


Study on the Persistent Photoluminescence of Trivalent Multiphoton($\text{Ce}^{3+}/\text{Ho}^{3+}$) doped YPO_4 Nanophosphors via Polyol Method

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Abstract: The use of persistent photoluminescence materials in low-light imaging, security labeling, and energy-efficient lighting has attracted much attention. This work examines the long-lasting photoluminescence characteristics of YPO_4 nanophosphors doped with Ce^{3+} and Ho^{3+} trivalent multiphoton ions. These ions' integration brings about distinctive energy shifts that support the glow phenomena. A controlled polyol method was used to create the YPO_4 nanophosphors, guaranteeing consistent doping amounts of Ce^{3+} and Ho^{3+} . The synthesized nanophosphors' crystalline nature was confirmed through X-ray diffraction analysis, and their well-defined nanostructure was revealed by scanning electron microscopy, Fourier-transform infrared spectroscopy (FTIR), UV study, and photoluminescence spectroscopy, which was used to examine their luminescent behavior.

Keywords: polyol method; cerium ion; holmium ion; yttrium ion; photoluminescence; charge transfer.

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1. Introduction

This section discusses how the successful applications of rare-earth (RE)-based materials in high-tech and biomedical fields have sparked increased interest in their research, particularly in the area of RE orthophosphates. The unique properties of RE orthophosphates, such as their good optical properties, restricted solubility, and exceptional chemical and thermal stability, make them valuable in various applications. Compounds such as $\text{YPO}_4:\text{Ce}/\text{Ho}$ have garnered attention in the quest for novel bright materials due to their exceptional optical and structural characteristics. The polyol-synthesized nanophosphor material $\text{YPO}_4:\text{Ce}/\text{Ho}$ has attracted much interest because of its potential uses in various technological fields [1-2]. The usage of YPO_4 doped with $\text{Ce}^{3+}/\text{Ho}^{3+}$ ions is becoming more popular as a viable option for the production of phosphors and scintillators due to its affordability and the fact that the bandgap provides its broad, enough room for the doped lanthanide ions' 4f and 5d orbitals [3-4]. The primary prerequisites for increasing luminescence intensity and scintillation efficiency in any host doped with rare earth ions are as follows: determining the ideal dopant ion concentration (activator) for maximum luminescence in a host material, doping the host material with the right amount of a sensitizer to maximize energy transfer to the activators, the host having low phonon vibrations, the host being transparent in the visible and near-infrared (NIR) regions, the host being chemically and thermally stable in

the ambient atmosphere, and selecting the right solvent/reaction medium. [5-8]. Since there not been many investigations published, we've studied the concentration influence of Ce^{3+}/Ho^{3+} doped YPO_4 host in powder form on their structural and optical properties in this work[9-10]. This work discusses the concentration impact of Ce^{3+}/Ho^{3+} doped YPO_4 production via polyol technique and other characterization and luminescence investigations.

2. Materials and Methods

2.1. Chemicals utilize.

Cerium acetate hydrate $(CH_3CO_2)_3Ce \cdot XH_2O$, Yttrium acetate hydrate, $(CH_3CO_2)_3Y \cdot XH_2O$, Holmium acetate hydrate $(CH_3CO_2)_3Ho \cdot XH_2O$, Hydrochloric acid in concentrated form Ammonium dihydrogen phosphate $(NH_4)_2HPO_4$, NaOH, ethylene glycol, Solvent Deionized water, acetone, a method for synthesizing $YPO_4:Ho^{3+}$ and Ce^{3+} luminescent nanoparticles by the polyol method. A certain number of processes and chemicals are used in the process. Here is an overview of the procedure:

2.2. Synthesis of $YPO_4: Ho^{3+}/Ce^{3+}$ nanoparticles via polyol method.

A strategy for synthesizing $YPO_4:Ho^{3+}$ and Ce^{3+} luminous nanoparticles utilizing the polyol approach. A certain number of processes and chemicals are used in the process. Here is an overview of the procedure.

