

### Experiment procedure:

The chemicals used in this work are all AR grade. Generally, 1 g of P123 ( $\text{EO}_{20}\text{PO}_{70}\text{EO}_{20}$ , EO= ethylene oxide, PO= propylene oxide) was dissolved in 20 ml anhydrous ethanol at room temperature (RT), then 1.6 ml 35wt% HCl, 0.143g salicylic acid, 0.1g citric acid, quantitative cerium acetate and zirconium acetate ( $n(\text{Ce})/n(\text{Zr})=1$ ), and 2.04 g (10mmol) aluminum isopropoxide were added into the above solution with vigorous stirring. The mixture was covered with polyethylene (PE) films and stirred vigorously for 5 h. Solvent evaporation was performed at 60 °C for 24 h and then 65 °C for another 48 h in air without stirring.

The resulting samples were calcined at 400 °C for 4h with a heating rate of 1 °C/min and calcined at 800 °C, 900 °C and 1000 °C respectively for 1 h with a heating rate of 10 °C/min.

Structural analysis of the obtained samples were carried out on a Phillips X'Pert SUPER powder X-ray diffractometer, Cu K $\alpha$  radiation,  $\lambda=1.5418$  Å). Transmission electron microscopy (TEM) images were obtained with a Tecnai F20 transmission electron microscope at 200 KV. The Barrett-Joyner-Halenda (BJH) pore-size distributions, Brunauer-Emmett-Teller (BET) surface area and total pore volume of the powders were examined via Nitrogen adsorption experiments. Nitrogen adsorption isotherms of the purified sample were determined at -196 °C using N<sub>2</sub> nitrogen in a conventional volumetric technique by Quantachrome Nova 4200.

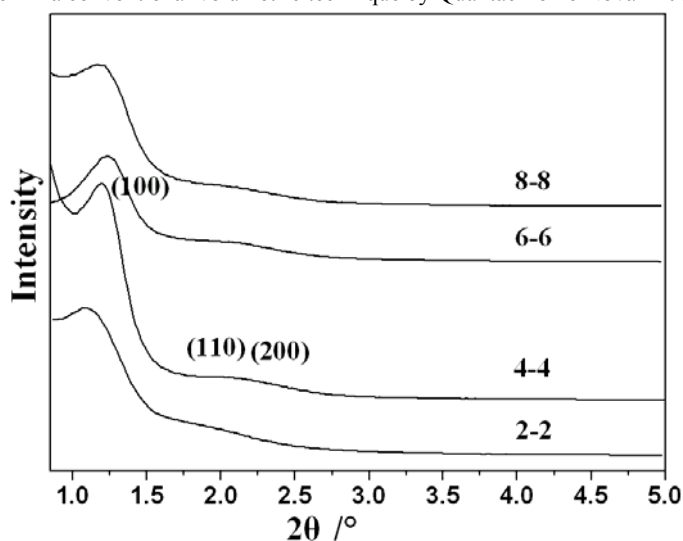


Figure S1 The small-angle XRD patterns of  $x\text{mol}\%\text{Ce}-x\text{mol}\%\text{Zr}/\text{Al}_2\text{O}_3$  ( $x=2-8$ ) calcined at 400 °C

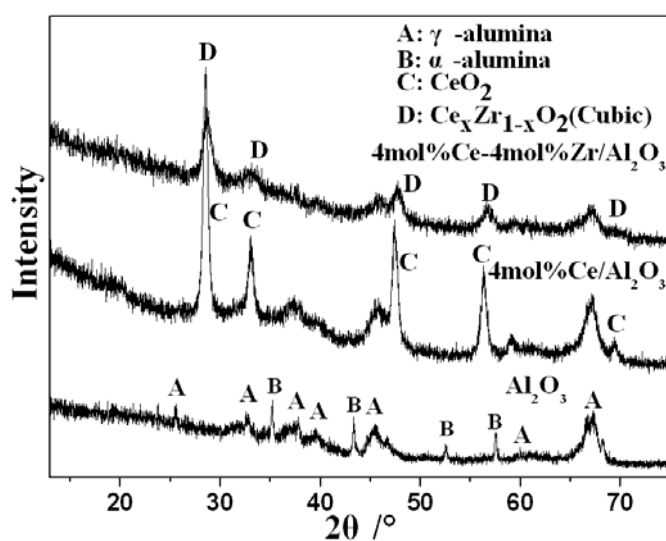


Figure S2 The wide-angle XRD patterns of pure  $\text{Al}_2\text{O}_3$ , 4mol%Ce/ $\text{Al}_2\text{O}_3$  and 4mol%Ce-4mol%Zr/ $\text{Al}_2\text{O}_3$  calcined at 1000 °C

