Supporting Information

Incorporating Three-Dimensional Ordered Macropores into High-Entropy Oxides for Catalytic Soot Combustion

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Synthesis of polystyrene spheres: Typically, 100 mL deionized water was added into a 250 mL three-neck flask with 800 rpm magnetic stirring. The N_2 bubbling, 75°C oil bath, and continuous condensate water were kept during the whole process. After N_2 deoxidizing for 30 mins, 13 mL styrene (99%, Adamas Reagent Co., Ltd.) was injected into the flask, and then a homogeneous liquid was observed after stirring for another 30 mins. 0.4 g potassium persulphate (KPS, Sinopharm Chemical Reagent Co., Ltd.) was dissolved in 20 mL of deionized water and added to the flask reactor as the polymerization initiator. The reaction lasted for 24 hours. Finally, the resulting solution was centrifugated under 4000 rpm for 90 mins by three times, and the precipitation was dried under 30°C. To study the influence of KPS amount on the size of PS spheres, the addition of KPS was changed to 0.2 g.

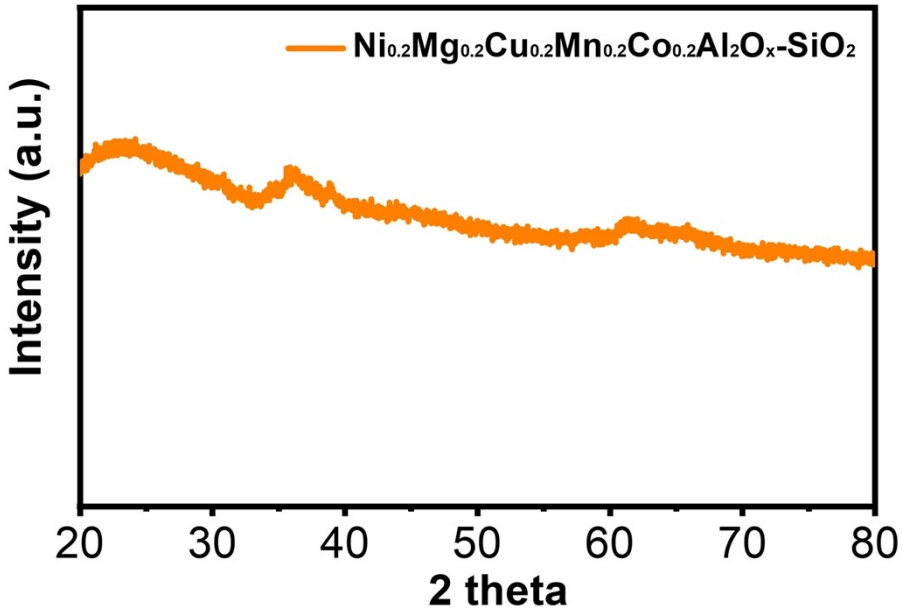

Fig. S1 The XRD pattern of $Ni_{0.2}Mg_{0.2}Cu_{0.2}Mn_{0.2}Co_{0.2}Al_2O_x$ templated by hard template commercial SiO_2.

Synthesis of $Ni_{0.2}Mg_{0.2}Cu_{0.2}Mn_{0.2}Co_{0.2}Al_2O_x$-$SiO_2$: The 3.33 mmol $Al(NO_3)_3$, 0.33 mmol $Ni(NO_3)_2$, 0.33 mmol $Fe(NO_3)_2$, 0.33 mmol $Cu(NO_3)_2$, 0.33 mmol $Mn(NO_3)_2$, 0.33 mmol $Co(NO_3)_2$ and 2 g commercial SiO_2 (Adamas, Hydrophobic, R812, 7nm,SSA:260 m^2/g) were added in 40 mL ethanol and stirred for 2h. Then the suspension was rotary evaporated under 40°C water bath until acquiring dried resultant. After calcined under 600°C for 5h (heating rate: 1.2°C/min), the product was added in 40mL 1mol/L NaOH solution to dissolve the SiO_2 template for 12h, followed by centrifuging and washing by water for three times. After desiccated in 80°C oven for 12h, the $Ni_{0.2}Mg_{0.2}Cu_{0.2}Mn_{0.2}Co_{0.2}Al_2O_x$-$SiO_2$ was acquired.

