Effects of doping Pr^{3+} and Gd^{3+} into YAG:Ce phosphors on the luminescence properties

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Abstract. A series of $(\text{Y}_{0.98-x-y}\text{Ce}_{0.02}\text{Pr}_x\text{Gd}_y)_3\text{Al}_5\text{O}_{12}$ ($x = 0-0.02$, $y = 0, 0.3$) phosphors were prepared by a modified co-precipitation method. The phases, luminescence properties, effect of calcination temperature on luminescence properties and energy transfer (ET) were investigated. The results indicated that the doping of Pr^{3+} and Gd^{3+} in all samples did not produce any new phases but caused a slight lattice expansion. The series of $(\text{Y}_{0.98-x}\text{Ce}_{0.02}\text{Pr}_x)_3\text{Al}_5\text{O}_{12}$ phosphors emit additional red light at 612 nm, and when $x = 0.005$, the red/yellow ratio of the emission spectrum is the largest. Compared with the $(\text{Y}_{0.975}\text{Ce}_{0.02}\text{Pr}_{0.005})_3\text{Al}_5\text{O}_{12}$ phosphor, continued doping 30 at% Gd^{3+} in the $(\text{Y}_{0.675}\text{Ce}_{0.02}\text{Pr}_{0.005}\text{Gd}_{0.3})_3\text{Al}_5\text{O}_{12}$ sample makes the peak wavelength of yellow light emission shift from 533 to 561 nm. Doping Pr^{3+} and Gd^{3+} results in the decrease of integrated emission intensity, but the emission intensity and the red/yellow ratio can be increased by raising the calcination temperature. In addition, there is only the ET from Ce^{3+} to Pr^{3+} in the $(\text{Y}_{0.675}\text{Ce}_{0.02}\text{Pr}_{0.005}\text{Gd}_{0.3})_3\text{Al}_5\text{O}_{12}$ sample.

Keywords. Co-precipitation; YAG:Ce phosphor; white-light LED; energy transfer.

1. Introduction

As a new generation of solid-state lighting source, the white light-emitting diodes (LEDs) have a good market prospect and have been widely used in general illumination, mobile telephones, signal lights, LCD backlights, car lights and so on, due to some advantages of long life, high efficiency, absence of mercury and energy saving [1–5].

White light LEDs are usually produced in two ways. The first is combining the blue light technology with the yellow phosphor to produce white light. The second method is a mixture of multiple monochromatic light. At present, the first way is the most popular and mature in application. The core technique of the first way is coating the yellow emitting YAG:Ce phosphor [6] on a blue InGaN LED [7], then the white light can be obtained by the combination of non-absorbed blue emission from blue LED and broad yellow emission from the $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}$ phosphor. As is well known, Ce^{3+} -doped YAG particularly plays an essential role as a colour converter in (In,Ga)N-based phosphor-converted white light LEDs [8], because the phosphor can strongly absorb the blue light and subsequently produce a broad emission in the yellow light region, due to the large crystal field splitting of $^2\text{D}(5\text{d})$ level of Ce^{3+} [9]. Thus, a bright white light by combining the converted yellow light with the residual blue light can achieve an overall energy efficiency close to a compact fluorescent lamp. However, the white LEDs based on the yellow-emitting YAG:Ce phosphor inherently suffer from the deficiencies of low colour-rendering index (CRI) and high correlated colour

temperature (CCT) due to relatively weak emission in the red spectral region [10,11]. Therefore, a lot of effort has been made to improve the colour rendering properties of YAG:Ce phosphor. By and large, there are two effective ways to enhance the red emission of YAG:Ce phosphors: one is to substitute Y^{3+} with larger ions (e.g., Gd^{3+}) to increase the crystal field splitting and regulate Ce^{3+} emission to the long wavelength regions [9,12–14], and the other is co-doping the red-emitting ions (e.g., Pr^{3+}) into the YAG:Ce phosphor [10,15]. However, the method of combining both ways to enhance the red emission of YAG:Ce phosphor has scarcely been published.

