

Review on Luminescence Spectroscopic Studies on Dy³⁺ Doped Nano Phosphors

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Abstract

In order to assess the luminescence spectroscopy experiments on Dy³⁺ doped nano phosphorus and metal oxides, this work includes a comprehensive literature analysis of final-results and discussion of prior research. In this review work, we take a look at what's already been written about Dy³⁺ doped nano phosphorus and metal oxides. The collected sample was analysed by means of Powder X-ray Diffraction (PXRD), Fourier Transform Infrared Spectroscopy (FTIR), Optical Absorption (AOD), and Photoluminescence (PL). In metal oxides, Characterization techniques gives information about quality and composition of the samples, they exhibit unique PL spectra depending on impurities, crystal defects and mineral associations. These samples may be used in mining companies to assess the economic viability. Compared to an undoped phosphorus PL spectrum, a Dy³⁺ doped Nano phosphorus PL spectrum shows a blue emission at 441 nm (2.81 eV). Flux was used in a solid-state reaction to create a phosphor that emits white light and contains Dy³⁺ ions. Analysis of the manufactured sample was performed using Powder X-ray Diffraction (PXRD), Optical Absorption (AOP), Photoluminescence (PL), and Fourier Transform Infrared Spectroscopy (FT-IR). Intense crystal fields and Zeeman interactions can affect Dy³⁺ ions, as evidenced by their abundant resonance signals in EPR spectra.

Keywords: Luminescence Spectroscopic, Nano Phosphorus, Trivalent Dysprosium Ion, X-ray Diffraction

1.0 Introduction

Rare Earth (RE) ion-based fluorescent materials have become increasingly important in recent years in domains such as lighting, solid-state lasers, bioimaging, etc. There have been numerous publications on White Light-Emitting Diode (W-LED) materials as researchers continue to explore materials doped with RE ions. Doping rare earth ions into a variety of inorganic host materials causes them to become an efficient luminophore and to function as efficient emitters in both the trivalent and divalent states due to the f-f and f-d transitions¹.

Phosphors doped with Dy³⁺ ions display white light emission with two peaks in the blue and yellow areas due to the f-f transitions characteristic of these ions. The luminous properties of these materials have been the subject of a great deal of research, but it is still appealing to create effective Dy³⁺ doped phosphors with excellent thermal stability for use in W-LEDs².

For this study, we used the standard melt method to introduce Dy³⁺ doping into phosphors at varying concentrations. The intensity and decline curves of blue and green emissions across their lives have been studied extensively. Under 350nm stimulation, the mechanism of

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interest has been studied, and a reasonable explanation has been proposed. Spectral sample CIE coordinates were calculated.

The luminescent cores of hosts are activated by activators in luminescent materials. When the Dy³⁺ ion is in the geometric centre of the symmetrical material, multilevel interactions are the primary source of luminescence, and the luminescent material emits white light of the usual spectrum when the transitions are of equal strength in both locations. Dy³⁺ has the ability to emit a wide range of wavelengths between its f-f transitions, making it one of the promising rare earth ion with the most diverse range of possible applications. Its distinctive colors blue and yellow intense emission bands in the visible range correspond to transitions $4F_{9/2}-6H_J$ ($J=15/2,13/2$) and play a crucial role in the fabrication of white light emitting phosphors. Optimizing the local field environment around Dy³⁺ ions with a sufficient yellow to blue (Y/B) intensity ratio is crucial to achieving white light emission. Luminescence studies on $\text{Li}_2\text{Sr}_2\text{Al}_2\text{PO}_4\text{F}_9:\text{Dy}^{3+}$ phosphor have been reported by Shinde *et al.*, photo luminescence characteristics of Dy³⁺ doped $\text{KLa}(\text{PO}_3)_4$ phosphor have been reported by Chemingui *et al.*, Ratnam *et al.* have reported white light emitting $\text{NaCaPO}_4:\text{Dy}^{3+}$ phosphor, and Zhang *et al.* have reported KSrBP_2O_8 . Sameway in metal oxides, PL spectra plays a very important role by analyzing the progress of beneficiation process by assessing the changes in the PL spectra, and from PI optimization mining operations can achieve maximum efficiency and minimize the waste.

2.0 Phosphor Preparations

Different techniques, such as solid-state reaction, solution combustion, sol-gel, etc., are used to generate Dy³⁺ doped phosphor. After the raw components were measured out using stoichiometric calculations for phosphors, they were thoroughly combined using an appropriate fuel. The chemical formula was used to determine the stoichiometric ratio of the reactants used to produce phosphor. Then after the weighed quantity of starting samples were placed in 25ml alumina crucibles and introduced into the programmable furnace for 3 hours. When the furnace has cooled to room temperature naturally, the finished product is extracted⁶.

