別紙第１号様式　　No.1

博　士　論　文　の　要　旨

専攻名 システム創成科学専攻

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博士論文題名 Upconversion Luminescence in ZnO-TiO_2 Composite Doped with Rare Earth Elements

（希土類元素をドープした ZnO-TiO_2 複合体のアップコンバージョン蛍光）

要旨（２，０００字程度にまとめること。）

In this dissertation, the upconversion (UC) phosphor fabricated using a new UC host material, ZnO-TiO_2 composite, is implemented. The practicable routes namely solid-state reaction and powder-solution mixing methods are contemplated for the synthesis of ZnO-TiO_2 composite UC phosphor activated with rare earth elements. This dissertation comprises of six chapters that cover essential background of UC phosphors, crucial aspects of three basic sensitizer-activator pairs of ions which are normally used for making UC phosphors, and the interesting and important results regarding the site preference of rare earth ions in the UC host material under the situation that the crystal structure of UC host material is the complex crystal matrix and the Rietveld refinement cannot be applied to estimate the substitution position of rare earth ions within target host crystal matrix. Initially, a brief and general background of phosphors, focusing mainly on fundamental knowledge of UC phosphor, mechanism models of the generation of UC luminescence (UCL), and the methods of preparation of UC phosphors is given in the first chapter. For the detailed research works, there are two main topics based on the synthetic route which can be described as follows.

Firstly, the pellet form of Ho^{3+} and Yb^{3+} co-doped ZnO-TiO_2 composite phosphor prepared by solid-state reaction method was successfully synthesized at 1200 °C for 4 h. The effect of synthesis parameters like UCL characteristics, ZnO/TiO_2 mixing ratios, and Ho^{3+} and Yb^{3+} concentrations were studied in detail. The most efficient product, ZnO:TiO_2 = 1:1 (in mole) doped with 0.05 mol% Ho^{3+} and 9 mol% Yb^{3+}, exhibited the green emission under a 980-nm laser excitation. The UC mechanism of this UC phosphor system was also suggested and discussed in detail based on the pump power dependent studies.

Secondly, the powder form of ZnO-TiO_2 composite phosphors containing different pairs of rare earth ions (Er^{3+}/Yb^{3+}, Ho^{3+}/Yb^{3+}, and Tm^{3+}/Yb^{3+}) prepared by a new synthetic method, namely powder-solution mixing method, was successfully carried out. In this research, the effect of firing temperature, ZnO/TiO_2 mixing ratio, and dopant concentration ranges on structural and UCL properties under a 980-nm laser excitation was investigated. The simple chemical formula equations, for explaining the site preference of rare earth ions in host crystal matrix, were generated by considering the target host crystal structure, its crystal properties, and the effect of dopants to its crystal matrix. Under optimum firing temperature (1300 °C) and fixed firing time (1 h), the most effective product of these three systems was ZnO:TiO_2 = 1:1 (in mole) doped with 3 mol% Er^{3+} and 9 mol% Yb^{3+} emitting bright red emission, ZnO:TiO_2 = 1:1 (in mole) doped with 0.03 mol% Ho^{3+} and 9 mol% Yb^{3+} exhibiting bright green emission, and ZnO:TiO_2 = 1:1 (in mole) doped with 0.125 mol% Tm^{3+} and 15 mol% Yb^{3+} emitting nearly pure NIR emission. The dependence of UC emission intensity on the excitation power was studied, and the UC mechanism was proposed and discussed in detail. Additionally, the comparison of UC emission intensity of the phosphor prepared by different synthetic methods was also investigated.

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In conclusion, it is believed that ZnO-TiO_2 composite phosphors containing Er^{3+}/Yb^{3+}, Ho^{3+}/Yb^{3+}, and Tm^{3+}/Yb^{3+} pairs of ions have the potential to be an effective UC phosphor not only because of the good properties of target host material but also the emission color and emission intensity observed. In addition, the new preparation method, powder-solution mixing, provides the homogeneous phase of the product which gives rise to the higher regularity of the distribution of UC emission intensity than the product synthesized by solid-state reaction method. Hence, this method might be one of the appropriate techniques for the fabrication of efficient UC phosphors.