Electronic supplementary information (ESI)

Enhanced near infrared-to-visible upconversion in CaTiO$_3$:Yb^{3+}/Er^{3+} phosphor via host lattice modification using co-doping of Mg^{2+} ions

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Upconversion quantum efficiency measurement and method. A homemade, optical-microscope (*Olympus IX93*)-based setup was used for measuring upconverted emission spectra and the upconversion efficiencies (UC-QEs) of solid films. The details of the setup were reported elsewhere.[1,2] The setup equipped two-color (785/980 nm) cw laser sources for excitation and an *Ocean Optics USB2000-FLG fiber spectrometer* as a detector. The excitation was designed to irradiated sample uniformly through a 20× objective lens (N.A. = 0.45). The excitation intensity was determined from the incident power divided by the irradiation area calculated with e^{-2} radius obtained by the spatial beam profile recorded with a COMS camera. The emission from the sample was collected by the same objective lens and transferred to the detector via a multimode optical fiber. A 700-nm short pass filter was used to block the excitation light. To calculate UC-QE (η_{UC}), which is 100% full scale, we employed the relative method by using the following equation,

$$\eta_{\mathrm{UC}} = 2\,\Phi_R\,\frac{F_S}{F_R}\,\frac{(1 - 10^{-A_R})}{(1 - 10^{-A_S})}\,\frac{P_R}{P_S}\,\frac{n_S^2}{n_R^2} \approx 2\,\Phi_{\mathrm{FL}}\,\frac{F_S}{F_R}\,\frac{A_R}{A_S}\,\frac{P_R}{P_S}\,\frac{n_S^2}{n_R^2}, \qquad (S1)$$

where F is the integrated emission intensity (spectral range 500-700 nm), A is absorbance, P is excitation power, and n is the refractive index of medium. Subscripts of S and R means the sample and reference, respectively. A was determined from the absorption spectrum measured at the same sample position where the emission spectrum measured with the same homemade setup by switching the internal optical paths. The value of A is smaller than 0.1, so the approximated relation was applied. As for the quantum yield standard Φ_R, we used the 100 µM solution of fluorescence dye IR-140 (Exciton Inc.) in ethylene glycol (FL-QY = 0.17) sandwiched between microscope slide and cover slip with a fixed-thickness spacer. For the sample, the same cover slip and spacer were used to keep the same configuration between the sample and reference. Though, it was difficult to exclude the scattering by sample grains for emission, tentatively apply refractive index of 1 for the sample is used in the calculation.

