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Structures and photoluminescence of another crown-typed polyoxotungstoeuropate $[H_2O\subset\{Eu(H_2O)_4\}_2\{Eu(H_2O)_2(AsW_9O_{33})\}_4]^{x-}$ in the presence of Na^+ or Ca^{2+}

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Abstract

Two novel polyoxotungstoeuropates $[H_2O\subset\{Eu_nNa_{1-n}(H_2O)_2\}_2\{Eu(H_2O)_2(AsW_9O_{33})\}_4]^{21-}$ ($n = 1/4$) **1a** and $[H_2O\subset\{Eu_nCa_{1-n}(H_2O)_4\}_2\{Eu_nCa_{1-n}(H_2O)_2(AsW_9O_{33})\}_4]^{20-}$ ($n = 2/3$) **2a** were formed by the reaction of α-B-$[AsW_9O_{33}]^{9-}$ and Eu^{3+} with Na^+ or Ca^{2+} as the counter cation. $Na_{18}H_3[H_2O\subset\{Eu_nNa_{1-n}(H_2O)_2\}_2\{Eu(H_2O)_2(AsW_9O_{33})\}_4]\cdot 48H_2O$ ($n = 1/4$) **1** crystallized in the tetragonal space group $P4/ncc$, with $a = 28.82(2)$ Å, $c = 26.45(2)$ Å, $V = 21974(23)$ Å^3 and $Z = 4$, while $Ca_9Na_2[H_2O\subset\{Eu_nCa_{1-n}(H_2O)_4\}_2\{Eu_nCa_{1-n}(H_2O)_2(AsW_9O_{33})\}_4]\cdot 63H_2O$ ($n = 2/3$) **2** crystallized in the triclinic space group $P\bar{1}$, with $a = 14.27(1)$ Å, $b = 20.26(2)$ Å, $c = 20.54(2)$ Å, $\alpha = 81.69(3)^\circ$, $\beta = 86.10(3)^\circ$, $\gamma = 72.93(3)^\circ$, $V = 5613(7)$ Å^3 and $Z = 1$. These anion structures consisted of tetramer of L-shaped units $[Eu(H_2O)_2(AsW_9O_{33})]^{6-}$ with water molecules encapsulated in the center. Interestingly, these tetramers also contained an apical attachment of two square-antiprismatic Eu^{3+} centers each of which is coordinated by one bridging O atom for each $[AsW_9O_{33}]^{9-}$ ligand and four aqua molecules. These apical Eu atoms had different coordination environments from Eu atom in L-shaped unit, and were disordered together with Na atoms with a quarter occupancy of Eu for **1**, while all the Eu atoms together with Ca atoms with a 2/3 occupancy of Eu for **2**. The photoluminescence spectra, excitation spectra and lifetimes for these two complexes were discussed based on crystal structures. Notably, for **1** the emission from two different sites of Eu^{3+} was isolated in photoluminescence spectra under two different $^7F_0 \rightarrow {}^5D_0$ excitation wavelengths at 579.8 and 580.5 nm.

Keywords: Polyoxotungstate; Nanostructures; Crystal structure; Photoluminescence; Europium

1. Introduction

Some polyoxometallolanthanoates, which can be regarded as lanthanide complexes coordinated by polyoxometalate ligands, attract considerable attention because of their properties originating from the lanthanides and have been intensively studied [1–5]. Much attention has also been focused on their synthetic and structural chemistry, since lanthanide cations play an important role of linking several polyoxometalate units in order to form new classes of material with a large anion cluster such as $[\{Pr(H_2O)_5\}_6Mo_{120}O_{366}H_{12}(H_2O)_{48}]^{6-}$ [6], $[Ce_{16}(H_2O)_{36}(WO_2)_4(W_2O_6)_8(W_5O_{18})_4(\alpha\text{-}AsW_9O_{33})_{12}]^{76-}$ [7]. We have also reported several crystal structures of luminescent polyoxometallolanthanoates for an insight into the photochemistry of lanthanide cations in molecular level [8–15].

