Supporting Information

Influence of Particle Size of CeO_2 Nanospheres Encapsulated in SBA-15 Mesopores on SO_2 Tolerance during NH_3-SCR Reaction

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1 Pore Structure of SBA-15

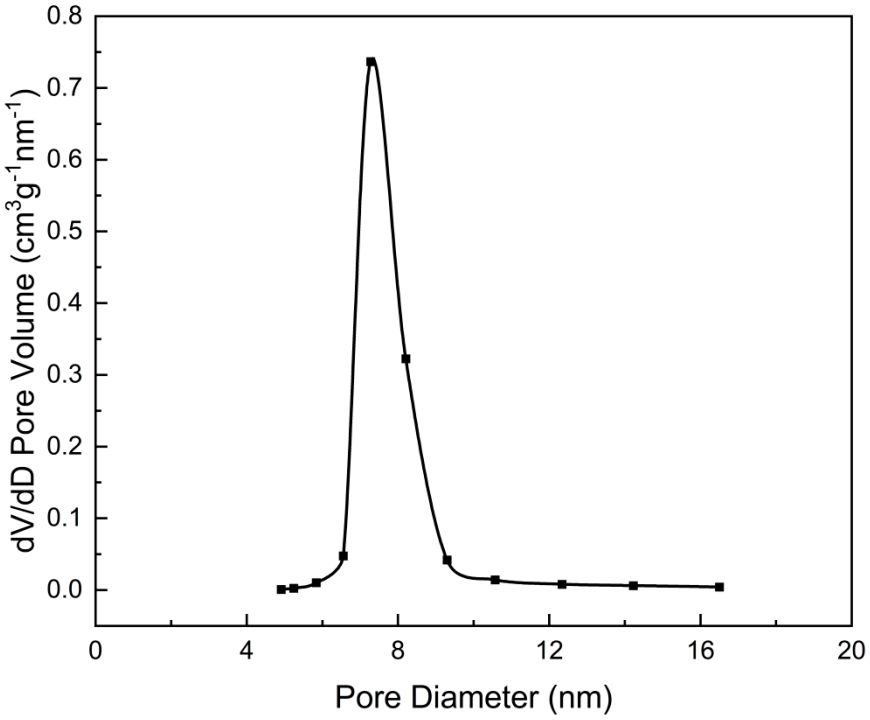

Fig S1. BJH pore distribution curves of pure SBA-15.

2 Particle size measured by laser particle size analyzer

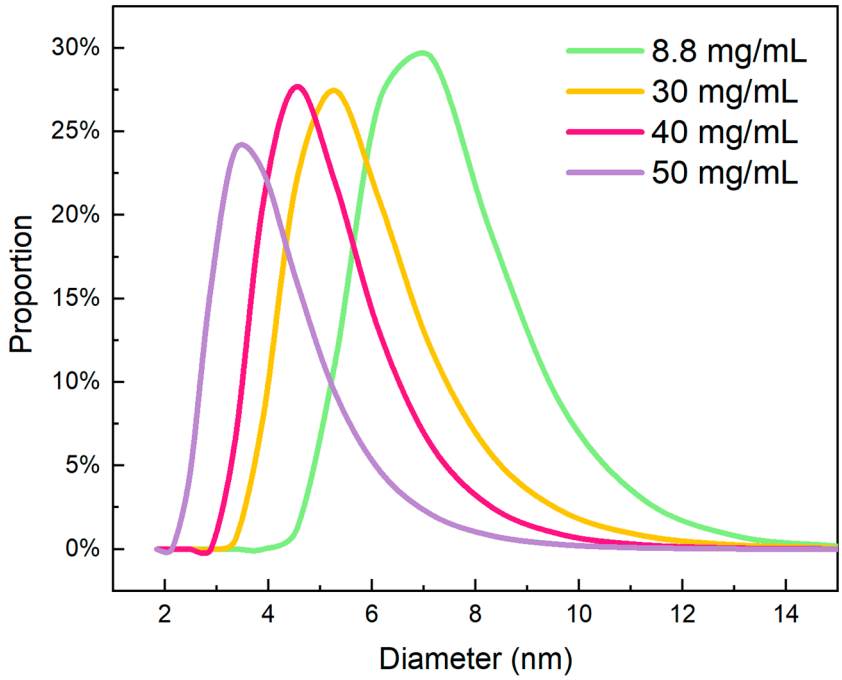

Fig S2. Particle size distribution curves of different PVP addition amounts.