Only when Pr^{3+} is doped in YAG:Ce phosphor, the increase of red emission component is not obvious, because Pr^{3+} is rarely doped. When the amount of the doped Pr^{3+} is more than 0.5 at.%, it is not beneficial to the luminescence properties of the sample [16]. Only if Gd^{3+} is co-doped in YAG:Ce phosphor, the paper [12] reported that the emission wavelength shifts to red with Gd^{3+} co-doping, but is accompanied by a 30% decrease of luminous efficacy. Considering the pros and cons of each side, both Pr^{3+} and Gd^{3+} are used for doping in YAG:Ce phosphor in this study. It is expected that the doping of Pr^{3+} and Gd^{3+} will not have any negative effect or have less negative effect on the luminescence properties of YAG:Ce phosphor except that the red emission component is enhanced. A series of YAG:Ce, Pr_x , Gd_y phosphors were prepared by the modified co-precipitated method reported by a previous work [17]. The phases, morphology, effects of doping Pr^{3+} and Gd^{3+} into the YAG:Ce

phosphor on the luminescence properties, effects of calcination temperature on luminescence properties, and energy transfer (ET) phenomenon among Ce^{3+} , Pr^{3+} and Gd^{3+} in YAG:Ce, Pr, Gd phosphor were investigated.

2. Experimental

The phosphor samples were prepared by the modified co-precipitated method. The rare earth and aluminium sources for phosphor synthesis are $\text{Pr}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.9%), $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.9%), $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.9%), $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.9%) and $\text{NH}_4\text{Al}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$ (99.9%). These salts weighted according to the chemical formula of $(\text{Y}_{0.98-x-y}\text{Ce}_{0.02}\text{Pr}_x\text{Gd}_y)_3\text{Al}_5\text{O}_{12}$ ($x = 0-0.02$, $y = 0, 0.3$) were dissolved into distilled water to obtain a mixed solution in which the concentration of Al^{3+} was 0.1 M. Ammonium hydrogen carbonate (NH_4HCO_3 , A.R.) was used as a precipitant. In the precipitating agent solution, the concentration of NH_4HCO_3 was 0.5 M, and 0.5% by volume triethanolamine (A.R.) which was used as a dispersant and pH regulator was added. Then the mixed solution was added into the precipitating agent solution at a speed of 200 ml h^{-1} accompanied by stirring at room temperature. After titration, the suspensions were aged for 24 h, and then filtered and washed with distilled water and alcohol. After drying the precipitates at 80°C for 24 h in a thermostatic drier, the obtained precursors were calcined at different temperatures for 2 h.

The phase formation process and microstructure of $\text{Pr}^{3+}/\text{Ce}^{3+}$:YAG powders were investigated by X-ray diffraction (XRD, D/max-rA model, using nickel-filtered $\text{Cu-K}\alpha$ radiation) and field emission scanning electron microscopy (SEM, Hitachi, S-4800). Photoluminescence (PL) spectra were measured with a Hitachi Spectra fluorimeter (F-7000).

3. Results and discussion

Figure 1 shows the XRD patterns of $(\text{Y}_{0.98}\text{Ce}_{0.02})_3\text{Al}_5\text{O}_{12}$, $(\text{Y}_{0.975}\text{Ce}_{0.02}\text{Pr}_{0.005})_3\text{Al}_5\text{O}_{12}$ and $(\text{Y}_{0.675}\text{Ce}_{0.02}\text{Pr}_{0.005}\text{Gd}_{0.3})_3\text{Al}_5\text{O}_{12}$ phosphors. All the XRD patterns are in good agreement with the standard profile of YAG (JCPDS 33-0040). No secondary phases are observed when the rare earth ions are doped, which confirms that Pr^{3+} , Ce^{3+} and Gd^{3+} successfully replaced Y^{3+} into the YAG lattice and did not induce significant changes in the host structure. Seen from the inset in figure 1 which shows the magnified picture of the strongest peak (420), the diffraction peak positions of these samples shifted to low angles with increase of the kinds of doped rare earth ions. The shift of the diffraction peaks is attributed to the lattice expansion arising from the replacement of Y^{3+} ($r = 1.019 \text{ \AA}$) by Ce^{3+} ($r = 1.143 \text{ \AA}$), Pr^{3+} ($r = 1.126 \text{ \AA}$) and Gd^{3+} ($r = 1.053 \text{ \AA}$) with larger ionic radii.