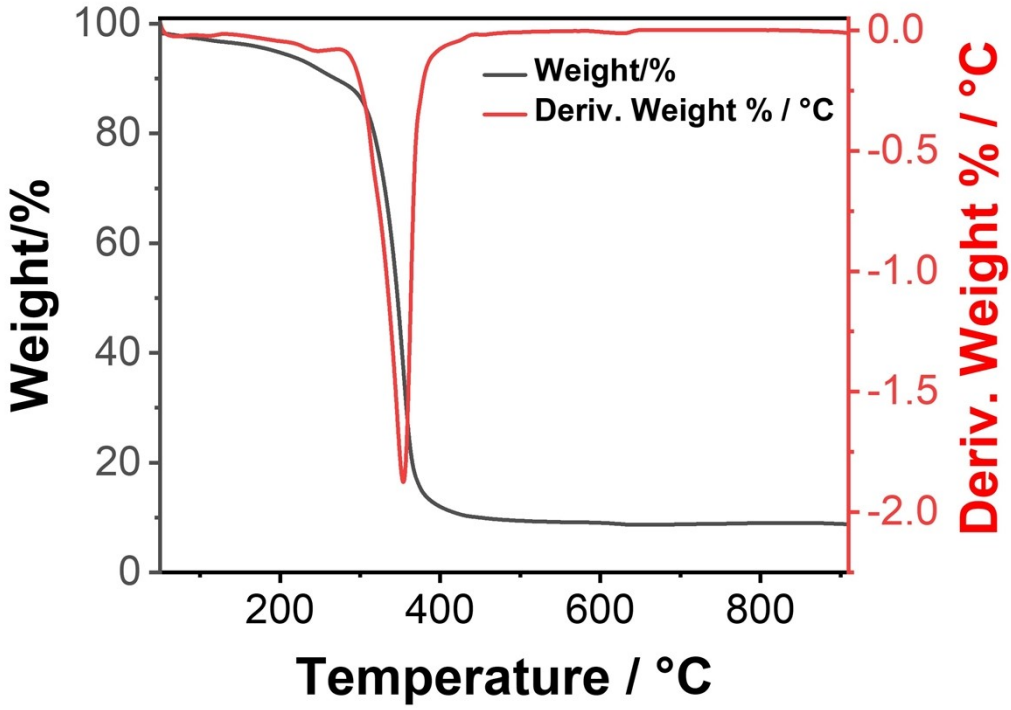

Fig. S2 The TGA and DTG curves of the precursor of 3DOM- $LaFe_{0.2}Co_{0.2}Ni_{0.2}Cr_{0.2}Mn_{0.2}O_x$ in the air atmosphere.

