ELLIOTT RESOLVED PHOTOLUMINESCENCE AND GUARDED HOT METHOD FOR OPTICAL AND CONDUCTIVITY PROPERTIES ANALYSIS OF SYNTHESIZED CEO$_2$ NANOPARTICLES

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Abstract

A cerium oxide (CeO_2) nanoparticle is an interesting material for a variety of applications in biotechnology and medicine. CeO_2 is the most important rare-earth element and the oxide has attracted much attention due to its promising applications in energy and environmental fields. Different structural characterization technique such as X-ray diffraction and transmission electron microscope has been studied. But the optical characterizations and conductivity of the synthesized CeO_2 play a major role. In this paper, the light emission characteristics and thermal conductivity properties of synthesized CeO_2 nanoparticles are analyzed by using Elliott resolved photoluminescence and steady state guarded hot method. The proposed method includes three different processes namely Photoluminescence studies of the samples, defect detection and evaluate the thermal conductivity property. The synthesized CeO2 nanoparticles are considered for identifying the structural and light emission characteristics. First, the Elliott resolved photoluminescence method is applied to analyze the light emission characteristics of the CeO2 nanoparticles by identifying the peak intensity at a particular wavelength of light. Then the point defects are identified in the electronic states inside the bandgap. The defect identification and refinement states improve the optical and optoelectronic performance of the CeO2 materials. Finally, the thermal conductivity properties of the prepared CeO2 nanoparticles are analyzed at room temperature by applying a steady state guarded hot measurement method. The thermal conductivity is evaluated with the different steady-state temperatures. The structural and light emission characteristics of the proposed method are analyzed by means of photoluminescence intensity, Commission Internationale de l'Elcairage (CIE) chromaticity coordinates and Correlated color temperature (CCT) values, photoluminescence intensity with defect and without defects, and thermal conductivity.

Keywords: Synthesized of CeO_2 nanoparticles**,** light emission and conductivity characteristics analysis, Elliott resolved photoluminescence method, steady state guarded hot measurement method

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1. Introduction

Cerium oxide is among the most promising transition metal oxides for a variety of applications such as nanomaterial, photocatalysis, gas sensors, fuel cells, polish materials, and automotive catalytic analysis. A photo-excited spontaneous emission process offers better optical and electronic properties of cerium oxide materials and has high chemical stability. This wide range of cerium oxide and nanoparticles applications is based on the unique properties of cerium oxide, such as optical characterization, thermal conductivity, Tip-enhanced photoluminescence (TEPL) nano-spectroscopy was developed in [1] to offer a better high sensitivity with less than 10 nm resolution, and it allows the preferred nano-scale characterizations. The near-field polarization of in-plane and out-of-plane components was not controlled for effective TEPL measurement. Optical and luminescence spectroscopy models were introduced in [2] for identifying the cerium oxide defects through the pulsed electron evaporation of ceramic oxide targets.

A Hybrid Nanofiber/Thin-Film Multilayer Tm3+-Doped SiO_2–HfO_2 was developed in [3] to improve the performance of intensity and spectral bandwidth. But, the defects in the intensity analysis were not identified. A Tailored optical and magnetic property was derived in [4] for identifying the structural characteristics by detecting the various defects. A surface structure identification and defect identification on the photocatalytic characteristics of Gd-doped CeO_2 nanoparticles were introduced in [5]. But the thermal conductivity of the doped CeO_2 nanoparticles was not evaluated.

Synthesis of hybrid nanostructures including dioxide and microcrystalline cellulose was arranged in [6] by the microwave-assisted hydrothermal route with different temperatures and pH values. Thermal conductivity of a constant nano-antifreeze containing cetyltrimethylammonium bromide coated cerium (IV) oxide nanoparticles was measured in [7]. The thermal conductivity of cerium oxide/ethylene glycol nanofluid was evaluated in [8] for different temperatures by applying an artificial neural network (ANN) and fitting method. But the accurate thermal behavior analysis was not performed.

The thermal conductivity of alumina, ceria, and their hybrid ratio was evaluated in [9] based on the correlation measurement. But the light emission characteristic was not analyzed. Thermal conductivity of spark plasma sintered CeO2 and Ce3Si2 combinations were measured in [10] from the estimated thermal diffusivity, exact heat, thickness, and uniform microstructure.

1.1 Contributions

From the analyses of existing work, novel different characteristics of the synthesized CeO_2 nanoparticles are analyzed with the following contributions.

- First, a novel Elliott resolved photoluminescence method is developed to analyze optical characteristics of the synthesized CeO_2 nanoparticles through the light emission approach. As a result, the peak photoluminescence intensity at a particular wavelength of light is identified.
- After that, the point defects are identified between the ground state and excited state. The defect identification and refinement states process are used to improve the optical characteristics of the CeO_2 materials.
- Finally, steady state guarded hot measurement method is employed to measure the thermal conductivity properties of the synthesized CeO_2 nanoparticles with room temperature.
- Finally, performance of different characteristics are analyzed and discussed.

