATION FOR OBTAINING CERAMIC GLAZES WITH PRE-DEFINITE COMPOSITION AND PROPERTIES

ANA-MARIA CORMOŞ[a], FIRUȚA GOGA[a], JOSEPH GASPAR[a], DAN IRIDON[a]

ABSTRACT. The paper presents a software application developed for fast determination of glaze or glass properties with a known composition, optimization of glaze composition using the pre-definite properties and calculation of necessary of raw materials using the composition of glaze and available raw materials. The software application was developed using Matlab software package. A friendly and easy to use interface of the software application was built using GUIDE (Graphical User Interface Development Environment) components of Matlab.

Keywords: *ceramic glazes, glaze properties, optimization of glaze composition*

INTRODUCTION

The ceramic glazes are glass thin films that cover the ceramic products in order to improve some properties. The properties of ceramic glazes have to be in a concordance with the ceramic support. The oxide composition of glaze gives the properties of glaze.

A number of characteristics of glaze are important for the calculation of composition of mixture of raw materials, such as:

- complexity of composition (the large number of oxides);
- glaze properties are correlated with properties and composition of the ceramic body;
- raw materials available.

This work presents a fast method for calculation of raw material mixture composition for glaze production using pre-definite properties, and oxides composition of raw materials. Using the developed software application, the user could choose the most suitable raw materials, in order to reproduce accurate the chemical composition of glaze and subsequently the product properties.

[a] *Universitatea Babeş-Bolyai, Facultatea de Chimie şi Inginerie Chimică, Str. Kogălniceanu, Nr. 1, RO-400084 Cluj-Napoca, Romania,* cani@chem.ubbcluj.ro

The calculation steps are followed:

- determination of glaze oxides composition, using the firing temperature of ceramic product (by reading the Norton diagram);
- calculation of glaze properties using a pre-definite composition;
- calculation of composition of mixture of raw materials;
- optimization of the composition of glaze in function of the thermal expansion coefficient.

The main importance of this software application is to decrease the calculation time and the number of practical experiments which must be done for obtaining an optimal composition glaze.

RESULTS AND DISCUSSION

Determination of ceramic glaze composition

The chemical composition of glazes is complex and it corresponds to an alkali-silico-aluminate glass. The composition is adapted to the support product and is correlated with the product final usage.

The ceramic support implies on pre-definite properties for the optimal of glaze composition. Ceramist researchers [1] tried to correlate the glazes composition with its fusibility and built a two part diagram (Norton). That diagram allows pursuing the variation of molar composition for glaze vs. firing temperature (Figure1).

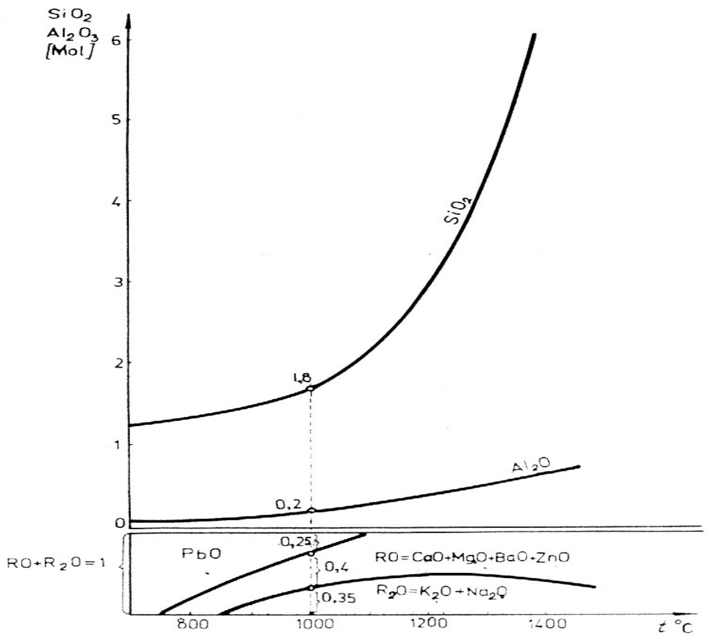

Figure 1. Norton Diagram.

Also, the glazes composition is characterized by certain indexes as follow:

- silica module - SiO_2/Al_2O_3 with value 4.2-6.2;
- acali module - Na_2O/K_2O with value 0.8 - 1.4;
- CaO/MgO with value with value 0.3 - 1.5;
- Acidity coefficient (acidity coefficient shows the activity of glaze vs. support product and allows estimation of stability of glaze in firing process).