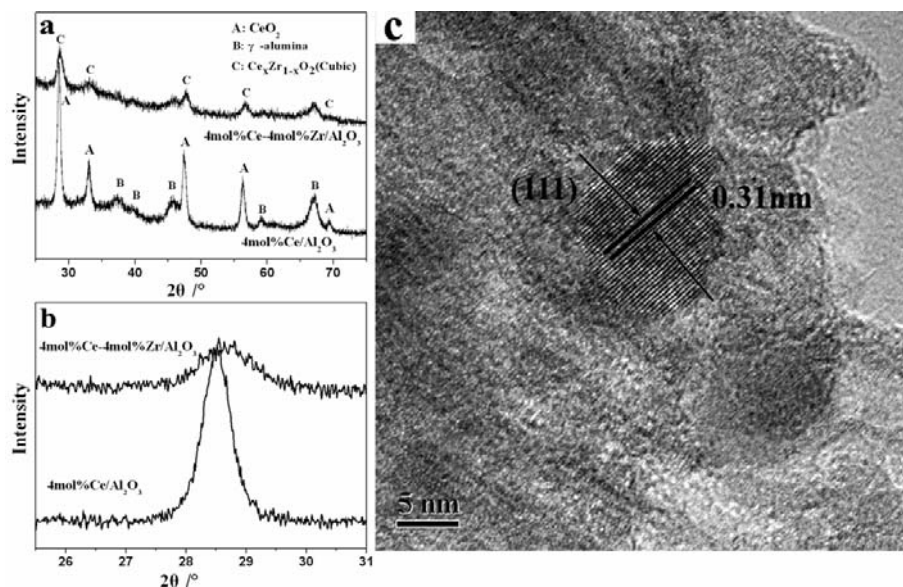


Figure S3 (a) and (b) : wide-angle XRD patterns of 4mol% Ce/Al<sub>2</sub>O<sub>3</sub> and meso-4CeZrAl calcined at 1000 °C; (c) HRTEM image of meso-4CeZrAl calcined at 1000 °C.

Figure S3a shows the wide-angle XRD patterns of 4mol% Ce/Al<sub>2</sub>O<sub>3</sub> calcined at 1000 °C. The appearance of crystalline CeO<sub>2</sub> particles can be expected according to the diffraction peaks correspond to a pure fluorite CeO<sub>2</sub> phase (JCPDS: 34-0394, space group *Fm3m*). The meso-4CeZrAl calcined at 1000 °C showed the similar patterns as 4mol% Ce/Al<sub>2</sub>O<sub>3</sub>, but all the ceria related peaks shifted to higher 2θ positions (Figure S3a and S3b). The peak due to reflection Ce (111) in the position 2θ=28.6 ° is the one with the higher intensity for CeO<sub>2</sub>, which shifted to 28.7 ° for meso-4CeZrAl (Figure S3b). These shifts are an indication that ceria and zirconia formed a solid solution. Since the shifted observed for meso-4CeZrAl was smaller and no diffraction peaks related to zirconia phases were detected, it is reasonable to be believed that only a little zirconium was incorporated into ceria lattice and the additional zirconia could be highly dispersed<sup>12</sup>. Lattice planes with a spacing of 0.31 nm are also present in the high-resolution TEM (HRTEM) image of an individual nanoparticle (Figure S3c), which is close to the  $d_{(111)}$  of CeO<sub>2</sub>, further confirming the results observed from wide-angle XRD dates.