1.2Organization of the article

The article is arranged into dissimilar sections as follows. Section 2 reviews the related works. Section 3 provides a brief description of the different processes such as Elliott resolved photoluminescence method, defect identification; thermal conductivity properties of the synthesized CeO_2 nanoparticles by applying a steady state guarded hot measurement method. Section 4 describes the performance results and discussion of different characteristics of the synthesized CeO_2 nanoparticles. At last, Section 5 concludes the paper.

2. Related works

An adaptive tip-enhanced nano-spectroscopy technique was developed in [11] for improving the sensitivity of ultrafast nano-spectroscopy. An innovative design and fabrication of cerium oxide nanoparticle-based optical sensors were developed in [12] to improve the sensing mechanisms as well as in-depth analysis.

The effect of cerium dioxide (CeO_2) nanoparticles on electrical conductivity properties were investigated in [13]. The developed model was used to increase the conductivity of the thermoplastic nanocomposite system. But the defect detection was not analyzed. The photocatalytic and corrosion inhibitor behaviors of cerium oxide-based nanomaterials were analyzed in [14] to investigate the effect of doping nanomaterials.

Oxygen vacancy defects on CeO_2 nanowires, synthesized through the hydrothermal method were analyzed in [15] through annealing under a variety of controlled atmospheres. An optical and electrical property of CeO_2 nanoparticles was analyzed in [16]. A density functional theory (DFT) was introduced in [17] for measuring the band structure before and after vacancy generation.

A pulsed laser deposition model was introduced in [18] for measuring the optical and dielectric properties of cerium oxide thin films. A Dielectric, magnetic and Raman spectroscopy analysis of CeO_2 nanoparticles was discussed in [19] through the precipitation approach. The influence of hydrothermal conditions within morphology tuning and physico-chemical constraints of ceria was discussed in [20].

3. Methodology

The thin film-deposited rare earth materials have expected much consideration due to their better optical and electrical properties which are highly attractive for applications such as electrochemical systems, fuel cells, and opto-electronic devices. Among the numerous nanomaterials, CeO_2 is a prominent nanomaterial due to its distinctive properties and a variety of engineering and biological applications. CeO_2 has attractive optical properties, such as a high refractive index, and good transmission in the visible and infrared regions with a wide band gap of 3.2 eV. The number of studies focused on the optical properties of cerium dioxide (CeO_2) nanoparticles (NPs) has recently increased due to their broad potential, e.g., in photocatalytic applications, fuel elements, and medical purposes. But in this paper, the synthesized CeO_2 NPs are subjected to photoluminescence analysis, defect detection of doped and undoped CeO_2, Finally, the conductivity properties of CeO_2 NPs are also analyzed in this paper.

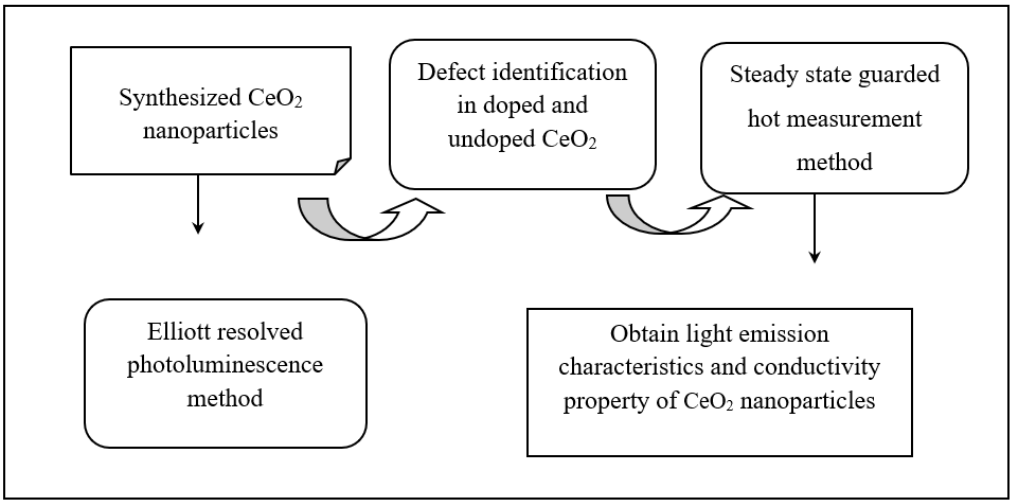

Figure 1 Architecture of the proposed Elliott resolved photoluminescence and steady state guarded hot method

Figure 1 illustrates an architecture diagram of the structural and light emission characteristics analysis of the proposed method which includes three different processes namely Elliott resolved photoluminescence method, Identify the defects in doped and undoped CeO_2, and analyze the thermal conductivity properties of CeO_2 using steady state guarded hot measurement method. The methodology considers the input as synthesized CeO_2. First, the light emission characteristics of the CeO_2 are analyzed by using Elliott resolved photoluminescence method. After that, the defect in the light emission characteristics of the CeO2 is detected to reduce the optical and optoelectronic performance of the CeO_2 materials. The steady state guarded hot measurement method is employed for measuring the thermal conductivity properties of the CeO_2. These processes are discussed briefly in the following sections.

