Supporting Information

Morphology and Phase Transformation from $NaCaSiO_3OH$ to $Na_2Ca_2Si_2O_7$ and Photoluminescence Evolution by Eu^{3+}/Tb^{3+} Doping

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Experimental Section

1. $NaCaSiO_3OH$ preparation

In a typical procedure, the starting solution I was prepared by mixing 0.472 g $Ca(NO_3)_2 \cdot 4H_2O$ into the solvent of H_2O and ethanol (EtOH) (the total volume of solution, 30 mL), and the volume ratio of EtOH/H_2O range from 0/30 to 30/0. The solution B was prepared by dissolved $Na_2SiO_3 \cdot 9H_2O$ (0.682 g) in 8 mL of deionized water. Then the solution II was added into the solution I dropwise to form a mixed soltion under vigorously magnetic stirring. After that, 6 g NaOH was quickly injected into the above solution, and after 10 min further stirring the resultant mixture was transferred into 50 mL stainless-steel autoclaves lined with poly(tetrafluoroethylene). The autoclave was sealed and maintained at 200 °C for 24 h and then cooled to room temperature. The products obtained at the bottom of the autoclave were collected,

washed several times with deionized water and ethanol , and dried in air. The detailed experimental parameters are listed in Table 1, and the products are denoted as S1-S8.

2. Preparation of Eu^{3+}/Tb^{3+} and Eu^{3+},Tb^{3+} co-doped in $Na_2Si_2O_7$

Fisrt, the precursor $NaCaSiO_3OH:Eu^{3+}$, $NaCaSiO_3OH:Tb^{3+}$ and $NaCaSiO_3OH:Eu^{3+}$, Tb^{3+} were prepared in a similar procedure except that $Eu(NO_3)_3$ or $Tb(NO_3)_3$ together with $Ca(NO_3)_2$ were the starting materials added to H_2O and EtOH. Then, the final products (Eu^{3+}/Tb^{3+} and Eu^{3+},Tb^{3+} co-doped in $Na_2Si_2O_7$) were obtained through a heat treatment for the corresponding precursor at 700 °C in air for 4 h.

3. Characterization

The powder X-ray diffraction (XRD) profile was measured using D8 Advance diffractometer (Bruker Corporation, Germany) with a monochromatized source of CuKα radiation (λ = 1.5406 Å) at 40 kV and 35 mA. The morphology and crystalline size of the $NaCaSiO_3OH$ samples were determined by scanning electron microscope (SEM, JEOL JSM-6510). Thermogravimetric analysis and differential scanning calorimetry (TG–DSC) were performed on a Setaram Labsys Evo at 10 $^oC\ min^{-1}$ in an air flow from room temperature to 650 ^oC. The situ variable temperature X-ray diffraction measurement (VT-XRD) were performed in X'Pert MRD diffractometer with a high-temperature reactor chamber (AntonPaar XRK 900) attached. Samples were measured from 2θ = 10° to 60° at a count rate of 3 s per step of 0.05°. Fluorescence spectrophotometer (F-4600, HITACHI, Japan), equipped with a

photomultiplier tube operating at 400 V, and a 150 W xenon lamp as the excitation source, was used to measure the room-temperature photoluminescence emission (PL) and photoluminescence excitation (PLE) spectra.

Table S1. Summary of the experimental conditions and the corresponding denotations for the final samples

Sample	EtOH /H_2O (mL)	NaOH (g)	Temp (^oC)	Time (h)
1	0/30	6	200	24
2	1/29	6	200	24
3	5/25	6	200	24
4	10/20	6	200	24
5	15/15	6	200	24
6	20/10	6	200	24
7	25/5	6	200	24
8	30/0	6	200	24

