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Europium (II)-Activated Oxonitridosilicate Yellow Phosphor with Excellent Quantum Efficiency and Thermal Stability - A Robust Spectral Conversion Material for Highly Efficient and Reliable White LEDs

Le Wang, Hong Zhang, Xiao-Jun Wang, Benjamin Dierre, Takayuki Suehiro, Takahashi Takeda, Naoto Hirosaki, Rong-Jun Xie

Clarifying physicochemical properties of a material is of great importance to design and utilize it in a right way. In this paper, we conduct a comprehensive survey of photoluminescence spectra, localized cathodoluminescence, temperature-dependent luminescence efficiency, and applications of Eu$^{2+}$-doped Sr$_2$Ba$_2$Si$_2$O$_7$N$_2$ in solid state lighting. This phosphor exhibits a broad emission band with a maximum at 560-580 nm and a full-width at half maximum of 92-103 nm upon the blue light excitation, whereas a dual-band emission (i.e., 470 nm and 550 nm) is observed under the electron beam irradiation due to the intergrowth of Ba$_2$Si$_2$O$_7$N$_2$:Eu$^{2+}$ and Sr$_2$Ba$_2$Si$_2$O$_7$N$_2$:Eu$^{2+}$ in each phosphor particle. Under the 450 nm blue light irradiation, this yellow phosphor exhibits excellent luminescence properties with absorption, internal and external efficiencies of 83.2, 87.7 and 72.6%, respectively. Furthermore, it also possesses high thermal stability, with the quantum efficiency being decreased by only 4.2% at 150°C and a high quenching temperature of 450°C.

High efficiency white LEDs using the title phosphor have the luminous efficacy, color temperature and color rendition of ~120 lm/W, 6,000 K and 61 respectively, validating its suitability for use in solid state white lighting
phosphors generally show lower absorption of blue light and conversion efficiency than YAG:Ce³⁺, searching for highly efficient yellow phosphors is thus continuously pursued by materials scientists and chemists.

![Figure 1](https://example.com/f1.png)  
*Figure 1* Crystal structure of Sr₁₋₀.₅Ba₀.₅Si₂O₅N₂ projected along the [100] (left) and along the [001] (right) directions (Sr/Ba: atoms: green, Sr/Ba: atoms: red, O: atoms: cyan, and N: atoms: orange).

Alkaline earth oxonitridosilicates, M₀.₅Si₂O₅N₂:Eu²⁺ (M = Ca, Sr, Ba), have been shown interesting blue-green (BaSi₂O₅N₂:Eu²⁺, λₘₐₓ = 495 nm), green (SrSi₂O₅N₂:Eu²⁺, λₘₐₓ = 540 nm) and yellow (CaSi₂O₅N₂:Eu²⁺, λₘₐₓ = 555 nm) emissions, exhibiting a broadband excitation spectrum covering the blue spectral region. The emission colour of these phosphors is tunable by forming solid solutions between the end members. Partial substitution of Sr²⁺ by Ca²⁺ or Ba²⁺ leads to a redshift of the Eu²⁺ emission due to the increased crystal-field splitting (Ca²⁺ → Sr²⁺) or a less restricted local lattice relaxation at the Eu²⁺ sites (Ba²⁺ → Sr²⁺). It crystallizes in a triclinic system with space group of P1, and lattice parameters are a = 7.2059(2), b = 7.3887(3), c = 7.3340(2) Å, α = 88.524(4), β = 84.454(3), γ = 75.980(4). There are two Sr/Ba crystallographic sites, and each Sr/Ba atom is coordinated to six oxygen atoms and one nitrogen atom. The average bond length of Sr-O is 2.68 Å. They also revealed the real structure of Sr₁₋₀.₅Ba₀.₅Si₂O₅N₂:Eu²⁺. It crystallizes in a triclinic system with space group of P1, and lattice parameters are a = 7.2059(2), b = 7.3887(3), c = 7.3340(2) Å, α = 88.524(4), β = 84.454(3), γ = 75.980(4). There are two Sr/Ba crystallographic sites, and each Sr/Ba atom is coordinated to six oxygen atoms and one nitrogen atom. The average bond length of Sr-O is 2.68 Å. They also discussed the relations between the crystal structure and the luminescence of Sr₁₋₀.₅Ba₀.₅Si₂O₅N₂:Eu²⁺ (2 mol% Eu²⁺), and clarified the abnormal redshift of the Eu²⁺ emission with the Ba substitution.