The XRD spectra of the samples were recorded employing an X-ray powder diffractometer and a CuK α radiation source to determine the material's physical phase and structure of the Dy³⁺ doped phosphors⁷. The transmission spectra of Dy³⁺ doped phosphors were measured using a UV spectrophotometer to learn more about the samples absorption at various wavelengths. In this review, we looked at a spectrum spanning from 200 to 820 nm in wavelength. The F4000 fluorescence spectrometer was used to examine the phosphors, while the FS5 model was used to evaluate the fluorescence lifetimes of the samples while being pumped by a xenon. This review presents the results of research into the structural and optical properties of phosphors that has been doped with Dy³⁺ ions. To this end, we will use powder X-ray diffraction to gain insight into the crystal structure and evaluate the lattice cell parameters, Fourier Transform Infrared Spectroscopy (FTIR) results the identification of the local structural groups, absorption spectroscopy to determine the Judd-Ofelt parameters, which will allow us to calculate the bonding and others parameters, photoluminescence spectroscopy to estimate the radiative properties and lifetime of the material^{8,9}.

3.0 Experimental Section

Here we have taken few experimental results which researchers have already published to understand the mechanism better. The solid state reaction technique was used to create a phosphor containing Dy³⁺ ions doped $\text{NaCaAlPO}_4\text{F}_3$ by choosing chemicals based on the stoichiometric calculations. Without further purification, all of the chemical reagents which they have employed in

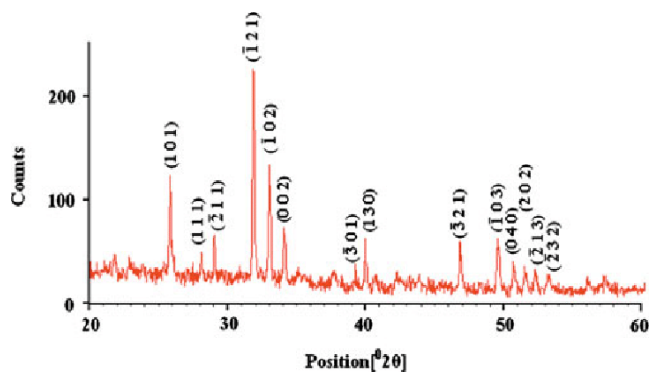


Figure 1. X-ray diffraction pattern of Dy³⁺ doped $\text{NaCaAlPO}_4\text{F}_3$ phosphor.

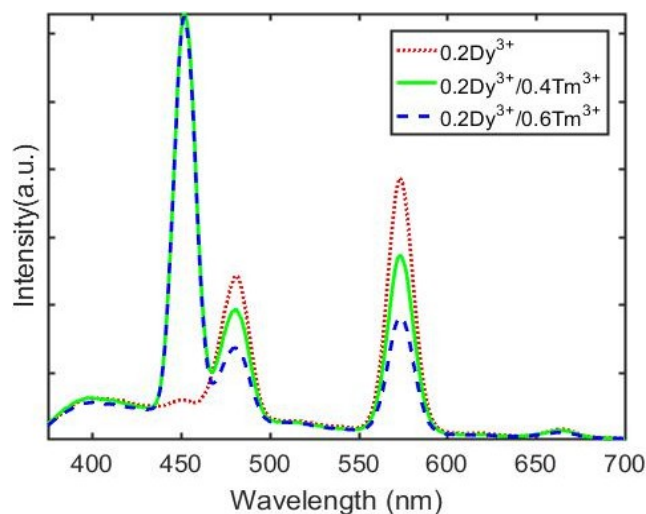


Figure 2. Emission spectra for different concentrations of $\text{Tm}^{3+}/\text{Dy}^{3+}$ co-doped phosphate glasses.

this experiment were of analytical grade. They used an agate mortar and pestle to reduce the beginning ingredients (Na_2CO_3 , $\text{AlF}_3 \cdot 3\text{H}_2\text{O}$, CaCO_3 , and $(\text{NH}_4)_2\text{HPO}_4$) to a fine powder over the course of 30 minutes. An hour was spent grinding the aforesaid concoction in an agate mortar after Dy_2O_3 was added. The acquired chemical mixture was sintered at different temperatures for different hours in a high temperature furnace with multiple intermediate grindings for 30 minutes. Finally, $\text{NaCaAlPO}_4\text{F}_3$ phosphor