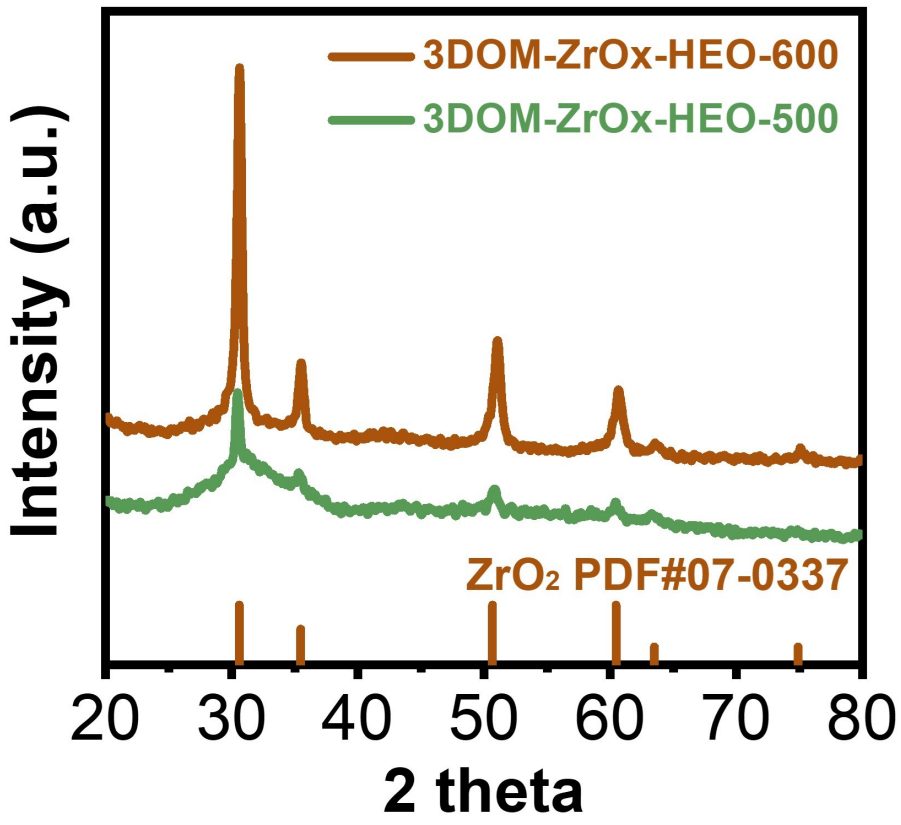

Fig. S3 The XRD patterns of 3DOM-HEO-ZrO_x prepared at 500°C and 600°C.

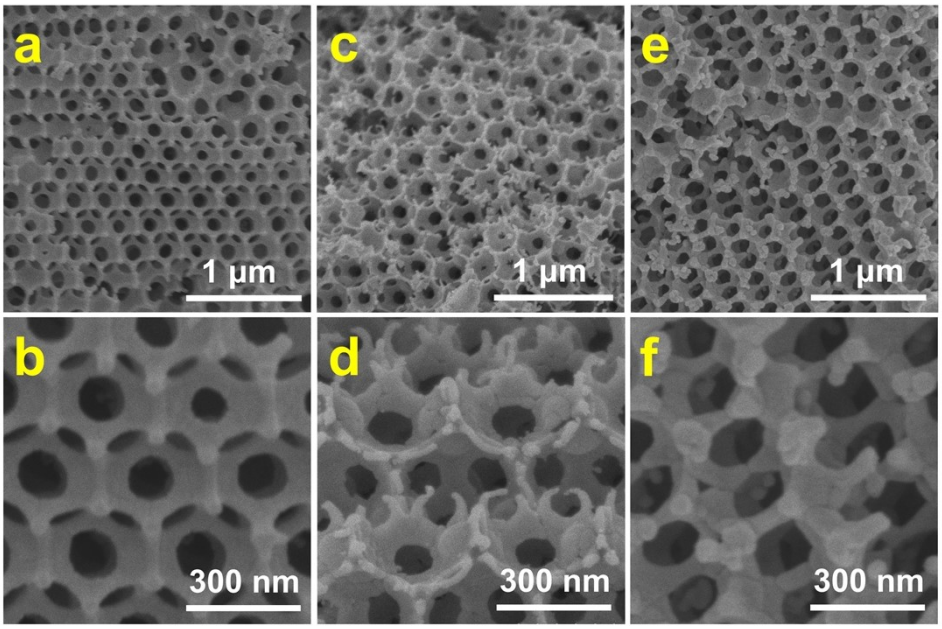

Fig. S4 The SEM images of the 3DOM-HEO-$LaMnO_x$ with different air calcination process: (a, b) 650°C for 5h with the 1.3°C/min heating rate; (c, d) 700°C for 3h with the 1.4°C/min heating rate; (e, f) 750°C for 3h with the 1.5°C/min heating rate.