In a typical synthesis, metal ions ($(CH_3CO_2)_3Y \cdot XH_2O$, $(CH_3CO_2)_3Ho \cdot XH_2O$, and $(CH_3CO_2)_3Ce \cdot XH_2O$) were dissolved in concentrated HCl to create a solution. Alternating additions of deionized water were made, and the solution was heated at $80^\circ C$ to evaporate any remaining HCl. There were at least five evaporations. Drop by drop, $(NH_4)_2HPO_4$ and NaOH were dissolved in water and added to the mixture. After adding ethylene glycol, the mixture was refluxed at $80^\circ C$ for 10 minutes. After two hours of heating at $120^\circ C$, acid evaporation was applied, and a light yellow solution became white, producing white precipitation. The mixture was let to cool to room temperature. The precipitate was collected, centrifuged, cleaned with acetone, and IR-lighted dried.

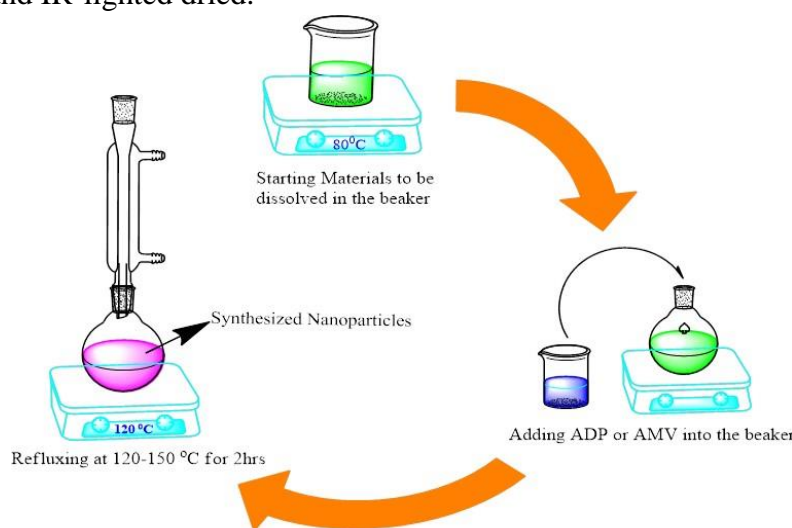


Figure 1. Schematic representation of the synthesis of $YPO_4:Ho^{3+}/Ce^{3+}$ nanophosphors by polyol method [16].

Doping of the YPO₄ nanoparticles was doped with Ho³⁺ 1% 10% and Ce³⁺ in various amounts. For the doped YPO₄ nanoparticles, fixed concentrations of Ho³⁺ (1%,3%) (3) and Ce³⁺ (10 at. percent) were utilised. Annealing was done for the samples, which underwent a 4-hour annealing procedure at 900°C following nanoparticle production. The current synthesis /appears to outline a challenging chemical synthesis procedure to produce doped YPO₄ nanoparticles from Figure 1.

3. Results and Discussion

3.1. XRD Analysis

From Figure 2, the (XRD) technique was used to examine the nanomaterial YPO₄:Ho³⁺/Ce³⁺. A lanthanum phosphate (YPO₄) host lattice doped with 1% holmium (Ho³⁺) and 10% cerium (Ce³⁺) ions makes up this nanomaterial. X-ray diffraction is a powerful technique for analyzing the crystal structure of materials. In this instance, the YPO₄:Ho³⁺/Ce³⁺ nanomaterial was examined using the XRD method. The nanomaterial was heated to 900°C using a procedure called annealing, then gradually cooled. The purpose of this annealing procedure is to fix flaws and stabilize the crystal structure. Unexpected information was discovered when examining the XRD diffraction pattern. It was evident from the observed patterns that the hexagonal phase gave way to the distinctive monoclinic phase. Notably, miller indices such as (-1 1 0), (2 0 0), (1 2 0), (0 1 2), (-1 0 3), and (2 1 2) demonstrated the successful integration of dopant components into the base material and suggested the existence of a unique monoclinic phase. The absence of impurity peaks in the XRD confirms a homogeneous distribution of the dopants (Ho³⁺ and Ce³⁺) inside the YPO₄ host lattice[11–12]. This suggests that dopants distribute evenly throughout the substance. Remarkably, the two most prominent peaks in the measured XRD pattern closely matched the pristine monoclinic structure of YPO₄, even when doped with Ce³⁺ and Ho³⁺ ions. This result implies that dopant addition had very little effect on the overall crystal structure of YPO₄. The combined XRD investigation findings confirmed the material's structural integrity, which persuasively demonstrated that Ho³⁺ and Ce³⁺ ions were effectively absorbed into the YPO₄ host lattice[13–14]. This accomplishment holds great potential for the synthesis of YPO₄:Ho³⁺/Ce³⁺ nanomaterial.