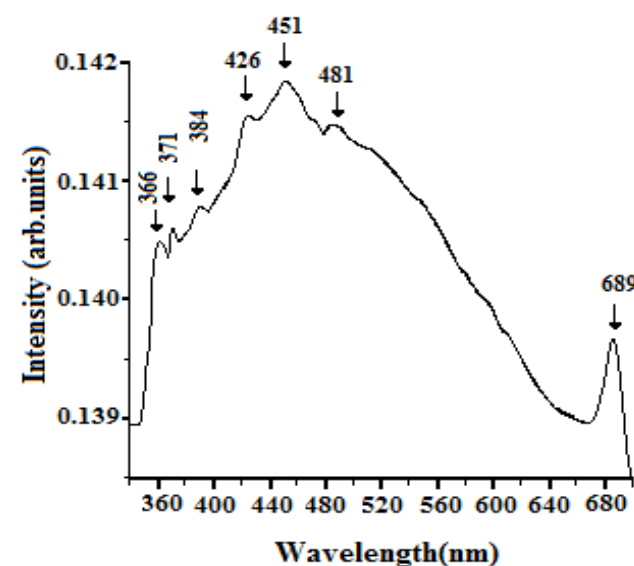


Figure 3. UV-Visible absorption spectrum of Dy^{3+} ions doped $\text{NaCaAlPO}_4\text{F}_3$ phosphor.

was obtained by doping it with Dy^{3+} ions and $3\text{H}_2\text{O}$ was employed as a flux^{10,11}.

Multiple methods were used to determine the precise nature of the produced phosphor. The average crystallite size of Dy^{3+} doped phosphor is calculated to be 82 nm using the Debye-Scherrer formula. The created phosphor material is a well crystalline substance, as seen by the crisp and intense peaks, which can lead to high luminous intensities¹⁴ that the Dy^{3+} doped phosphors are crystalline in design^{12,13}. From the XRD spectra, there are exceptionally sharp diffraction peaks of the samples, suggesting that the Dy^{3+} doped phosphors are crystalline in design^{12,13}.

Figure 2 reveals the emission spectra at 453nm ($^1\text{D}_2 \rightarrow ^3\text{F}_4$), although the 480nm, 573nm, and 665nm emission intensities, which can be attributable to Dy^{3+} , all reduced considerably as the concentration of Dy^{3+} ions was raised, the opposite was true for Tm^{3+} . Dy^{3+} is responsible for the energy transfer, as shown.

At 352nm, 682nm, and 794nm, corresponding to the $3\text{H}_61\text{D}_2$, 3F_3 , and 3H_4 transitions, respectively, it is evident that the light is being significantly absorbed. Absorption of UV radiation at 352nm is particularly strong. Dy^{3+} transition (absorptions at 348nm, 365nm, 387nm, and 800 nm): $6\text{H}_{15/2}$ ($4\text{M}_{15/2}$, $6\text{P}_{7/2}$), $6\text{P}_{5/2}$, $4\text{I}_{13/2}$, and $6\text{F}_{5/2}$. Clear increases in 348nm UV absorption strength can be seen¹⁵.

Figure 3 demonstrates the visible and near-infrared (NIR) optical absorption spectra of a $\text{NaCaAlPO}_4\text{F}_3$ phosphor sample that has been doped with Dy^{3+} ions.

Mono-doped 0.2 Tm^{3+} and 0.2 Dy^{3+} phosphors' excitation and emission spectra. Intensities of Tm^{3+} blue emission at 453nm and 352nm excitation, it is not difficult to determine that 352nm and 453nm originate from Tm^{3+} transitions ($3\text{H}_6 \rightarrow 1\text{D}_2$ and $1\text{D}_2 \rightarrow 3\text{F}_4$ respectively). The strongest green emission of Dy^{3+} at 573nm and 348nm excitation, corresponding to the Dy^{3+} transitions: $6\text{H}_{15/2} \rightarrow 6\text{P}_{7/2}$ and $4\text{F}_{9/2} \rightarrow 6\text{H}_{13/2}$ respectively. Therefore, UV light at 350nm is the preferred pump source for $\text{Dy}^{3+}/\text{Tm}^{3+}$ doped phosphors¹⁶.

