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Synthesis and characterization of a new ceramic pigment based on the pyroxene structure

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The traditional and unavoidable source of the different tone from blue to pink in the ceramic pigments is usually ionic divalent cobalt. This element maintains an optimal colouring performance that depends on thermal and chemical stability. Recent focus was pointed to the synthesis of ceramic pigment based on cobalt in order to minimize the environmental impact (cobalt is hazardous and toxic) and the production cost (cobalt is both scarce and expensive).

The coordination chemistry of cobalt complexes has fascinated chemists for a long time due to the spectacular range of colours they produce. The colour variation depends on the different coordination number: when cobalt is in tetrahedral coordination (as in cobalt spinel CoAl2O4, the well known "cobalt blue") the colour is a deep blue, while when the coordination number is higher, six or eight (as in Co-olivine Co2SiO4 or in pyroxene CaCoSi2O6) the hue becomes pink or violet.

Since now little attention has been paid to Co pyroxenes as inorganic pigments. Co-pyroxenes are solid solution along the join CaCoSi2O6 - Co2Si2O6. This series is interesting in ceramic field, both for lower cobalt content in formula, respect to olivine and spinel, and for the remarkable stability to the alteration, which makes it useful also for the immobilization of cobalt containing sludge. The pyroxene structure allows a replacement in two sites M1 and M2 at different coordination, being M1 a regular octahedron and M2 a distorted polyhedron with 6-8 fold coordination.

In order to verify the potential use in ceramics pyroxenes along the series CaCoSi2O6 - Co2Si2O6 were therefore synthesized, at room pressure and characterized in order to obtained the best condition for producing a pink pigment. The syntheses were done between 1000 and 1200°C with ceramic route, and annealing from 2 to 96 h.

SEM-EDS, XRD and Raman analysis show that for Co-poor samples and temperatures lower than 1100°C the presence of Co-akermanite (Ca2CoSi2O7), cristobalite and Ca,Co-pyroxene while in Co-rich samples we found Co-olivine (Co2SiO4), cristobalite and Co-pyroxene. This is also confirmed by the color analysis: from the samples with the composition CaCoSi2O6 to Ca0.5Co1.5Si2O6 we have a change in the colorimetric parameter that correspond to the presence of the deep blue hue of Co-akermanite (where the cobalt is in tetrahedral coordination). At temperature higher or equall 1150°C, Co-akermanite disappeared. Between CaCoSi2O6 and Ca0.8Co1.2Si2O6 the pyroxene is the only phase present while in Co-rich samples there is the coexistence of Co-olivine, Ca,Co-pyroxene and cristobalite. Colorimetric analysis was used to clarify the relation between phase content, composition and the resulting colour.

The possible use of Co-pyroxene as a pink-purple ceramic pigment has been discussed.