The acidity coefficient depends of molar fraction of oxides from glaze:

$$A = \frac{y + 3 \cdot z}{1 + 3 \cdot x}$$

where:

y - molar concentration of SiO_2;

z - molar concentration of B_2O_3;

x - sum of molar concentration of oxides: Al_2O_3 and Fe_2O_3.

The variation of glaze characteristics vs. acidity coefficient is presented in Table 1 [2, 3].

Table 1. Variation of glazes characteristics vs. acidity coefficient

Acidity coefficient (A)	Glazes characterizations
0.7 - 1.25	Useable mat glazes if are rich in PbO
1.45	Mat glazes
1.90 – 2.0	Possible glazes
2.60	Normal glazes
2.90	Possible glazes
2.90	De-vitrification is produced

The developed software application allows determination of the oxide composition of glaze for a known temperature, using Norton diagram. Molar formula read from Norton diagram is converted in mass or molar percentage. Using the percentage composition of oxides, the desirable glaze properties are calculated: thermal expansion coefficient, density, and the refractive index and dispersion index. In order to validate the glaze composition, the developed software application could also be used to calculate the acidity coefficient.

Properties of the glazes in the solid state

The glaze properties are very important for reliability, prediction and design of the ceramic material.

The physical-mechanical properties are calculated using an additive relation described in next relation [4, 5]:

$$P = \sum_{i=1}^{n} p_i c_i \ , \tag{1}$$

where:

P - property;

p_i - additive coefficient of the “i” oxide property;

c_i - concentration of “i” component in glaze, [mol %].

In practice, for some oxides (B_2O_3, SiO_2, PbO, TiO_2) the additive coefficients are strong influenced by the existence and concentration of the other oxides from the system.

In this article the following properties of glazes are calculated using the developed software: the thermal coefficient of expansion, the refractive index and dispersion index and density.

Glaze composition optimization

To obtain a glaze with a pre-define properties; an optimization of glazes composition is necessary. In this software application, the group multi-dynamic optimization method was used [6]. The application of this method is characterized by following elements: an initial solution (the initial glaze composition), the precision of determination of extreme point.

The developed software application optimizes the composition of glaze in function of the thermal expansion coefficient with preservation of acidity coefficient in a specific limit for glaze. The difference between thermal coefficient of expansion of glaze and support layer have to be smaller than 10^{-7}, in order that the glaze not cracks during the cooling.

Beside the two above mentioned coefficients, the oxide ratio is equally important to obtain a glaze ($4.2 < SiO_2/Al_2O_3 < 6.2$; $0.8 < Na_2O/K_2O <1.4$; $0.3 < CaO/MgO <1.5$). The developed application maintains the oxide ratio between the limits.

The calculation of composition of mixture of raw materials

The composition of mixture of raw materials is calculated using the equations of mass balance for any raw materials. The mass balance equations for “i” component is described by formula:

$$\sum_{j=1}^{n} c_{i,j} m_j = c_{i,am} m_{am} \tag{2}$$

where: m_j -“j” raw material weight;

m_{am} – the total mass of blend;

$c_{i,j}$ – the concentration of “i” component in a “j” raw material;

$c_{i,am}$ – the concentration of “i” component in mixture.

Software application – Case Study

Solving of mathematical model and finding the optimum of raw materials composition was done using Matlab software [7]. For the end-users, the GUIDE component (Graphical User Interface Development Environment) of Matlab was used in order to create an interactive interface which allows an easy handling of application. The interface application is easy to use because is using the standard components of the graphic interface of Windows Operation System, and the users do not need to know Matlab syntax.

The main application window [8] (see Figure 2) contains the graphic elements for input data (temperature and composition) and commands for calculation of glaze properties, optimization of glaze composition and calculation of glaze formula. The glaze composition can be introduce in two ways using the Norton diagram (using the firing temperature) or read the glaze's composition from keyboard.

Using the "display" bottom the program can show the glaze composition (Figure 3).

