



Luminescence properties of Pr^{3+} doped $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramics for potential applications in white LEDs and scintillators



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ABSTRACT

$(\text{Pr}_x\text{Y}_{0.9-x}\text{La}_{0.1})_2\text{O}_3$ ($x = 0.05, 1, 5$) transparent ceramics were fabricated by conventional ceramic processing. The absorption, emission spectra excited by ultraviolet and X-ray as well as the excitation spectra have been investigated as a function of the Pr^{3+} concentration. The emission under ultraviolet excitation shows the ability of Pr^{3+} to downconvert ultraviolet light to blue emission. The strong excitation band at 430–520 nm is in good agreement with the emissions of InGaN blue chips ($\lambda_{\text{em}} = 450\text{--}470$ nm), which can be efficiently excited by the 470 nm light and generate bright red emission at around 630 nm. The radioluminescence spectra demonstrate the possible application of Pr^{3+} : $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ in scintillator. In addition, the concentration quenching observed in the case of the P1 and P5 ceramics leads us to conclude that an optimum Pr^{3+} concentration to generate the best luminescence properties is achievable. Given overall luminescence properties, the low Pr^{3+} content (0.5 mol%) doped $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramic is a potential candidate for the applications in the ultraviolet-based and InGaN blue chip based white light-emitting diodes, as well as the scintillators.

1. Introduction

White light-emitting diodes (LEDs) are considered to be the next-generation lighting source due to their excellent advantages of power saving, long lifetime, high brightness and environmental friendliness [1]. In order to generate white light from LEDs, two typical methods are commonly used: first, mixing different color components from LED chips and second, using phosphors to downconvert the emission from a blue or ultraviolet (UV) LED to a longer wavelength, generating a higher efficient solid-state light [2]. In recent years, the combination of a blue-emitting LED and a yellow-emitting Ce^{3+} : $\text{Y}_3\text{Al}_5\text{O}_{12}$ (Ce^{3+} : YAG) phosphor has been much studied and developed which is relatively easy to fabricate white LEDs [3]. However, the white LEDs based on the Ce^{3+} : YAG phosphor exhibit a poor color rendering index ($\text{CRI} < 80$) because of the lack of a red light component (above 600 nm), which is not suitable for applications requiring high a CRI, such as medical lighting [4]. Therefore a red light source are currently needed to compensate LED for the red deficiency of output light [5]. Among many candidates, Pr^{3+} doped materials have been researched for many years owing to its broad red emission band at around 600–700 nm [6], which makes it a suitable candidate of Pr^{3+} ion as the red emission center in the combination of white LED. On another level, Pr^{3+} ion with strong absorption in the near UV region of 250–400 nm [7] downconvert the UV light to blue emission, which is also favorable for the application in

the UV converted white LEDs.

Scintillator materials have been widely studied due to its applications in medical imaging techniques, high energy and nuclear physics detectors and so on. Pr^{3+} ion also provides fast 5d-4f emission and a short decay time, which are of interest for numerous scintillator applications requiring high counting rates and high time resolution [8].

Those luminescence properties of Pr^{3+} for relevant applications require high chemical and physical properties and thermal stabilities of host, which are among the properties of Y_2O_3 transparent ceramics, together with their low-cost and easy preparation. Y_2O_3 displays a high thermal conductivity (13.6 W mK^{-1} , higher than YAG: 11 W mK^{-1}), low phonon energy (591 cm^{-1} , lower than YAG: 857 cm^{-1}) indicating a low multi-phonon relaxation property, high density (5.04 g cm^{-3} , higher than YAG: 4.56 g cm^{-3}) which makes it a more favorable host than YAG [9]. Our previous work shows that the sintering temperature of Y_2O_3 transparent ceramics can be much decreased to 1700°C (lower than the melting point of Y_2O_3 single crystal at 2430°C) without influence on optical properties by doping 10 mol% La_2O_3 as additive [10]. Previous work has fabricated a high-transparent Pr^{3+} -doped yttrium lanthanum oxide transparent ceramic, but it only studied the absorption spectrum among optical properties [7].

