Supporting Information

The influence of H_2O or/and O_2 introduction during the low-temperature gas-phase sulfation of organic COS+CS_2 on the conversion and deposition of sulfur-containing species in the sulfated CeO_2-OS catalyst for NH_3-SCR

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Catalyst characterization

The BET specific surface area, pore diameter and pore volume of catalysts were determined by N_2 adsorption-desorption analysis at 77 K using a micromeritics ASAP 2460 system instrument. The crystal phase diffraction pattern of catalysts was identified on the 6100 X-ray diffraction Analyzer with Cu Ka radiation (model D/max RA, Rigaku Co., Japan). The scanning range (2θ) was collected from 10 ° to 80 ° at a scanning velocity of 5 °/min. The Raman spectra of samples were collected at a Raman Spectrometer (InVia Reflex, Renishaw), using a laser at 532 nm line as the excitation source. The surface morphology and structure of catalysts were observed by scanning electron microscopy (SEM) on a ZEISS SIGMA HD instrument. Transmission electron microscopy (TEM) was performed on a Thermo Fischer Talos F200x instrument with an operating voltage of 200 kV.

X-ray photoelectron spectroscopy (XPS) was performed to acquire the surface element information of the samples, which was conducted on ThermoFisher Scientific Escalab 250Xi using an Al Kα X-ray source with an excitation energy of 1486.7 eV. The energy calibration of all elements were calibrated by normalizing the C 1s line of adsorbed amorphous hydrocarbons to 284.8 eV, and the spectra was fitted by XPS PEAK software with Gaussian-Lorentz function.

H_2 temperature programmed reduction (H_2-TPR) and NH_3 temperature programmed desorption (NH_3-TPD) experiments were carried out on AutoChem II with TCD detection (Micromeritics, USA). For H_2-TPR, 100 mg samples were pretreated at 300 °C for 30 min in Ar

atmosphere and then cooled to 50 °C. Subsequently, the data were collected at a heating rate of 10 °C/min from 50 to 900 °C in a 10% H_2-Ar (40 mL/min) atmosphere. In NH_3-TPD, 100 mg samples were pretreated at 500 °C for 1 h in He atmosphere and then cooled to 35 °C. The sample was purged with 10% NH_3/He (50 mL/min) for 2 h at 35 °C, and then purged with high purity He for 1 h to remove the physical adsorption species. Finally, the desorption processes were initiated from 35 to 500 °C with a ramp rate of 10 °C/min.

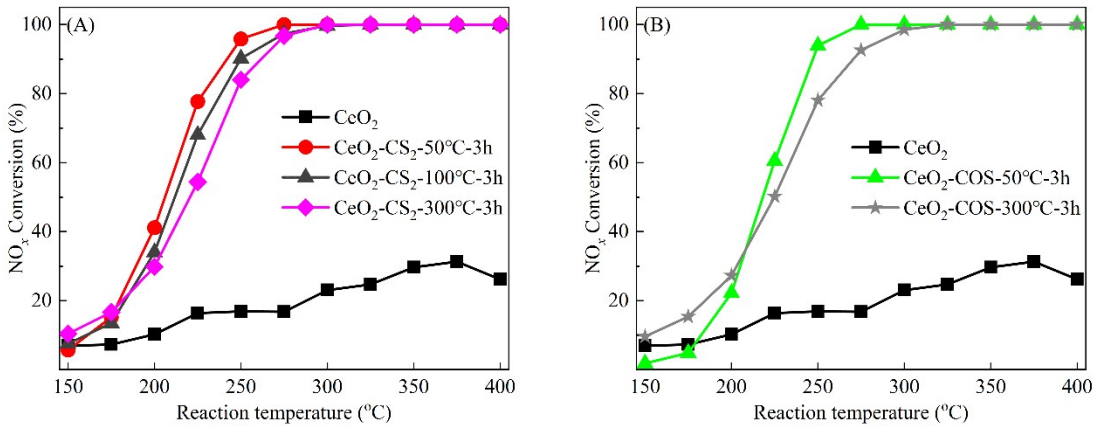

Fig. S1. Influence of sulfation temperature on the NH_3-SCR activity of the gas-phase sulfated CeO_2 catalysts by organic sulfur ((A) CS_2, (B) COS).