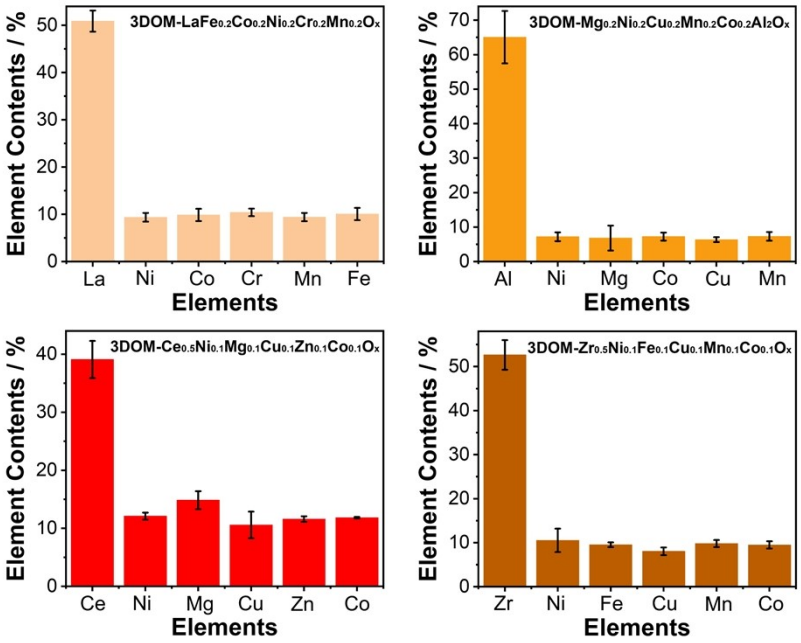

Fig. S5 The ICP results of: a) 3DOM-$LaNi_{0.2}Fe_{0.2}Co_{0.2}Cr_{0.2}Mn_{0.2}O_x$; b) 3DOM-$Ni_{0.2}Mg_{0.2}Cu_{0.2}Mn_{0.2}Co_{0.2}Al_2O_x$; c) 3DOM-$Ce_{0.5}Ni_{0.1}Mg_{0.1}Cu_{0.1}Zn_{0.1}Co_{0.1}O_x$; d) 3DOM-$Zr_{0.5}Ni_{0.1}Fe_{0.1}Cu_{0.1}Mn_{0.1}Co_{0.1}O_x$ (Error bar was determined based on the ICP results of three samples).

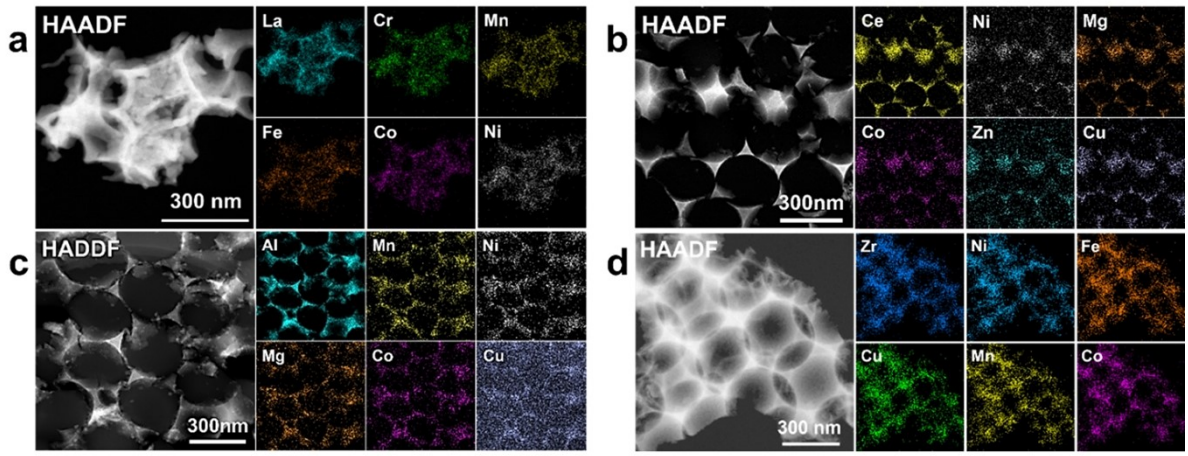

Fig. S6 STEM-HAADF and EDS mapping images (300 nm scale) of (a) perovskite 3DOM-HEO-$LaMnO_x$; (b) cubic 3DOM-HEO-CeO_x; (c) cubic 3DOM-HEO-ZrO_x; (d) spinel 3DOM-HEO-$MgAl_2O_x$.