Table S1. The amount of pvp added and the D_{10}, D_{50} and D_{90} size data obtained from the laser particle size analyzer.

PVP amounts in ethanol	D_{10} (nm)	D_{50} (nm)	D_{90} (nm)	Abbreviation
8.8 mg/mL	5.22	6.52	8.72	9 nm
30 mg/mL	4.06	4.96	7.02	7 nm
40 mg/mL	3.49	4.41	6.14	6 nm
50 mg/mL	2.71	3.55	5.20	5 nm

3 NH_3-SCR performance of different loading percentage of CeO_2 nanospheres on SBA-15

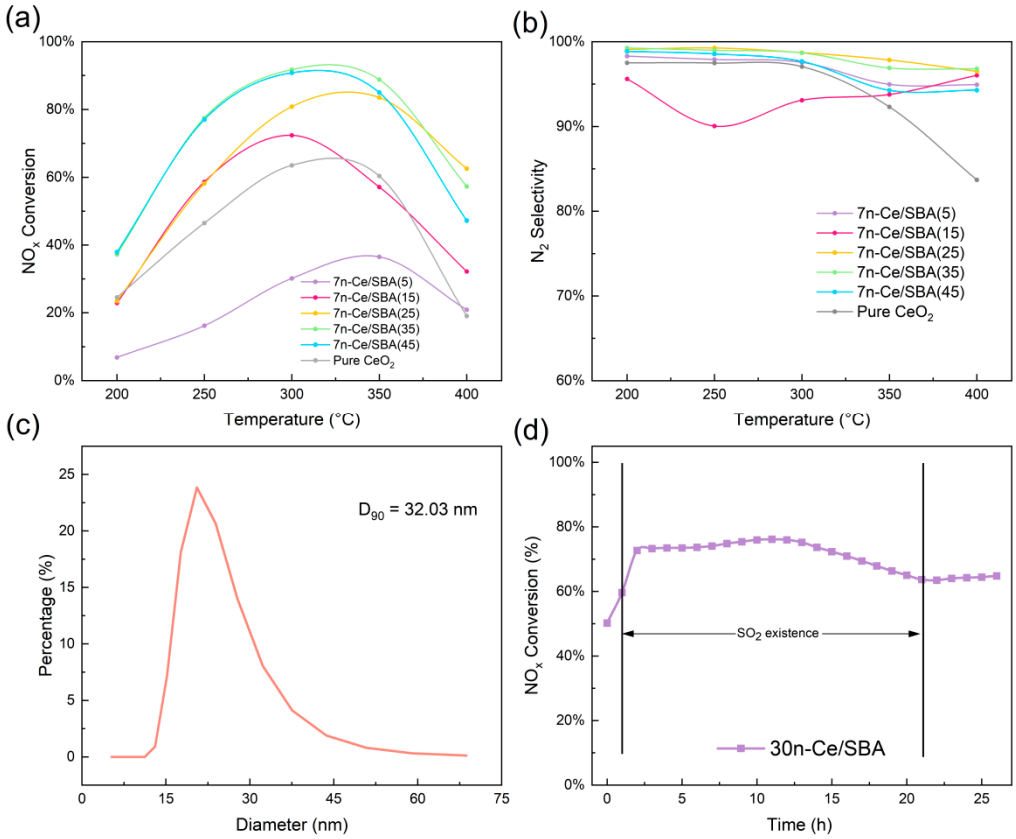

Fig S3. NO_x conversion (a) and N_2 selectivity (b) curves of five different loading percentages of catalysts and pure CeO_2. (c) particle size distribution curves of PVP addition amounts of 7 mg/mL and (d) NO_x conversion of 30n-Ce/SBA catalyst in 100 ppm SO_2 existing for 20 h.