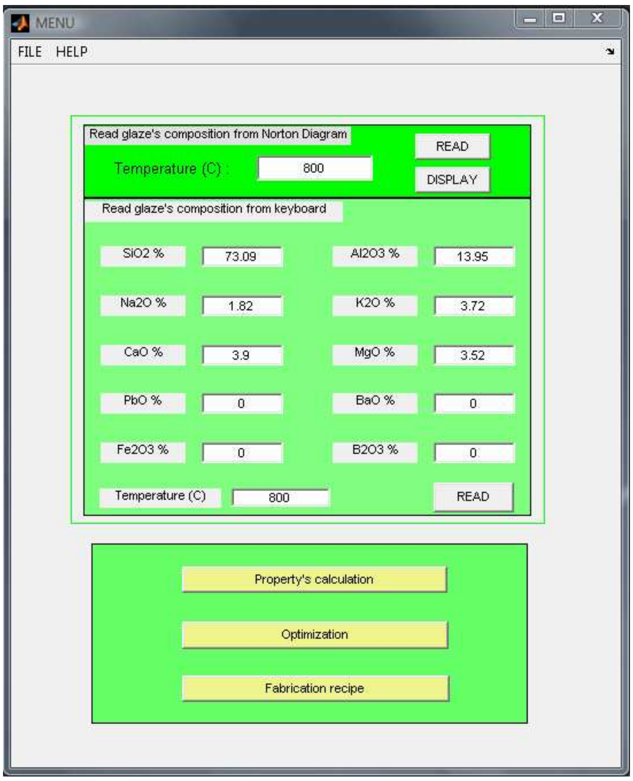

Figure 2. Main application window

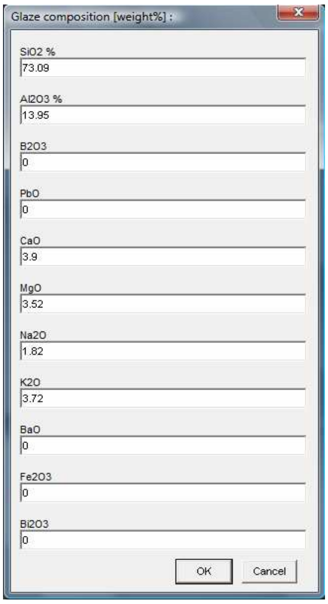

Figure 3. Glaze composition (weight %)

The optimization of the composition needs to know the glaze properties, because any numeric optimization method expects an initial solution. The glaze properties can be visualized using „Property's calculation" button from main window. The properties of glaze are showed in a new window (Figure 4).

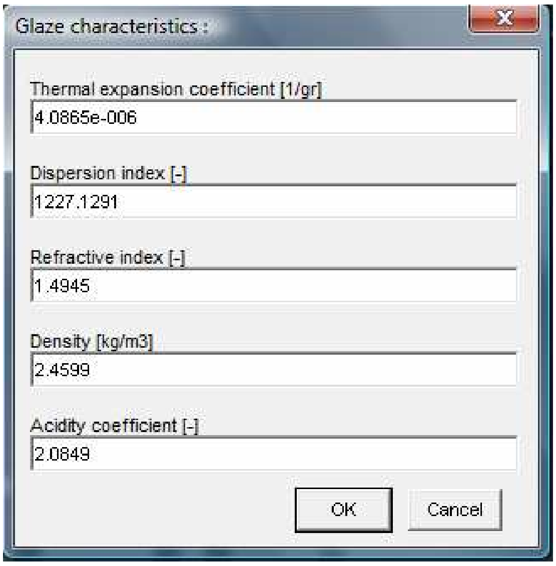

Figure 4. Glaze characteristics

Using the "Optimization" button from main application interface, a new window is open (Figure 5). The value of parameters for optimization: lower value and upper value of thermal coefficient of expansion and acidity coefficient are defined using this window. The calculation of optimal composition is done using the "Run" button and the composition result is display in a new window (Figure 6). The glaze properties after optimization are presented in Figure 7.

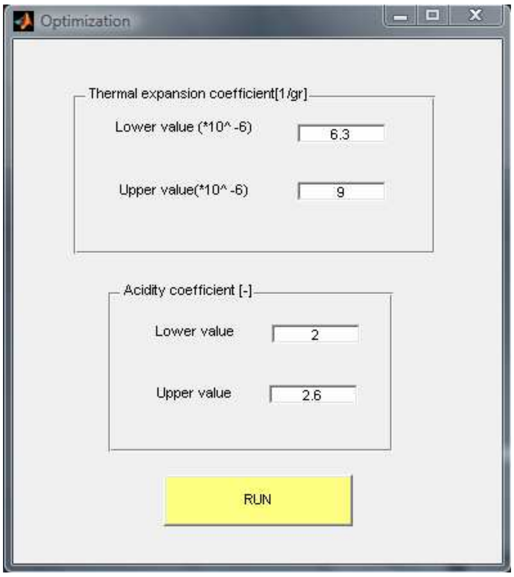

Figure 5. Optimization window.

