Journal of Alloys and Compounds 509 (2011) 4209–4213

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Investigation on temperature dependence of photoluminescence in $Sr_{1.7}Eu_{0.3}M_xCeO_{4.15+x/2}$ ($M = Li^+$, Na^+, K^+, $x = 0, 0.3$) red phosphors

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ARTICLE INFO

Article history:
Received 14 September 2010
Received in revised form
10 November 2010
Accepted 20 November 2010
Available online 1 December 2010

Keywords:
Thermal quenching
$Sr_2CeO_4:Eu^{3+}$
Li^+
Na^+
K^+

ABSTRACT

The temperature-dependent luminescence of $Sr_{1.7}Eu_{0.3}M_xCeO_{4.15+x/2}$ ($M = Li^+$, Na^+, K^+, $x = 0, 0.3$) samples was investigated and discussed in the temperature range from 303 to 573 K. It is found that the thermal quenching temperature of samples decreases with Li^+-/Na^+-doping but increases with the incorporation of K^+. We suggest that these observations are resulted from two factors. One is that the incorporation of $Li^+/Na^+/K^+$ ions reduces the strength of potential field at the O^{2-} sites, and then results in a red-shift of the Eu–O charge transfer band. The other is that Δr expands with Li^+-/Na^+-doping but shrinks with K^+-doping. We consider that it is a feasible way to adjust the temperature-dependent luminescence properties of materials by introducing appropriate impurities.

1. Introduction

Danielson et al. discovered a new efficient blue–white rare earth phosphor, Sr_2CeO_4, which can be excited by UV light, cathode ray and X-ray [1]. Its luminescence is generally considered to originate from a ligand-to-metal Ce^{4+}–O^{2-} charge transfer state (CTS) [1,2]. Most studies on Sr_2CeO_4 in recent years were fixed on the luminescence properties of this host doped with trivalent luminescence centers such as Eu^{3+}, Sm^{3+}, Dy^{3+}, Ho^{3+}, Er^{3+}, Tm^{3+}, etc. [3–5]. In these phosphors, the luminescence of rare earth ions originates from the energy transfer from Ce^{4+}–O^{2-} CTS. Sr_2CeO_4 activated with high content of Eu^{3+} has been studied extensively as a promising red emitting material, in which the luminescence from the Ce^{4+}–O^{2-} CT band disappears [6,7]. However, there are few papers published on the temperature-dependent luminescence of $Sr_2CeO_4:Eu^{3+}$, which is an essential property for its application. It has been reported that Li^+ and Na^+ could produce a change of symmetry and vibration modes around the luminescence center to result in the enhancement of the luminescence intensity [8–10]. In our present work, we introduced photoluminescence properties of M^+-free or M^+ co-doped $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ (M = Li, Na, K) samples as a function of temperature, and calculated the thermal quenching temperature of samples. In addition, we discussed the thermal quenching mechanism of samples, especially the mechanism of alkali metal ions varying the thermal quenching temperature of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$. Based on our discussions, we found a possible way to change thermal quenching luminescence properties of fluorescence materials by adding appropriate impurity ions.

2. Experimental

All the powder samples were prepared by the high-temperature solid-state reaction method. The compositions of the as-calcined samples are $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ (SCOE), $Sr_{1.7}Eu_{0.3}Li_{0.3}CeO_{4.3}$ (SCOEL), $Sr_{1.7}Eu_{0.3}Na_{0.3}CeO_{4.3}$ (SCOEN), $Sr_{1.7}Eu_{0.3}K_{0.3}CeO_{4.3}$ (SCOEK), respectively. The Sr:Eu:M:O ratio shows the appropriate stoichiometric proportion of these compounds. The mixtures of the raw materials, such as analytical reagent (AR) grade $SrCO_3$, Li_2CO_3, Na_2CO_3, K_2CO_3, CeO_2 (99.99%) and Eu_2O_3 (99.99%), were ground thoroughly in an agate mortar and fired at 1323 K in air in a muffle furnace for 20 h with one intermediate grindings and then cooled down to room temperature.