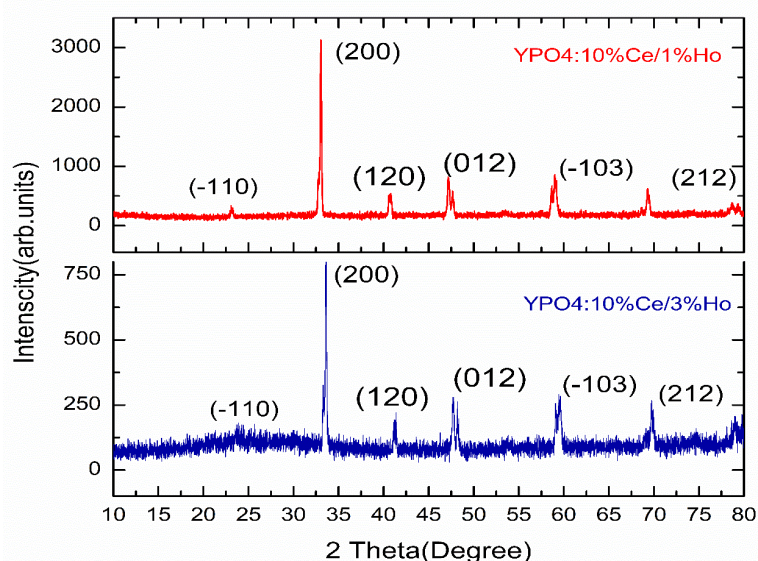


Figure 2. XRD pattern of YPO₄:Ho³⁺/Ce³⁺ samples.

3.2. SEM analysis.

The SEM images and data are used to infer the crystal structure, dopant distribution, crystal defects, and other relevant properties of the $\text{YPO}_4:\text{Ce}/\text{Ho}$ samples (Figure 3). The physical structure, chemical composition, and quality of crystal samples like YPO_4 doped with Ce and Ho are better understood by using SEM examination. YPO_4 crystals that have been carefully doped with Ho^{3+} and Ce^{3+} ions are depicted in this detailed SEM image. At 900°C , these crystals underwent a revolutionary annealing process that led to the development of an interesting nanophosphor material. A wide variety of characteristics and structures appear inside this visual tapestry, showing a vivid microscopic environment. The multitude of atypical forms that adorn the material's surface are the main focus of this SEM picture[15]. The most noticeable are the recognizable spherical-shaped particles that dominate the image and give the nanophosphor substance a characteristic appearance. Several particles that take on more specialized forms, such as cones and cuboids balance out this overwhelming presence.

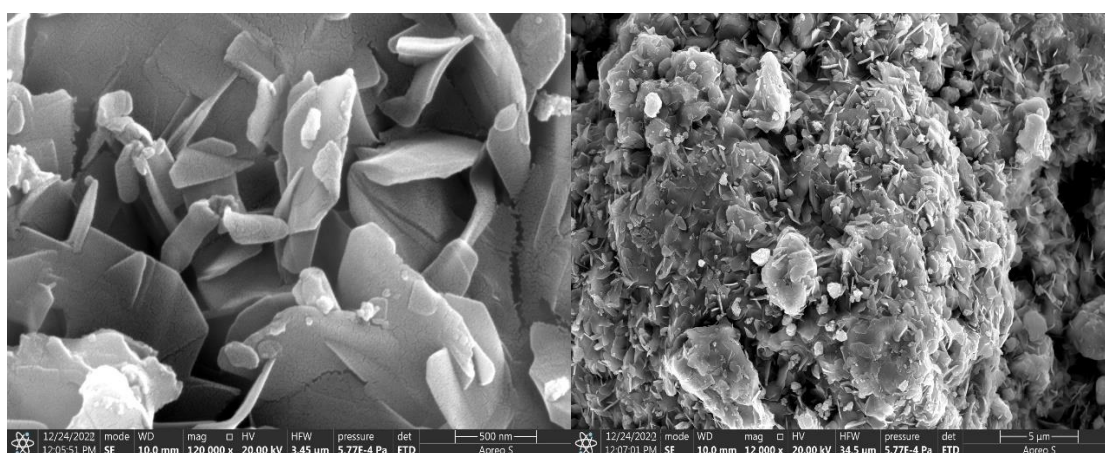


Figure 3. SEM images of $\text{YPO}_4:\text{Ho}^{3+}/\text{Ce}^{3+}$ samples.