3.1 Elliott resolved photoluminescence method for light emission characteristics analysis

Photoluminescence (PL) is the method of spontaneous emission of light from a material. It is an efficient technique to investigate discrete energy levels and to extract valuable information about semiconductor composition. The main advantage of the Photoluminescence its self is fast, contactless, and nondestructive. Therefore, it helps to apply the optoelectronic properties of synthesized CeO_2 nanomaterials of various sizes in nanometer during the manufacturing process without complex sample preparation. The Elliott resolved photoluminescence method is developed in this paper to measure purity and crystalline quality, and identify certain impurities or defects in the CeO_2 nanoparticles. The photoluminescence intensity is computed by means of the Elliott formula. Hence the name of the method is called as Elliott resolved photoluminescence method. The block diagram of Elliott resolved photoluminescence set-up is shown in figure 2.

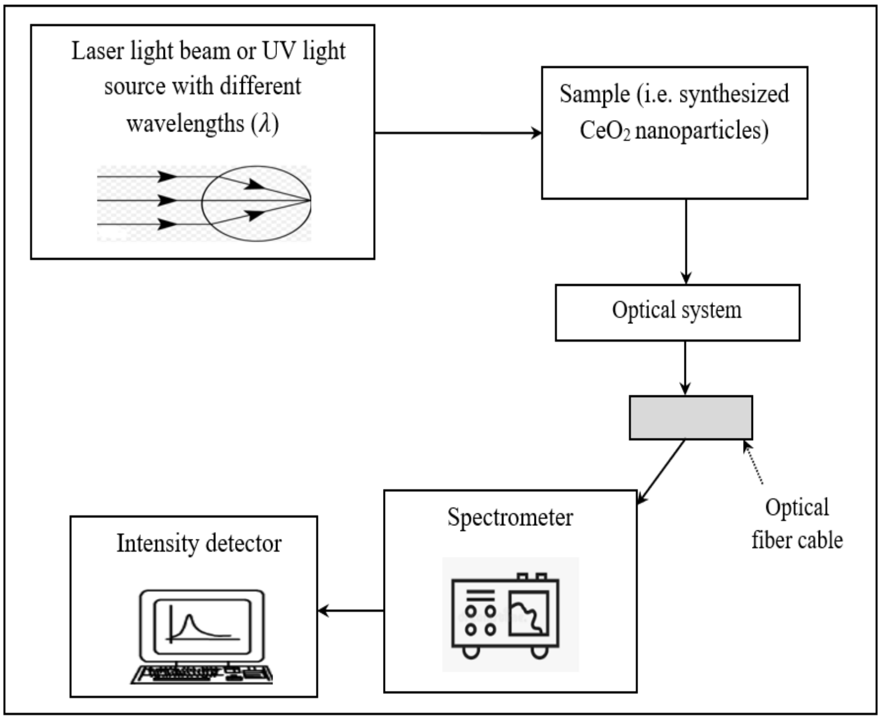

Figure 2 Block diagram of Elliott resolved photoluminescence set-up

As shown in above figure 2, the process of Elliott resolved photoluminescence setup is constructed for identifying the optical characteristics of the synthesized CeO_2 nanoparticles. First, the monochromatic light source typically a laser or UV light source is used to emit lights with different wavelength (λ). Then the light falls on the sample (i.e. synthesized CeO_2 nanoparticles), it is absorbed and initiates the excess energy. This excess amount of energy is dissipated through the emission of light, which is called as photoluminescence process.

The emitted light energy is given into a fiber optic cable and then direct into a spectrometer. A filter is placed in fiber optic input to eliminate any incident laser light. In the spectrometer, a diffraction grating diffracts dissimilar wavelengths in various directions to an array of photo-detectors that compute the intensity of each wavelength component. Finally, the digital information is captured by the Intensity detector or computer, which displays a photoluminescence intensity spectrum. The observed spectrum designates the relative intensities of light with the different wavelengths. As a result, the peak intensity is identified at a particular wavelength of light.

3.1 Linear Absorption based Elliott resolved photoluminescence method

In this subsection, light emission process of Elliott resolved photoluminescence method is described. This process offers a dominant light emitted characterization of CeO_2 nanoparticles. First, a monochromatic source of light, typically a laser or UV light source is used to stimulate the sample. Photoluminescence process is depending on the light interacts with the synthesized CeO_2 nanoparticles. This method occurs as the light is absorbed and emitted at a range of longer wavelengths. The light emission process of Elliott resolved photoluminescence method is shown in figure 3.

