



## A strategy to design lanthanide doped dual-mode phosphor mediated spectral convertor for solar cell applications

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### ABSTRACT

Herein, we demonstrate the synthesis of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor by a facile high-temperature solid-state method as a spectral convertor for the enhancement of conversion efficiency of solar cell. The dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor has cubic crystal structure with space group  $Ia\bar{3}$ . The surface morphology of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor has been investigated by scanning electron microscopy. The spectroscopic features of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor have been examined by photoluminescence (PL) and time-resolved photoluminescence (TRPL). The dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor gives strong red emissions at 611 nm and 663 upon excitation wavelengths of 200–500 nm and 980 nm, respectively. The dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor has average lifetime  $\sim 598.7 \mu\text{s}$ . Further, PL intensity distribution in dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor have been examined by PL confocal microscopy. The spatially resolved PL mapping results reveal that dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor has PL emission throughout the sample at excitation wavelengths of 375 nm and 980 nm, respectively. Hence, the obtained results suggest that dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}, \text{Er}^{3+}, \text{Yb}^{3+}$  phosphor could be a promising luminescent material for spectral convertor to improve the efficiency of next generation silicon solar cell.

### 1. Introduction

Globally the demand for energy has increased rapidly in recent times due to overpopulation and people opting for a luxurious lifestyle. It was predicted that the energy consumption could be double by 2015 as compared to reports in 2001 (globally) [1]. With less availability and higher demand for energy supply, the need to opt green and renewable energy resources have gained worldwide attention [2,3]. The most abundant form of energy, “Solar energy” involves no cost as a source and is present in almost all parts of the earth. Thus, solar energy can meet the upcoming demand acting as a source of energy. A solar cell, which converts the solar energy into electrical energy, is expected to be a promising candidate for green and renewable energy has already grabbed the attention of various scientific and industrial communities. It can tackle the energy demand by contributing a large portion in generating it and contribute to the overall sustainable development of society [4,5]. In past few decades, numerous efforts have been done in order to develop photovoltaic industry further. Still, the development of highly efficient solar cell at low cost and making it economically

feasible is still a big challenge [4].

The spectrum of Sun ranges from ultra-violet to infrared (280–2500 nm) which falls on the Earth’s surface. However, the silicon solar cell utilizes only a small portion of sun spectrum for its conversion to electrical energy. Theoretically, the single junction silicon solar cell has bandgap 1.1 eV with the efficiency of 30% [1]. While, 70% energy losses are due to a spectral mismatch between the solar cell response and solar spectrum. There are mainly two types of energy losses in solar cell; the thermal losses which are due to the relaxation of the hot charge carrier generated by absorption of high energy photons and transmittance of low energy photon (having energy lower than bandgap silicon i.e. 1.1 eV) [1]. When solar cell absorbs the photon having high energy, the excess energy is dispersed in the form of heat which is responsible for the increase in temperature and decrease in efficiency of the solar cell. On the other hand, the photons having lower energy than the bandgap of silicon are transmitted through the solar cell without electron-hole pair generation. The transmittance losses are dominant than thermal losses because a large part of the solar spectrum cannot be absorbed by the solar cell [1]. Both thermalization and transmittance

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losses can be minimized by using materials that can convert the high or low energy photons into a spectral range that can be utilized by the solar cell to generate an electron-hole pair. The use of luminescent materials to overcome the losses due the spectral mismatch is a promising approach, in order to enhance the conversion efficiency of solar cells. Therefore, many efforts have been directed towards the development of down-conversion/shift (DC/DS) and up-conversion (UC) luminescent materials for the spectral converter in photovoltaic applications. The down-conversion/shift (DC/DS) materials absorb high energy photons (UV photon) and convert them into lower energy photons (generally in the visible spectral region) which are absorbed by solar cell without any heat dissipation. Up-conversion materials absorb two or more photons of lower energy (NIR or IR photons) convert them into high energy photons (generally in visible region) which are absorbed by solar cell for generation of photocurrent [3,4]. The 50–60% of sun spectrum is distributed the NIR spectrum region (800–1700 nm) which is limits the efficiency of silicon solar cell [1,4,6–8]. Therefore, the upconversion materials are promising candidates as spectral converter. Theoretically, the spectral conversion efficiency of solar cell can be improved up to 50.7% by idea up-converter [8]. Many materials like quantum dots, nanocrystals, core-shell nanoparticles, hybrid nanomaterials (organic and inorganic) and lanthanide doped materials have been investigated for solar spectral converter [1,4,6–24]. But lanthanides doped luminescent materials have attracted more attention due to their electronic structure which permits it for facile photon management [4].

The lanthanide doped materials have been investigated extensively for many potential applications like solid state lighting, display panel, anti-counterfeiting application, biomedical and luminescent spectral converter due their ladder-like energy levels (known as Dieke diagram) [3,4,25–30]. The ladder-like energy levels of the lanthanide ions permits them to emit photons through various processes such as down-conversion/shift (DC/DS), quantum cutting (QC) and up-conversion (UC) [10]. Therefore, lanthanide doped materials are a promising candidate for the spectral converter applications. However, phosphor doped with  $\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$  and  $\text{Yb}^{3+}$  ions (for down-conversion and up-conversion) have been studied more as solar spectral converter [1,4,18]. Moreover, the fluorides are considered as a potential host for UC and DC phosphor due to their low phonon energy. But, the oxide host lattices have some advantages over fluoride host lattices such as high absorption efficiency in VUV region, high stability, easy synthesis and ease to form transparent layers which make them more suitable for practical applications [2]. The  $\text{Gd}_2\text{O}_3$  is also a good host matrix for down-conversion (DC) or up-conversion (UC) phosphors due to its good chemical durability, thermal stability, low toxicity, high photochemical stability and low phonon energy (phonon cutoff  $\sim 600 \text{ cm}^{-1}$ ) [31–35].