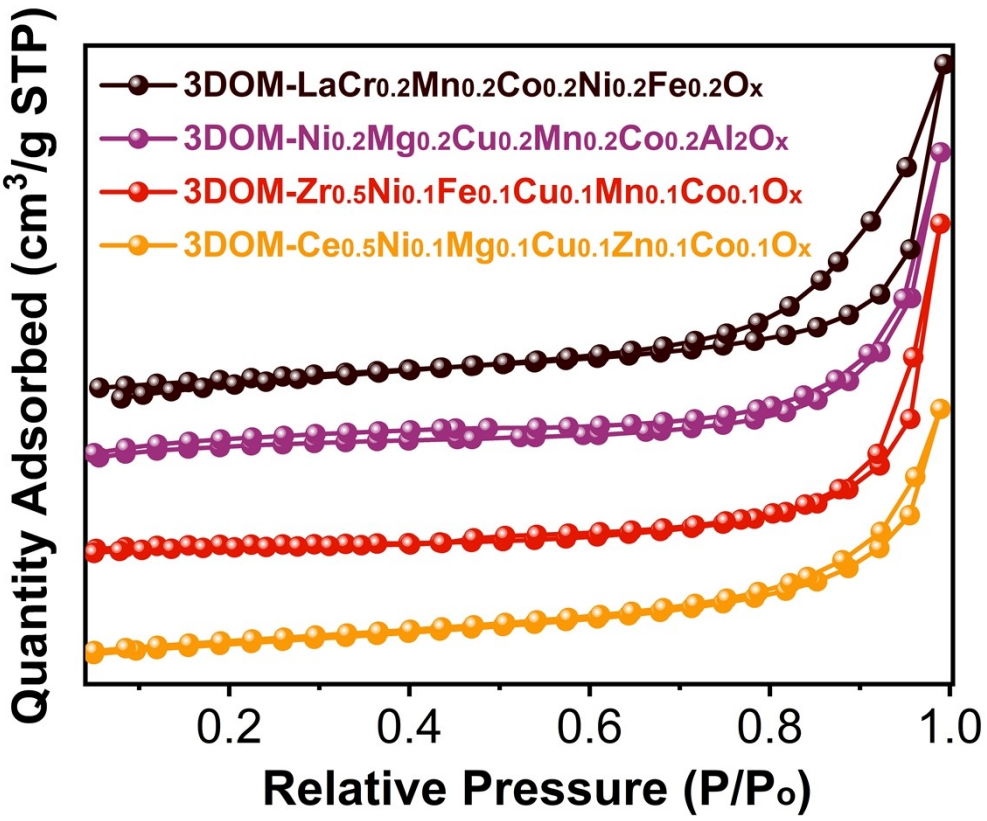

Fig. S7 The N_2 isotherm adsorption-desorption curves of 3DOM-HEO-metal oxides. The quantity absorbed of 3DOM-$Zr_{0.5}Ni_{0.1}Fe_{0.1}Cu_{0.1}Mn_{0.1}Co_{0.1}O_x$, 3DOM-$Ni_{0.2}Mg_{0.2}Cu_{0.2}Mn_{0.2}Co_{0.2}Al_2O_x$, and 3DOM-$LaCr_{0.2}Mn_{0.2}Co_{0.2}Ni_{0.2}Fe_{0.2}$-$O_x$ were moved upward by 10, 20, and 30 units respectively.