1. N. Tripathi, M. Ando, T. Akai, K. Kamada, J. Mater. Chem. C. 2022, 10, 4563.

2. A. Abulikemu, Y. Sakagami, C. Heck, K. Kamada, H. Sotome, H. Miyasaka, D. Kuzuhara, H. Yamada, ACS Appl. Mater. Interfaces. 2019, 11, 20812.
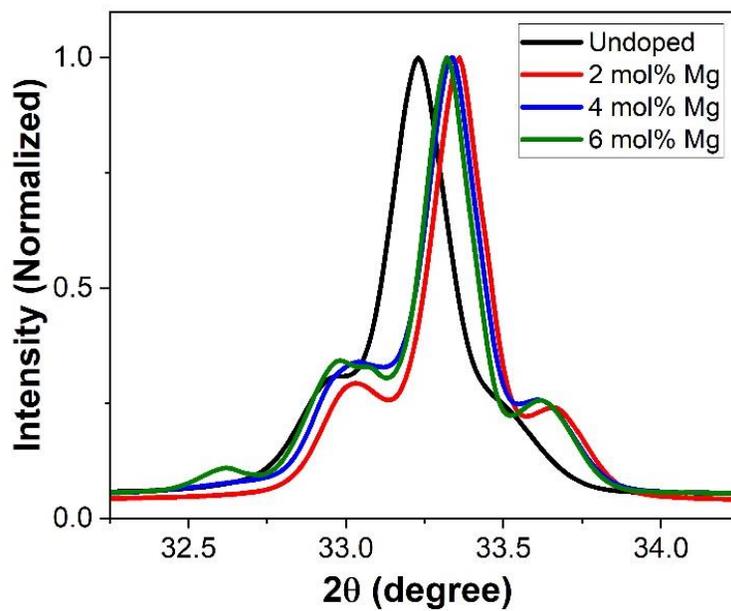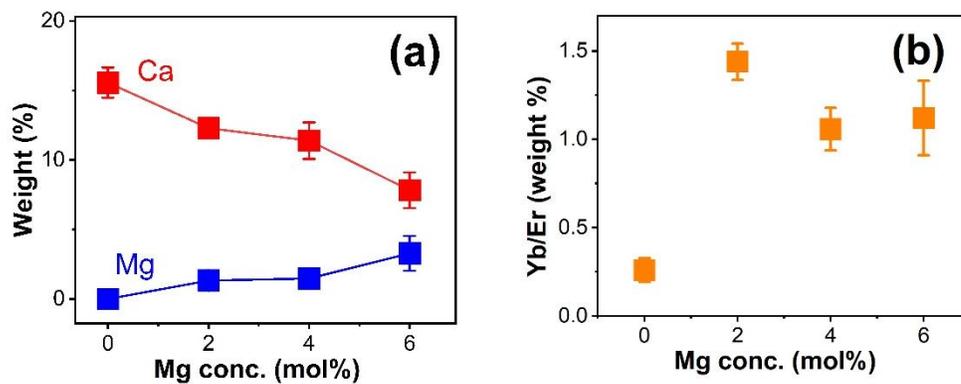
Figure S1: XRD peak corresponding to the $CaTiO_3$ phase, showing 2θ shift toward higher angle in Mg^{2+} doped $Ca_{1-x}Mg_xTiO_3:Yb^{3+}/Er^{3+}$ samples.
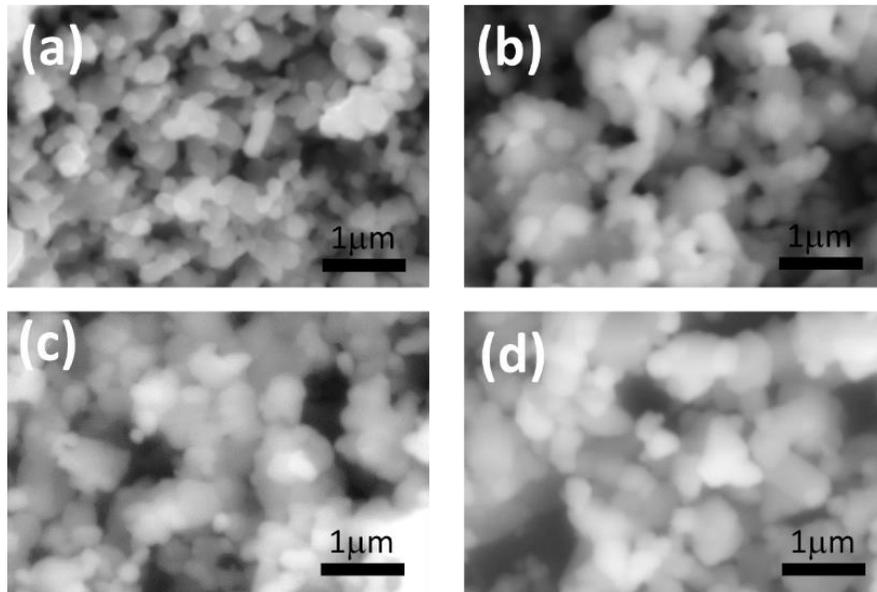
Figure S2. SEM micrographs of $Ca_{1-x}Mg_xTiO_3$:Yb^{3+}/Er^{3+} samples at Mg doping concentration of (a) x= 0, (b)x= 0.02 (c) x= 0.04. and (d) x= 0.06.
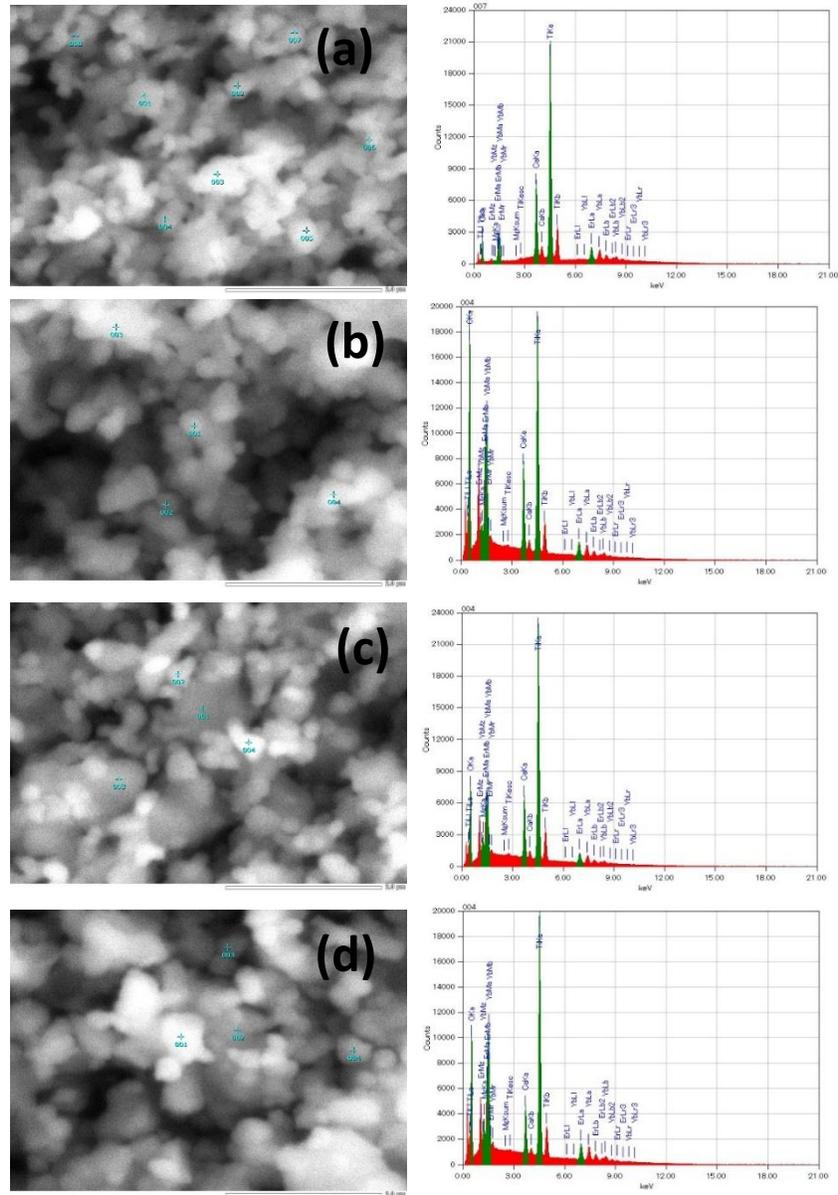
Figure S3. EDS spectrum for $Ca_{1-x}Mg_xTiO_3$:Yb^{3+}/Er^{3+} samples at Mg^{2+} doping concentration of (a) x= 0, (b)x= 0.02 (c) x= 0.04. and (d) x= 0.06.