4 EDS mapping of Ce/SBA-S series catalyst

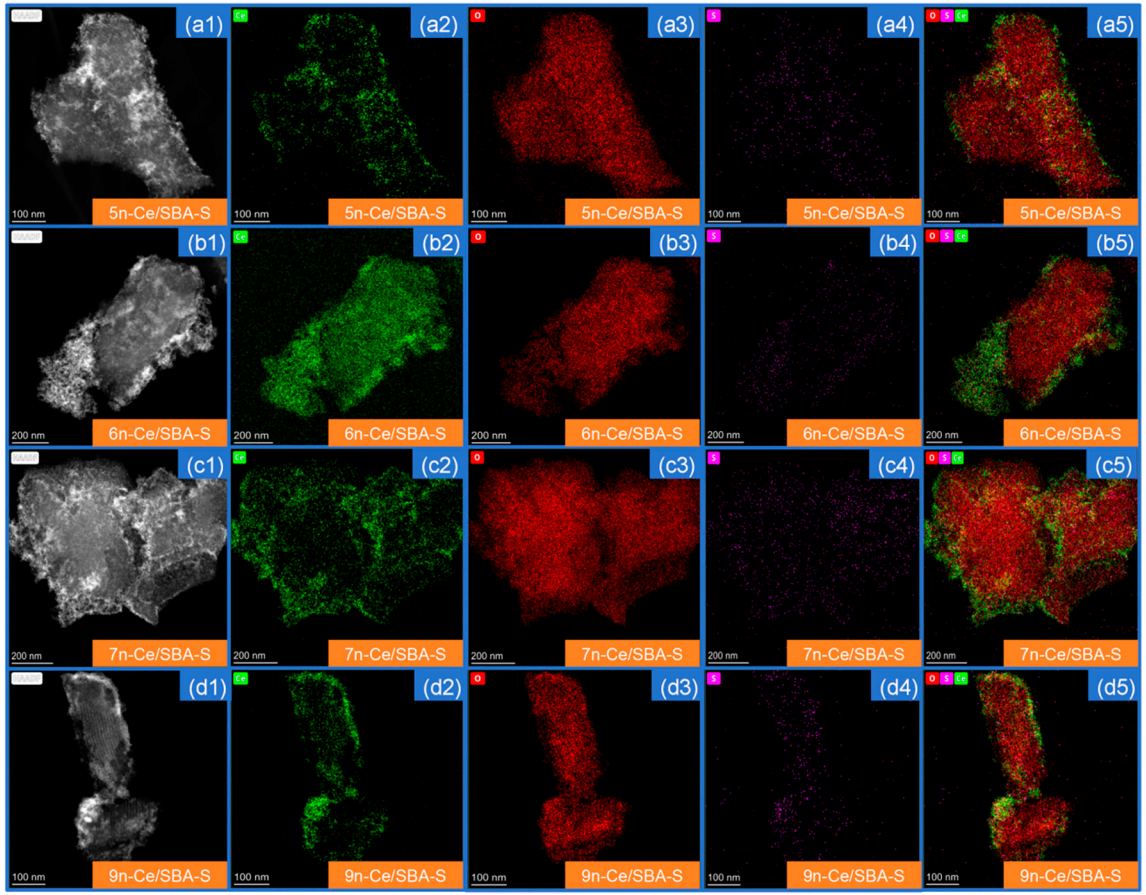

Fig. S4. EDS Mapping of Ce/SBA-S series catalysts.

5 NH_3 and H_2 consumption of catalysts

Table S2. The fitted NH_3-TPD data of catalysts.