Figure 2 illustrates the mechanics of down-conversion luminescence and a population diagram. Since the energy of the ($4\text{M}_{15/2} \rightarrow 6\text{P}_{7/2}$) level of Dy^{3+} is close to that of the $^1\text{D}_2$ level of Tm^{3+} in $\text{Dy}^{3+}/\text{Tm}^{3+}$ energy resonance transmission between co-doped systems Dy^{3+} and Tm^{3+} ions occur readily¹⁷. The blue (453nm) emission is generated when Dy^{3+} ions absorb energy under 350nm

excitation, populating the excited state ($4M_{15/2} \rightarrow 6P_{7/2}$), and then transmit some of that energy to Tm³⁺ ions in the $3H_6$ state, which are excited to the $1D_2$ energy level. Thus, the fluorescence intensity associated with Dy³⁺ drops and that of Tm³⁺ increases when Tm³⁺ concentration rises.

The blue (480nm), green (573nm), and red (665nm) emission is enhanced by increasing Dy³⁺ concentration, whereas the 453nm ($1D_2 \rightarrow 3F_4$) emission of Tm³⁺ decreases to do. From this, we can easily conclude that energy transfer from Tm³⁺ to Dy³⁺ occurs. Furthermore, the intensity of Dy³⁺ (blue, green, red) starts to decrease when the concentration of Dy³⁺ changes from 0.7 to 0.8 mol%. Later they have concluded that the increased probability of cross-relaxation of the 0.2 mol% Tm³⁺/0.8 mol% Dy³⁺ ions in the samples caused fluorescence quenching of Dy³⁺. The emission spectra in 0.2Dy³⁺/xTm³⁺ (x=0, 0.4, 0.6) co-doped phosphors. The 453nm ($1D_2 \rightarrow 3F_4$) emission intensity of Tm³⁺ significantly improved with increasing Tm³⁺ ion concentration. In contrast, the 480nm, 573nm, and 665nm emission intensities assigned to Dy³⁺ decreased dramatically. From this, we conclude that the energy is transferred from Dy³⁺ to Tm³⁺ (Iopscience.iop.org, 2022).

Down-conversion light emission mechanism and assembly schematics are depicted. In the Dy³⁺/Tm³⁺ co-doped system, the energy of the ($4M_{15/2} + 6P_{7/2}$) level of Dy³⁺ is close to the energy, facilitating the energy resonance transfer process between the Dy³⁺ and Tm³⁺

ions. Happen $1D_2$ state lies in Tm³⁺. On this side, the Dy³⁺ ion absorbs energy under 350 nm excitation, occupies the excited state ($4M_{15/2} + 6P_{7/2}$), transfers part of the energy to the Tm³⁺ ion in the $3H_6$ state, and into the $1D_2$ state. Finally, the Tm³⁺ ion at the $1D_2$ energy level relaxes by radiation to the intermediate $3F_4$ state, producing blue (453 nm) emission. Therefore, increasing Tm³⁺ concentration decreases the fluorescence intensity associated with Dy³⁺ and increases the fluorescence intensity of Tm³⁺.

The fluorescence lifetimes of Tm³⁺ emission at 453 nm and Dy³⁺ emission at 573 nm were each tested under 350 nm excitation. They have reported average decay time (τ) and energy transfer efficiency (η) were calculated as $0(2) \cdot \tau$. As the number of Tm³⁺ ions in a sample grows, the ion's fluorescence lifetime shortens from 0.80 ms to 0.61 ms. As this is the case, we can conclude that Dy³⁺ transfers its energy to Tm³⁺. Lifetime Diminution of Luminescence Due to $1D_2 \rightarrow 3F_4$ (Tm³⁺) at room temperature. By increasing the concentration of Dy³⁺ in the sample, the fluorescence lifetime decreases from 19.42 μ s to 9.80 μ s. This provides evidence for energy transfer from Tm³⁺ to Dy³⁺ (14-16).

4.0 DRS Spectra for Dy³⁺

The Figure 4 and 5 displays the translated absorption data alongside the original DRS spectra of produced compounds $Sr_{1.75}Ca_{1.25}(PO_4)_2$ doped with different concentrations of Dy³⁺ ions. The f-f electronic transitions

