

## **Electronic Supplementary Information**

### **Tunable Color Generation of Transparent Composite Films Reinforced with Luminescent Nanofillers**

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## Experimental details

### 1. Preparation of *c*-LGdH:RE (RE = Eu and Tb).

Aqueous suspensions of  $\text{Gd}_{2.00-x}\text{RE}_x(\text{OH})_5\text{Cl}\cdot n\text{H}_2\text{O}$  (RE = Eu and Tb for LGdH:Eu and LGdH:Tb, respectively) were prepared at room temperature. The concentration of activator was fixed to 10 % ( $x = 0.20$ ) for both LGdH:Eu and LGdH:Tb. Typically, stoichiometric amounts of  $\text{GdCl}_3\cdot 6\text{H}_2\text{O}$ ,  $\text{EuCl}_3\cdot 6\text{H}_2\text{O}$ , and  $\text{TbCl}_3\cdot 6\text{H}_2\text{O}$  were dissolved in water to prepare aqueous solutions (5.0 mM, 100 mL). After a clear solution was formed by uniform stirring, an aqueous KOH solution (10.0 mM, 100 mL) was added drop-wise. The resulting mixture was stirred for 1 h at room temperature, and the product was centrifuged and washed with water several times. The recovered slurry was dispersed in water ( $\sim 20 \text{ g L}^{-1}$ ) and dried at 50 °C under > 50 % humidity. Then, the dried powder was heated at 600 °C for 1 h under a 4 %  $\text{H}_2$  + 96 % Ar mixed gas flowing to produce calcined layered nanofillers (*c*-LGdH:RE). The solid product was dispersed in water by sonication (*caution; the increase in temperature can induce a reconstruction of the layered hydroxide structure*), and the precipitate was separated from the aqueous solution after 1 h. A slurry of *c*-LGdH:RE was obtained by centrifugation of the supernatant suspension thereafter. The last procedure was repeated with the precipitate for the maximum recovery of the *c*-LGdH:RE nanosheet slurry. To measure the X-ray diffraction patterns and SEM and AFM images, the slurry was dried at 40 °C for a day. Dopant (Eu and Tb) content and chemical compositions of the obtained products were confirmed using ICP and TG analyses. Experimental values were in agreement with the nominal compositions within experimental errors, i.e.,  $\text{Gd}_{1.80}\text{Eu}_{0.20}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$  (LGdH:Eu) and  $\text{Gd}_{1.80}\text{Tb}_{0.20}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$  (LGdH:Tb).

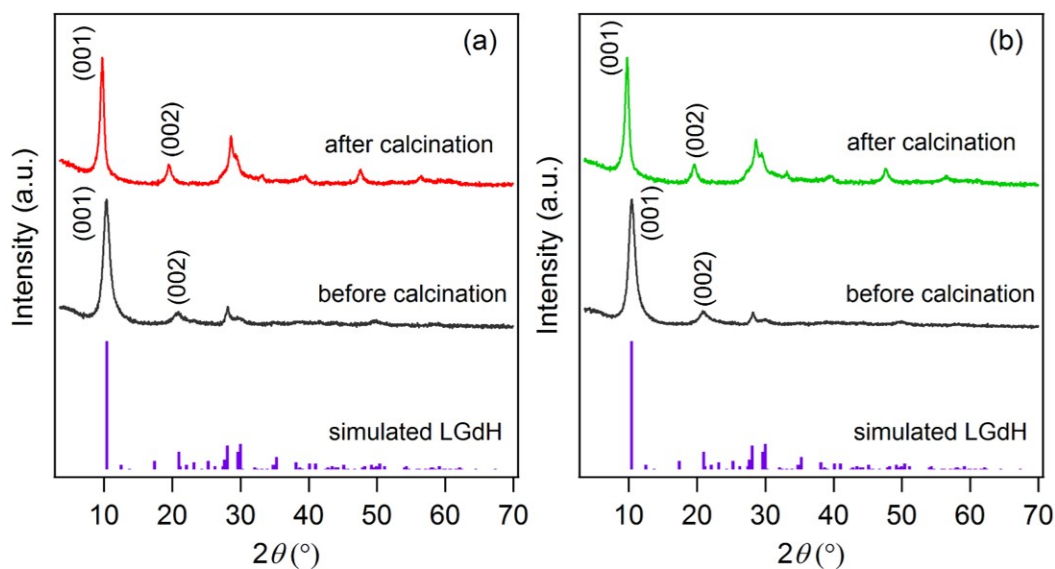
## 2. Fabrication of c-LGdH:RE/PVA Composite Films.