Recently, we have reported the crown-shaped ring cluster polyoxotungstatoeuropates, $[K\subset\{Eu(H_2O)_2(AsW_9O_{33})\}_6]^{35-}$ and $[Cs\subset\{Eu(H_2O)_2(AsW_9O_{33})\}_4]^{23-}$ by the self-assembly of the L-shaped $[Eu(H_2O)_2(AsW_9O_{33})]^{6-}$ building blocks, which is composed of Eu^{3+} and α-B-$[AsW_9O_{33}]^{9-}$, in the presence of alkali-metal cations [16]. These anion encapsulate a single alkali-metal cation (K^+ and Cs^+) in the center, to form cyclic hexamer and tetramer composed of the L-shaped $[Eu(H_2O)_2(AsW_9O_{33})]^{6-}$ in about 2.6

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and 2.2 nm diameter, respectively. These crown-shaped ring anions structures result from the polymerization governed by alkali-metal cation.

We are interested in this system and attempt to prepare this type polyoxometallolanthanoates. This paper describes novel $[H_2O\subset\{Eu(H_2O)_4\}_2\{Eu(H_2O)_2(AsW_9O_{33})\}_4]^{x-}$ polyoxotungstoeuropates formed by the reactions of L-shaped $[Eu(H_2O)_2(AsW_9O_{33})]^{6-}$ with different counter cations, Na^+ and Ca^{2+}. The photoluminescence property of $Na_{18}H_3[H_2O\subset\{Eu_nNa_{1-n}(H_2O)_2\}_2\{Eu(H_2O)_2(AsW_9O_{33})\}_4]\cdot 48H_2O$ ($n = 1/4$) **1** and $Ca_9Na_2[H_2O\subset\{Eu_nCa_{1-n}(H_2O)_4\}_2\{Eu_nCa_{1-n}(H_2O)_2(AsW_9O_{33})\}_4]\cdot 63H_2O$ ($n = 2/3$) **2** were investigated and compared with the results of crystallographic structure analysis.

2. Experimental

2.1. Synthesis

$Na_9[AsW_9O_{33}]\cdot 19.5H_2O$ was synthesized according to published procedures [17], and characterized by infrared spectroscopy.

$Na_{18}H_3[H_2O\subset\{Eu_nNa_{1-n}(H_2O)_2\}_2\{Eu(H_2O)_2(AsW_9O_{33})\}_4]\cdot 48H_2O$ ($n = 1/4$) **1**. A solution of $Eu(NO)_3\cdot 6H_2O$ (0.46 g, 1.0 mmol) in water (10 mL) was added to a solution of $Na_9[AsW_9O_{33}]\cdot 19.5H_2O$ (2.8 g, 1.0 mmol) in water (30 mL). The solution was warmed to approximately 80 °C. A solid of NaCl (5.0 g, 85.5 mmol) was added to the hot solution and the resultant solution was left to stand at room temperature. After a few days, the colorless crystals of **1** were obtained as Na salts. Yield: 1.50 g. Elemental analysis (%) calculated: Na 3.98, Eu 6.07, W 58.76, As 2.66; found: Na 3.94, Eu 5.63, W 57.50, As 2.66.

$Ca_9Na_2[H_2O\subset\{Eu_nCa_{1-n}(H_2O)_4\}_2\{Eu_nCa_{1-n}(H_2O)_2(AsW_9O_{33})\}_4]\cdot 63H_2O$ ($n = 2/3$) **2**. A solution of $Eu(NO)_3\cdot 6H_2O$ (0.23 g, 0.5 mmol) in water (10 mL) was added to a solution of $Na_9[AsW_9O_{33}]\cdot 19.5H_2O$ (1.4 g, 0.5 mmol) in water (50 mL). The solution was warmed to approximately 80 °C. As soon as a solution of $CaCl_2\cdot 2H_2O$ (0.20 g, 1.4 mmol) in water (10 mL) was added to the hot solution, the resultant solution was left to stand at room temperature. In a few hours, the colorless crystals of **2** were obtained as Na/Ca-mixed salts. Yield: 1.02 g. Elemental analysis (%) calculated: Na 0.40, Ca 3.81, Eu 5.26, W 57.22, As 2.59; found: Na 0.38, Ca 3.91, Eu 4.85, W 56.71, As 2.53.