Figure S4. Elemental distribution of (a) Ca, Mg and (b) Yb/Er, estimated from of EDS spectrum for $Ca_{1-x}Mg_xTiO_3{:}Yb^{3+}/Er^{3+}$ samples as a function of Mg^{2+} doping concentration.
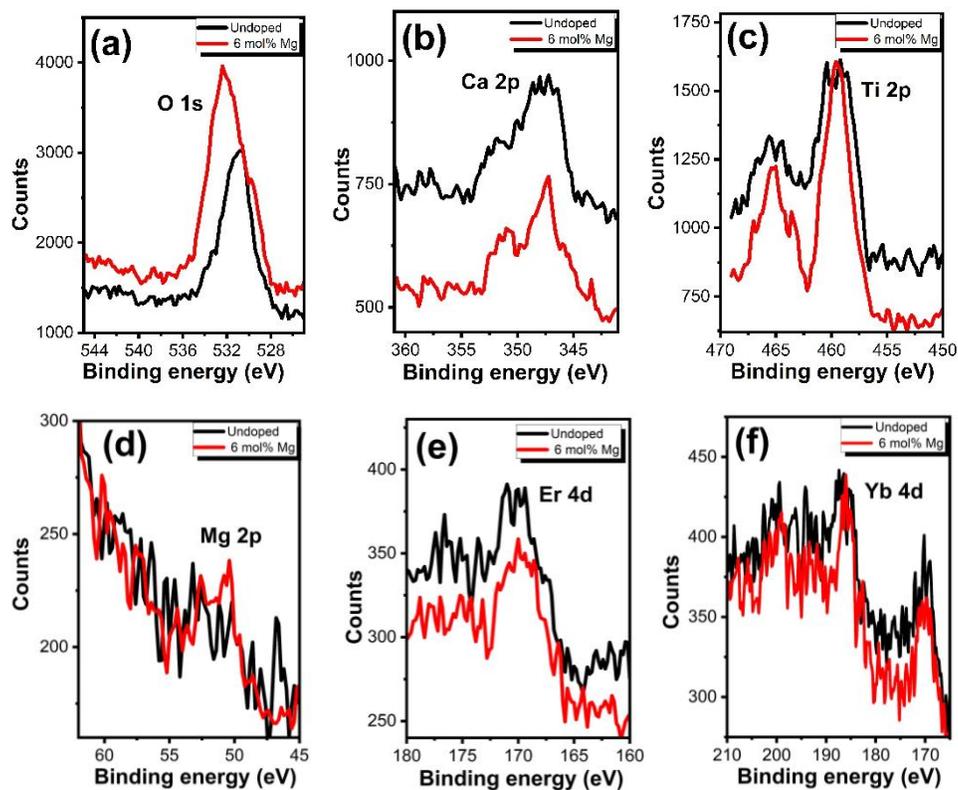
Figure S5: XPS spectra for Undoped and 6 mol% Mg doped $Ca_{1-x}Mg_xTiO_3$:Yb^{3+}/Er^{3+} samples