The X-ray powder diffraction analyses were carried out with a Rigaku D/max 2500 X-ray powder diffractometer (Cu Kα radiation, 40 kV, 20 mA) at room temperature (RT). The photoluminescence excitation spectra (PLE) at room temperature (RT) were performed by a Fluorolog 3-21 spectrofluorometer (Jobin Yvon Inc/specx) equipped with a 450 W Xe lamp and double excitation monochromators. Temperature-dependent luminescence spectra were obtained by a system which consists of a computer controlled CCD detector and a heater. During the measurement, an UV lamp with maximum wavelength of 365 nm was adopted as the excitation source.

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doi:10.1016/j.jallcom.2010.11.159

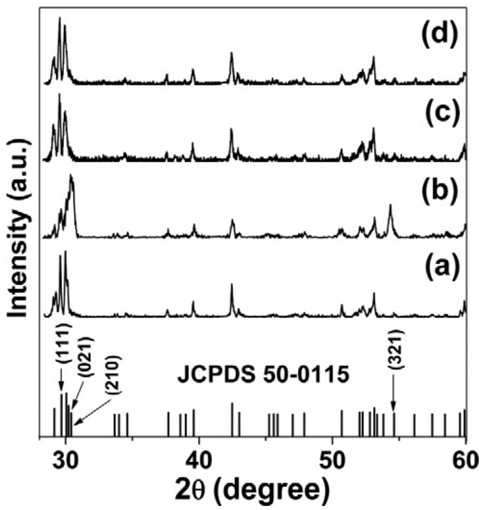

Fig. 1. XRD Patterns of samples (a) $Sr_{1.7}Eu_{0.3}CeO_{4.15}$, (b) $Sr_{1.7}Eu_{0.3}Li_{0.3}CeO_{4.3}$, (c) $Sr_{1.7}Eu_{0.3}Na_{0.3}CeO_{4.3}$ and (d) $Sr_{1.7}Eu_{0.3}K_{0.3}CeO_{4.3}$.

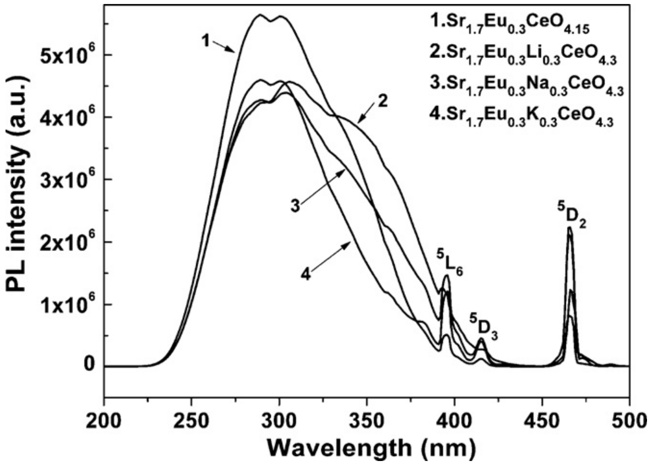

Fig. 2. Room-temperature photoluminescence excitation (PLE) spectra of samples $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ and $Sr_{1.7}Eu_{0.3}M_{0.3}CeO_{4.3}$ ($M = Li^+$, Na^+, K^+). The monitored wavelength is 616 nm.

3. Results and discussion

In order to characterize the phase purity of the samples, X-ray powder diffraction (XRD) measurements were performed for the as-synthesized samples. These XRD patterns of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$, and $Sr_{1.7}Eu_{0.3}M_{0.3}CeO_{4.3}$ ($M = Li^+$, Na^+, K^+) were plotted in Fig. 1, in good agreement with the reported powder pattern in JCPDS standard card numbered 50-0115 [Sr_2CeO_4], confirming the formation of single-phase crystalline products. Sr_2CeO_4 is indexed to an orthorhombic cell in space group *Pbam* [1]. The structure consists of linear chains of edge-sharing CeO_6 octahedra. The terminal Ce–O distance is about 0.1 Å shorter than the equatorial distance [1]. In the Sr_2CeO_4 structure, both the Ce^{4+} and Sr^{2+} ions are surrounded by six oxygen ions. As we know, when coordination number is equal to 6, the ionic radii of cations Ce^{4+}, Eu^{3+}, Sr^{2+}, Li^+, Na^+, K^+ are 87, 94.7, 118, 76, 102 and 138 pm, respectively [11]. Therefore, we believe that Eu^{3+} ions prefer to occupy both the Sr^{2+} sites and Ce^{4+} sites because of the similar radius and charge.