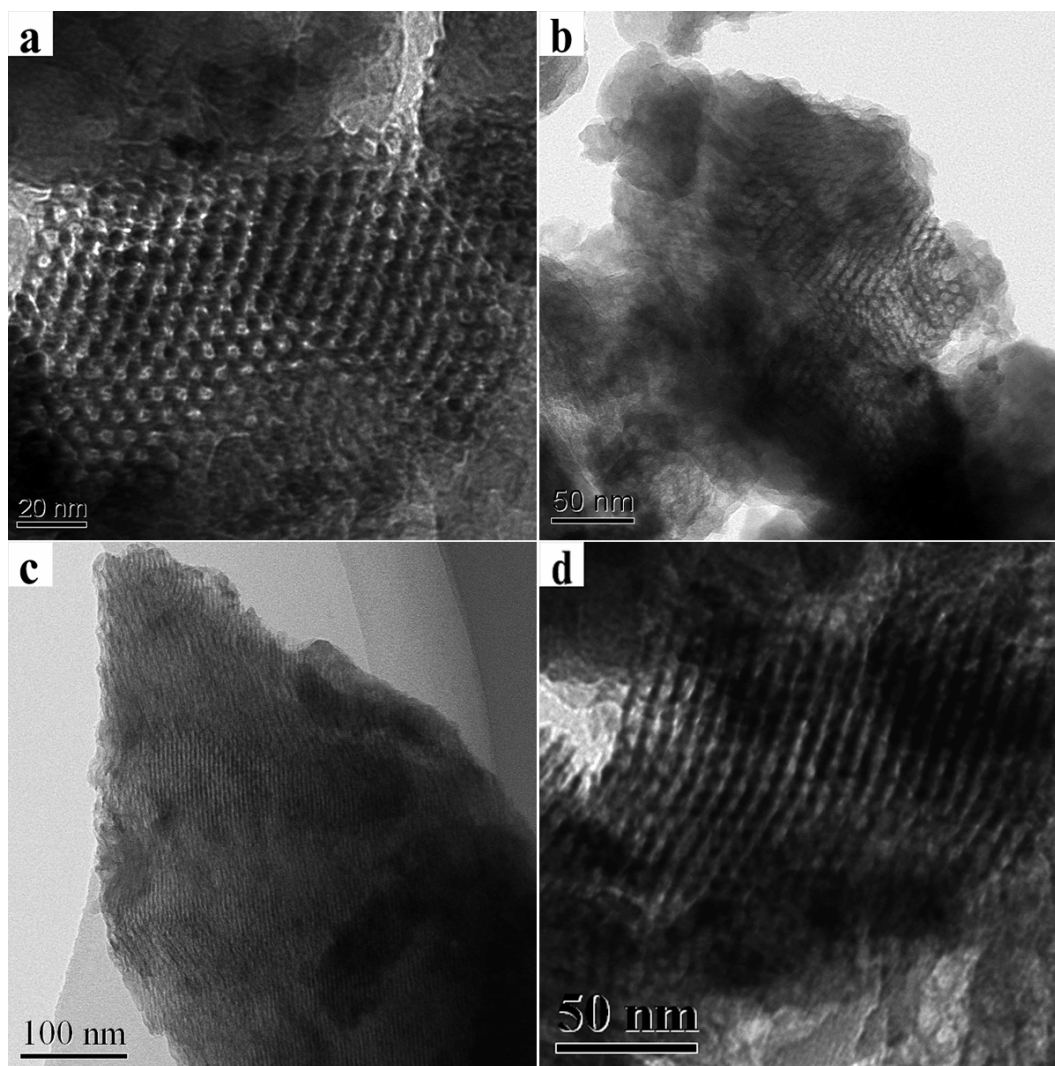


Figure S4 TEM images of meso-4CeZrAl calcined at 400 °C viewed along the (a, b) [001] and (c, d) [110] direction.

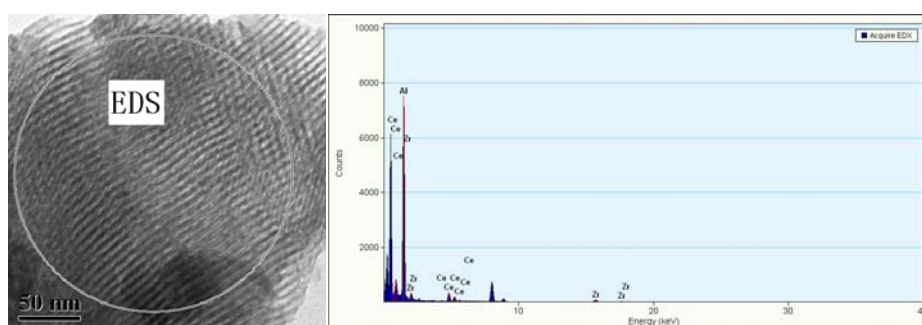


Figure S5 TEM images of meso-4CeZrAl calcined at 400 °C viewed along the [001] direction and its EDS.

There is no bulk ceria and zirconia nanoparticles can be seen from the TEM images (Figure S4 and S5) for the sample calcined at 400 °C. However, the ceria and zirconia were detected by the Energy Dispersive Spectrometer (EDS). So it is reasonable to conclude that the ceria and zirconia are highly dispersed on the surface of alumina.