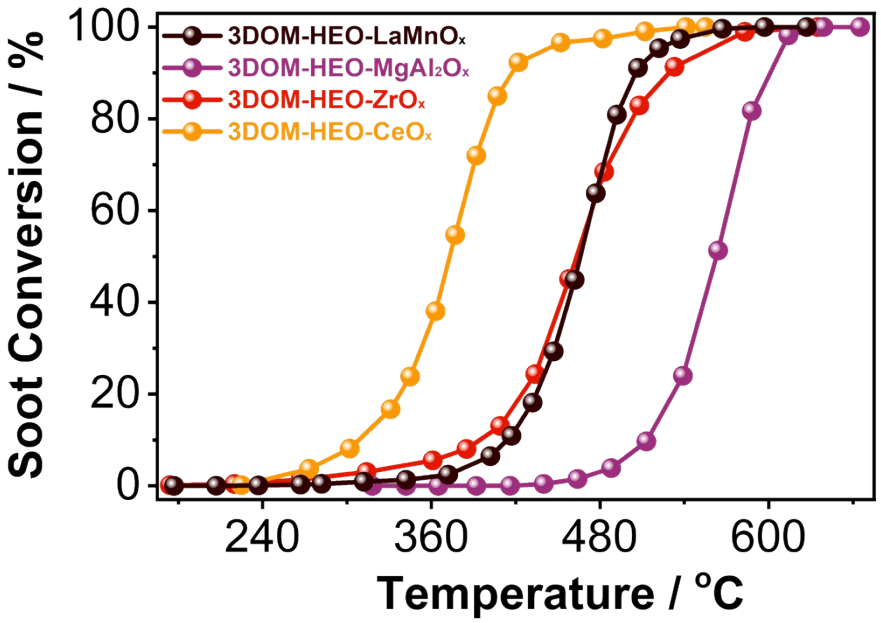

Fig. S8 The comparison of catalytic soot combustion tests for 3DOM-HEO-CeO_x, 3DOM-HEO-ZrO_x, 3DOM-HEO-$LaMnO_x$, and 3DOM-HEO-$MgAl_2O_x$, reaction condition: tight contact of 10mg soot and 0.2g catalysts by hand-milling for 15 min, taking 10mg mixture of soot and catalyst for soot combustion reaction, dispersed by 0.1g silica sand, 30 mL/min continuous pure air flow, heating from room temperature to 750°C with the 5°C/min heating rate.

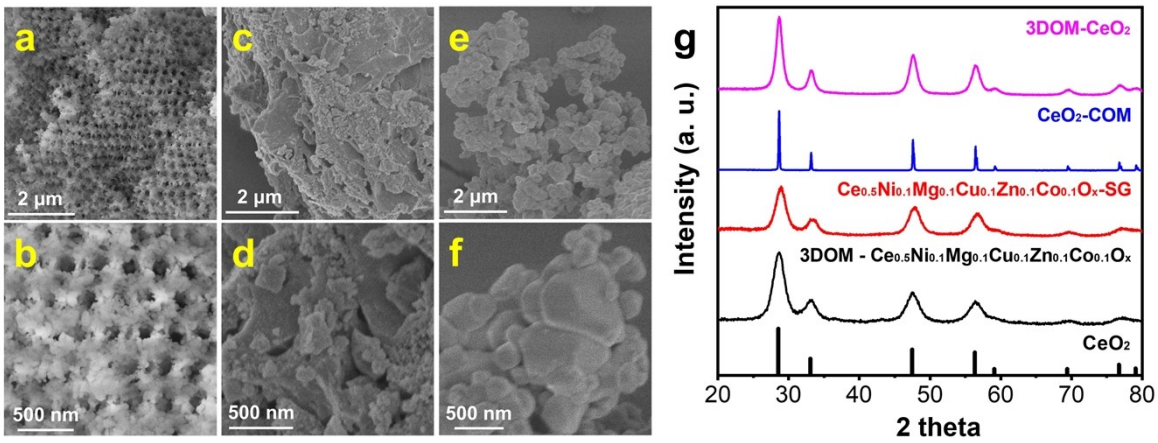

Fig. S9 The SEM images of (a, b) 3DOM-CeO_2; (c, d) HEO-CeO_x-SG; (e, f) CeO_2-COM. (g) the XRD patterns of 3DOM-HEO-CeO_x, CeO_2-COM, 3DOM-CeO_2, and HEO-CeO_x-SG.

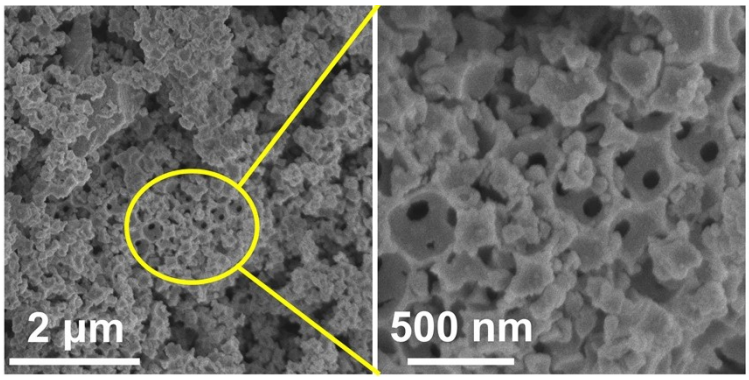

Fig. S10 The SEM images of 3DOM-HEO-CeO_x-reacted.