Effect of the incorporation of Li^+ ions on $SrTiO_3$ structure has been investigated in Ref. [12]. The authors considered it is not likely for Li^+ ion to be located in Ti^{4+} site for the big charge difference between them. However, we believe that Li^+ ion is small enough to occupy any crystal lattice site. In our present research, when introduced into Sr_2CeO_4, the Li^+ ions could be located at the sites of Sr^{2+} and Ce^{4+}, moreover, there is a possibility that some Li^+ ions resides in interstitial sites between or among the host ions. In addition, for Na^+ ions, they could be located at Sr^{2+} sites more easily than Ce^{4+} sites, but it is difficult for K^+ ions to replace Sr^{2+} or Ce^{4+} because of its bigger radius.

The corresponding cell constants and unit cell volumes of the samples are listed in Table 1. As we know, if the ions with smaller radius substitute the larger cations in the crystalline lattice, the cell volume of the host compound will decrease [13,14]. Therefore, as shown in Table 1, the cell volume of sample SCOEN decreases with the doping of Na^+ ions, because the ionic radii of Na^+ ions (102 pm) are smaller than that of Sr^{2+} ions (118 pm). This conclusion confirms that Na^+ ions prefer to occupy Sr^{2+} sites than Ce^{4+} sites, as discussed above. Following the same rule, when the K^+ ions are added as charge compensation, the cell volume of Sr_2CeO_4:Eu^{3+}, K^+ increases, because of its bigger radius than Sr^{2+}. However, the cell volume of Sr_2CeO_4:Eu^{3+} doped with Li^+ increases, despite the fact that the radius of Li^+ is smaller than that of Sr^{2+} or Ce^{4+}. The larger size of Li^+ ion than that of interstitial site would be responsible for this phenomenon. Besides, for Sr_2CeO_4:Eu^{3+}, Li^+, the relative intensity of crystal faces (0 2 1), (2 1 0) and (3 2 1) increase, but the relative intensity of crystal face (1 1 1) decreases, as observed in Fig. 1(b). These changes in the relative intensity of crystal faces can be attributed to the expansion of the two cell constants *a*, *c* and the contraction of the cell constant *b* of the sample.

Table 1
The calculated lattice parameters of all samples.

Phosphors	*a* (Å)	*b* (Å)	c (Å)	*V* (Å^3)
$Sr_{1.7}CeO_4$:$Eu_{0.3}{}^{3+}$	6.1174	10.3968	3.6024	228.52
$Sr_{1.7}CeO_4$:$Eu_{0.3}{}^{3+}$, $Li_{0.3}{}^+$	6.1242	10.3591	3.6057	228.75
$Sr_{1.7}CeO_4$:$Eu_{0.3}{}^{3+}$, $Na_{0.3}{}^+$	6.1014	10.3717	3.5976	227.66
$Sr_{1.7}CeO_4$:$Eu_{0.3}{}^{3+}$, $K_{0.3}{}^+$	6.1210	10.3714	3.6055	228.89