As a potential yellow luminescent material for solid state lighting, a deeper insight into the photoluminescence properties is thus continuously pursued by materials scientists and chemists.

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3. RESULTS AND DISCUSSION

3.1 Photoluminescence Spectra

As seen in Fig. 2, the excitation spectrum of Sr_{0.5-x}Eu_{x}Ba_{0.5}Si_{2}O_{5}N_{2} is quite broad, covering the spectral range of 250 to 500 nm. This broad excitation band can be deconvoluted into five sub-bands by Gaussian fitting, which are centered at 260, 304, 350, 406, and 472 nm, respectively. These sub-bands can be considered as five 5d energy levels of Eu^{2+}, so that the crystal field splitting and the center of gravity are then calculated as 17,200 and 36,400 cm^{-1} respectively. The emission spectrum exhibits an unsymmetrical broad excitation band can be deconvoluted into five sub-bands by Gaussian fitting, which are centered at 260, 304, 350, 406, and 472 nm, respectively. The Stoke shift is thus calculated as 3,600 cm^{-1}, which is quite close to 3,573 cm^{-1} reported in the literature.\(^{25}\) The larger Stokes shift observed in the Sr_{0.5-x}Eu_{x}Ba_{0.5}Si_{2}O_{5}N_{2} samples is attributed to the Sr-O band due to the larger Pauling electronegativity of Eu\(^{2+}\) (Sr: 0.95, Eu: 1.2, O: 3.44).\(^{35}\)

![Figure 2](image1.png)

**Figure 2** Excitation (left) and emission (right) spectra of Sr_{0.5-x}Eu_{x}Ba_{0.5}Si_{2}O_{5}N_{2} (x = 0.04). The emission spectrum was measured under the 450 nm irradiation, and the excitation spectrum was monitored at 570 nm.

**Figure 3** Excitation and emission spectra of Sr_{0.5-x}Eu_{x}Ba_{0.5}Si_{2}O_{5}N_{2} with varying Eu concentrations.

The Stoke shift of Eu\(^{2+}\) is about 94 nm. The Stoke shift is thus calculated as 17,200 and 36,400 cm^{-1} respectively. The emission spectrum exhibits an unsymmetrical band with the maximum at 570 nm (x = 0.04), and the full width at half maximum (FWHM) is about 94 nm. The Stoke shift is thus calculated as 3,600 cm^{-1}, which is quite close to 3,573 cm^{-1} reported in the literature.\(^{25}\) The larger Stokes shift observed in the Sr_{0.5-x}Eu_{x}Ba_{0.5}Si_{2}O_{5}N_{2} samples is attributed to the Sr-O band due to the larger Pauling electronegativity of Eu\(^{2+}\) (Sr: 0.95, Eu: 1.2, O: 3.44).\(^{35}\)

![Figure 4](image2.png)

**Figure 4** Variations of the emission peak and the full width at half maximum (FWHM) of Sr_{0.5-x}Eu_{x}Ba_{0.5}Si_{2}O_{5}N_{2} as a function of the Eu concentration (x).

With increasing the Eu concentration, both of the excitation and emission bands are red-shifted, as shown in Fig. 3. The emission maximum is linearly increased, shifting from 560 to 580 nm when the Eu concentration increases from x = 0.005 to x = 0.15, as shown in Fig. 4. At the same time, the emission band broadens with increasing the doping level, resulting in the increase of FWHM (Fig. 4). The right wing of the excitation spectrum is intensified gradually as the Eu doping level increases. These increases are mainly ascribed to the enhanced covalence of the Eu-O band with respect to the Sr-O band due to the larger Pauling electronegativity of Eu\(^{2+}\).