Neat PVA and composite c-LGdH:RE/PVA (RE = Eu and Tb) films were prepared by the solution casting method. PVA ( $M_w = 85,000\text{--}124,000$ , 99+% hydrolyzed, 2.0 wt%) was dissolved in water under stirring at 70–80 °C. Aqueous colloidal solutions containing c-LGdH:Eu and c-LGdH:Tb nanosheets ( $1.0\text{ gL}^{-1}$ ) were prepared by the ultrasonic treatment of c-LGdH:RE slurries in water (*caution; extensive ultrasonic treatment recovers the structure of LGdH:RE and reduces the photoemission intensity*). Appropriate amounts of c-LGdH:RE colloidal solution (c-LGdH:RE/PVA ratios = 0.5, 1.0, 2.0, 4.0 wt%) were taken and diluted to 15.0 mL with water. A PVA solution (2.0 wt%, 15.0 mL) was then added to these colloidal solutions. Resulting aqueous mixtures were stirred for 30 min and finally sonicated to remove air bubbles. The temperature of the solution was maintained near room temperature during this step. Composite films were prepared by pouring these mixtures on a Petri dish (8 cm in diameter) and allowing the water to evaporate at 50 °C. After drying, the film was readily removed from the plate.

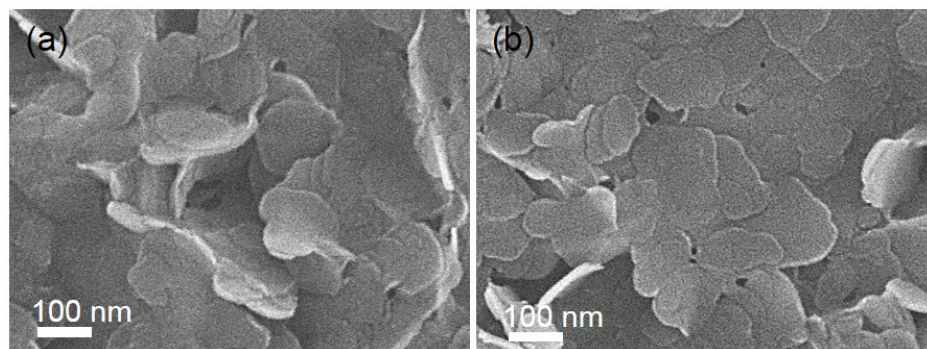
## 3. Characterization.

Powder X-ray diffraction (XRD) patterns were recorded with a Bruker D8 Advance diffractometer. Inductively coupled plasma (ICP) and thermogravimetric (TG) analyses were performed using Thermo Elemental Thermo ICAP 6000 and Seiko Instruments TG/DTA320, respectively. A TG curve was obtained in air at a heating rate of 5 °C/min. Field emission scanning electron microscopy (FE-SEM) was carried out with a Carl Zeiss LEO SUPRA 55 electron microscope operating at 30 kV. Specimens for the FE-SEM were coated with Pt-Rh for 180 s under vacuum. Atomic force microscopy (AFM) was carried