Further, various DC and UC phosphor have been explored as the solar spectral converter for solar cell applications. But, the dual-mode phosphors having both DC and UC in single host lattice are not much investigated for photovoltaic applications. It is also hard to design a lanthanide doped material which has emission in the visible region via both DC and UC processes. Although their few reports on the dual-mode hybrid materials as spectral converter are available in literature [10]. While, they have their own disadvantage like the stability of the organic compound in hybrid materials. Therefore, the synthesis of dual-mode luminescent materials for spectral converter is still a challenging task which is addressed in present investigation.

In present investigations, we reported the synthesis of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor for spectral converter for solar cell applications. The phosphor was synthesized by a facile solid-state method which can be easily produce at large scale. This phosphor has the ability to absorb UV and IR photons in single host lattice and emit photons in visible region which are absorbed by the solar cell for generation of an electron-hole pair. The dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor emit strong red color at 611 nm upon excitation with a wavelength of 251 nm while strong red color at 663 nm on excitation

wavelength of 980 nm. Further, the surface morphology of phosphor was investigated using scanning electron microscopy. Moreover, photoluminescence (PL) mapping results legitimate that dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor has PL emission throughout the sample at excitation wavelengths 375 nm and 980 nm, respectively. Thus, obtained optical results suggest that the dual-mode phosphor provides a new direction for the next generation solar cell to improve the solar cell efficiency via a broad spectral converter.

## 2. Experimental

### 2.1. Materials

The precursors;  $\text{Gd}_2\text{O}_3$  (99.99%),  $\text{Eu}_2\text{O}_3$  (99.99%),  $\text{Er}_2\text{O}_3$  (99.99%),  $\text{Yb}_2\text{O}_3$  (99.99%), were purchased from Sigma-Aldrich. All reagents were of analytical (AR) grade and used as received without further purification.

### 2.2. Synthesis of $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ , $\text{Er}^{3+}$ , $\text{Yb}^{3+}$ phosphor

The  $\text{Gd}_{1.4}\text{O}_3:\text{Eu}_{0.3}^{3+}$ ,  $\text{Er}_{0.1}^{3+}$ ,  $\text{Yb}_{0.2}^{3+}$  phosphor ( $\text{Eu}^{3+} = 15 \text{ mol\%}$ ,  $\text{Er}^{3+} = 5 \text{ mol\%}$  and  $\text{Yb}^{3+} = 10 \text{ mol\%}$ ) was synthesized by a facile high-temperature solid-state method which can be easily scaled-up in large quantity [3,36]. According to the stoichiometric ratio, the starting materials;  $\text{Gd}_2\text{O}_3$ ,  $\text{Eu}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ , and  $\text{Yb}_2\text{O}_3$  were taken. After precise weighting of  $\text{Gd}_2\text{O}_3$ ,  $\text{Eu}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ , and  $\text{Yb}_2\text{O}_3$ , the materials were crushed well in an agate mortar for homogeneous mixing. Further, the homogeneous mixture was kept into an alumina crucible and heated in a box furnace at the temperature of 1000 °C for 3 h. Using this method, the yield of synthesized phosphor obtained is more than 90% with a high degree of homogeneity.

### 2.3. Characterizations

The crystal structure identification of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor was performed by using X-ray powder diffraction (XRD) with Bruker AXS D8 Advance X-ray diffractometer, using  $\text{Cu K}\alpha_1$  radiation ( $\lambda = 1.5406 \text{ \AA}$  and angle 10–80° at a scanning rate of 2°/min). The surface morphology was examined by using Carl ZEISS EVOR-18 equipment at 10 kV operating voltage equipped with X-ray energy-dispersive spectroscopy. The photoluminescence (PL) and time-resolved photoluminescence (TRPL) spectroscopy were explored by using Edinburgh spectrometer (model no. FLS980) where xenon lamp and 266 nm diode laser act as sources of excitations, respectively. The NIR PL emission and PL mapping of phosphor were performed using a WITec alpha 300R+ confocal PL microscope system, where 375 nm and 980 nm diode lasers act as sources of excitations. The spot size of excitation beam is in confocal microscopy is  $1.38 \times 1.28 \text{ mm}$ . The scanning resolution of confocal microscopy is 160 nm.

## 3. Results and discussions

The crystal structure analysis of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor was investigated using XRD diffraction. The XRD pattern of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor is shown in Fig. 1a. The XRD result exhibits that the dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor has cubic crystal structure with space group  $\text{Ia}\bar{3}$  (JCPDS card no. 86-2477). The calculated lattice parameters for dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor are  $a = b = c = 10.77531 \pm 0.00030 \text{ \AA}$ , which is comparable with stranded parameters  $a = b = c = 10.809 \text{ \AA}$  (JCPDS card no.86-2477). Further, the estimated unit cell volume for dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor is  $1251 \pm 0.0929 \text{ \AA}^3$ . The Fig. 1b show the proposed crystal structure of  $\text{Gd}_2\text{O}_3$ , where Gd atoms are replaced by Eu, Er and Yb atoms. Further, the surface morphology of dual-mode  $\text{Gd}_2\text{O}_3:\text{Eu}^{3+}$ ,  $\text{Er}^{3+}$ ,  $\text{Yb}^{3+}$  phosphor was investigated by using scanning electron

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