Fig. 2 presents the PLE spectra of $Sr_{1.7}Eu_{0.3}M_xCeO_{4.15+x/2}$ ($M = Li^+$, Na^+, K^+, $x = 0$, 0.3) samples, monitored at the strongest emission peaks at 616 nm. The weak sharp lines at 395 nm, 415 nm and 466 nm are ascribed to the transitions between the $^7F_0$ and the $^5L_6$, $^5D_3$, $^5D_2$ levels of Eu^{3+} ions. For the strong broad band of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$, with a maximum at 300 nm and two shoulders at 289 and 338 nm, some authors have ascribed it to the Ce^{4+}–O^{2-} CT band [15], however, some other authors have considered that the CT band of Ce^{4+} inhibits the CT band of Eu^{3+} in this lattice [6]. In the present case, we consider this broad band as the overlap of the Ce^{4+}–O^{2-} CT band and the Eu^{3+}–O^{2-} CT band. Because the Eu^{3+}–O^{2-} CT band is also around 300 nm, it is reasonable that we suppose the Eu^{3+}–O^{2-} CT band to be the middle part of this broad band, which peak is located at 300 nm. This assumption can simplify the question that how M^+ ions (M = Li, Na, K) influence energy of the Eu^{3+}–O^{2-} CT band, which is important for the thermal luminescence properties of samples in our work. Based on this assumption, it is clear that there is a red-shift for the Eu^{3+}–O^{2-} CT band with the addition of Li^+, Na^+ or K^+ ions, from 300 to 306, 304 or 301 nm, respectively. In other words, the red-shift of the Eu–O CT band decreases in the following order: SCOEL > SCOEN > SCOEK > SCOE.

The mechanism for the influence of Li^+, Na^+ or K^+ on the Eu^{3+}–O^{2-} CT band is discussed in detail as follows: Jörgensen has issued that energy of the Eu^{3+}–O^{2-} CTS depends on the potential field strength at the anion O^{2-}-site, in Cs_2O, BaO and La_2O_3 [16]. This potential field strength will be influenced by the charge and electronegativity values of the surrounding cations around O^{2-}. Su et al. [17–19] have carried out thorough researches on the Eu^{3+}–O^{2-} CTS in $A_2YM'O_6$ ($A = Ca^{2+}$, Sr^{2+}, Ba^{2+}, $M' = Sb^{5+}$, Nb^{5+}), $REM'SbO_6$ and $RE_2M'_2O_7$ ($RE = La^{3+}$, Gd^{3+} and Y^{3+}, $M' = Ti^{4+}$, Zr^{4+}) systems,