The concentration quenching occurs when the Eu concentration reaches x = 0.05, as shown in Fig. 5. The critical distance (R\(_{c}\)) for the energy transfer can be estimated by using the formula suggested by Blasse:\(^{36}\)

\[
R_{c} = \left( \frac{3 \gamma}{4 \pi \kappa N} \right)^{1/3}
\]

Here \(\gamma\) is the critical concentration, \(N\) the number of cation sites in the unit cell, and \(V\) the volume of the unit cell. For Sr_{0.5-x}Ba_{0.5}Si_{2}O_{5}N_{2}, \(\gamma_{c} = 0.05\), \(N = 4\), and \(V = 377.07\ \text{Å}^{3}\). By using the equation, the critical distance for energy transfer is calculated to be 15.33 Å. The decay time for Sr_{0.5-x}Ba_{0.5}Si_{2}O_{5}N_{2}:Eu is 95 - 1.07 µs, which is in the range of decay time usually for Eu\(^{2+}\) (i.e., 0.2 - 2 µs).\(^{37, 38}\) A tendency in the reduction of decay time with increasing the dopant concentration is seen in Fig. 6, although the decline is quite small. Taking into account of the experimental error, it is noted that the decay time nearly keeps invariable when the Eu concentration reaches x ≈ 0.5. The variation in decay time is consistent with the concentration quenching shown in Fig. 5.

As we may know, the excited Eu\(^{2+}\) centres can decay to the ground state by radiative and nonradiative processes, giving a so-called decay time signal. The total decay rate \(A_{T}\) is the sum of the radiative \(A_{R}\) and nonradiative \(A_{NR}\) rates, which can also be written as

\[
A_{T} = A_{R} + A_{NR}
\]

Or

\[
\frac{1}{\tau} = \frac{1}{\tau_{R}} + \frac{1}{\tau_{NR}}
\]

Where \(\tau\), \(\tau_{R}\) and \(\tau_{NR}\) is the luminescence decay time, radiative and nonradiative decay time, respectively. Given that the nonradiative decay time of a very diluted sample is zero, the radiative decay time of Sr_{0.5-x}Ba_{0.5}Si_{2}O_{5}N_{2}:Eu can be obtained from the sample with x =
0.005, i.e., $\tau_0 = 1.072\ \mu$s. Therefore, the decrease of the luminescence decay time with increasing the Eu concentration definitely leads to the enhanced nonradiative rate (Fig. 6), indicative of the luminescence loss caused by the radiativeless energy transfer.

![Image 69x123 to 297x329]

Figure 6 Decay time of $\text{Sr}_{1.5}\text{Eu}_{0.5}\text{Ba}_{2}\text{Si}_2\text{O}_2\text{N}_2$ as a function of the Eu concentration. The samples were excited at 370 nm and the 568 nm emission was monitored.

3.2. Dual-Band Cathodoluminescence

Differing from photoluminescence that only excites electrons into energy levels with energies lower than incident photons, cathodoluminescence promises to pump the electrons directly into the conduction band of host crystals, thus enabling to detect the emissions that cannot be appreciably found in the PL measurement, and allowing to understanding the localized luminescence of phosphors. A great difference between photoluminescence and cathodoluminescence one is clearly seen in Fig. 7, where the CL spectrum of one phosphor particle exhibits two emission bands centred at 470 and 550 nm, respectively. These two bands cannot be assigned to the emissions from two different phases rather than one phase with two luminescence centers. This is also evidenced by the great difference in the band width of the emission spectra. The full width at half maximum of the emission band is 37 and 82 nm for 470 and 550 nm, respectively. The very narrow emission band centered at 470 nm is equivalent to that of $\text{BaSi}_2\text{O}_2\text{N}_2$ that has a different crystal structure from $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{Si}_2\text{O}_2\text{N}_2$. It is thus supposed that the intergrowth of $\text{Sr}_{0.5-x}\text{Ba}_{0.5-x}\text{Si}_2\text{O}_2\text{N}_2$ (it has a shorter emission wavelength than $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{Si}_2\text{O}_2\text{N}_2$, so $\sigma = 0$) and $\text{BaSi}_2\text{O}_2\text{N}_2$ occurs in each particle, forming an interesting and special woven structure. As seen in Fig. 8, the elemental mapping of Si, O, N and Eu is very uniform in a phosphor particle investigated, whereas that of Sr and Ba is obviously position-dependent. It evidences the intergrowth of a Ba-enriched phase ($\text{BaSi}_2\text{O}_2\text{N}_2$) and a Sr/Ba-contained phase ($\text{Sr}_{0.5}\text{Ba}_{0.5}\text{Si}_2\text{O}_2\text{N}_2$). Schnick et al. addressed that crystallites of $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{Si}_2\text{O}_2\text{N}_2$ were built up of small anti-phase domains within larger twin domains, and Li et al. also confirmed the nanophase segregation and mixing in some compositions. Both observations support the proposed intergrowth in this work. The discussion of the intergrowth structure is beyond the scope of this work, and will be presented elsewhere.