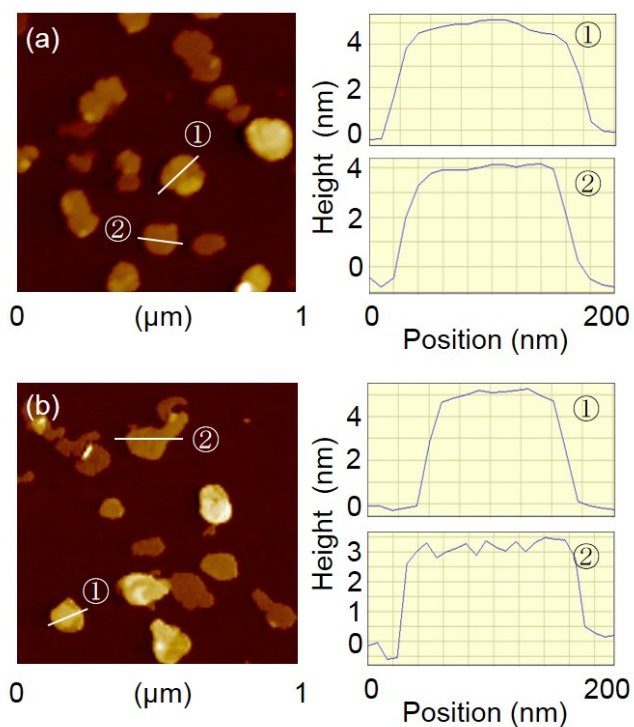
out using a Pucostation STD (Pucostation). Ultrasonicated Si substrates were dipped into the suspension in aqueous solutions ( $0.1 \text{ gL}^{-1}$ ) for 10 min and then washed with water. Optical transmittance of films was measured with UV–Vis spectrophotometers, Shimadzu Multispec-1501 and a LAMBDA 35. The mechanical properties of films were measured using a UTM (Universal Testing Machine) Model 5844 (Instron Co., USA). The testing film was cut into a ribbon ( $5 \text{ mm} \times 50 \text{ mm}$ ) with an average thickness of  $65 \pm 8 \mu\text{m}$ . The initial distance between instrument grips was kept at 10 mm, and samples were tested at a cross-head speed of  $5 \text{ mm min}^{-1}$ . Using the obtained stress-strain curves, Young's modulus and ultimate tensile strength were calculated. Photoluminescence spectra were measured at room temperature using a FP-6600 spectrophotometer (JASCO) with a Xenon flash lamp.



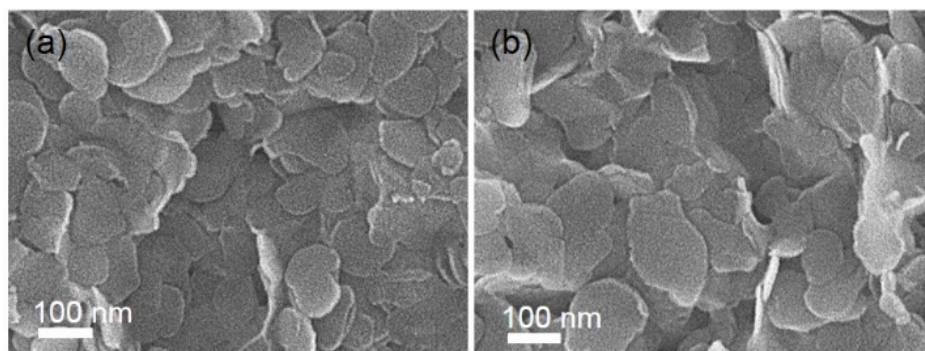
**Fig. S1** Simulated and observed powder X-ray diffraction patterns before and after calcining (a)  $\text{Gd}_{1.98}\text{Eu}_{0.02}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$  (LGdH:Eu) and (b)  $\text{Gd}_{1.98}\text{Tb}_{0.02}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$  (LGdH:Tb).