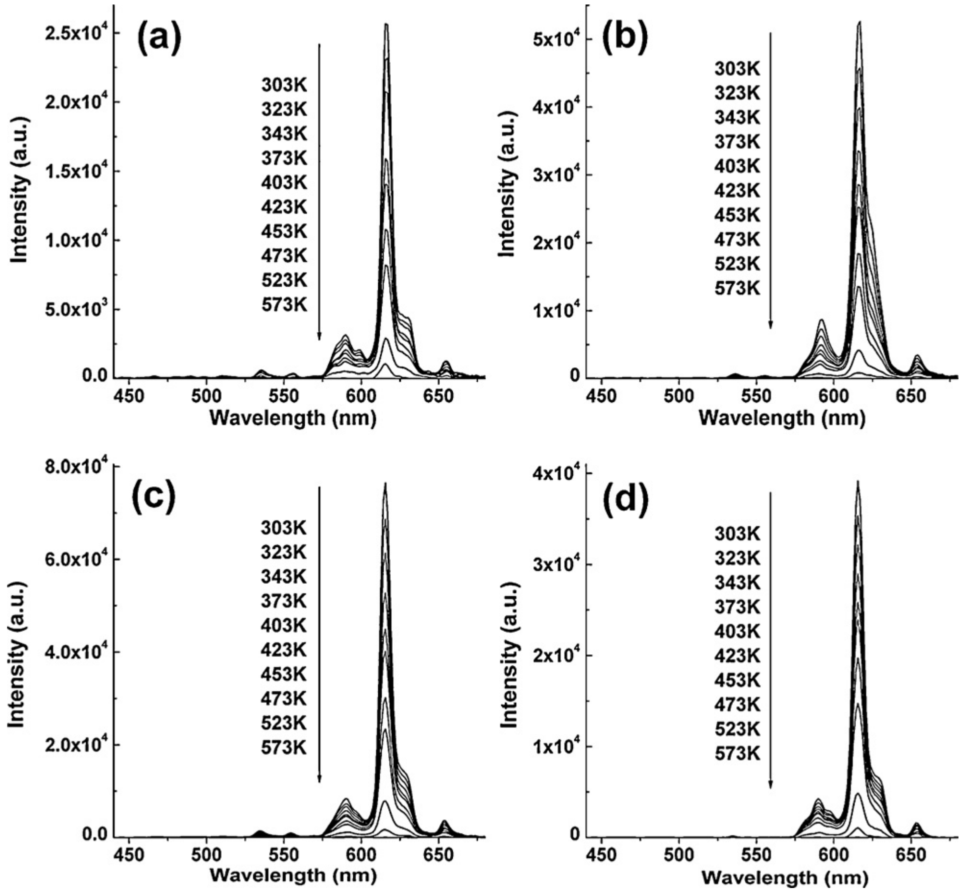

Fig. 3. The temperature-dependent photoluminescence emission spectra of all samples (a) $Sr_{1.7}Eu_{0.3}CeO_{4.15}$, (b) $Sr_{1.7}Eu_{0.3}Li_{0.3}CeO_{4.3}$, (c) $Sr_{1.7}Eu_{0.3}Na_{0.3}CeO_{4.3}$ and (d) $Sr_{1.7}Eu_{0.3}K_{0.3}CeO_{4.3}$. The excitation wavelength is 365 nm.

and then have drawn a conclusion that the M′ with lower electronegativity value will make the Eu^{3+}–O^{2-} CT band (in Eu^{3+}–O–M′) have a bigger red-shift. In the present study, when coordination number is equal to 6, electronegativity (EN) values of cations Ce^{4+}, Sr^{2+}, Li^{+}, Na^{+}, K^{+} are 1.608, 1,139, 1.009, 1.024 and 0.998, respectively, i.e., the sequence is $Ce^{4+} > Sr^{2+} > Na^{+} > Li^{+} > K^{+}$ [20]. Therefore, according to the above issues offered by Q. Su, the red-shift of the Eu–O CT band is expected with the addition of Li^{+}, Na^{+} or K^{+}. This deduction is in line with the excitation spectral data observed in Fig. 2. However, a further deduction that the red-shift of the Eu–O CT band decreases in the order K^{+}, Li^{+}, Na^{+} is inconsistent with the decreasing order Li^{+}, Na^{+}, K^{+} observed from the excitation spectra in Fig. 2. What is the reason for this inconsistency? According to the theory proposed by Jörgensen, in the Eu^{3+}–O^{2-}–M′, if the lattice ion M′ with higher charge is replaced, the potential field at O^{2-} will become weaker, resulting in the electrons of O^{2-} moving to Eu^{3+} more easily, and as a result, the Eu^{3+}–O^{2-} CT band has a larger red-shift. For Li^{+}-doping, it influences not only Eu^{3+}–O^{2-}–Sr^{2+} but also Eu^{3+}–O^{2-}–Ce^{4+}. For Na^{+}-doping, it influences Eu^{3+}–O^{2-}–Sr^{2+} more than Eu^{3+}–O^{2-}–Ce^{4+}. For K^{+}-doping, it influences only Eu^{3+}–O^{2-}–Sr^{2+}. As a result, the red-shift of the Eu^{3+}–O^{2-} CT band varies in a simple decreasing order $Li^{+} > Na^{+} > K^{+}$.

The temperature-dependent photoluminescence spectra of the samples under excitation of 365 nm are given in Fig. 3. As shown in parts (a)–(d) of Fig. 3, the main emission peaks located at about 591, 616 and 654 nm, are observed for these samples, which are attributed to the $^5D_0 \rightarrow {}^7F_1$, $^7F_2$ and $^7F_3$ transitions of Eu^{3+}, respectively. With an increase in the temperature ($T \geq 300$ K), the emission intensity of all samples decreases gradually without the change of the peak positions and shapes, indicating obvious thermal quenching behaviors. The integrated emission intensity of these samples at different temperatures is shown in Fig. 4. If the thermal quenching temperature (T_q) is defined as the temperature at which the integrated luminescence intensity is 50% of its original value [21–23], it can be deduced from the intensity of emission peaks at different temperature in Fig. 3. The calculated T_q value for $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ is 446 K, for Li^{+}, Na^{+} or K^{+} co-doped sample is 414 K, 436 K and