Figure 2 shows the SEM image of YAG:Ce, Pr, Gd phosphors calcined at 1000°C . As shown in figure 2, the morphology of the sample is approximately spherical. The particle

Figure 1. XRD patterns of (a) $(\text{Y}_{0.98}\text{Ce}_{0.02})_3\text{Al}_5\text{O}_{12}$, (b) $(\text{Y}_{0.975}\text{Ce}_{0.02}\text{Pr}_{0.005})_3\text{Al}_5\text{O}_{12}$ and (c) $(\text{Y}_{0.675}\text{Ce}_{0.02}\text{Pr}_{0.005}\text{Gd}_{0.3})_3\text{Al}_5\text{O}_{12}$.

Figure 2. SEM image of $(\text{Y}_{0.675}\text{Ce}_{0.02}\text{Pr}_{0.005}\text{Gd}_{0.3})_3\text{Al}_5\text{O}_{12}$.

size is homogeneous. The mean size is about 40 nm. There is only a slight adhesion between the particles, which shows that the phosphors have good dispersity. Obviously, such powders are beneficial to the luminescence properties of the phosphors.

Figure 3 shows the PL excitation spectra and PL emission spectra of the sample a $(\text{Y}_{0.995}\text{Pr}_{0.005})_3\text{Al}_5\text{O}_{12}$, sample b $(\text{Y}_{0.98}\text{Ce}_{0.02})_3\text{Al}_5\text{O}_{12}$, sample c $(\text{Y}_{0.975}\text{Ce}_{0.02}\text{Pr}_{0.005})_3\text{Al}_5\text{O}_{12}$ and sample d $(\text{Y}_{0.675}\text{Ce}_{0.02}\text{Pr}_{0.005}\text{Gd}_{0.3})_3\text{Al}_5\text{O}_{12}$, respectively. The PL excitation spectrum and PL emission spectrum of Pr^{3+} singly doped sample are shown in figure 3a. It can be seen that the PL emission spectrum of Pr^{3+} upon 288 nm excitation mainly consists of one strong emission band and two intense emission lines in the Pr^{3+} singly doped sample. The strong UV region emission band at 381 nm originates from the transition from the lowest-lying $4f5d$ state to $^3\text{F}_J$ ($J = 2, 3, 4$) state of Pr^{3+} . The group intense blue emission line at 488 nm originates from $^3\text{P}_0 \rightarrow ^3\text{H}_4$ transition. Another intense red emission line at 612 nm with a weak satellite line at 640 nm originates from $^1\text{D}_2 \rightarrow ^3\text{H}_4$ transition. The PL

Figure 3. PL excitation spectra and PL emission spectra of (a) $(Y_{0.995}Pr_{0.005})_3Al_5O_{12}$, (b) $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$, (c) $(Y_{0.975}Ce_{0.02}Pr_{0.005})_3Al_5O_{12}$ and (d) $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$.