2.2. X-ray structure determination

All single crystals were mounted with Paraton-N oil in a fiber loop. Intensity data of **1** and **2** were collected on a Rigaku RAXIS-RAPID imaging plate diffractometer with Mo Kα graphite monochromatized radiation ($\lambda = 0.71069$ Å) at −100 °C. Both structures were solved by direct methods using SHLEXS-97 and refined by a full-matrix least-squares refinement on F^2 using the CrystalStructure software package [18]. Lorentz polarization effect and numerical absorption corrections [19,20] were applied to the intensity data, and H atoms were not indicated in the calculation. The crystal data and details for the refinement are listed in Table 1. The W, As, Eu and Ca atoms (except for disordered atoms) were refined anisotropically, while the rest were refined isotropically. The residual maximum and minimum Fourier peaks for **1** were 5.42 and −2.00 eÅ^{-3}, and for **2** 2.44 and −2.39 eÅ^{-3}, respectively. Since crystal structures for **1** and **2** show disorder in the range of water molecules and Na^+ cations, their occupancy of 1/2 determined based on refinement of their temperature factors. In addition, two water O atoms O37 and O38 coordinating to Eu2 and Eu3 in **1** were the occupancy of 1/2 because of their large thermal displacement parameters. For **1** Eu2 and Na1, and Eu3 and Na2 atoms, which occupancies were fixed at 1/4 and 3/4 were refined as lying on same site, respectively. Similarly, for **2** Eu1 and Ca1, Eu2 and Ca2, and Eu3 and Ca3 atoms which occupancies were fixed at 2/3 and 1/3 were refined as lying on same site, respectively. The occupancy of Ca4 was also fixed at 1/2 due to the short distance (3.42(4) Å) between symmetric atoms.

2.3. Photoluminescence spectra

Luminescence and excitation spectra were obtained by using powder pellet of the sample. The light source for the photoluminescence measurements was a Continuum 90300

Table 1
Crystal data, structure determination and refinement data for **1** and **2**

	1	**2**
Formula	$Na_{19.5}Eu_{4.5}W_{36}As_4O_{193}H_{125}$	$Ca_{11}Na_2Eu_4W_{36}As_4O_{212}H_{160}$
Formula weight	11264.28	11566.12
Crystal system	Tetragonal	Triclinic
Space group	$P4/ncc$	$P\bar{1}$
a (Å)	28.82(2)	14.27(1)
b (Å)	26.45(2)	20.26(2)
c (Å)		20.54(2)
α (°)		81.69(3)
β (°)		86.10(3)
γ (°)		72.93(3)
V (Å^3)	21974(23)	5613(7)
Z	4	1
D_c (g cm^{-3})	3.405	3.421
F(0 0 0)	19852	5146
μ (cm^{-1})	207.78	204.26
$2\theta_{max}$ (°)	55.0	55.4
Crystal dimensions (mm)	0.10 × 0.10 × 0.10	0.10 × 0.05 × 0.03
Temperiture (°C)	−100.0	−100.0
No. of data measure	219463	56744
R (int)	0.048	0.105
No. of unique data	13741	25001
No. of observed data	4682[$I > 2.0\sigma(I)$]	8323[$I > 3.0\sigma(I)$]
No. of variables	368	668
GOF	1.007	1.000
R1[a]	0.050	0.043
wR2[b]	0.103	0.081
Maximum shift/error	0.000	0.000

[a] $R1 = \sum||Fo|-|Fc||/\sum|Fo|$.

[b] $wR2 = \{\sum w(Fo^2-Fc^2)^2/\sum(wFo^2)^2\}^{1/2}$.

YAG (355 nm, 400 mJ/pulse) laser. The luminescence was collected at an angle of 90° to exciting light and focused onto the entrance slit of a Spex 750 M spectrometer, which was equipped with Hamamatsu Photonix R636 photomultiplier tube. Luminescence at low temperatures was measured using an Oxford Instruments CF 204 cryostat. The time profiles of the luminescence were measured on LeCloy 9361 digital storage oscilloscope.

3. Results and discussion

3.1. Anion structures

The two polyoxotungstoeuropates, **1a** and **2a** were isostructural. These anion structures were a tetramer of $[Eu(H_2O)_2(AsW_9O_{33})]^{6-}$ L-shaped units similar to the Cs^+-ion-encapsulated $[Cs\subset\{Eu(H_2O)_2(AsW_9O_{33})\}_4]^{23-}$ anion [16]. In **1a** and **2a**, the central Cs^+ cation in $[Cs\subset\{Eu(H_2O)_2(AsW_9O_{33})\}_4]^{23-}$ is replaced by water molecule, and they have an apical attachment of two square-antiprismatic Eu^{3+} centers (Fig. 1), each of which is coordinated by one terminal O atom for each $[AsW_9O_{33}]^{9-}$ ligand and four aqua molecules. As for **1a**, the O atoms of these four water molecules are disordered with half occupancy each, therefore these four positions are occupied by only two water molecules.