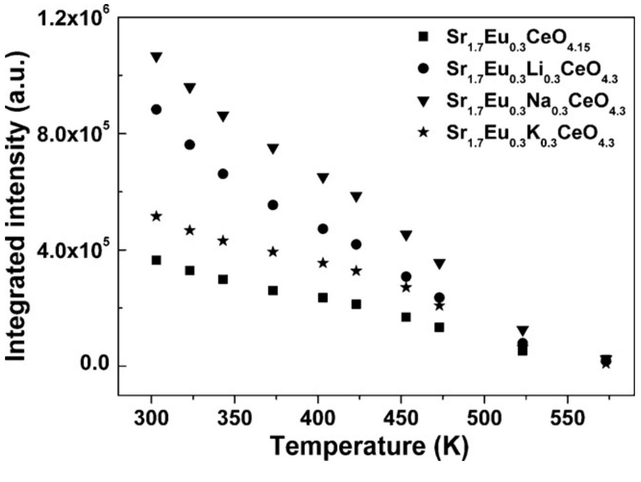

Fig. 4. The integrated emission intensity of samples $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ and $Sr_{1.7}Eu_{0.3}M_{0.3}CeO_{4.3}$ ($M = Li^{+}$, Na^{+}, K^{+}) at different temperatures.

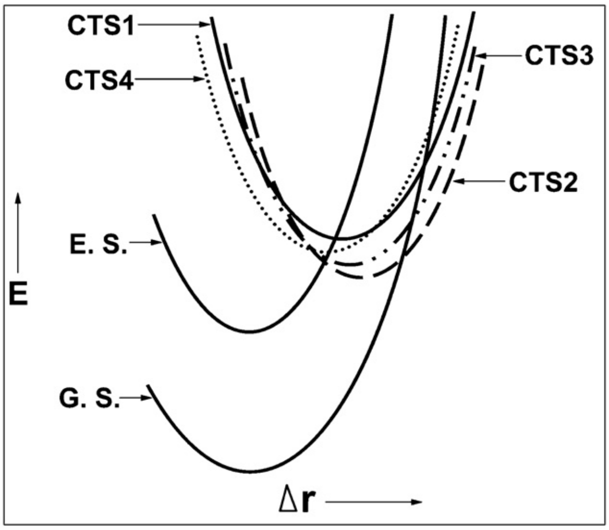

Fig. 5. A simple configuration coordinate model for the thermal quenching mechanism of the samples. The CTS1 (solid line) represents the Eu^{3+}–O^{2-} charge transfer state in $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ phosphor without Li^+, Na^+ or K^+ ions. The CTS2 (dashed line), CTS3 (dash-dot line), CTS4 (dotted line) refers the Eu^{3+}–O^{2-} charge transfer state in the phosphor added with Li^+, Na^+ or K^+, respectively. G. S. and E. S. are the ground and excited states of Eu^{3+}, respectively.

456 K, respectively. Therefore, the T_q value of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ decreases with the presence of Li^+, Na^+ but increases with K^+-doping, and the T_q values of samples reduce in the following order, SCOEK > SCOE > SCOEN > SCOEL.

The temperature-dependent luminescence of Eu^{3+} has been explored in many systems in the past several decades. Fönger and Struck [24,25] observed the thermal quenching phenomenon of Eu^{3+} luminescence in LaOCl, Y_2O_2S, and La_2O_2S. They interpreted the phenomenon taking into account the Franck–Condon shift between the Eu^{3+}–O^{2-} CT band and the 4f states and the resultant resonance crossovers between these states. That is, with increasing temperature, the excited electrons in ^5D states go back to the ground state nonradiatively via the crossovers. In the present study, we think that the thermal quenching behavior of the luminescence of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ can be attributed to the similar mechanism.

The quenching temperature of emission from luminescent centers in different host lattices has been studied by Blasse and Bril [26,27]. They have argued that there are two factors influencing on the T_q value of emission, for the luminescent centers that can be excited by charge transfer from the anion to the central cation. One is the energy of the charge transfer state (CTS); the other is Δr, the difference between the equilibrium configuration of the excited state and that of the ground state. In the present case, we will explain in detail how the incorporation of Li^+, Na^+ or K^+ ions influences the T_q value of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$, based on the above two factors.