excitation spectrum of Pr^{3+} singly doped sample by monitoring 612 nm emission consists of two groups of excitation bands. The group in the UV region consists of two strong excitation bands located at 288 and 238 nm, which correspond to the transitions from the ground state to two lowest energy levels of excited 4f5d state of Pr^{3+} . The group in the range of 420–500 nm originates from ground state to the $^3\text{P}_J$ ($J = 0, 1, 2$) levels of Pr^{3+} transitions. Figure 3b shows the PL excitation spectrum and PL emission spectrum of the Ce^{3+} singly doped sample. The emission spectrum of the Ce^{3+} singly doped sample exhibits a well-known yellow emitting broad band due to the transition from the lowest-lying 5d state to the 4f ground state, peaking at around 530 nm. The excitation spectrum of the Ce^{3+} singly doped sample consists of two excitation bands at 340 and 455 nm, which correspond to the two lowest energy levels of the crystal field splitting bands of excited 5d state of Ce^{3+} . Figure 3c and d shows the PL excitation spectra and PL emission spectra of YAG:Ce, Pr phosphor and YAG:Ce, Pr, Gd phosphor, respectively. It can be seen that the PL emission spectra of the YAG:Ce, Pr phosphor and YAG:Ce, Pr, Gd phosphor contain not only the yellow band of Ce^{3+} but also the intense red lines of Pr^{3+} only when Ce^{3+} is excited at 470 nm. Furthermore, the PL excitation spectra of the two samples by monitoring 612 nm emission contain not only the excitation bands of Pr^{3+} but also the excitation bands of Ce^{3+} . In the YAG:Ce, Pr, Gd phosphor, the emission of Gd^{3+} is not detected, which indicates that Gd^{3+} is not involved in ET between these rare earth ions. With the $^4\text{F}_7$ electronic configuration, Gd^{3+} has a stable electronic structure. The host environment has little effect on the energy levels of Gd^{3+} . In addition, the lowest excitation energy of Gd^{3+} is very high. The direct ET from Ce^{3+} to Gd^{3+} is hardly possible in the YAG system. Therefore, there is only the ET from Ce^{3+} to Pr^{3+} in the phosphors. Moreover, it can be seen from figure 3b–d that the peak wavelength of

Ce^{3+} shifts greatly towards long wavelength with the increase of the kinds of the doped rare earth ions. Especially, the peak wavelength of the YAG: Ce, Pr phosphor is located at 533 nm. while doping 30 at% Gd^{3+} in the YAG:Ce, Pr, Gd phosphor makes the peak wavelength shift to 561 nm. The redshift of the emission spectrum is up to about 28 nm. It indicates that when Y^{3+} was replaced by the larger Pr^{3+} and Gd^{3+} , Pr^{3+} and Gd^{3+} made the crystal field around Ce^{3+} ions stronger. According to the crystal field theory, the d-d orbital splitting of Ce^{3+} becomes larger under the influence of stronger crystal field [18]. However, compared with the Ce^{3+} singly doped sample, the redshift produced by the YAG: Ce, Pr phosphor is not obvious, which is due to the fact that Pr^{3+} is rarely doped. When the amount of the doped Pr^{3+} is more than 0.5 at%, it is not beneficial to the luminescence properties of the sample [16]. That is why Gd^{3+} is usually used to make the PL emission spectrum of Ce^{3+} redshift and improve the colour rendering properties of YAG:Ce phosphor.

In order to resolve the pathway of $\text{Ce}^{3+}-\text{Pr}^{3+}$ ET and avoid direct excitation into the $^3\text{P}_J$ ($J = 0, 1, 2$) levels of Pr^{3+} , we use 340 nm to excite the upper 5d state of Ce^{3+} . At the same time, in order to obtain the optimum doping concentration of Pr^{3+} , the PL emission spectra of the $(\text{Y}_{0.98-x}\text{Ce}_{0.02}\text{Pr}_x)_3\text{Al}_5\text{O}_{12}$ phosphors with various Pr^{3+} concentrations upon 340 nm excitation are shown in figure 4a. Besides a rapid relaxation down to the lowest 5d state, the excited upper 5d state of Ce^{3+} may have additional two possible pathways for de-excitation, which are radiative transition to the ground state and ET to the $^3\text{P}_J$ states of Pr^{3+} . However, the $^3\text{P}_0-^3\text{H}_4$ blue emission line of Pr^{3+} was not detected at 488 nm in all samples. This behaviour indicates that the pathway of $\text{Ce}^{3+}-\text{Pr}^{3+}$ ET hardly starts from both the upper 5d state and the lowest 5d state of Ce^{3+} to the $^3\text{P}_0$ level of Pr^{3+} . This conclusion is also supported by the fact that both the purple and yellow emission bands of Ce^{3+} do not spectrally overlap with the $^3\text{H}_4-^3\text{P}_J$ absorption lines of

Figure 4. PL emission spectra of (a) $(Y_{0.98-x}Ce_{0.02}Pr_x)_3Al_5O_{12}$ and (b) the indicated pathway of $Ce^{3+} \rightarrow Pr^{3+}$ ET.