Both the apical Eu atoms are disordered together with Na atoms with a quarter occupancy of Eu for **1**, and all the Eu atoms together with Ca atoms with a 2/3 occupancy of Eu for **2** (these disordered Na and Ca atoms can not occupy equal

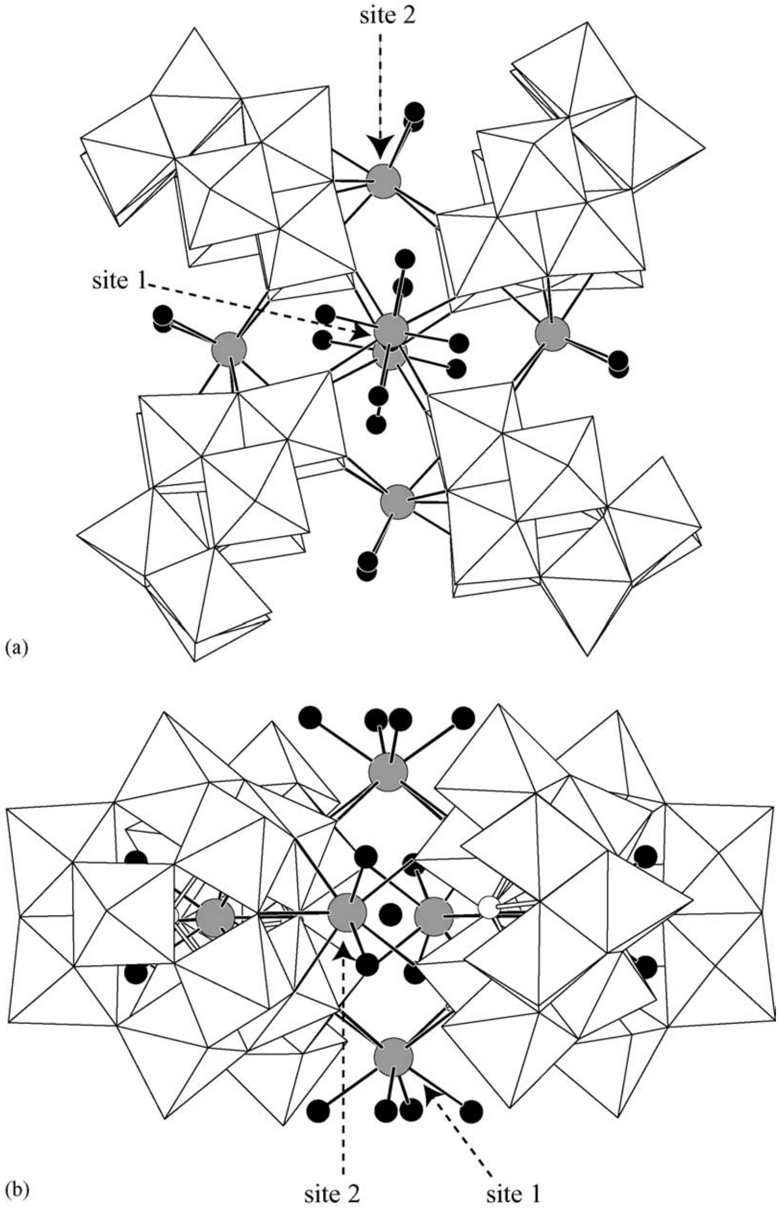

Fig. 1. Structure of **1** and **2** viewed from the top (a) and from the side (b) (octahedron: WO_6; gray: Eu; black: H_2O; open circle: As).

site with Eu atoms, however, these atoms in these molecular structures were refined as same position with fixed at these occupancies, due to be undecided positions by the X-ray structure analysis).

The separations of the two apical Eu atoms (site 1) in each anion are 7.3(6) and 7.370(2) Å on the four-fold axis of these anions involving O atom of central water molecules. The equatorial Eu atom (site 2) linking between α-B-$[AsW_9O_{33}]^{9-}$ units coordinates by two terminal and four bridging oxygen atoms which belong to neighboring unit and the coordination by additional two aqua molecules results in distorted square antiprismatic ligand field. The distances between adjacent Eu atoms in site 2 for **1a** is 6.660(2) Å and **2a** is 6.623(2) and 6.639(2) Å, and the distances between them and Eu atoms of site 1 in **1a** range from 5.920(2) to 5.986(2) Å and **2a** range from 5.942(2) to 5.983(2) Å. In **1a**, the distances from the central water O atom to each Eu atoms for site 1 are 3.44(5) and 3.84(5) Å and for site 2 are 4.765(1) and 4.712(2) Å, and in **2a** are 3.685(1) Å for site 1 and 4.675(1) and 4.702(1) Å for site 2. The arrangement of these Eu^{3+} ions in **1a** and **2a** give an octahedral conformation, respectively.