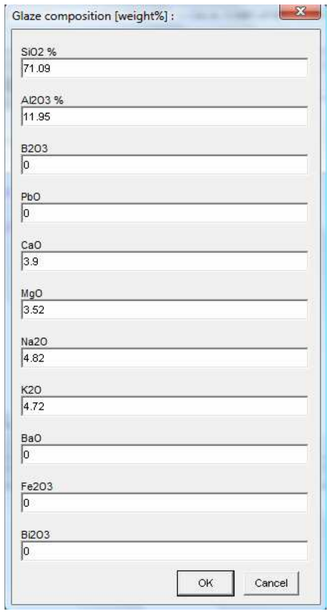

Figure 6. Glaze [%weight] composition, after optimization.

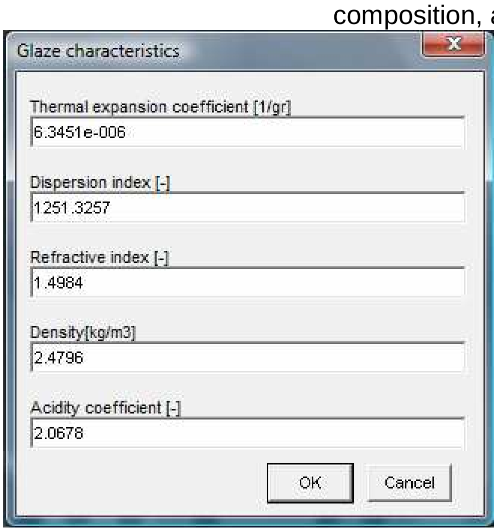

Figure 7. Glaze characteristics, after optimisation.

Other option of application developed is the determination of fabrication formula using a raw material database. The raw materials database of the software application is presented in Table 2. The opening of the window for the calculation of composition of mixture of raw materials (Figure 8) is doing by using the „Fabrication recipe” button from main application window.

Table 2. Raw materials composition

Raw Materials	SiO_2 %weight	Al_2O_3 %weight	CaO %weight	MgO %weight	Na_2O %weight	K_2O %weight	LOI* %weight
Feldspatr	71.6	16.48	1.15	0.20	1.50	8.57	0.50
Kaolin	53.5	32.78	0.47	0.38	0.15	0.47	12.18
Sand	98.38	0.92	0.40	0.04	0.06	0.20	0.00
Talc	61.36	0.63	0.83	30.65	0.08	0.03	6.42
Wollastonite	51.82	1.01	44.30	1.13	0.20	0.32	1.22
Limestone	0.00	1.47	53.90	0.00	0.00	0.00	44.64
Soda ash	0.00	0.00	0.00	0.00	57.20	0.00	42.50
Dolomite	0.20	0.38	26.10	24.32	0.00	0.00	66.10
Alumina	0.00	1.70	32.00	0.00	0.00	0.00	66.10

* LOI = Loss on ignition

For determination of raw materials quantity, the user must specify: raw materials used for glazes production and the bases oxides that are used for calculations. The results, the quantity of raw materials are available in a text file, „compmp.txt”

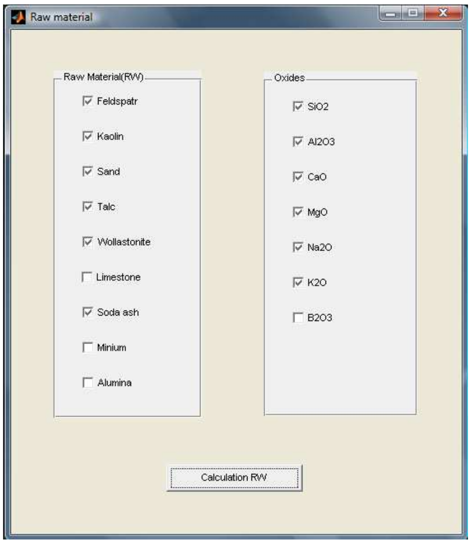

Figure 8. The calculation of composition of mixture of raw materials

In order to validate the software application, the raw materials used and calculated formula for a glaze was determinate, Table 3.

Table 3. Raw materials for glazes

Raw Material	Kg/Kg
Feldspatr	0.5389
Kaolin	0.0843
Sand	0.1808
Talc	0.1075
Wollastonite	0.0695
Soda ash	0.0693

The raw materials database of software application developed could be improved by adding new components and new oxides according to Seger formula.

CONCLUSIONS

The software application allows a fast determination of glazes or glass properties with a known composition, the optimization of glazes composition using the pre-definite properties and calculation of necessary of raw materials using the composition of glazes and available raw materials.

The main importance of this software application is to decrease the calculation time and the number of experiments which must be achieved for obtaining an optimal composition glaze.

The developed software application is helpful for students, chemical engineering, and scientific researchers who work in oxide materials science and not only. The using of application reduces significantly the necessary time for preparation of fabrication recipe.

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