Little attention has been paid to the potential applications of Pr^{3+} -doped yttrium lanthanum oxide transparent ceramics in white LED. Besides, compared with powder phosphors, ceramic phosphors exhibit

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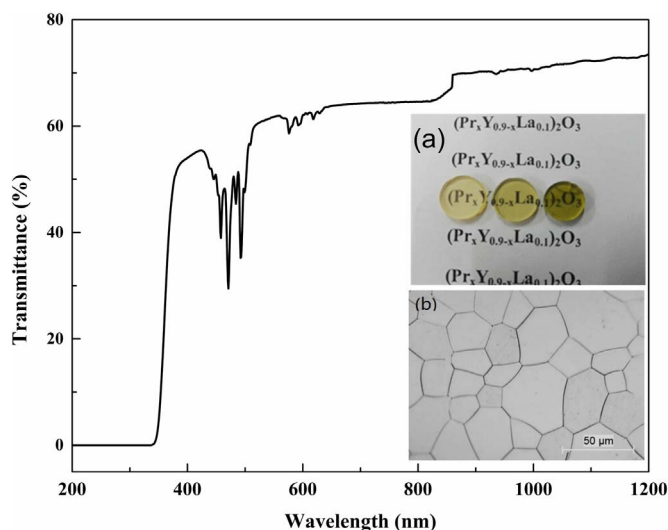


Fig. 1. In-line transmittance spectrum of P0.5 ceramic. Inset shows the (a) photograph of P0.5, P1 and P5 ceramics and (b) microstructure of the P0.5 ceramic.

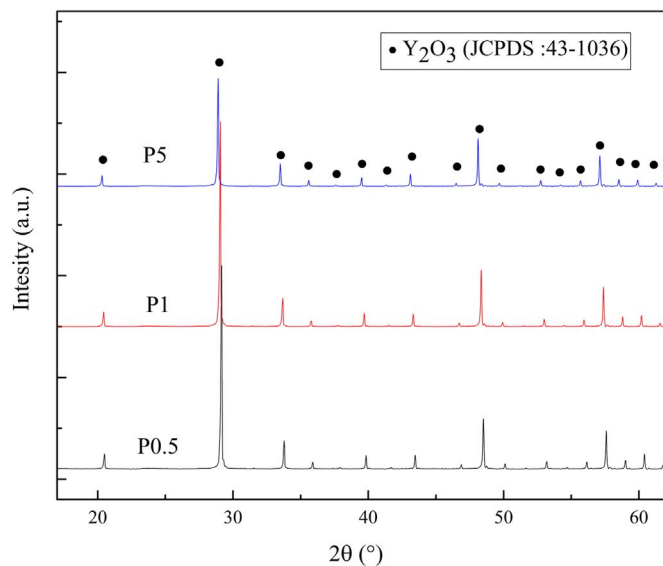


Fig. 2. XRD patterns of Pr^{3+} : $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramics.

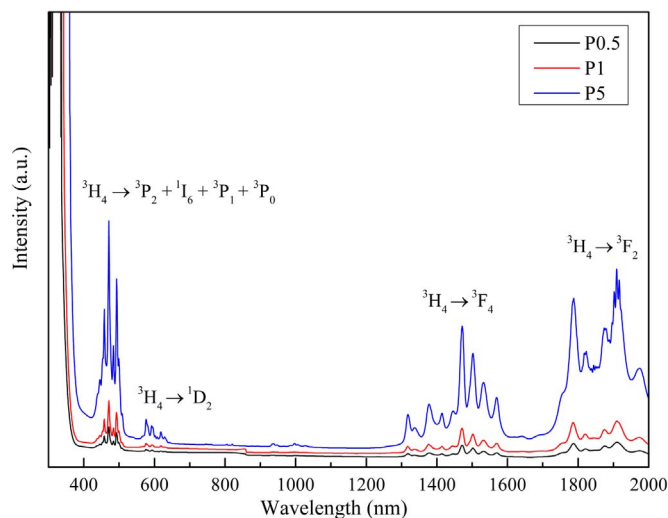


Fig. 3. Absorption spectra of Pr^{3+} : $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramics.