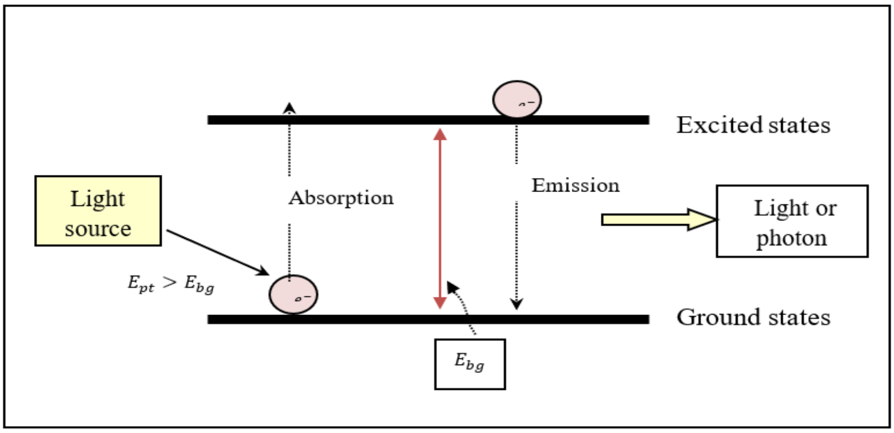

Figure 3 Schematic diagram of light emission process of Elliott resolved photoluminescence method

Figure 3 illustrates the light emission process of Elliott resolved photoluminescence method. By applying an Elliott resolved photoluminescence method, there are two states are employed for light emission process such as higher energy state and lower energy state. The higher energy state is called as excited states whereas the lower energy states are called as ground states.

As the light is directed onto a sample or synthesized CeO$_2$ nanoparticles, the electrons inside the CeO$_2$ nanoparticles travelled into higher energy excited states which called as excitation. This only takes places when the energy of the photon or light is greater than the wide band gap energy levels (E_{bg}). The band gap energy of the samples is in the range from 3.2 to 3.5 eV.

The energy level of the incident photon or light is measured as follows,

$$E_{pt} = \frac{h_{pt} * C_{pt}}{\lambda_{pt}} \qquad (1)$$

Where, E_{pt} denotes an energy level of the photon, h_{pt} denotes a Planck's constant ($6.626 * 10^{-34}\,Js$), C_{pt} denotes a speed of the photon or light ($3 * 10^{8}\,m/s$), λ_{pt} indicates a wavelength of the photon or light in meter.

After a few nanoseconds the electron moved down from the excited states to ground state and releases or emits the absorbed energy in the form of a photon which called as relaxation. Therefore, the spontaneous emission due to optical excitation is called photoluminescence.

The emitted light is collected and analyzed with spectrometer to yield important structural information about the CeO$_2$ nanoparticles properties. Typically, the emitted light is lower energy than the energy in the excitation, since some energy is lost to non-radiative processes. The energy of the emitted light is related to the difference between the two energy levels involved in the transition between an excited state and a ground state.

$$E_L = E_{es} - E_{gs} \qquad (2)$$

Where, E_L denotes an energy of the emitted light, E_{es} denotes an energy of emitted light in excited state, E_{gs} indicates of emitted light in ground state. The observed intensity curve indicates that the linearly increased while increasing the wavelength of the light. But the peak intensity is identified at a particular wavelength of light. After reaches the peak value, then the intensity gets reduced linearly while increasing the wavelength. The intensity is estimated by applying an Elliott resolved photoluminescence method.

$$I_{PL} = Im\left[\sum \frac{f_\lambda * s_\lambda}{E_\lambda - \hbar\omega - i\,\gamma_\lambda\,(\omega)}\right] \qquad (3)$$

Where, I_{PL} denotes a Elliott resolved photoluminescence intensity, f_λ indicates a oscillator strength of bound state of an electron and an electron hole which are attracted to each other, s_λ denotes a spectral response function, E_λ denotes a photon energy, $\hbar\omega$ denotes a probe-photon energy, $\gamma_\lambda\,(\omega)$ dephasing constant.

3.2 Defect detection doped and undoped CeO2

The presence of defects in optical characteristics of the CeO$_2$, the light intensity becomes distorted in specific manner that depends on the shape and size of the defects. Different types of intrinsic defects are very common in nanoparticles structure analysis which is done by Elliott resolved photoluminescence method. The defect in the doped and undoped CeO2 plays an important role in modifying the photoluminescence property. The controlling of the defects in CeO2 materials is to improve the device properties and reliability.