The first influencing factor, the variation of Eu^{3+}–O^{2-} CTS of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ caused by $Li^+/Na^+/K^+$, has been discussed via the excitation spectra in Fig. 2. Then, we discuss how Li^+, Na^+ or K^+ influences the second influencing factor Δr. In Sr_2CeO_4, for the characteristic 4f–4f transitions of Eu^{3+}, Δr is zero; but for the Eu^{3+}–O^{2-} charge transfer transition, Δr has a positive value ($\Delta r > 0$) [26]. Namely, Δr of the Eu^{3+}–O^{2-} CT transition will expand when the excitation takes place in the Eu–O charge transfer state. van Pieterson et al. [2] studied the luminescence properties of Sr_2CeO_4 doped with the smaller Ca^{2+}. They considered the possibility of expansion of the excited Ce–O CT state is expected to be larger as a result of the presence of a smaller Ca^{2+} ion on a larger Sr^{2+}-site. According to this conclusion, in our present study, the introduction of doping ions with smaller radii into the crystalline lattice leads to larger expansion of the excited Eu–O CT state. Namely, Δr will expand with the addition of Li^+ or Na^+ ions, but will shrink with the doping of K^+ ions. Moreover, there is a decreasing order as follows: Δr (Li^+-doped) > Δr (Na^+-doped) > Δr (Li^+-, Na^+- or K^+-free) > Δr (K^+-doped).

Based on the above discussion, a configuration coordinate (CC) model for the mechanism of the present case is given in Fig. 5. The CTS1 indicates the Eu–O charge transfer state of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$, which intersects with the excited and the ground states of Eu^{3+}. With an increase in the temperature, the electrons in excited 5D states are thermally agitated to the crossover points and then nonradiatively go back to the ground state via the CTS1, leading to the thermal quenching of characteristic luminescence in $Sr_{1.7}Eu_{0.3}CeO_{4.15}$. In addition, it can be easily deduced from CC model that the lower energy of the CTS or the larger Δr makes the intersection point lower. Therefore, according to the influence of Li^+, Na^+ or K^+ on energy of the Ce–O CTS and the Δr discussed above, with the doping of Li^+, Na^+ or K^+ into the lattice, the Eu^{3+}–O^{2-} CTS of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ moves from CTS1 to CTS2, CTS3 or CTS4, respectively. The electrons in the CTS2, CTS3 or CTS4 can go back nonradiatively to the ground state via the intersection point at a lower temperature than in CTS1, that is, the quenching temperature (T_q) of $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ will vary with the doping of Li^+, Na^+ or K^+. Specifically, for samples without or with M^+ (M = Li, Na, K) ions, the T_q values of samples decreases in the following order, SCOEK > SCOE > SCOEN > SCOEL.

4. Conclusions

The temperature-dependent luminescence of $Sr_{1.7}Eu_{0.3}M_xCeO_{4.15+x/2}$ (M = Li^+, Na^+, K^+, $x = 0, 0.3$) is introduced in our present researches. The thermal quenching of the samples is due to the excited electrons are thermally moved to the crossover points and then nonradiatively go back to the ground state via the Eu–O CTS. The addition of $Li^+/Na^+/K^+$ will change the position of the Eu–O CTS, consisting of energy and Δr. Firstly, the CTS moves to longer wavelength. Secondly, the difference between the equilibrium configuration of the excited Eu^{3+}–O^{2-} CT state and that of the ground state of Eu^{3+}, i.e., Δr, becomes larger with the doping of Li^+ or Na^+ but smaller with that of K^+. Eventually, these two variations result in the decreasing of the T_q values. We think that it is a feasible method to adjust the temperature-dependent luminescence properties of one material by adding appropriate impurities. A further research on $Sr_{1.7}Eu_{0.3}CeO_{4.15}$ red phosphor co-doped with other cations, which own various charge, EN values and ionic radii, can be carried out to confirm our deduction.

Acknowledgment

The work was financially supported by National Natural Science Foundation of China (Grant No. 20921002).

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