The dual-band cathodoluminescence is not solely limited to one or several phosphor(s) but a universal phenomenon for the title phosphor. On the other hand, such a dual-band emission is not observed in samples under the 450 nm blue light irradiation, which can be attributed to reabsorption as a result of the broadband...
excitation spectrum covering the UV-to-blue spectral region. Therefore, the dual-band cathodoluminescence will not affect the title phosphor to be used as a LED phosphor. In addition, Li et al. observed a similar dual-band emission for phosphors with $y > 0.65$ in $\text{Sr}_2\text{Ba}_2\text{Si}_2\text{O}_5\text{N}_2$:Eu when excited at 405 nm, so that the color tuning could be realized by just controlling the Sr/Ba ratio.\(^{32}\)

![Figure 9](image)

Figure 9 Diffuse reflectance spectra of $\text{Sr}_2\text{Ba}_2\text{Si}_2\text{O}_5\text{N}_2$ and $\text{Sr}_0.5\text{Eu}_0.5\text{Ba}_2\text{Si}_2\text{O}_5\text{N}_2$ ($x = 0.04$).

3.3. Diffuse Reflectance, Absorption and Quantum efficiency

The diffuse reflectance spectra of samples of $x = 0$ and 0.04 are given in Fig. 9. The optical absorption edge of the $\text{Sr}_2\text{Ba}_2\text{Si}_2\text{O}_5\text{N}_2$ host is situated at ~213 nm, indicative of the band gap of 5.82 eV which is a little bit larger than 5.6 eV for $\text{SrSi}_2\text{O}_5\text{N}_2$.\(^{33}\) The onset of the optical absorption edge of the phosphor is located at 444.5 nm (~2.79 eV), which is ascribed to the 4f $\rightarrow$ 5d electronic transition of Eu\(^{2+}\). As the doping amount of Eu increases, the absorption band is clearly seen to redshift (see Fig. 10a). The redshift is in a good agreement with the changes in excitation spectrum with the dopant concentration. Meanwhile, the absorption of the phosphor is enhanced with increasing the Eu content. Upon the 450 nm excitation, the absorption efficiency increases obviously from 60 to 85% for $x = 0.01$ and 0.12, respectively. For phosphors in solid state lighting, the blue light absorption is required to be as high as possible, enabling more photons to be involved in the luminescence process.

The effect of the Eu concentration on the external quantum efficiency of the yellow phosphors is shown in Fig. 10b. Upon the blue light irradiation, the efficiency is optimized at $x = 0.08$ (66%). It is not consistent with the critical concentration for concentration quenching \(i.e., x = 0.05\). The reason for this difference can be explained by the different variations of absorption efficiency and internal quantum efficiency with the Eu concentration. As we may know, quantum efficiency, including internal and external quantum ones, is a key parameter for understanding the luminescence efficiency of a phosphor. The internal quantum efficiency is the ratio of the number of photons emitted by the sample to the number of photons absorbed by the sample, whereas the external one is the ratio of the number of photons emitted by the sample to the number of incident photons to the sample. Therefore, the external quantum efficiency is equal to the internal one multiplied by the absorption efficiency, which is thus usually smaller than the internal one. This loss is ascribed to the nonradiative photophysical processes such as scattering, trapping or energy transfer. The internal quantum efficiency is proportional to the integrated emission spectra that has the maximum at $x = 0.05$, which is also in agreement with concentration quenching (or decay time), whereas the absorption efficiency continuously increases with increasing the Eu concentration. This finally results in an optimized dopant concentration \(i.e., x = 0.08\) for the external quantum efficiency that is different from the one for internal quantum efficiency.

