COLOUR IN OPAQUE CERAMIC GLAZES: FORMULATION WITH KUBELKA-MUNK MODEL

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ABSTRACT

In this study the efficiency of the Kubelka-Munk model has been evaluated to predict the colour of an opaque ceramic glaze obtained by a mixture of black pigment (spinel Ni-Fe-Cr) and zircon opacifier ($ZrSiO_4$). After the physical and chemical characterization of the glaze components (frit, pigment and opacifier) suggestions were made for the adaptation of the Kubelka-Munk model to facilitate the experimental analysis procedure. The reflectance curves predicted by the adapted Kubelka-Munk model were in good agreement with the experimental reflectance curves, with deviations of less than 2,0%. Thus, the proposed Kubelka-Munk model permits reproducing a desired colour and evaluating its behaviour with the concentration of the added pigments.

1. INTRODUCTION

The formulation, as well as the adjustment of glaze colour in the tile industry is made using the CIELab system, through the measurement of the L*, a*, b* parameters. However, this system has some limitations. In fact there is no systematic relation between the L*, a*, b* values and the concentration of the added pigments. The Kubelka-Munk model allows relating colour with the concentration of the added pigments. In this work the efficiency of the Kubelka-Munk model has been evaluated for the colour of opaque ceramic glazes (zircon) obtained by a mixture of black pigment (spinel Ni-Fe-Cr) and zircon opacifier ($ZrSiO_4$). In particular the model was adapted considering the peculiarity of the studied system. The opaque glaze was considered one component of the mixture in the Kubelka-Munk (Equation 1):

$$\left(\frac{K}{S}\right)_M = \frac{c_g K_g + c_p K_p + c_o K_o}{c_g S_g + c_p S_p + c_o S_o} = \frac{(1-R)^2}{2R} \qquad \text{(Equation 1)}$$

where the subscripts *g*, *p* and *o* refer to glaze, pigment and opacifier respectively. Taking into account that the scattering of the opaque glaze and that of the opacifier addition is the same ($S_g=S_o=1$):

$$\left(\frac{K}{S}\right)_M = \frac{c_g K_g + c_p K_p + c_o K_o}{c_g + c_o + c_p S_p} \qquad \text{(Equation 2)}$$

To determine the coefficients of Equation 2 four samples are needed: one sample prepared only with the glaze, one sample with a fixed percentage of the opacifier, one sample with a fixed percentage of pigment and one sample with a mixture of pigment and opacifier. Hence, with these four samples it is possible to determine the coefficients of Equation 3 and to predict the colour of the black glazes as a function of the concentration of black pigment (spinel) and the opacifier added in the glazes.

2. PROCEDURE

The coloured glazes were prepared by wet milling of 92 wt% frit, 8 wt% kaolin and different wt% of pigment spinel Ni-Fe-Cr (0.5; 1.0; 2.5; 3.0 and 5.0) and zircon opacifier (0.5; 1.0; 2.5; 3.0 and 5.0). Cylindrical samples of the glazes (Figure 1) were prepared pressing the powder (6 wt% water) with a laboratory press. The samples were fired in a semi-industrial kiln at 1175°C ± 10°C with a cycle of 35 minutes. The reflectance curves and the L*,a*,b* parameters were measured by a Datacolor Spectraflash 600 spectrophotometer with geometry d/8, illuminant D65 and observer at 10°.

Figure 1. Example of the black glazes prepared.

3. RESULTS

The L*,a*,b* coordinates of the coloured glazes are shown in Table 1. The L* parameter decreases as the pigment concentration is increased, as expected. The a* and b* parameters, in contrast, have random changes that are difficult to interpret, underlining the difficulty of using these parameters for the formulation of colours. The reflectance curves predicted by the Kubelka-Munk model are shown in Figure 2. Excellent agreement is observed between the experimental curves and the model curves. The deviations are less that 2.0%.

Ni-Fe-Cr Pigment	L*	a*	b*
0.5	65.7	- 0.2	- 3.4
1.0	59.6	- 0.2	- 3.8
2.5	45.2	0.1	- 4.4
3.0	43.6	0.1	- 4.5
5.0	33.9	- 0.4	- 3.8

Table 1. L, a*, b* values of the coloured glazes with different percentages of black pigment.*

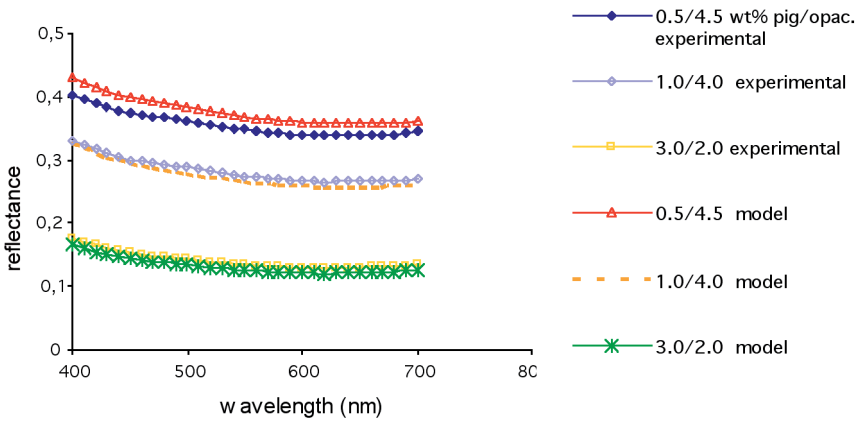

Figure 2. Reflectance curves obtained experimentally compared with those determined by the Kubelka-Munk model of the prepared coloured glazes.

4. CONCLUSIONS

Even though the L*, a*, b* parameters are widely used in the control and formulation of colour in ceramic glazes, it is difficult to evaluate their behaviour with the concentration of pigments added. With the proposed Kubelka-Munk model it is possible to relate colour with the concentration of the pigment and added opacifier, and predict the resulting colour with a good accuracy.

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