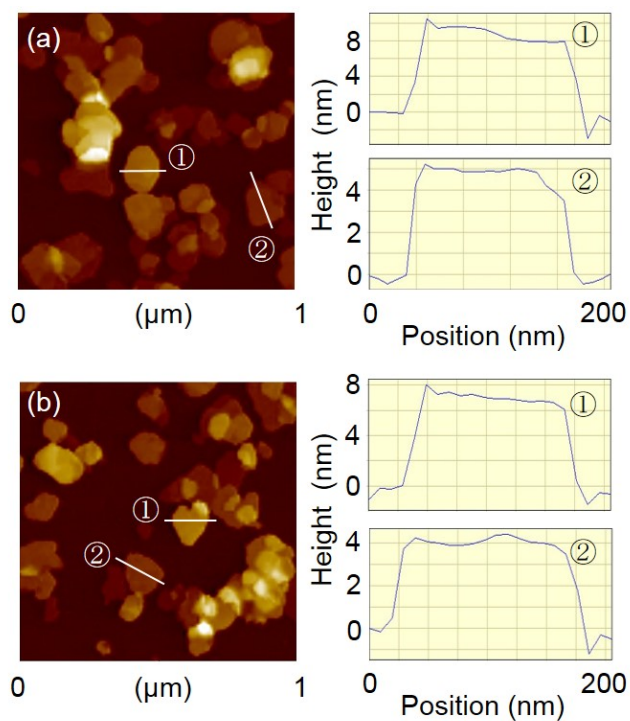
**Fig. S2** SEM images of as-prepared (a) Gd<sub>1.98</sub>Eu<sub>0.02</sub>(OH)<sub>5</sub>Cl·1.5H<sub>2</sub>O (LGdH:Eu) and (b) Gd<sub>1.98</sub>Tb<sub>0.02</sub>(OH)<sub>5</sub>Cl·1.5H<sub>2</sub>O (LGdH:Tb).



**Fig. S3** AFM images and height profiles of as-prepared (a) Gd<sub>1.98</sub>Eu<sub>0.02</sub>(OH)<sub>5</sub>Cl·1.5H<sub>2</sub>O (LGdH:Eu) and (b) Gd<sub>1.98</sub>Tb<sub>0.02</sub>(OH)<sub>5</sub>Cl·1.5H<sub>2</sub>O (LGdH:Tb) deposited on the Si wafer.



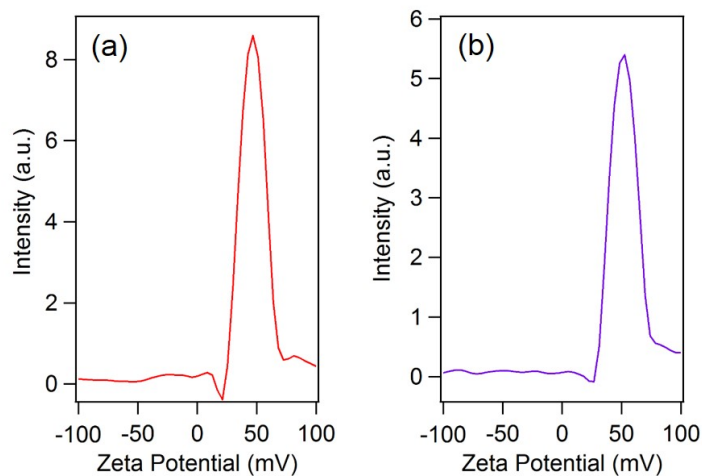
**Fig. S4** SEM images of (a) c-LGdH:Eu and (b) c-LGdH:Tb obtained after calcining  $\text{Gd}_{1.98}\text{Eu}_{0.02}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$  and  $\text{Gd}_{1.98}\text{Tb}_{0.02}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$ , respectively, at 600 °C for 1 h.