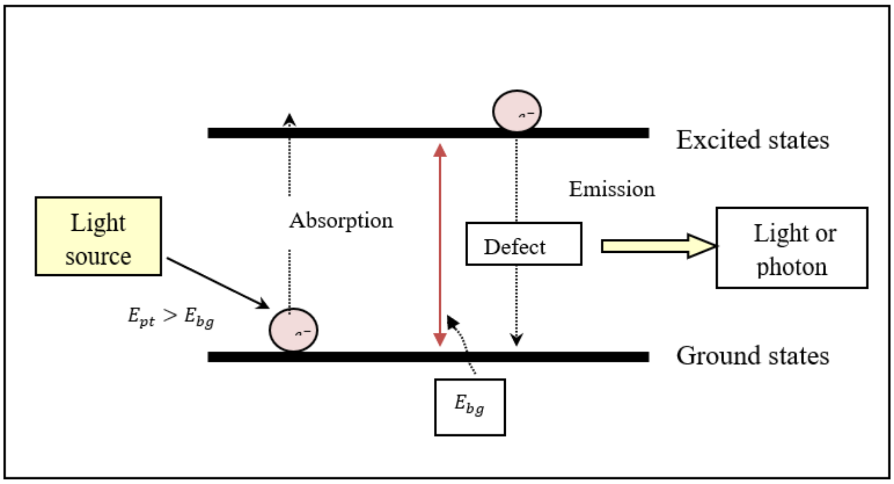

Figure 4 Schematic diagram of defect detection in CeO2 materials

Figure 4 depicts the typical relaxation processes of the photoluminescence method. The defect states causing nonradiative transitions which did not detected directly as emission peaks. In the nanomaterial, radiative recombination takes places in band-to-band transition. The predictably formed defects are identified in additional electronic states inside the bandgap. The defects are identified during the relaxation processes of the photoluminescence method. When the electron transit down from the excited states to ground state, the defect is identified and releases or emits the energy in the form of a photon. Therefore, the defect states reduces the optical and optoelectronic performance of the CeO_2 materials,

The relative concentration of defects is evaluated by comparison with the total photoluminescence intensity since defects decreases the emission intensity of luminescence. The defects expected to significantly influence the physical properties of the CeO_2 nanoparticles.

3.3 Conductivity properties of the CeO2 nanoparticles

In this subsection, the conductivity properties of the prepared CeO_2 nanoparticles are analyzed at room temperature. The conductivity property indicates that the electricity or heat pass through a CeO_2 nanoparticles. The conductivity is measured depending on temperature. It represents a nanoparticles ability to conduct the electric current.

The steady state measurement method is applied to measure the conductivity properties of the prepared CeO_2 nanoparticles by using the electrical power output of a hot plate with heat conduction. In order to perform an accurate estimation of the thermal conductivity of nanoparticles, steady-state temperature boundary states are preserved on the top and the down planes of the samples or nanoparticles. It helps to maintain a one-dimensional temperature gradient and heat flux between the hot surface plate and a cold surface plate. The schematic design of steady state measurement method is given below,

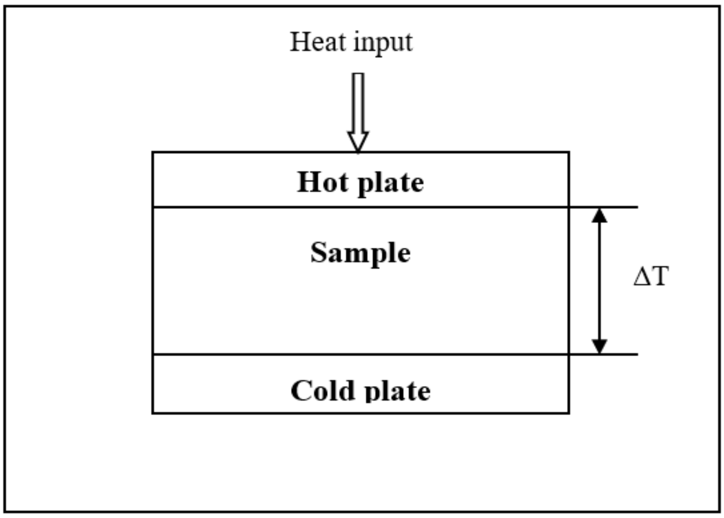

Figure 5 Schematic design of steady state guarded hot measurement method

Figure 5 indicates a schematic design of steady state measurement method where the two plates such as hot plate, cold plate and samples are utilized. The sample is positioned between two plates held with different temperatures, with one being heated and the other plate being cooled. The temperature of the hot plate, cold plate, the heat flux supplied to the sample and the thicknesses 'Δt' of the sample are continuously monitored.