Some counter cations observed in these compounds are disordered. However, the counter cations (Na3 in **1** and Ca7 and Ca8 in **2**), which is coordinated by water O atoms for $[Eu(H_2O)_2]^{3+}$ in site 2, are not observed for the disorder (the K atom with 5/6 occupancy was observed in $Cs_{22}K_{13}[K\subset\{Eu(H_2O)_2(AsW_9O_{33})\}_6]\cdot 44H_2O$ [16]).

3.2. Photoluminescence property

The photoluminescence spectra for **1** and **2** at 4.2 K were measured under various excitation wavelengths and the excitation spectra at 4.2 K were also obtained by monitoring Eu^{3+} $^5D_0 \rightarrow {}^7F_1$ or $^5D_0 \rightarrow {}^7F_2$ emissions at various wavelengths. Fig. 2 shows the $^7F_0 \rightarrow {}^5D_0$ excitation spectra of these complexes observed at 4.2 K. In **1** and **2**, the $^7F_0 \rightarrow {}^5D_0$ excitation transitions occur at about 579.8 and 580.4 nm and 579.7 and 580.1 nm, respectively. For the different $^5D_0 \rightarrow {}^7F_2$ emissions (617.6 and 614.7 nm) of **1**, the variation of relative intensity of each $^7F_0 \rightarrow {}^5D_0$ peak is shown obviously. While the two $^7F_0 \rightarrow {}^5D_0$ peaks of **2** cannot be well separated. The existence of some peaks of the $^7F_0 \rightarrow {}^5D_0$ transition

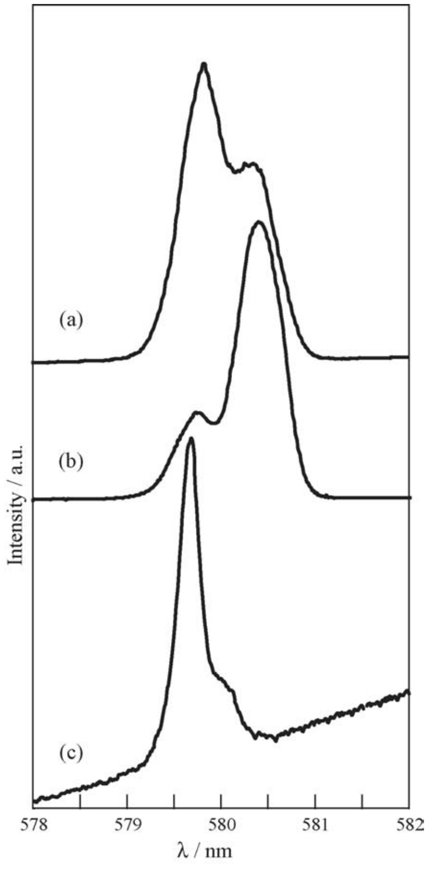

Fig. 2. $^5D_0 \rightarrow {}^7F_0$ excitation spectra of **1** and **2** solids at 4.2 K. The spectra were obtained by monitoring $^5D_0 \rightarrow {}^7F_2$ at the 617.6 (a) and 614.7 nm (b) for **1**, and 589.2 nm (c) for **2**.