3.3. FTIR analysis.

Here are some crucial considerations as from Figure 4 that might be made when interpreting FTIR data for $\text{YPO}_4:\text{Ce}/\text{Ho}$ samples phosphate group vibrations. Since YPO_4 is a phosphate molecule, the FTIR spectrum should exhibit the typical phosphate group vibrations. The phosphate (PO_4^{3-}) group's symmetric stretching vibrations are thought to cause the prominent peak at $1000\text{--}1200\text{ cm}^{-1}$. The phosphate (3 PO_4^{3-}) group's asymmetric stretching vibrations are what cause the peak to appear about $560\text{--}600\text{ cm}^{-1}$. Stretching of Yttrium-Oxygen, in the FTIR spectrum, yttrium-Oxygen (Y-O) stretching vibrations commonly occur between 400 and 700 cm^{-1} [16,17]. If the YPO_4 crystal lattice is doped with cerium and holmium ions, there could be minute shifts or modifications in the FTIR peaks associated with these dopants. These could result from modifications in lattice vibrations brought on by the presence of certain ions. Broad bands in the spectrum could be caused by hydroxyl (OH^-) groups that have been adsorbed onto the sample surface or by water vibrations. To differentiate between inherent crystal vibrations and surface adsorbed species, careful interpretation is required. FTIR spectra often occupy a large range, frequently ranging from 4000 cm^{-1} (wavenumber) down to about 400 cm^{-1} different portions of the spectrum, revealing details about various vibrational characteristics[18]. In order to identify individual peaks in the FTIR spectrum, it is necessary to contrast the observed peaks with the vibrational modes of well-known functional

groups. FTIR can provide information about a substance's crystal structure. Peak variations or shifts might point to modifications in crystal lattice symmetry brought on by dopant inclusion.

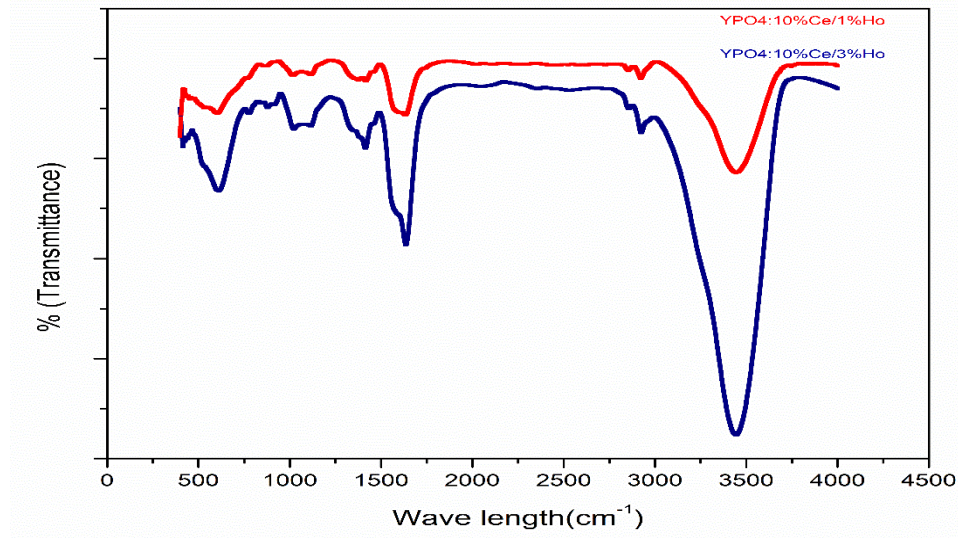


Figure 4. FTIR pattern of YPO₄:Ho³⁺/Ce³⁺ samples.