The measured heat power rate is transferred across the samples due to guarded heaters. After thermal equilibrium has attained and the heating and cooling plates are kept in stable temperatures, the thermal conductivity is calculated from the input values. By applying a Fourier's law in the heat conduction process, material thermal conductivity at a given temperature is calculated.

$$\sigma = Q\, \frac{\Delta t}{(T_H - T_c) * a} \quad (4)$$

Where, σ thermal conductivity, Q denotes a Heat flux at the center of the sample, Δt indicates thicknesses, T_H denotes a temperature at hot plate, T_c denotes a temperature at cold plate, a indicates a heat transfer area. In this way, thermal conductivity properties of the synthesized CeO_2 nanoparticles are measured.

4. RESULTS AND DISCUSSIONS

In this section, performance of the Structural and light emission characteristics of CeO_2 synthesis is analyzed with different metrics such as Photoluminescence intensity, Photoluminescence intensity with defect, and the PXRD pattern, Commission Internationale de l'Elcairage (CIE) chromaticity co-ordinates and Correlated color temperature (CCT) values and electrical conductivity. The performance are analyzed with the help of graphical representation.

4.1 Performance analysis of Photoluminescence intensity

Photoluminescence intensity is the measure the intensity of emitted light as a function of wavelength by using an optical spectrometer. The intensity of emitted light is analyzed with the varying wavelength in nanometer (nm). The Photoluminescence intensity of emitted light is measured in terms of absorption unit (a.u).

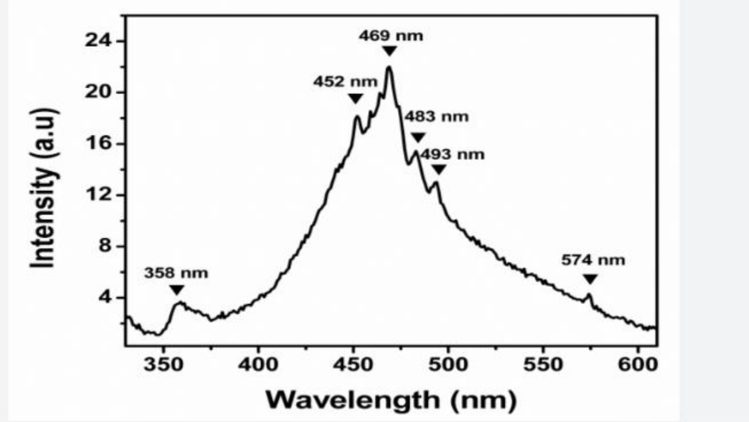

Figure 6 performance of photoluminescence intensity with respect to wavelength

The performance of photoluminescence intensity with respect to wavelength is depicted in figure 6. The photoluminescence intensity is used for characterizing the optical and photochemical properties of CeO_2 nanoparticles and the photoluminescence peak intensity correlates directly with the defect densities in materials. As shown in figure 6, wavelength on the spectrum is taken in 'x' axis and the photoluminescence intensity observed at 'y' axis. From the figure, it is evident that the intensity of the light is plotted against the wavelength on the spectrum. As shown in figure 6, the peak intensity occurs at a wavelength of 469nm. There are six emission peaks were observed at 358, 452, 469,483,493 and 574 nm excitation corresponding to wavelength. The Elliott resolved photoluminescence method is applied for achieving the different intensity level.

As the laser light beam or UV light is directed onto synthesized CeO_2 nanoparticles, the electrons travelled from higher energy excited states. After a few nanoseconds the electron moved down from the excited states to ground state and emits different absorbed energy. These spontaneous emissions of the light is collected and analyzed with spectrometer to yield peak intensity level.

4.2 Performance analysis of Commission Internationale de l'Elcairage (CIE) chromaticity co-ordinates

The CIE chromaticity coordinates is measured in this section based on the excitation wavelength observed from the Elliott resolved photoluminescence method. This CIE chromaticity has applied for different fields including luminescent materials, color science and technology, color related product design. The color scale was constructed based on the data obtained by commission Internationale de l'eclairage 1931. The color matching functions are used to measure the CIE coordinates X, and Y. nanophosphors on excitation with 358, 452, 469,483,493 and 574 nm wavelengths.

Table 1 Calculated CIE coordinates nanophosphors on different excitation wavelengths

Excitation wavelength (nm)	CIE coordinates (X,Y)
358	(0.175, 0.005)
452	(0.154, 0.019)
469	(0.126, 0.053)
483	(0.078, 0.170)
493	(0.031, 0.363)
574	(0.471, 0.527)

Table 1 reports the performance results of CIE coordinates of the different excitation wavelengths considered as 358, 452, 469,483,493 and 574 nm. The obtained results indicate that the CIE coordinates is measured and plotted the value.

For each coordinate the color scale is constructed based on the data obtained by commission Internationale de l'eclairage 1931. For each wavelength, various color scale results are obtained.