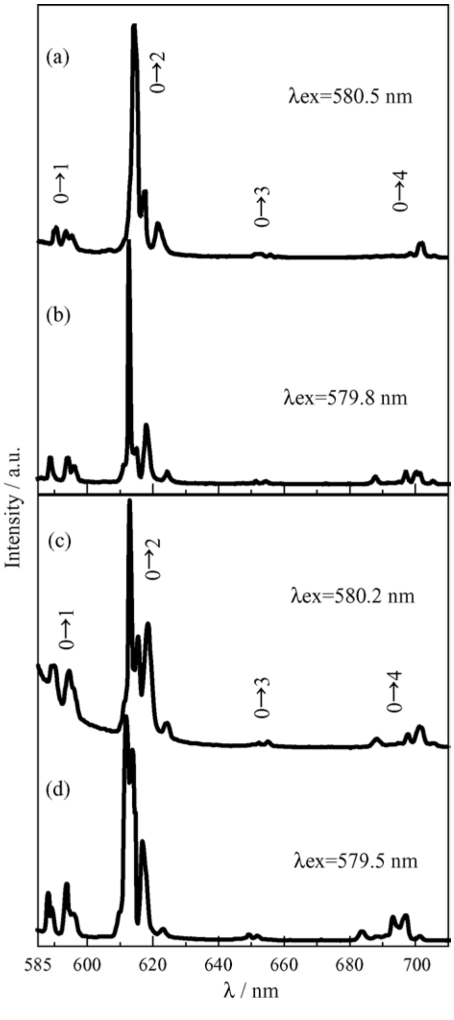

Fig. 3. Low-resolution emission spectra of **1** and **2** solids at 4.2 K. The spectra were obtained by excitation into the $^7F_0 \rightarrow {}^5D_0$ at 580.5 (a) and 579.8 nm (b) for **1**, and 580.2 nm (c) and 579.5 nm (d) for **2**. Numbers indicate $J \rightarrow J'$ for the $^5D_J \rightarrow {}^7F_{J'}$ transitions.

is taken as evidence that at least two different Eu^{3+} sites are responsible for the series of luminescence transition, since the $^7F_0 \rightarrow {}^5D_0$ transition at about 580 nm cannot be split by any crystal field. These results suggest the presence of two Eu^{3+} sites, which are sites1 and 2 in anion structures, as determined by single-crystal X-ray structure analyses.

Fig. 3 shows the low-resolution emission spectra of **1** and **2** at 4.2 K under excitation by each $^7F_0 \rightarrow {}^5D_0$ transitions (579.8 and 580.5 nm for **1**, and 579.5 and 580.2 nm for **2**). The $^5D_0 \rightarrow {}^7F_{1\text{–}4}$ lines are visible, and clearly exhibit different patterns between each excitation wavelength, respectively. These high-resolution emission spectra $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ band system for **1** and **2** are shown in Figs. 4 and 5. In **1**, the emission spectra by the excitation at 579.8 nm exhibits the $^5D_0 \rightarrow {}^7F_1$ emission lines at about 588.2, 593.9 and 596.0 nm, and the excitation at 580.5 nm exhibits at about 590.6, 593.9 and 595.5 nm.

These each three lines exhibit the pattern depended on excitation wavelengths, respectively. Similarly, $^5D_0 \rightarrow {}^7F_2$ emission lines show each five lines at about 611.0, 612.5, 614.3, 617.8 and 624.4 nm by the excitation at 579.8 nm, and about 612.0, 614.0, 614.8, 617.5 and 622.0 nm by the excitation at 580.5 nm. Emission spectra for the two sites were isolated with selective $^7F_0 \rightarrow {}^5D_0$ excitation at 579.8 and 580.5 nm. In the case of **2**, the $^5D_0 \rightarrow {}^7F_1$ emission lines on the 579.5- and 580.2-nm light excitation consists of four lines at about 587.8, 589.1, 593.8 and 596.8 nm for $\lambda = 579.5$ nm, and 589.2, 590.0, 594.0 and 595.5 nm for $\lambda = 580.2$ nm. Similarly, $^5D_0 \rightarrow {}^7F_2$ emission lines show five lines at about 610.4, 612.5, 614.4, 618.0 and 624.4 nm on the $\lambda = 579.5$ nm light excitation, and about 611.4, 613.1, 615.5, 618.5 and 624.3 nm on the $\lambda = 580.2$ nm light excitation.

