Electronic Supplementary Information

Protected-annealing regulated defects to improve optical properties and luminescence performance of Ce:YAG transparent ceramic for white LED

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1 Detailed Fabrication of Transparent Ceramics

In this work, high purity powders Y_2O_3 (99.99% purity, Alfa Aesar, Ward Hill, America), α- Al_2O_3 (99.99% purity, Alfa Aesar, Ward Hill) and CeO_2 (99.99% purity, Alfa Aesar, Ward Hill) were selected as the starting material. The mean particle sizes of the starting Y_2O_3, Al_2O_3 and CeO_2 powders were 5.0 μm, 300 nm and 5 μm, respectively. They were weighted precisely in stoichiometric

proportions to obtain $(Ce_xY_{1-x})_3Al_5O_{12}$, ($x$=0.0005 and 0.002). Tetraethyl orthosilicate (TEOS, 99.99%, Alfa Aesar) and MgO (99.999%, Alfa Aesar, Ward Hill, MA) were selected as sintering additives.

These powders were mixed with 0.3 wt.% DS005 (99.9 % purity, Alfa Aesar, Ward Hill, America) as a dispersing agent in anhydrous ethyl alcohol in a ball milling jar, and then planetary ball milled for 15 h to obtain powders with a uniform particle size distribution and mean particle size of 0.39 μm. The mixture was dried at 60 ^oC in an oven for 24 h, and then meshed and sieved through a 100-mesh screen. After that, the sieved powder mixture was initially uniaxially pressed at 20 MPa in a stainless-steel mold with a diameter of 22 mm, and then cold isostatic pressed at 200 MPa for 300 s, to obtain compacted powder pellets with a relative density of ~53 % of the theoretical value of YAG. The pressed pellets were then calcined at 800 ^oC for 4 h in air in a muffle furnace to remove the volatile organic residues. The calcined green bodies were then sintered in a tungsten mesh heated vacuum furnace at 1780 ^oC for 8 h, Subsequently, air annealing treatment was carried out in a muffle furnace after vacuum sintering. Hydrogen annealing (5% H_2/95% N_2) was also performed using a tube furnace under the gas flow rate was 20 ml/min. After that, the specimens were mirror polished on both surfaces to 1.0 mm-thick.

2 XPS spectra

Chemical states of Ce atoms of 0.2 at.% Ce:YAG transparent ceramics

following different annealing conditions were measured by X-ray photoelectron spectroscopy (XPS), as shown in Fig. S1. It is well known that the special peaks at 885 eV and 917 eV are corresponding to the characteristic peaks of Ce(III) and Ce(IV), respectively. Unfortunately, no obvious special peaks were observed from the figure. For Ce:YAG ceramics, Ce^{3+} ions are embedded in YAG lattice and only few Ce ions are floated on the surface of tested specimens. Accordingly, no obvious XPS peaks were observed from XPS spectrum.

3 Supplementary Figures

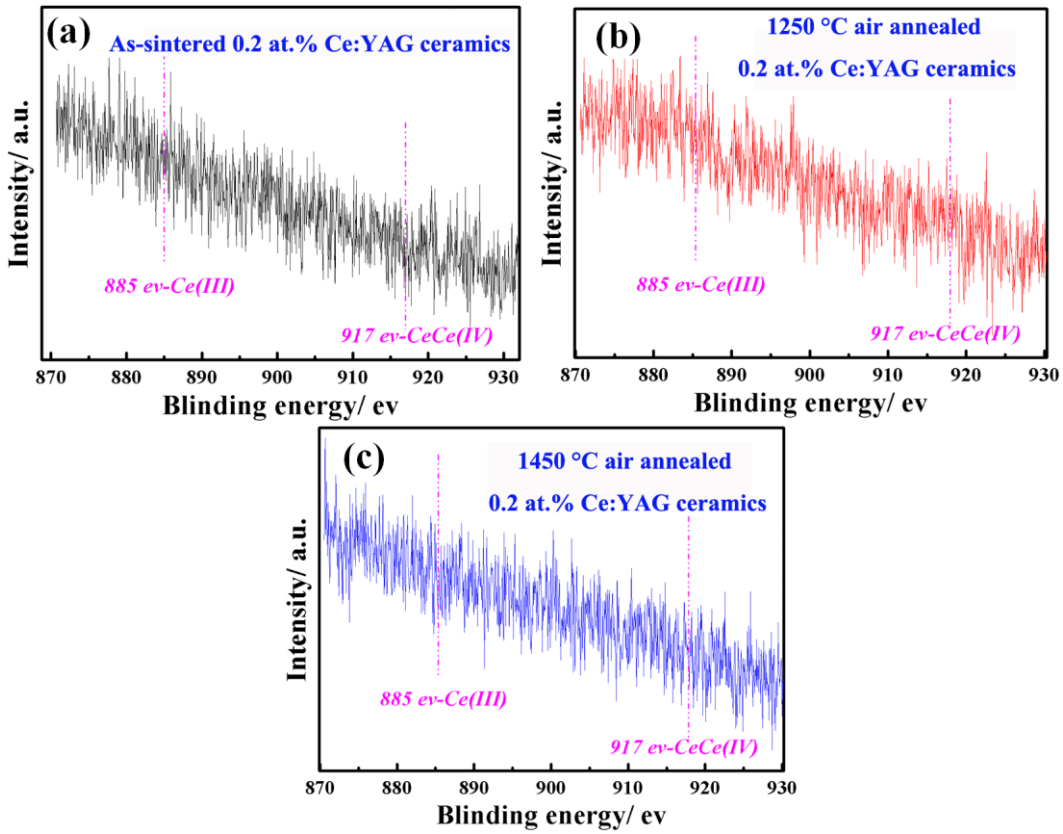

Fig. S1 The Ce 3d photoemission spectra in XPS for (a) as-sintered, (b) 1250 °C air annealed, and (c) 1450 °C air annealed 0.2 at.% Ce:YAG ceramics