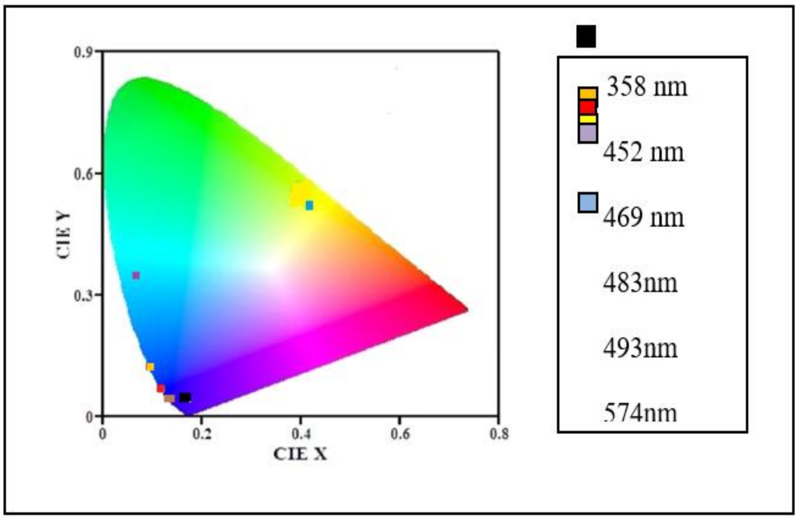

Figure 7 Performance results of CIE chromaticity co-ordinates

Figure 7 given above demonstrates the graphical illustrations of CIE chromaticity co-ordinates versus the number of wavelength that are varied from 358, 452, 469,483,493 and 574 nm. As shown in the graphical plot, the CIE X chromaticity co-ordinates and CIE Y chromaticity co-ordinates. Further, the CIE chromaticity values of the CeO_2 nanoparticles include the combination of both blue and red components for display devices and other optoelectronic applications. It reveals that the CeO_2 nanoparticles products are useful for display solid state lightning applications.

4.3 Performance analysis of Correlated Color Temperature (CCT)

The Correlated Color Temperature (CCT) is measured based on the X, Y coordinates calculated from the CIE chromaticity function. The CCT s measured in terms of Kelvin (K). The CCT is measured by using McCamy's approximation as given below,

$$CCT = (437 * Q^3) + (3601 * Q^2) + 6861Q + 5517 \quad (5)$$

Where,

$$Q = \frac{(X-0.3320)}{(0.1858-Y)} \quad (6)$$

From (5) (6), CCT is measured in Kelvin (K). The different results of the CCT is listed in table 2 .

Table 2 Calculated CCT

Excitation wavelength (nm)	CIE coordinates (X,Y)	CCT (K)
358	(0.175, 0.005)	1988
452	(0.154, 0.019)	1765
469	(0.126, 0.053)	1908
483	(0.078, 0.170)	-989715
493	(0.031, 0.363)	26704
574	(0.471, 0.527)	3290

Table 2 illustrates the overall calculated results of the CCT with respect to the CIE coordinates. It describes the color not brightness of a light source and is measured in Kelvin. A warm light is around 2700K and moving to neutral white at around 4000K, and to cool white, at 5000K or more. Therefore, the formed samples are negative CCT value which lies below the black body locus. The CCT measurement results are used to clearly indicate the feasible potential application of CeO_2 nanophosphor in display devices.\

4.4 Performance analysis of Defect detection in CeO_2 nanoparticles

In this section, defect in the photoluminescence intensity is identified with respect to the energy in terms of electron volt (eV). The defect is identified with doped CeO_2 nanoparticles and without doped CeO_2 nanoparticles. The two different graphical plots are illustrated in figure 8 (a) and figure 8(b).