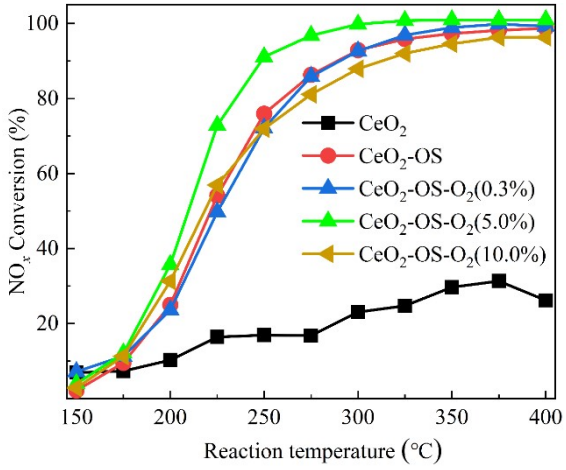

Fig. S2. Influence of oxygen concentration on the NH_3-SCR activity of the sulfated CeO_2-OS catalyst by the organic CS_2+COS at 50 °C. The simulated flue gas components during the sulfation: CS_2 30 ppm + COS 140 ppm (when used, COS:CS_2=7:3); O_2=0.3 vol.%, 5.0 vol.%, 10.0 vol.% (when used); pretreated at 50 °C for 3 h.

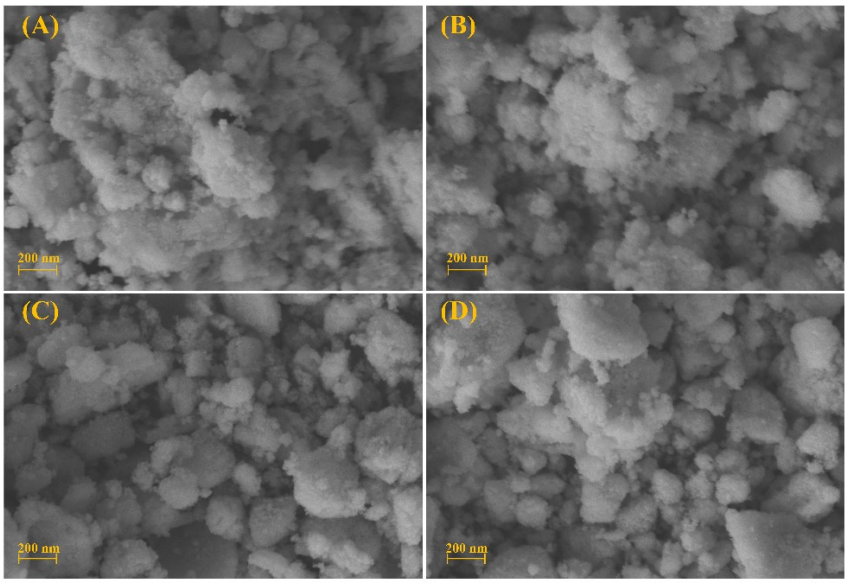

Fig. S3. The SEM images of (A) CeO_2-OS, (B) CeO_2-OS-O_2, (C) CeO_2-OS-H_2O and (D) CeO_2-OS-O_2+H_2O catalysts.