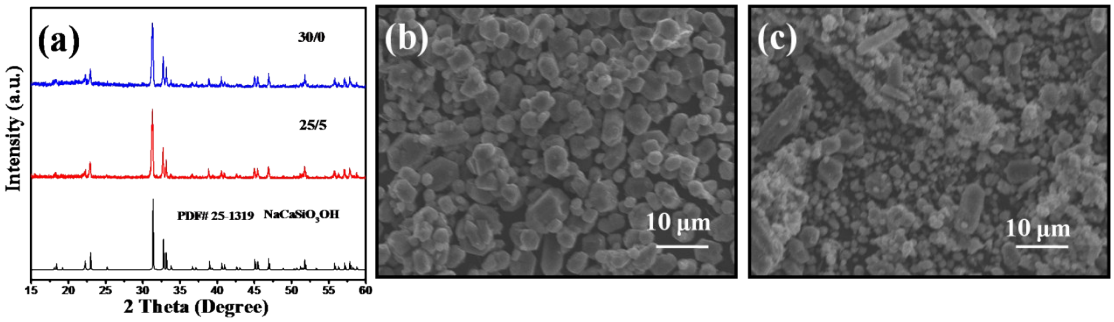

Fig.S-1 XRD patterns of as-prepared $NaCaSiO_3OH$ (a) with various volume ratios of EtOH to H_2O, 25/5 and 30/0. The standard data for $NaCaSiO_3OH$ (JCPDS card no. 25-1319) is shown as a reference. The SEM images of $NaCaSiO_3OH$ samples prepared in EtOH/H_2O = 25/5 (b) and 30/0 (c).

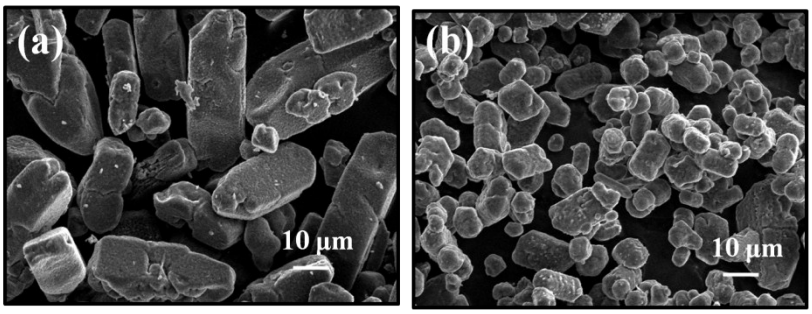

Fig.S-2 The SEM images for synthesis $Na_2Ca_2Si_2O_7$ via heat-treatment in air of $NaCaSiO_3OH$ precursors $EtOH/H_2O$ = 15/20 (a) and 20/10 (b) at 1000 °C

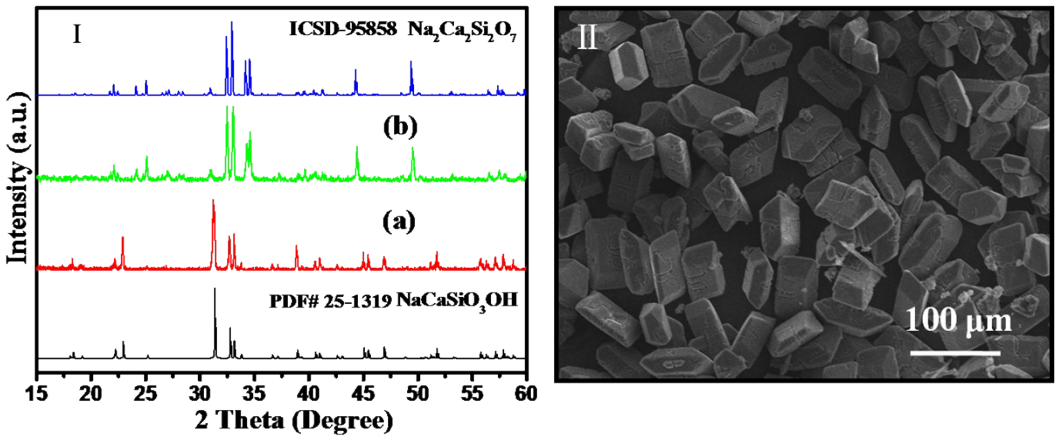

Fig.S-3 XRD patterns-I of as-prepared $NaCaSiO_3OH:0.03Tb^{3+},0.08Eu^{3+}$ (a) and $Na_2Ca_2Si_2O_7:0.03Tb^{3+},0.08Eu^{3+}$ (b) phosphors. The standard data for $NaCaSiO_3OH$ (JCPDS card no. 25-1319) and $Na_2Ca_2Si_2O_7$ (ICSD card no. 95858) are shown as a reference. The SEM image-II of $NaCaSiO_3OH:0.03Tb^{3+},0.08Eu^{3+}$ sample prepared in $EtOH/H_2O$ = 5/25.