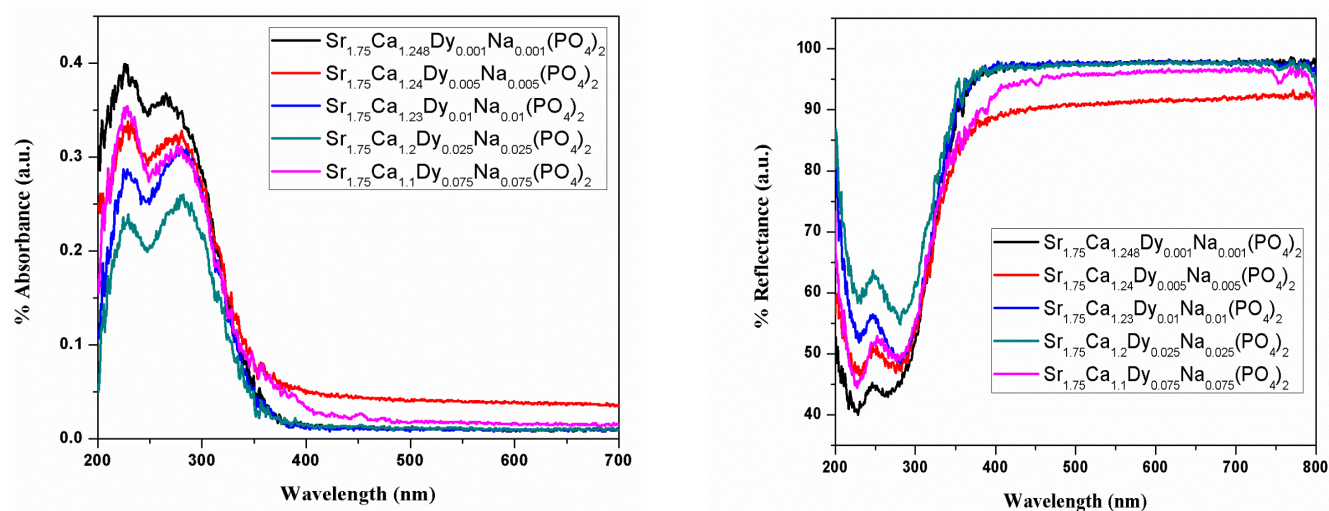


Figure 4. DRS spectra of $Sr_{1.75}Ca_{1.25}(PO_4)_2$ doped with different concentrations of Dy³⁺.

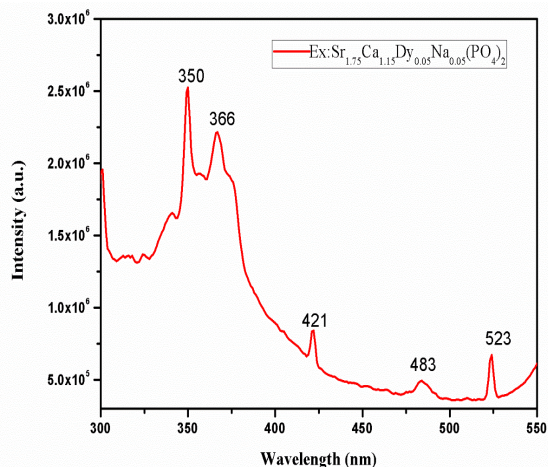


Figure 5. DRS spectra (% of Abs Vs. wavelength (nm) of $\text{Sr}_{1.75}\text{Ca}_{1.25}(\text{PO}_4)_2$ doped with different concentrations of Dy^{3+}

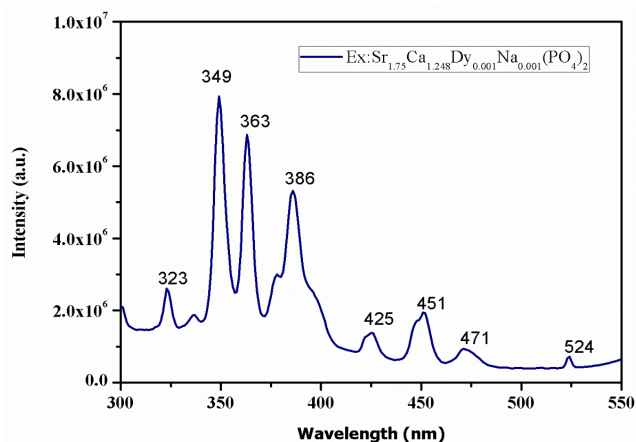


Figure 8. The emission spectrum of $\text{Sr}_{1.75}\text{Ca}_{1.15}\text{Dy}_{0.05}\text{Na}_{0.05}(\text{PO}_4)_2$ under 350 nm excitation.