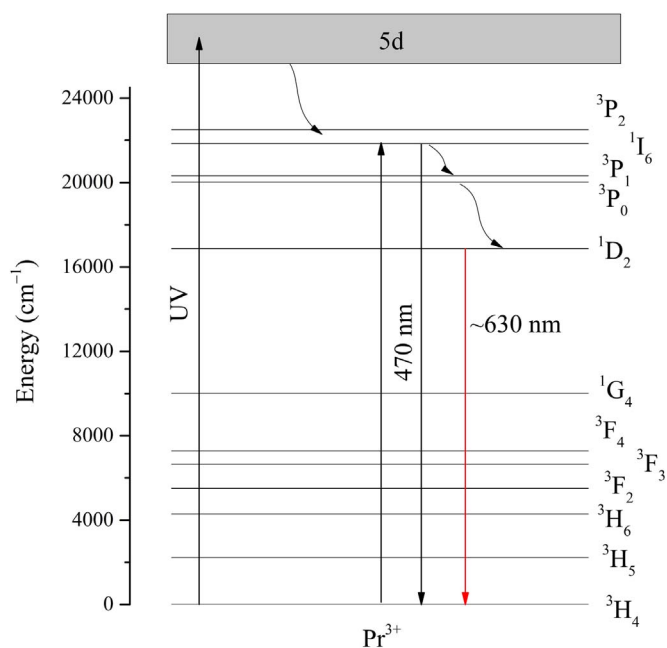


Fig. 4. Partial energy diagram of Pr^{3+} in the $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramics.

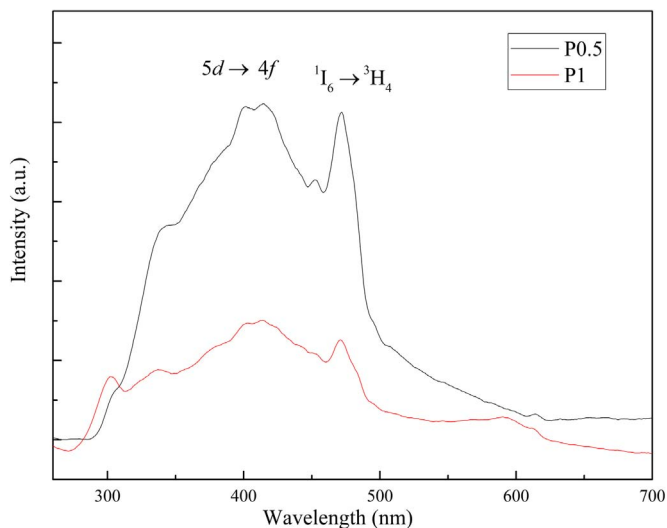


Fig. 5. The emission spectra of Pr^{3+} : $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramics under the UV (246 nm) excitation.

a longer lifetime of white LED devices without using organic resin [11]. In this work, the luminescence properties of Pr^{3+} : $(\text{Y}_{0.9}\text{La}_{0.1})_2\text{O}_3$ transparent ceramics as a function of Pr^{3+} concentration were investigated for the application of white LEDs and scintillators.

2. Experimental procedure

Starting materials Y_2O_3 (99.99 wt%), La_2O_3 (99.99 wt%) and Pr_6O_{11} (99.99 wt%) nanopowders, obtained from Rare-Chem. Hi-Tech Co., Ltd., China, were weighed with the composition (in mol%) of $(\text{Pr}_x\text{Y}_{0.9-x}\text{La}_{0.1})_2\text{O}_3$ ($x = 0.5\%$, 1% , 5%), which are denoted according to the nomenclature P0.5, P1 and P5 respectively. The compound were milled with ZrO_2 balls in alcohol for 5 h, dried and calcined at $1100\text{--}1200^\circ\text{C}$ for 20–25 h in air atmosphere. Pellets isostatically pressed under 200 MPa with 15 mm in diameter and 6 mm in thickness were sintered at $1500\text{--}1700^\circ\text{C}$ for 30–50 h in H_2 atmosphere.

Each specimen was double-polished with 2 mm for optical measurement. The in-line transmission and absorption spectra were

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