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Control of optical properties of oxynitride pigments through stoichiometries

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We have demonstrated that a color of the perovskite-type $LaTiO_2N$ oxynitride could be tuned from orange through yellow and green to white by proper adjustment of O/N and Ti/La ratios. However, we have not obtained real red oxynitrides in this system yet. $LaTaON_2$, which contains more nitrogen amount than $LaTiO_2N$, can be expected as the redder pigment. However, the ionic size of La^{3+} seems to be so small for accommodating the ideal perovskite that the crystal structure may be deformed to be monoclinic system. Distortions to bond angle of Ta-(O,N)-Ta were reported to make the bandgap wider. In this study, we prepared solid solutions of $La_{1-x}Ba_xTa(O,N)_3$ and the compositional variation of optical properties as well as structural ones were examined. We also studied effects of addition of NaCl flux during nitridation on the color due to particle size distributions. Perovskite-type $La_{1-x}Ba_xTa(O,N)_3$ oxynitrides showed slight redshift of absorption edge and deteriorated reflectivity in the longer wavelength regions after absorption edge with increasing Ba content x due to relaxing Ta-(O,N)-Ta distortion. Addition of NaCl flux in the oxide precursor of $La_{1-x}Ba_xTa(O,N)_3$ during the nitridation led to improvement of the reflectivity after absorption edge without changing the anion ratio. $LaNbON_2$ should be one of the redder pigments. Substitution of titanium by niobium in $LaTiO_2N$ would enhance covalency in bonding between the cations and anions because electronegativity of niobium is slightly larger than that of titanium. We also examined anion composition and optical properties of solid-solution of $LaTi_{1-x}Nb_x(O,N)_3$ and will discuss difference in ways of variation of optical properties against the substitutions mentioned above. Perovskite-type $LaTi_{1-x}Nb_x(O,N)_3$ oxynitrides showed redshift of the absorption edge due to enhancement of covalent character and increased absorption in red region due to anion defects, with increasing Nb content x. The absorption would possibly affect the gradient of reflectivity curve after the absorption edge to assist to show the redder color. The oxynitride solid-solutions with x=0.2 and 0.3 possessed the color coordinate same as that of red iron oxide.

Biography

Toshihiro Moriga graduated from Department of Chemistry, Osaka University in 1988 and received his Doctor of Science from Osaka University in 1996. He is now a Professor of Department of Advanced Materials, Institute of Technology and Science, Tokushima University, Vice-dean of Faculty of Engineering of the University and Director of Center for International Cooperation in Engineering Education of the Faculty. He has published more than 130 papers in peer-reviewed journals and an organizer of the special session, “Advanced structure science and pioneering novel materials” in fall meetings of the Ceramic Society of Japan since 2007.

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