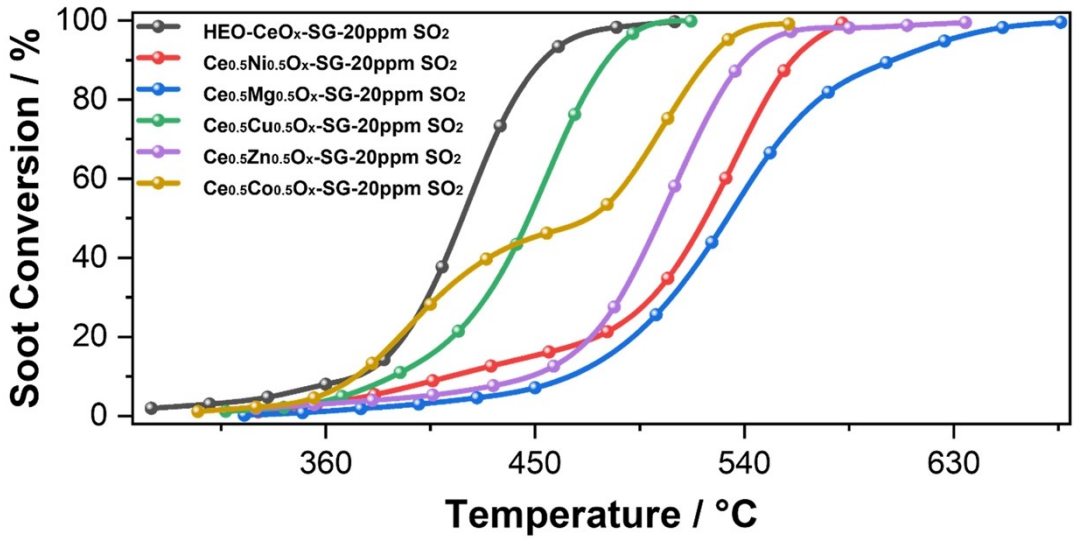

Fig. S11 the comparison of catalytic soot combustion tests under 30 mL/min air containing 20 ppm SO_2 for $Ce_{0.5}Ni_{0.1}Mg_{0.1}Cu_{0.1}Zn_{0.1}Co_{0.1}O_x$-SG, $Ce_{0.5}Mg_{0.5}O_x$-SG, $Ce_{0.5}Cu_{0.5}O_x$-SG, $Ce_{0.5}Zn_{0.5}O_x$-SG, and $Ce_{0.5}Co_{0.5}O_x$-SG.

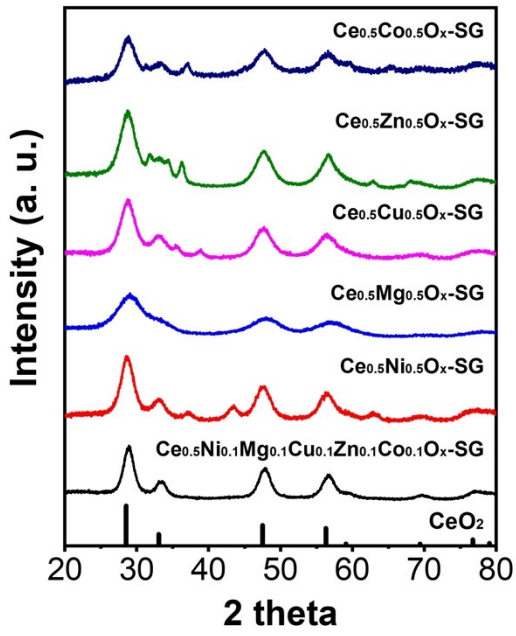

Fig. S12 the XRD patterns of the $Ce_{0.5}Ni_{0.1}Mg_{0.1}Cu_{0.1}Zn_{0.1}Co_{0.1}O_x$-SG, single-elemental doped $Ce_{0.5}Ni_{0.5}O_x$-SG, $Ce_{0.5}Mg_{0.5}O_x$-SG, $Ce_{0.5}Cu_{0.5}O_x$-SG, $Ce_{0.5}Zn_{0.5}O_x$-SG, $Ce_{0.5}Co_{0.5}O_x$-SG.