3.4. Temperature-Dependent Luminescence Efficiency

Thermal stability is one of key technological parameters for phosphors used in solid state lighting because the GaN-based chip generates heat that will quench the luminescence of spectral conversion materials. It can be evaluated by measuring the temperature dependent luminescence efficiency including photoluminescence intensity and quantum efficiency. As shown in Fig. 11, the photoluminescence intensity tends to decline as the temperature rises, due to the thermally-induced non-radiative energy transfer. The photoluminescence intensity at 150 and 200°C maintains 90 and 84% of the initial intensity (25°C) respectively, indicative of very small thermal quenching and of good thermal stability of $\text{Sr}_2\text{Ba}_2\text{Si}_2\text{O}_5\text{N}_2$:Eu\(^{2+}\). The thermal quenching temperature ($T_{0.5}$) is about 450°C, which is much higher than 288°C \(i.e., 560\mathrm{K})\ reported by Bachmann.\(^{34}\) This difference is ascribed to the variations in firing temperature, the higher temperature (1650°C vs 1500°C) used at this work results in a pure phase with less orthosilicate impurities.\(^{25}\) The activation energy for thermal quenching is calculated as 0.161 ± 0.005 eV for $\text{Sr}_{0.46}\text{Ba}_{0.54}\text{Si}_2\text{O}_5\text{N}_2$:Eu\(^{2+}\) ($x = 0.04$). Moreover, with increasing temperature the emission spectrum is blueshifted slightly, with the left wing enhanced. This is ascribed to the increased probability of radiative transitions from higher energy levels of the 5d electron orbitals of Eu\(^{2+}\). In addition, the thermal quenching temperature decreases a little with increasing the Eu\(^{2+}\) concentration.

The temperature dependent quantum efficiency of $\text{Sr}_{0.46}\text{Ba}_{0.54}\text{Si}_2\text{O}_5\text{N}_2$:Eu\(^{2+}\) was measured for the first time, as shown in
Fig. 12. Under the 450 nm excitation, both internal and external quantum efficiency slightly decreases with increasing temperature, reduced by 4.1 and 2.6% at 150°C respectively. The reduction is much less than that in the peak emission intensity (by 10% at 150°C). The small temperature-dependent luminescence and quantum efficiency imply that Sr$_{0.5}$Ba$_{0.5}$Si$_2$O$_5$N$_2$:Eu$^{2+}$ is a thermally stable yellow phosphor.

3.5. Luminescence Efficiency Enhanced by Post-Treatment

In most cases, phosphors are usually post-treated for achieving higher efficiency and tailoring the particle size as well as particle size distribution. The sample of $x = 0.04$ was selected for post-treatments such as a dilute HCl acid washing. It is clearly seen that the treated sample has coarsened and smoothed particles, of which the average particle size ($D_{50}$) is around 24.7 μm (vs 27.7 μm for the untreated sample) and the degree of dispersion ($S$) is 1.46 (vs 1.82) (Fig. 13). These enhancements are ascribed to the removal of tiny particles as well as residual impurities associated with the acid cleaning.

The acid cleaning also leads to the improvements in luminescence efficiency, as shown in Fig. 14. Upon the 450 nm excitation, the absorption, internal and external efficiencies of the treated sample are 83.2, 87.7 and 72.6%, increased by 6.4, 5.6 and 10% respectively. Less scattering and surface defects are considered to account for the enhanced efficiency of the treated phosphor. In addition, the post-treatment does not alter the thermal quenching behaviour of the sample. As shown in Table 1, except for the well-known YAG:Ce the Sr$_{0.5}$Ba$_{0.5}$Si$_2$O$_5$N$_2$:Eu phosphor exhibits higher luminescence efficiency and smaller thermal quenching compared to other promising yellow nitride or orthosilicate phosphors, enabling it to be an excellent colour converter for white LEDs with high luminous efficiency.