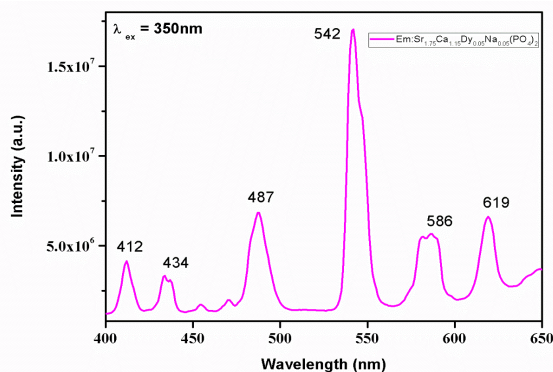


Figure 6. Photoluminescence Study of $\text{Sr}_{1.75}\text{Ca}_{1.25}(\text{PO}_4)_2$: $x\text{Dy}^{3+}$.

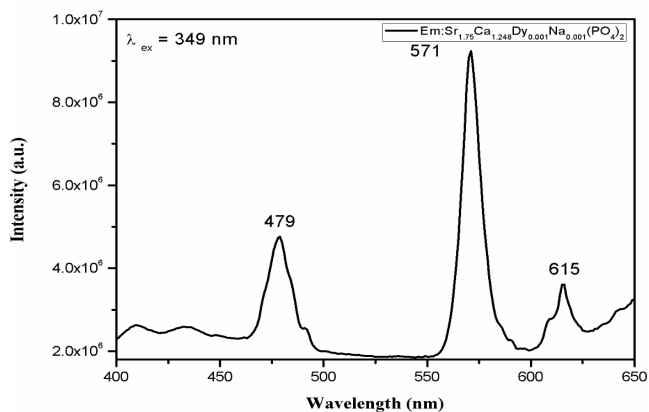


Figure 7. The excitation spectrum of $\text{Sr}_{1.75}\text{Ca}_{1.248}\text{Dy}_{0.001}\text{Na}_{0.001}(\text{PO}_4)_2$

of Dy^{3+} are responsible for the unique absorption band observed in all of the composition¹⁸.

In Figure 6, we see the emission spectra of $\text{Sr}_{1.75}\text{Ca}_{1.25}(\text{PO}_4)_2$: $x\text{Dy}^{3+}$ phosphors when they are excited by UV light at 348nm and 387nm. Similar patterns can be seen in the emission spectra of $\text{Sr}_{1.75}\text{Ca}_{1.25}(\text{PO}_4)_2$: Dy^{3+} , with two major emissions at 481.6 (487) and 575 nm (4F9/2-6H15/2 and 4F9/2-6H13/2 transitions, respectively). Due to its nature as a forced electric dipole transition, the 4F9/2-6H13/2 phase change can only take place when the corresponding Dy^{3+} ions are positioned at local sites with non-inversion centre symmetry. The spectrum of radiation emitted by $\text{Sr}_{1.75}\text{Ca}_{1.25}(\text{PO}_4)_2$: Dy^{3+} shows a yellowish glow when excited^{18,19}.

The structural analysis's conclusion that Dy^{3+} is positioned in a more non-centrosymmetric position in the $\text{Sr}_{1.75}\text{Ca}_{1.25}(\text{PO}_4)_2$ host lattice is supported by the fact that the red emission (4F9/2-6H13/2) is more intense than the blue emission (4F9/2-6H15/2).

5.0 Conclusions

Different stoichiometric preparations of dysprosium-doped phosphors were made in the present study using a solid-state synthesis and co-precipitation and other methods. The above literature was analyzed and outcome was produced compounds using DRS and XRD, with identical results for characterization obtained using both techniques. The researchers also estimated that CIE values indicate a distinct progression of colors from an organo-yellow to a pure white as Dy concentrations are varied.

In conclusion, a high temperature melting procedure was used to successfully create phosphors doped with varying amounts of Dy³⁺/Tm³⁺. Under 350 nm excitation, tunable white light down conversion emission was produced. The energy transfer method is investigated, and the fluorescence decay lifespan and its CIE coordinates at various RE ion concentrations are reported in great detail. The research in the study shed light on how to create a desired color by down-converting emissions in phosphors by adjusting the concentration of RE ions depending on experimental data. Dy³⁺/Tm³⁺ co-doped phosphate glasses show promise for W-led applications under UV stimulation, as seen by changes in their associated CIE color coordinates, making them a potential material for use in solid-state lighting and other sectors²². Unlike in phosphors, PL studies play a key role in applications of metal oxides in mining industry, identification of minerals to ore processing and how it impacts on environment by detecting impurities or trace elements, concentration of impurities, crystal structure and defects within the metal oxides. By all these companies can plan to achieve material with required standards and specifications.

6.0 References

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