3.4. UV analysis.

From Figure 5, the following peak types might be observed in YPO₄:Ce/Ho UV studies: peaks of absorption, you could see peaks in the UV absorption spectra that were caused by electronic transitions in the host YPO₄ lattice, the Ce and Ho dopants, and both. These peaks might indicate the energies needed to absorb UV radiation. Peaks of Ce³⁺ and Ho³⁺ Luminescence: When exposed to UV irradiation, luminescent materials like YPO₄:Ce/Ho display distinctive emission peaks. The ions Ce³⁺ and Ho³⁺ have a unique luminescent behavior. While the Ho³⁺ dopant may emit at longer wavelengths, such as between 540 nm and (green to red emission), the Ce³⁺ dopant may produce broadband emission with peaks about 460 nm (blue emission).

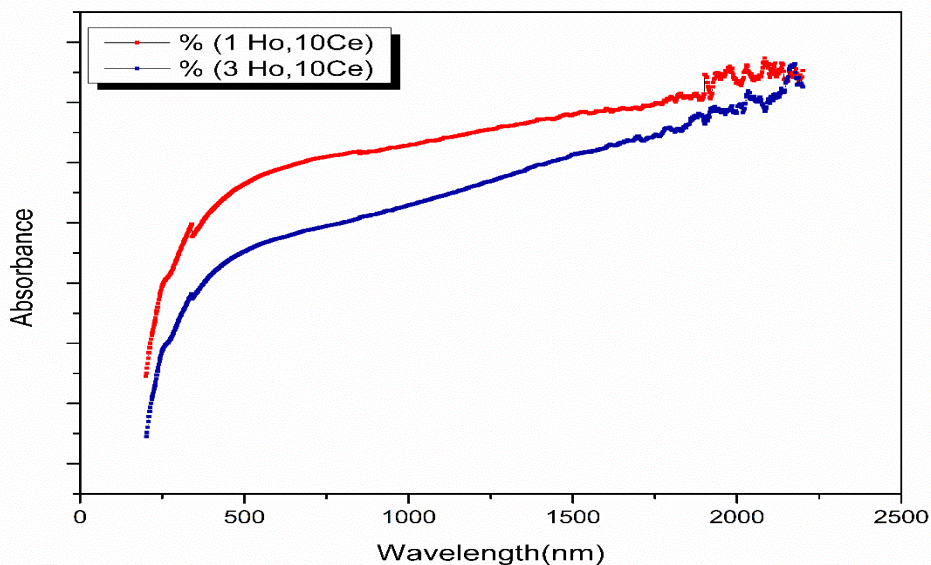


Figure 5. UV pattern of YPO₄:Ho³⁺/Ce³⁺ samples.

Energy Transfer Peaks: If the Ce³⁺ and Ho³⁺ ions in the material contact, energy transfer processes may occur [19]. It is possible to spot emission spectrum energy transfer peaks, which

show that energy absorbed by one dopant is being transferred to another dopant and changing the luminous characteristics. Stark splitting, peaks that can result from interactions between dopants or the crystal lattice, is the splitting of energy levels in a crystal due to an external electric field. Additional peaks in the emission or absorption spectra may result from this splitting.

3.5. Luminescence analysis.

In the investigation of $\text{YPO}_4:\text{Ho}^{3+}$, Ce^{3+} luminescent materials, making use of upconversion analysis, upconversion luminescence is produced when higher-energy photons absorb lower-energy photons via a mechanism known as Suppressive-Stokes luminescence. Figure 6 shows the emission spectra of YPO_4 (Ho^{3+} , Ce^{3+}) under down-conversion (DC) and up-conversion (UC) conditions, respectively. We created the nanophosphor compound $\text{YPO}_4:0.01\text{Ho}^{3+}/0.1\text{Ce}^{3+}$ in our study by adding 1% Ho^{3+} and 10% Ce^{3+} dopants into YPO_4 . Further research was carried out on this optimized nanophosphor material to investigate its upconversion and down-conversion luminous characteristics [20].

3.5.1. UC study.

Figure 6 shows the upconversion spectra of $\text{YPO}_4:\text{Ho}^{3+}/\text{Ce}^{3+}$ (doped with 1% Ho^{3+} and 20% Ce^{3+}), which illustrates the reactions to various laser excitations above 980 nm. Emission bands at 540 nm (green) and 650 nm (red) are produced as a result of the interaction between Ce^{3+} , Ho^{3+} , and PO_4^{3-} ions[21-22]. The intensity of the 650 nm emission band (R = red) is affected by the co-dopants' and host material's respective concentrations. It is important that the upconversion spectra show green emission with a notable wavelength range of about 360 nm. Ce^{3+} ions operate as sensitizers under 980 nm excitation because they have greater absorption rates than Ho^{3+} ions.