Table I Photoluminescence properties of several yellow phosphors for solid state lighting (E.Q.E and T.Q. is the external quantum efficiency and thermal quenching, respectively)

<table>
<thead>
<tr>
<th>Phosphors</th>
<th>$\lambda_{max}$ /nm</th>
<th>FWHM /nm</th>
<th>E.Q.E /%</th>
<th>T.Q. /%</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Y$_2$Al$<em>5$O$</em>{12}$:Ce$^{3+}$</td>
<td>555</td>
<td>122</td>
<td>81</td>
<td>87</td>
<td>5</td>
</tr>
<tr>
<td>La$_3$Si$_3$N$_9$:Ce$^{3+}$</td>
<td>580</td>
<td>120</td>
<td>60</td>
<td>84</td>
<td>24</td>
</tr>
<tr>
<td>Sr$_2$Ba$_2$(AlO$_3$)$_2$(SiO$_2$)$_5$:Ce$^{3+}$</td>
<td>539</td>
<td>125</td>
<td>63</td>
<td>82</td>
<td>15</td>
</tr>
<tr>
<td>CaAlSi$_3$N$_3$:Ce$^{3+}$</td>
<td>580</td>
<td>135</td>
<td>54</td>
<td>87</td>
<td>22</td>
</tr>
<tr>
<td>Sr$_2$Ba$_2$(SiO$_2$)$_3$:Eu$^{2+}$</td>
<td>552</td>
<td>92</td>
<td>63</td>
<td>85</td>
<td>9</td>
</tr>
<tr>
<td>Ca-$\alpha$-sialon:Eu$^{2+}$</td>
<td>585</td>
<td>94</td>
<td>59</td>
<td>90</td>
<td>5</td>
</tr>
<tr>
<td>Sr$<em>{0.5}$Ba$</em>{0.5}$Si$_2$O$_5$N$_2$:Eu$^{2+}$</td>
<td>560</td>
<td>93</td>
<td>72</td>
<td>90</td>
<td>This work</td>
</tr>
</tbody>
</table>
3.6. Applications in White LEDs

The suitability of the Sr$_{0.5}$Ba$_{0.5}$Si$_2$O$_2$N$_2$:Eu yellow phosphor in white LEDs was also testified in this work. White LED lamps were fabricated by mounting a blend mixture of about 0.1500 g phosphor powders and 2.3916 g silicone resin on a blue LED chip ($\lambda_{em} = 450$ nm). Figure 15 shows the electroluminescence spectrum of a white LED driven at a 20 mA current. The lamp has a correlated color temperature (CCT) of 5970 K, color rendering index (Ra) of 61, and CIE chromatic coordinates (x, y) of (0.323, 0.328). The luminous efficiency of the white LED is as high as 117 lm/W. These results demonstrate that Sr$_{0.5}$Ba$_{0.5}$Si$_2$O$_2$N$_2$:Eu has strong potential applications in developing highly efficient, thermally stable cool white LEDs.

![Electroluminescence spectrum of white LEDs](image)

**Figure 15** Electroluminescence spectrum of white LEDs prepared by combining Sr$_{0.5}$Eu$_{0.5}$Ba$_{0.5}$Si$_2$O$_2$N$_2$ ($x = 0.04$) with a blue LED chip, measured under a 20 mA drive current at room temperature.

4. CONCLUSIONS

Knowing of the detailed physiochemical properties of a material is of great importance for properties optimization and practical applications. In this work, a thorough investigation has been carried out to elucidate photoluminescence properties and the temperature-dependent luminescence efficiency of a yellow-emitting Sr$_{0.5}$Ba$_{0.5}$Si$_2$O$_2$N$_2$:Eu$^{2+}$ phosphor for solid state lighting. This oxonitridosilicate phosphor shows high absorption and quantum efficiency of 83 and 88% upon the 450 nm excitation, respectively. It also possesses excellent thermal stability, with the thermal quenching temperature as high as 450°C. The wavenumber observed by the CL mapping does not affect the photoluminescence properties, although it remains unclarified how it forms. White light LEDs with the luminous efficacy of ~120 lm/W and the colour temperature of ~6,000 K can be fabricated by combining this yellow phosphor with a blue LED chip ($\lambda_{em} = 450$ nm), indicative of a promising robust yellow colour converter for high efficiency and high reliability solid state lighting.

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Notes and references