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Robert Ianoş, Radu Lazău, Roxana Băbu□ă, Eliza Muntean, Elena-Alina Moacă, Cornelia Păcurariu

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#### ACCEPTED MANUSCRIPT

# Solution combustion synthesis: a straightforward route for the preparation of chromium-doped lanthanum aluminate, LaAl<sub>1-x</sub>Cr<sub>x</sub>O<sub>3</sub>, pink red pigments

Robert Ianoș<sup>a,\*</sup>, Radu Lazău<sup>a,\*</sup>, Roxana Băbuță<sup>a,b</sup>, Eliza Muntean<sup>a</sup>, Elena-Alina Moacă<sup>a,b</sup>, Cornelia Păcurariu<sup>a</sup>

a. Politehnica University of Timişoara, Faculty of Industrial Chemistry and Environmental Engineering, P-ţa Victoriei No. 2, Timişoara 300006, Romania b. "Victor Babes" University of Medicine and Pharmacy Timişoara, Faculty of Pharmacy, Eftimie Murgu Square No. 2, Timişoara 300041, Romania

\*Corresponding author: robert\_ianos@yahoo.com; radulazau@gmail.com

#### **Abstract**

The high exothermicity of the combustion reactions in the  $La(NO_3)_3$ – $Al(NO_3)_3$ – $Cr(NO_3)_3$ –urea–glycine system allows the obtaining of  $LaAl_{1-x}Cr_xO_3$  (x=0.05) pink red pigments directly from the combustion reaction, without any additional annealing. The  $Al^{3+}$  by  $Cr^{3+}$  partial substitution causes a decrease in the combustion temperature from 1721 °C to 1563 °C. The presence of 2 %  $CaF_2$  mineralizer causes colour intensification and red hue increase. The  $LaAl_{1-x}Cr_xO_3$  crystallites size varies between 51 nm and 71 nm and the BET specific surface area varies between 3.0 m²/g and 7.3 m²/g. The characteristics of the obtained pigments are correlated with the combustion gases release rate, which in turn depends on the combustion temperature, the resulting volume of gases and the duration of the combustion reactions. Pigment testing has shown that it dissolves in the enamel melt but can be used to obtain water-based acrylic paints.

Keywords: combustion synthesis; pigments; red; pink; perovskite

#### 1. Introduction

LaAlO<sub>3</sub> perovskite is a highly valued material due to its excellent chemical and thermal stability up to 2100 °C, high dielectric constant and low toxicity [1-3]. Thus, it is currently used as host crystalline network for ceramic pigments [4,5] and pigments with high near-infrared reflectance [6,7], phosphor material [8,9], electrolyte [10] and electrode material [11] for SOFCs or catalytic material [12,13].

There is a growing need for pink red pigments in the ceramic industry and scientists developed therefore a number of pink pigments, such are: zircon-based [14], Cr-doped cassiterite  $(Sn,Cr)O_2$  [15], Ca,Co-pyroxenes [16], Cr-doped malayaite CaSnSiO<sub>5</sub> and sphene CaTiSiO<sub>5</sub> [17-20], Co-doped tialite  $(Al_2TiO_5)$  [21], pink ruby  $(Al_2O_3:Cr)$  [22,23], Fe-ZrSiO<sub>4</sub> [24], Al<sub>2</sub>O<sub>3</sub>:Mn [25] and Cr-doped spinel solid solutions  $M(Al_{2-x}Cr_x)O_4,M=Mg$ , Zn (for x up to 0.4) [26].

The use of such pink red pigments is mainly oriented towards low firing glazes, up to  $1200^{\circ}$ C [15,19,24,26]. There are very few information presented in the literature regarding the synthesis and properties of Cr-doped LaAlO<sub>3</sub> pigments. For instance, Ianoşev et al. [4] obtained the LaAl<sub>0.95</sub>Cr<sub>0.05</sub>O<sub>3</sub> perovskite pigment via thermal decomposition of the complex combinations, with 1% CaF<sub>2</sub> addition, after annealing at 1200 °C. Another group of authors [5] obtained a similar pigment by flame-spraying synthesis, starting from oxides and annealing the sprayed product up to

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