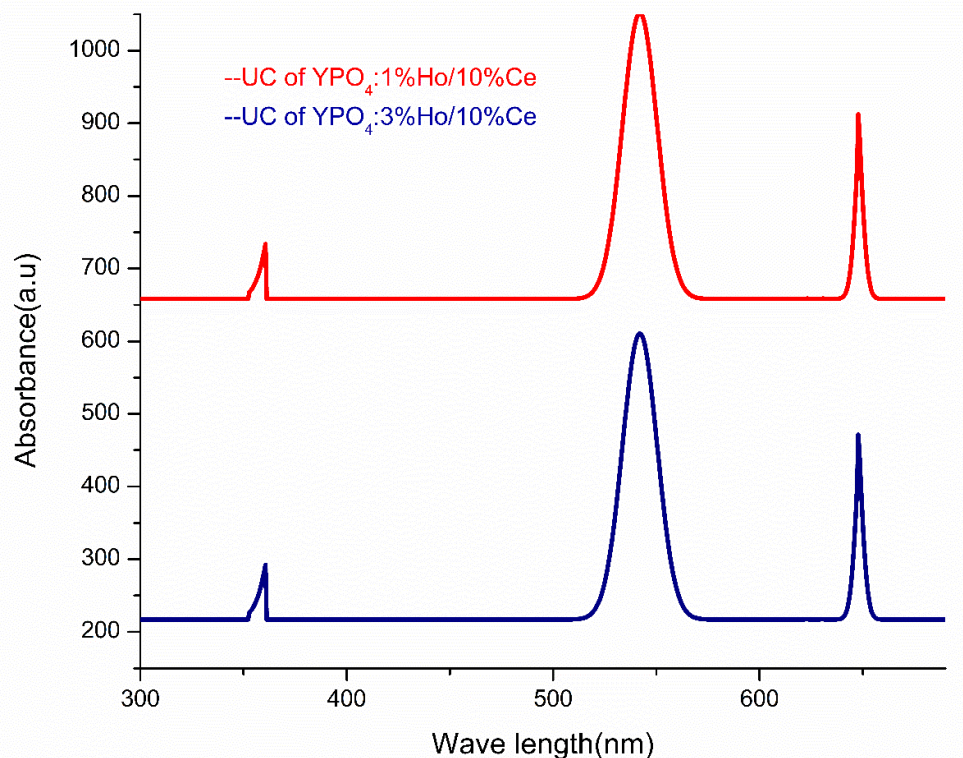


Figure 6. UC Study of $\text{YPO}_4:\text{Ho}^{3+}/\text{Ce}^{3+}$ samples at 980nm laser excitations.

3.5.2. DC study.

From Figure 7, Down-Conversion (DC) Phenomenon Analysis, The Stokes shift phenomenon results from lower energy light (Eem) being emitted while higher energy light rays (Eexe) are absorbed. The DC emission spectra of $\text{YPO}_4:\text{Ho}^{3+}/\text{Ce}^{3+}$ (with 1 at.% Ho) after ultraviolet (UV) light at 279 nm are shown in Figure 6. Energy transfers (ET) are suggested by emission bands at 360 nm (green), 460 nm, 542 nm, and 650 nm (red). For the Ho^{3+} ion, notable transitions include $^5\text{F}_4$ and $^5\text{S}_2$ $^5\text{I}_8$, and for the Ce^{3+} ion, $^5\text{F}_5$ $^5\text{I}_8$, $^2\text{F}_{5/2}$ and $^2\text{D}_4$ $^7\text{F}_5$. Excitation wavelengths of about 279 nm were used to generate emission spectra, which showed different Ho^{3+} emission peaks corresponding to each excitation. The intensity of Ho^{3+} emission is lower when directly stimulated at 460 nm ($^5\text{I}_8$ $^5\text{G}_6$) compared to indirect excitation at 279 nm because Ho^{3+} ion-mediated f-f transitions have a naturally low absorption cross-section. A wide emission band with a center at 460 nm develops when it is stimulated at 279 nm, along with observable peaks related to PO_4^{3-} and Ho^{3+} emissions. The smaller absorption cross-section of transitions involving Ho^{3+} is to blame for the reduced intensity of Ho^{3+} emission under direct excitation at 460 nm. Excitation at 279 nm, however, results in the generation of strong peaks for Ho^{3+} and the wide PO_4^{3-} emission band at 460 nm.