Figure 5. PL emission spectra of (a) $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$, (b) $(Y_{0.975}Ce_{0.02}Pr_{0.005})_3Al_5O_{12}$ and (c) $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$.

Pr^{3+} in the blue spectral region. The appearance of the red emission line of Pr^{3+} is only the result of ET from the lowest 5d of Ce^{3+} to the 1D_2 of Pr^{3+} , as illustrated in figure 4b, because the yellow emission band of Ce^{3+} has a spectral overlap with the red line (zero-phonon line) of Pr^{3+} . For the series of $(Y_{0.98-x}Ce_{0.02}Pr_x)_3Al_5O_{12}$ phosphors, compared with the $x = 0.0025$ sample, although the yellow emission intensity of the $x = 0.005$ sample is decreased slightly, the red emission intensity is increased. Thus the PL emission intensity ratio of the red line from Pr^{3+} as an acceptor to the yellow band from Ce^{3+} as a donor (red/yellow ratio) is increased. It indicates that the efficiency of $Ce^{3+} - Pr^{3+}$ ET increases with the increase of Pr^{3+} concentration. When $x > 0.005$, not only does the integrated emission intensity decrease seriously, but the red/yellow ratio also decreases, which is attributed to the concentration quenching of Pr^{3+} as well as the lattice distortion and instability caused by Y^{3+} being replaced by Pr^{3+} ions with large ion size. The conclusion can also be used to explain why doping Pr^{3+} and Gd^{3+} results in the decrease in integrated emission intensity, as shown in figure 5. Therefore, the optimum doping concentration of Pr^{3+} in the YAG:Ce phosphor is $x = 0.005$.

Figure 5 shows that the integrated emission intensity of $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ (a), $(Y_{0.975}Ce_{0.02}Pr_{0.005})_3Al_5O_{12}$ (b) and $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ (c) decrease successively. Compared with $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ phosphor, the decrease of emission intensity of YAG:Ce, Pr phosphor is mainly attributed to the ET from Ce^{3+} to Pr^{3+} . For doping Gd^{3+} , according to the analysis above, the direct ET from Ce^{3+} to Gd^{3+} is hardly possible in the YAG system. However, the substitution of Gd^{3+} ions with large ion size can cause the lattice distortion and instability resulting in a decrease in photon energy. And doping Gd^{3+} ions can bring more lattice defects, such as anti-site cation etc. [12]. And because of the large amount of doped Gd^{3+} , these problems are amplified in the $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ phosphor. These problems are disadvantageous to the luminescence properties of the sample. That is why Gd^{3+} -doping can lead to the

Figure 6. PL emission spectra of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ calcined at different temperatures.

decrease of emission intensity. Of course, when Pr^{3+} is doped in YAG:Ce phosphor, these problems also exist.

Although the intensity of emission spectrum decreases with the increase of the kinds of doped rare earth ions, the emission wavelength of Ce^{3+} shifts to long wavelength, and the red/yellow ratio is obviously increased, which is beneficial to improve the CRI of YAG:Ce phosphors. In addition, the decreased PL emission intensity can be improved by increasing calcination temperature, as shown in figure 6. Figure 6 shows the PL emission spectra of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ calcined at different temperatures. When the calcination temperature was raised from 1000 to 1300 °C, it did not bring about any appreciable change of the peak position, but yielded a significant increment in integrated emission intensity. It indicates that the integrated emission intensity can be increased by raising the calcination temperature. The enhanced luminescence is primarily owing to the improved crystallinity of the sample. Meanwhile, it can be seen that the red/yellow ratio of the emission spectra of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ phosphors has been obviously increased with the increase of calcination temperature. This implies that the efficiency of $Ce^{3+} - Pr^{3+}$ ET increases with the increase of calcination temperature. This conclusion can be proved by the fluorescence decay curves.