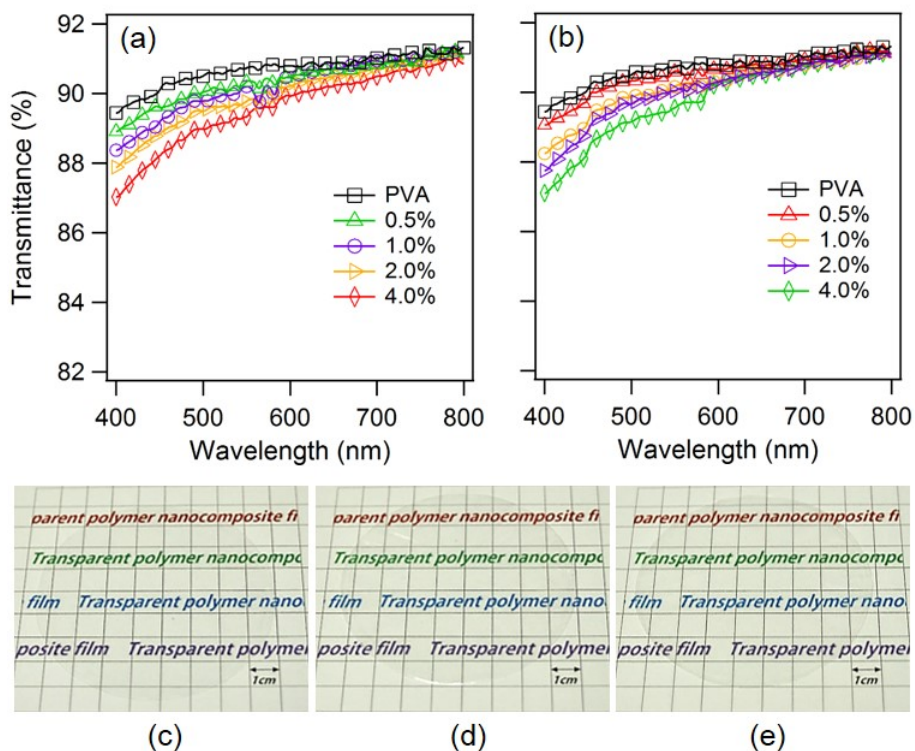
**Fig. S5** AFM images and height profiles of (a) c-LGdH:Eu and (b) c-LGdH:Tb obtained after calcining  $\text{Gd}_{1.98}\text{Eu}_{0.02}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$  and  $\text{Gd}_{1.98}\text{Tb}_{0.02}(\text{OH})_5\text{Cl}\cdot 1.5\text{H}_2\text{O}$ , respectively, at 600 °C for 1 h. Nanosheets were randomly chosen from a deposit of the aqueous colloid on a Si wafer.

**Table S1** Mechanical properties obtained from stress-strain responses for the composite films prepared by dispersing c-LGdH:Eu nanosheets with different ratios in PVA.

	PVA	0.5 %	1.0 %	2.0 %	4.0 %
Young's Modulus (GPa)	$0.53 \pm 0.07$	$1.42 \pm 0.08$	$2.39 \pm 0.26$	$2.20 \pm 0.12$	$1.63 \pm 0.14$
Yield Strength (MPa)	$9.14 \pm 1.85$	$14.81 \pm 1.08$	$16.24 \pm 2.13$	$27.54 \pm 1.92$	$17.61 \pm 2.42$
Tensile Strength (MPa)	$42.58 \pm 2.13$	$49.52 \pm 2.06$	$51.28 \pm 1.17$	$56.06 \pm 1.42$	$53.03 \pm 0.82$

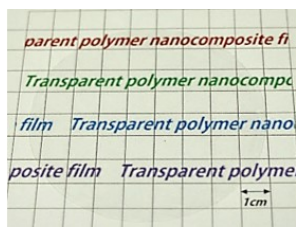


**Fig. S6** Zeta potentials of (a) c-LGdH:Eu (46.8 mV) and (b) LGdH:Eu (51.9 mV).

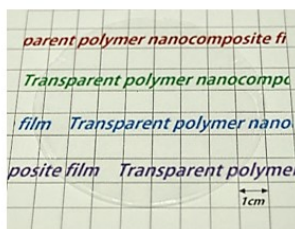


**Fig. S7** Transmission spectra of (a) c-LGdH:Eu/PVA and (b) c-LGdH:Tb/PVA composite films ( $\sim 30 \mu\text{m}$  thickness) as a function of c-LGdH:RE concentration in PVA. Photographs in daylight of (c) neat PVA film and (d) c-LGdH:Eu/PVA and (e) c-LGdH:Tb/PVA composite films containing 4.0 wt% c-LGdH:RE nanofillers (grid =  $1 \text{ cm} \times 1 \text{ cm}$ ). The free-standing films were obtained by peeling from a Petri dish (8 cm in diameter).