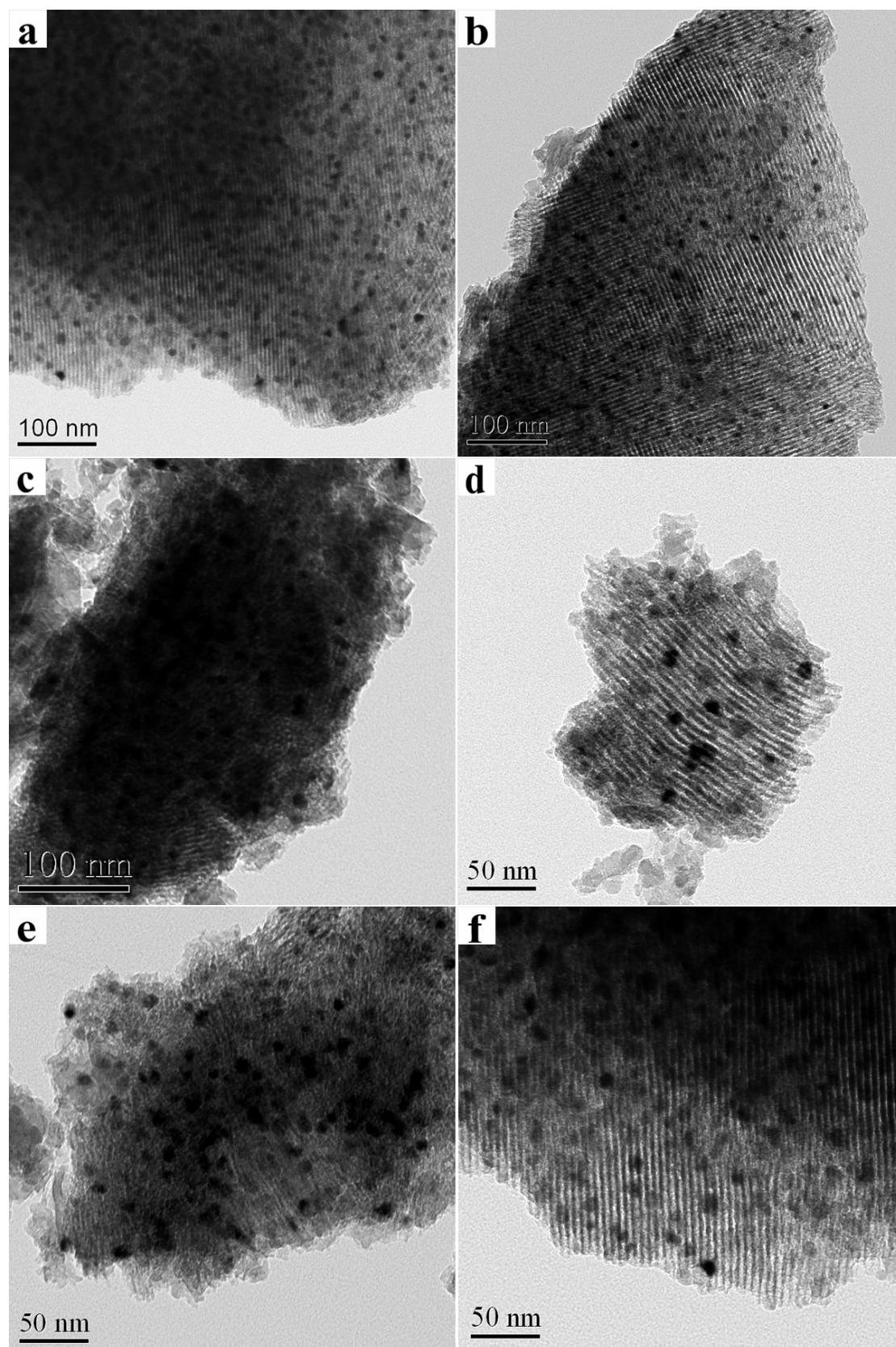


Figure S6 TEM images of meso-4CeZrAl calcined at 1000 °C.

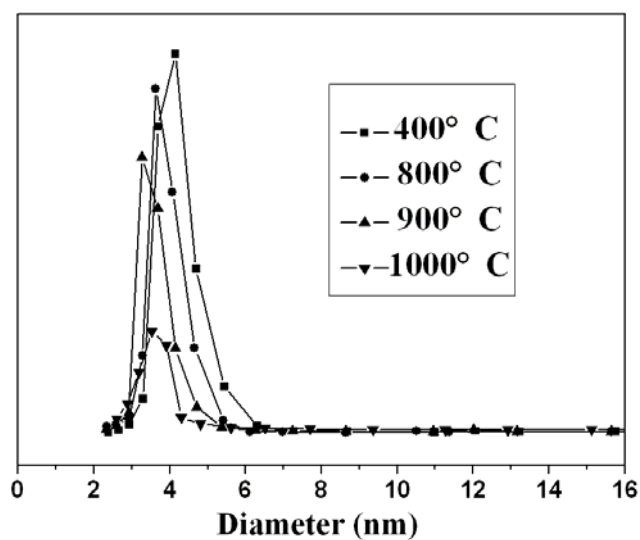


Figure S7 Pore-size distribution curves of meso-4CeZrAl calcined at different temperatures

Table S1: Textural parameters of calcined meso-4CeZrAl.

Calcined temperature (°C)	Surface area ( $S_{\text{BET}}$ :m <sup>2</sup> /g)	Pore volume ( $P_V$ :cm <sup>3</sup> /g)	Average pore diameter ( $D_V$ : nm)
400	284	0.44	4.4
800	240	0.33	3.9
900	210	0.28	3.9
1000	115	0.14	4.2