Figure 7 shows the fluorescence decay curves of $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ phosphor and $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ phosphors calcined at 1000, 1100 and 1300 °C, respectively. All the fluorescence decay curves were measured by monitoring the yellow emission of Ce^{3+} at 561 nm upon 340 nm pulsed excitation. It is found that the fluorescence of $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ phosphor calcined at 1000 °C deviated slightly from the single exponential decay, and the average lifetime obtained from the decay pattern is 53.7 ns (τ_{Ce0}) which is not equal the intrinsic lifetime (61 ns), due to the fact that the relatively high Ce^{3+} concentration produced various non-radiative quenching processes in $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ phosphor. Compared with $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ phosphor calcined at 1000 °C, the fluorescence decay of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$

Figure 7. Fluorescence decay curves of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ obtained at different calcination temperatures.

phosphors calcined at 1000°C speeds up, which should be due to the $Ce^{3+}-Pr^{3+}$ ET process and various non-radiative quenching processes. When the calcination temperature of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ phosphor is increased from 1000 to 1300°C, the fluorescence decay speeds up continuously, and the corresponding average lifetimes obtained from these decay patterns are 42.2 (τ_{Ce1}), 38.5 (τ_{Ce2}) and 36.6 ns (τ_{Ce3}), which should be attributed to the fact that the efficiency of $Ce^{3+}-Pr^{3+}$ ET is increased with increasing calcination temperature. The relative $Ce^{3+}-Pr^{3+}$ ET efficiency of $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ phosphors which can be estimated using $\eta_{(Ce \rightarrow Pr)} = 1 - \langle \tau_{Ce} \rangle / \langle \tau_{Ce0} \rangle$ [16] are 21.4% (1000°C), 28.3% (1100°C) and 31.8% (1300°C), respectively. With the increase of calcination temperature, the sample has better crystallinity and fewer defects which are beneficial to the ET of $Ce^{3+}-Pr^{3+}$. Therefore, when the calcination temperature is increased from 1000 to 1300°C, the red/yellow ratio will be increased.

The above analysis shows that the YAG:Ce, Pr, Gd yellow phosphors developed in this work have more red portions in emission spectra than $(Y_{0.98}Ce_{0.02})_3Al_5O_{12}$ and $(Y_{0.975}Ce_{0.02}Pr_{0.005})_3Al_5O_{12}$ phosphors. Because a large red/yellow ratio is significant for the fabrication of white LEDs with a high CRI, the YAG:Ce, Pr, Gd yellow phosphors are desired for application in LEDs for warm-white lighting.

4. Conclusions

A series of $(Y_{1-x-y}Ce_{0.02}Pr_xGd_y)_3Al_5O_{12}$ phosphors were prepared by the modified co-precipitated method. The doping of Pr^{3+} and Gd^{3+} in all samples did not produce any new phases but caused a slight lattice expansion. For the series of $(Y_{1-x}Ce_{0.02}Pr_x)_3Al_5O_{12}$ phosphors, under excitation at either 340 or 470 nm, they both show a broad yellow emission peak at 530 nm and a narrow red emission peak at

612 nm. The optimum doping concentration of Pr^{3+} in YAG:Ce phosphor is $x = 0.005$. Compared with doubly doped sample $((Y_{0.975}Ce_{0.02}Pr_{0.005})_3Al_5O_{12})$, continued doping 30 at% Gd^{3+} in the $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ sample makes the peak wavelength of yellow emission shift from 530 to 561 nm. The appearance of the red emission peak at 612 nm and the redshift of the yellow emission are regarded helpful for improving the CRI of the phosphor. Doping Pr^{3+} and Gd^{3+} results in an overall decrease in the intensity of emission spectra, but the emission intensity and the red/yellow ratio can be increased by raising calcination temperature. In addition, the ET phenomenon from Ce^{3+} to Pr^{3+} was proved, and Gd^{3+} is not involved in ET in $(Y_{0.675}Ce_{0.02}Pr_{0.005}Gd_{0.3})_3Al_5O_{12}$ samples. Also, the efficiency of $Ce^{3+}-Pr^{3+}$ ET is increased with increasing calcination temperature.

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