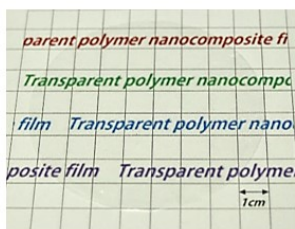




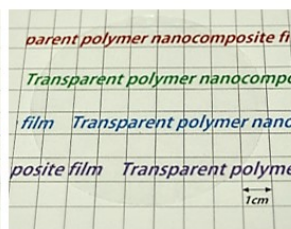
(a) neat PVA



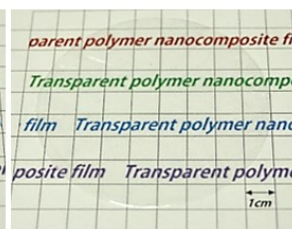
(b) c-LGdH:Eu 0.5 %



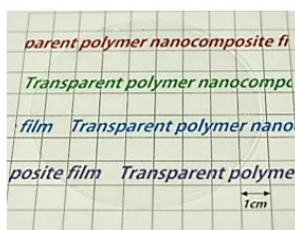
(c) c-LGdH:Eu 1.0 %



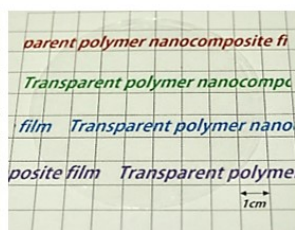
(d) c-LGdH:Eu 2.0 %



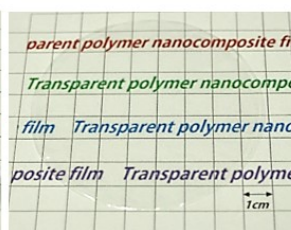
(e) c-LGdH:Eu 4.0 %



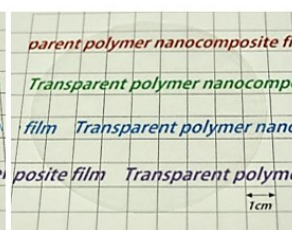
(f) c-LGdH:Tb 0.5 %



(g) c-LGdH:Tb 1.0 %

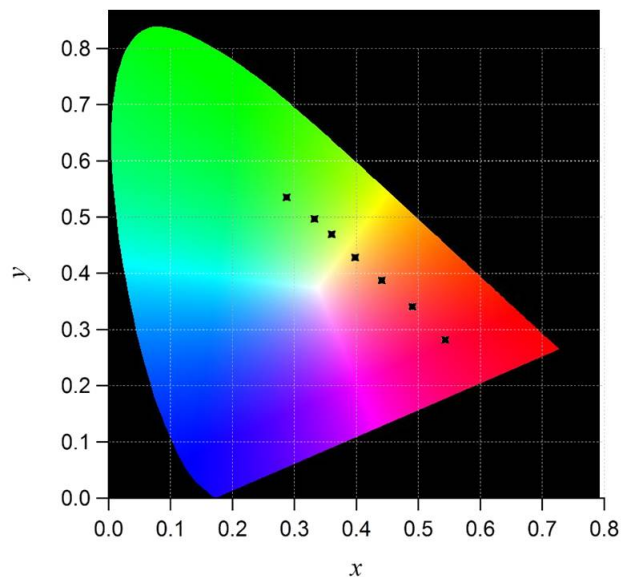


(h) c-LGdH:Tb 2.0 %



(i) c-LGdH:Tb 4.0 %

**Fig. S8** Photographs under daylight of (a) neat PVA film, (b–e) c-LGdH:Eu/PVA and (f–i) c-LGdH:Tb/PVA composite films ( $\sim 30 \mu\text{m}$  thickness) with different concentrations of c-LGdH:RE nanofillers. The free-standing films were obtained by peeling from a Petri dish (8 cm in diameter).



**Fig. S9** CIE 1931 chromaticity diagram. The black marks indicate the luminescent color coordinates of composite films as a function of c-LGdH:Eu/c-LGdH:Tb concentration ratio in PVA. From bottom, the films containing c-LGdH:Eu only (0.54, 0.28), c-LGdH:Eu/c-LGdH:Tb mixing ratio of 5.0 (0.49, 0.34), 2.0 (0.44, 0.39), 1.0 (0.40, 0.43), 0.5 (0.36, 0.47), and 0.2 (0.33, 0.50), and c-LGdH:Tb only (0.29, 0.54).