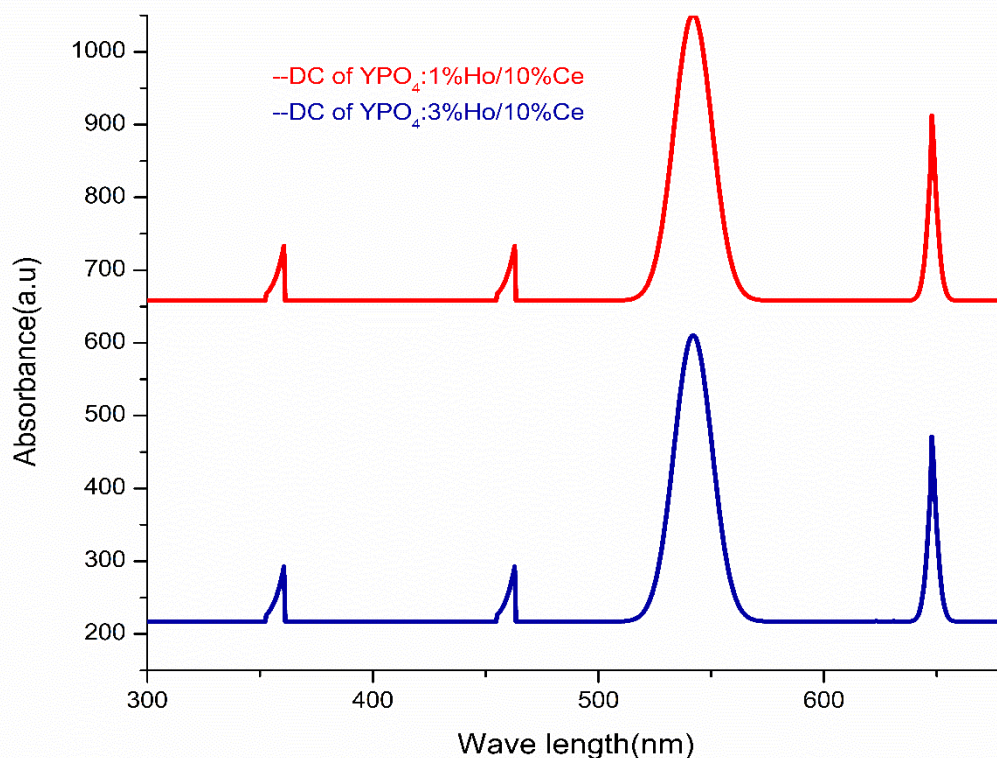


Figure 7. DC pattern of $\text{YPO}_4:\text{Ho}^{3+}/\text{Ce}^{3+}$ samples.

4. Conclusion

In summary, a thorough understanding of the energy transition properties of YPO_4 nanophosphors doped with Ce^{3+} and Ho^{3+} ions using the polyol method has been obtained through our extensive investigation, which included X-ray Diffraction (XRD), UV analysis, FTIR spectroscopy, SEM analysis, as well as Up-conversion and Down-conversion studies. Unique energy shifts were produced when Ce^{3+} and Ho^{3+} ions were incorporated into the YPO_4 host lattice, which helped to explain why photoluminescence events persisted. The work notably revealed the exceptional UV absorption behavior that Ce^{3+} ions cause in $\text{YPO}_4:\text{Ce}/\text{Ho}$

nanophosphors through efficient sensitization processes. Our investigation greatly expands the understanding of luminous materials by providing new insights into YPO₄:Ce/Ho nanophosphors synthesized by the polyol method. This new understanding has significant potential for various uses, including cutting-edge lighting systems and optoelectronic devices. Owing to its remarkable optical and structural characteristics, YPO₄:Ce/Ho is a material of great importance that will propel future developments in a wide range of technological and scientific fields.

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Conflicts of Interest

The authors declare no conflict of interest. This research work (synthesis part) is completely carried out in our laboratory at GITAM Deemed to be University, Hyderabad.

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