Table S1: Composition estimation from XPS data

Sample	Elements					
	O 1s [at.%]	Ca 2p [at.%]	Ti 2p [at.%]	Mg 2p [at.%]	Er 4d [at.%]	Yb 4d [at.%]
Undoped	33.5	2.3	8.3	<0.1	0.7	4.1
6 mol% Mg doped	48.0	3.6	12.3	8.1	1.5	7.3
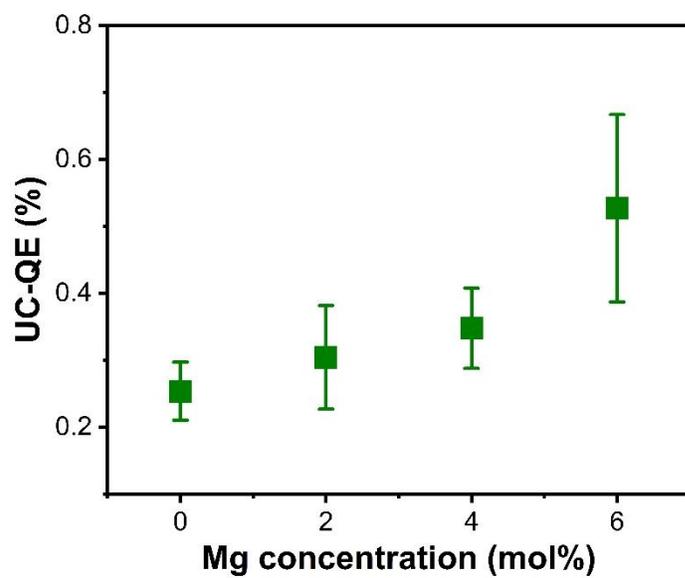
Figure S6. Upconversion efficiency graph for $Ca_{1-x}Mg_xTiO_3:Yb^{3+}/Er^{3+}$ samples as a function of Mg^{2+} doping concentration, measured at 980 nm excitation (Excitation intensity 1 $W\text{-}cm^{-2}$).