Multicolor and white light emission of GdPO₄ nanophosphors with a novel 3D architecture

Solid state lighting in the form of light emitting diodes (LEDs) has emerged in recent years as a high competent field and is therefore a subject of intensive research. LEDs combined with a phosphor can be used to produce visible light. Most white LEDs in production today are modified blue LEDs coated with a vellow emitting phosphor (YAG:Ce) [1]. However, such white LEDs present a low color rendering index and no full range of visible light. Another approach involves using a UV LED chip coated with three emitting blue, green, and red phosphors to generate warm white light. This, however, has a drawback that the blue emission efficiency is poor because of the strong reabsorption of the blue light by the green and red phosphors. The development of a single-phased white-lightemitting phosphor operating under UV excitation is of prime importance for a better performance of such LEDs. More importantly, compared to the multiple emitting components of the white LED system, the single-phased white-light-emitting phosphor for a UV-pumped white LED would enable easy fabrication with perfect stability and color. On the other hand, there has been recently an increase in interest and research into nanophosphors based on rare earth (RE) compounds including LnF₃[2], Ln₂O₃[3], Ln(OH)₃[4], LnVO₄[5] and LnPO₄[6], to mention a few. Among the large number of lanthanide compounds, lanthanide orthophosphates (LnPO4) represent an important class of materials because they possess a variety of favorable properties such as very high thermal stability $(2300 \degree C)$, low solubility in water (Ksp = $10^{-25} - 10^{-27}$) and high refractive index (n=1.5) that make them very useful in a large variety of applications as luminescent or laser materials, magnets, ceramics, catalysts, and also in medicine. The present contribution shows the synthesis and characterization of a single-phased GdPO₄ phosphor with a novel morphology, which emits white light under UV excitation.

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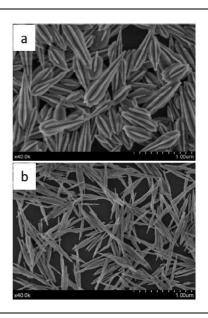


Figure 1: SEM micrographs of twin –like and rod-like GdPO4 particles synthesized using an EG/H_2O ratio of 90/10 (a) and 80/20 (b).

Homogeneous. monoclinic GdPO₄ particles consisting of three intersecting lance-shaped crystals forming a penetration (Figure 1a) twin were synthesized following a very simple and fast method consisting in the hydrothermal reaction of Gadolinium acetylacetonate with H₃PO₄ in a mixture of ethylene glycol and water at 180°C. The experimental conditions were found to have a strong effect on the morphological and structural features of the prepared particles. Thus, slightly increasing the amount of water in the solvent mixture leads to hexagonal rod-like GdPO₄·0.5H₂O nanoparticles (Figure 1b), while decreasing it only produces an amorphous gel-like precipitate. The synthesis procedure is also successful for the preparation of Eu^{3+} , Tb^{3+} - and Dy^{3+} -doped $GdPO_4$ particles with the same morphology and crystalline structure as the undoped materials. The effect of the doping level on the luminescent properties of the twin-like nanophosphors was evaluated (Figure 2). Likewise, the monoclinic twin-like GdPO4 nanophosphors were found to be much efficient than the hexagonal rod-like GdPO₄ ones in terms of emission intensity, especially the Eu- and Dy-doped particles. Finally, a solid state single-phase white light emitting nanophosphor has been fabricated for the first time in this system by triply doping the monoclinic GdPO₄ twined particles with appropriate concentrations of Eu³⁺, Tb³⁺ and Dy³⁺ (Figure 3).

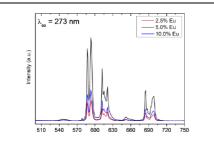


Figure 2: Emission spectra of different Eu-containing twin-like GdPO₄ nanophosphors recorded after exciting at 273 nm

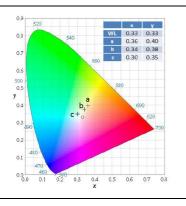


Figure 3: CIE chromaticity diagram of Eu/Tb/Dy triply doped twined GdPO4 particles and table displaying the corresponding CIE coordinates (Eu/Tb/Dy = 1.0/1.0/1.0 (a), 1.0/0.7/1.0 (b) and 0.5/0.7/1.0 (c).

References

- Bachmann, V.; Ronda, A.; Meijerink, C. Chem. Mater., 21 (2009) 2077.
- [2] Lorbeer, C.; Cybinska, J.; Mudring, A.V. Chem Comm. 46 (2010) 571.
- [3] Pedersen, H.; Ojamae, L. Nano Lett. 6 (2006) 2044.
- [4] Mu, Q.Y.; Wang, Y.D. J. Alloys Comp. 509 (2011) 2060.
- [5] Thirumalai, J.; Chandramohan, R.; Vijayan, T.A. Mater. Chem. Phys. 127 (2011) 259.
- [6] Rodriguez-Liviano, S.; Aparicio, F.J.; Rojas, T.C.; Hungria, A.B.; Chinchilla, L.E.; Ocaña, M. Cryst. Growth Design 12 (2012) 635.