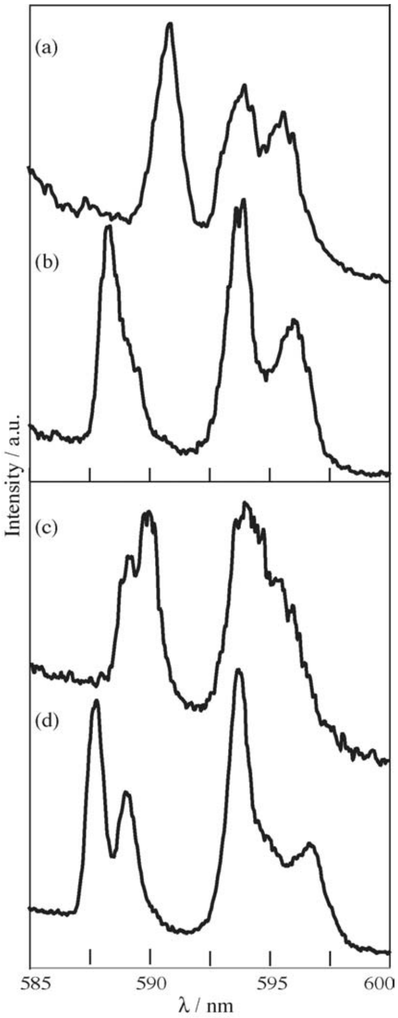

Fig. 4. High-resolution $^5D_0 \rightarrow {}^7F_1$ emission spectra of **1** and **2** solids at 4.2 K. The spectra were obtained by excitation at 580.5 (a) and 579.8 nm (b) for **1**, and 580.2 (c) and 579.5 nm (d) for **2**.

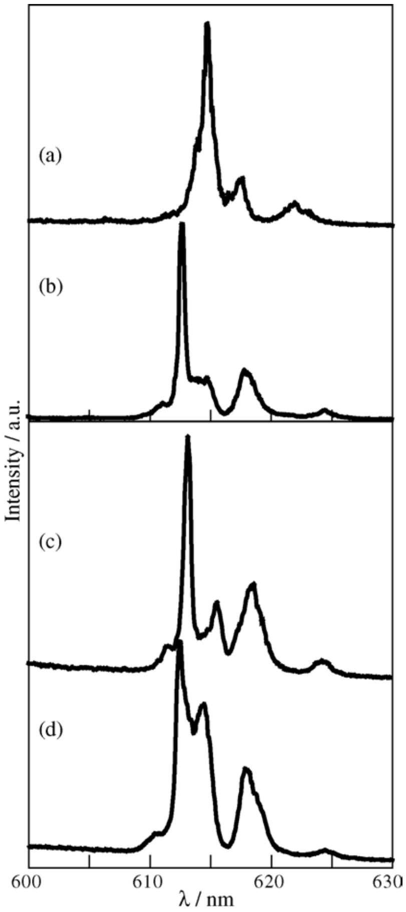

Fig. 5. High-resolution $^5D_0 \rightarrow {}^7F_2$ emission spectra of **1** and **2** solids at 4.2 K. The spectra were obtained by excitation at 580.5 (a) and 579.8 nm (b) for **1**, and 580.2 (c) and 579.5 nm (d) for **2**.

For further characterization of two sites, the lifetime of the corresponding $^5D_0$-states were measured at 4.2 K. The decay time of **1** monitored around $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ lines under 579.8 or 580.5 nm were single exponential which indicate the presence of two different decay time, and their luminescence lifetimes were 0.41 ± 0.03 ms at 579.8 nm and 0.50 ± 0.03 ms at 580.5 nm, respectively. For the $^5D_0$ lifetime method for the determination of the number of bonded water molecules [21], both values correspond to the number of two water ligands coordinating to Eu^{3+}, which is in good agreement with the result of single-crystal X-ray structure analysis. This lifetime value 0.41 ± 0.03 ms under 579.8 nm is also corresponding to the value of $^5D_0$ lifetime measured in $Cs_{16.75}Na_{6.25}[Cs\subset\{Eu(H_2O)_2(AsW_9O_{33})\}_4]\cdot 24.5H_2O$, which consists of only the Eu^{3+} linking between $[AsW_9O_{33}]^{9-}$ ligands [22]. Thus, it is concluded that the emission under the excitation at 579.8 nm is associated with Eu^{3+} of site 2, and another may be associated with Eu^{3+} of site 1. For **2**, the decay pattern of $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ under 579.5 and 580.2 nm were not single exponential, and their luminescence lifetimes for each lines were observed in the wide range of approximately 0.20–0.40 ms. It is suggested that the overlapped emission of several sites was exhibited under the both excitation wavelengths, and it was difficult to distinguish the emission sites only by the using $^7F_0 \rightarrow {}^5D_0$ excitation wavelengths.

Furthermore, the disordering of Eu^{3+} with Ca^{2+} atoms resulted in additional sites consisting of similar coordination environments in the anion, thereby leads to a difficulty in high resolution of each emission pattern.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jallcom.2004.11.086.

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