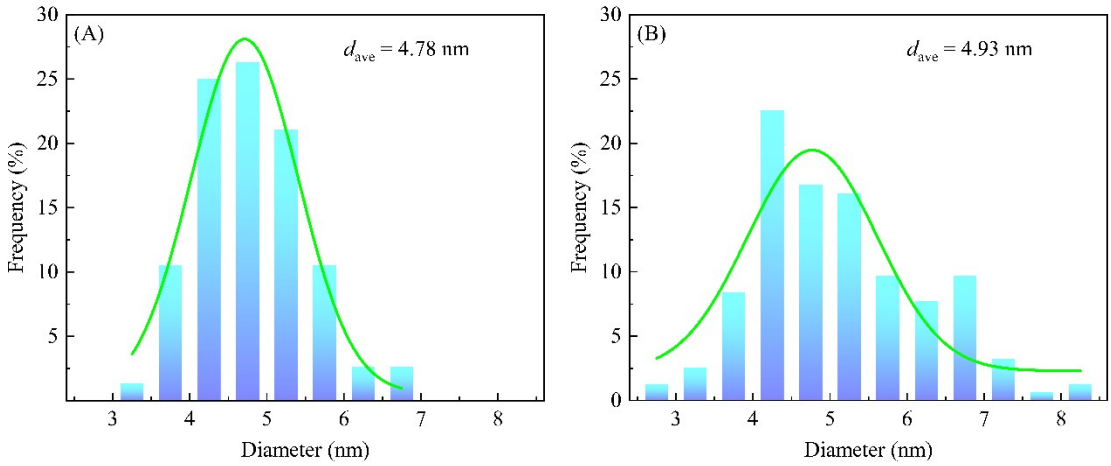

Fig. S4. Particle size distributions of CeO_2 and CeO_2-OS-O_2+H_2O catalysts.

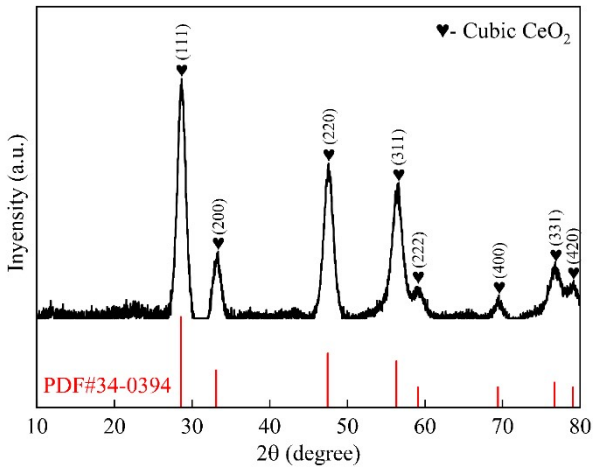

Fig. S5. The XRD patterns of the as-prepared CeO_2 catalyst.

Table S1. The chosen O_2 concentrations for the hydrolysis of organic COS/CS_2

Catalysts	O_2 concentrations	Optimal O_2 concentrations	GHSV	Ref.
Cu-Fe/TSAC	0%, 0.5%, 1%, 5%	0.5%	10000 /h	[1]
Cu-K-Co/AC	0%, 0.1%, 0.2%	0.1%	30000 /h	[2]
Al_2O_3-K/CAC	0%, 1.5%, 5.5%	1.5%	10000 /h	[3]
La_2O_2S, Nd_2O_2S	0%, 0.25%, 0.5%, 1%, 1.5%, 2%	0.5%, 1%, 1.5%	10000 /h	[4]
NaOH/Al_2O_3	0%, 1%, 2%, 3%	0%	220000 /h	[5]

Table S2. Hydrolysis conditions and properties of different catalysts for organic COS/CS_2

Catalysts	Test condition	t_{90} (min)	Ref.
$MgAlCeO_x$	COS=470 ppm, T=50 °C, GHSV=5000 /h	100	[6]
ZnAl-20Sm-Na MMO	COS=400 ppm, T=60 °C, WHSV=7500 mL/(g·h)	160	[7]
Ni-mAl_2O_3	COS=300 ppm, T=70 °C, GHSV=9000 /h	150	[8]
5%Fe/MCAC	COS=400 ppm, T=70 °C, GHSV=6000 /h	210	[9]
10-KA	COS=400 ppm, T=50 °C, GHSV=20000 /h	300	[10]
$K_{0.1}Al_2O_3$-PA	COS=200 ppm, T=50~150 °C, GHSV=24000 /h	-	[11]
K@Al	COS=150 ppm, CS_2=50 ppm, T=80~160 °C, GHSV=20000/h	-	[12]
Ni(5)ACF(400)	COS=400 ppm, CS_2=50 ppm, T=60 °C, GHSV=10000 /h	-	[13]
CSB-XXY	COS=450 ppm, CS_2=40 ppm, T=150 °C, GHSV=10000 /h	-	[14]

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