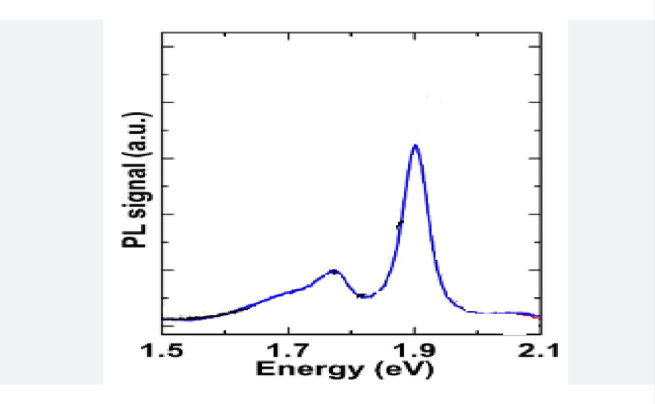

Figure 8(a) doped CeO_2 nanoparticles with defects

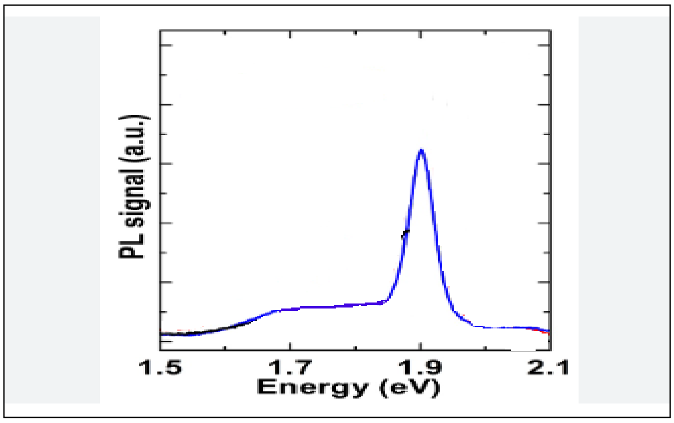

Figure 8(b) doped CeO_2 nanoparticles without defects

Figure 8(a) (b) illustrates the performance results of defect detection in the photoluminescence (PL) intensity using with and without doped CeO_2 nanoparticles . In order to calculate the defect detection, the energy level is taken in the range from 1.5 to 2.1. As shown in the graph, the horizontal axis represents the energy level and the vertical axis provides the outcomes of the photoluminescence (PL) intensity in absorption unit (a.u). In this analysis, the point defects in CeO_2 nanoparticles result in an increase in the overall room-temperature photoluminescence intensity. In addition, the defects create a new emission peak that lead to a better identifying the defect in CeO_2 nanoparticles.

As the number of defects in the nanoparticles increases, the overall brightness of the light that is emitted by the nanoparticles gets increased. From the figure 8(a), the light has a photoluminescence peak at $1.90\,eV$ without doped CeO_2 nanoparticles, which determines its wavelength and color. But the point defects also created a new photoluminescence peak at $1.78\,eV$ as shown in figure 8(b).

4.5 Performance analysis of thermal conductivity

Thermal conductivity is defined as the ability of a given CeO_2 nanoparticles to conduct/transfer heat. It is measured in the SI units of W/mK (Watts per meter Kelvin). The thermal conductivity is measured using equation (4).

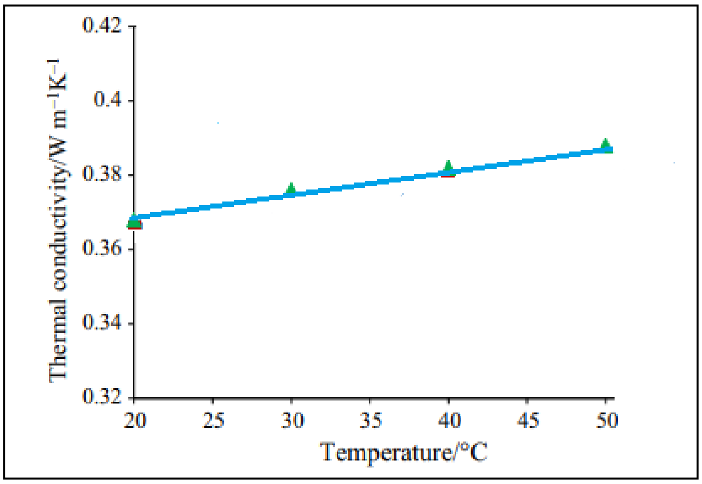

Figure 9. Performance results of thermal conductivity of CeO_2 nanoparticles with different temperature

The graphical results illustrates that the performance results of the thermal conductivity of synthesized CeO_2 nanoparticles with respect to a different temepratiure in degree celcious (℃). The different temperatures are taken the 'x' direction and the results of the thermal conductivity obtained at the 'y' axis. The thermal conductivity is measured by applying a steady state guarded hot measurement method. The synthesized CeO_2 nanoparticles placed between hot plate and cold plate. Then the hot plate is heated with the different temperature and the other plate being cooled. The heat power rate is transferred across the nanoparticles due to guarded heaters. After reaching the thermal equilibrium, the heat and cooling plates are kept in stable temperatures and the corresponding thermal conductivity is calculated. As shown in figure 9, with the increase of temperature, the thermal conductivity gets increased.

5. Conclusion

In this paper, enhance optical characterizations of is synthesized CeO_2 nanoparticles are successfully analyzed through the structural and light emission process. First, the Elliott resolved photoluminescence method is introduced for identifying the light emission characteristics of the synthesized CeO_2 nanoparticles and identifying the peak intensity at a particular wavelength of light. After that, the point defect in the photoluminescence intensity is detected to improve the optical and optoelectronic performance of the CeO_2. Steady state guarded hot measurement method is applied for measuring the thermal conductivity properties of the CeO_2 nanoparticles with respect to different temperatures. The performance analysis indicates that the synthesized CeO_2 nanoparticles improve the luminescence efficiency in terms of efficient better intensity and thermal conductivity.

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