Catalysts		5n-Ce/SBA	6n-Ce/SBA	7n-Ce/SBA	9n-Ce/SBA	9nm CeO_2	SBA-15
Total NH_3 consumption (mmol/g)		0.242	0.257	0.277	0.315	0.241	0.240
Peak 1	Temperature (°C)	95	94	94	98	92	99
	Area (%)	19.01	13.61	17.46	16.43	33.38	18.05
	NH_3 consumption (mmol/g)	0.046	0.035	0.048	0.052	0.080	0.043
Peak 2	Temperature (°C)	164	143	144	164	131	154
	Area (%)	48.89	25.80	34.34	43.49	22.62	13.66
	NH_3 consumption (mmol/g)	0.118	0.066	0.095	0.137	0.055	0.033
Peak 3	Temperature (°C)	281	254	274	292	206	330
	Area (%)	32.10	60.59	48.20	40.08	44.00	68.29
	NH_3 consumption (mmol/g)	0.078	0.156	0.134	0.126	0.106	0.164

Table S3. The fitted H_2-TPR data of catalysts.

Catalysts		5n-Ce/SBA	6n-Ce/SBA	7n-Ce/SBA	9n-Ce/SBA	9nm CeO_2
Total H_2 consumption (mmol/g)		1.023	0.882	1.225	1.144	1.391
Peak 1	Temperature (°C)	433	429	426	450	344
	Area (%)	74.89	59.31	69.78	79.16	20.11
	H_2 consumption (mmol/g)	0.766	0.523	0.855	0.906	0.280
Peak 2	Temperature (°C)	623	609	596	605	417
	Area (%)	23.37	40.20	23.49	10.93	37.25
	H_2 consumption (mmol/g)	0.239	0.355	0.288	0.125	0.518
Peak 3	Temperature (°C)	741	778	674	673	671
	Area (%)	1.74	0.49	6.73	9.91	42.64
	H_2 consumption (mmol/g)	0.018	0.004	0.082	0.113	0.593

6 XPS data of 30n-Ce/SBA-S

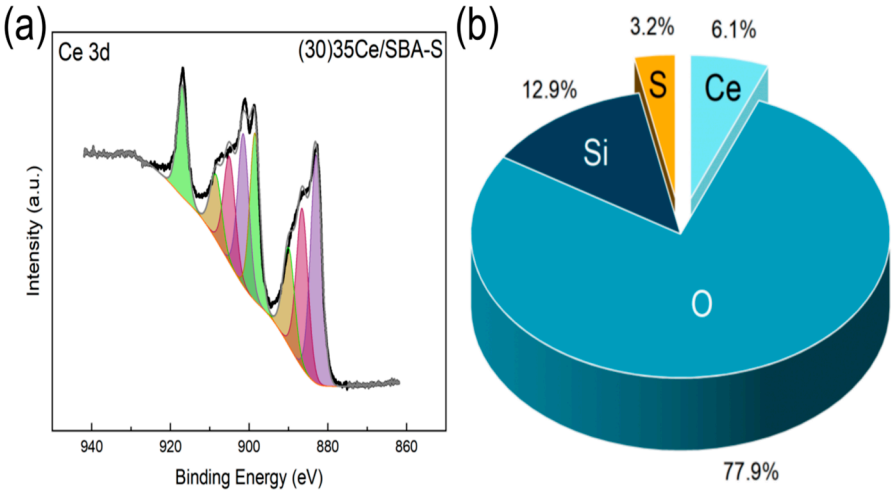

Fig. S5. (a) XPS spectrum of Ce 3d orbital of 30n-Ce/SBA-S and (b) content of elements on the surface of 30n-Ce/SBA-S from XPS results.

Table S4. The binding energy of the peak U of Ce 3d orbital of catalysts after SO_2 poisoning.

Catalyst	5n-Ce/SBA	6n-Ce/SBA	7n-Ce/SBA	9n-Ce/SBA	30n-Ce/SBA
Binding Energy (eV)	883.19	883.30	883.43	**883.50**	882.99

7 N_2 isotherm adsorption and desorption curves

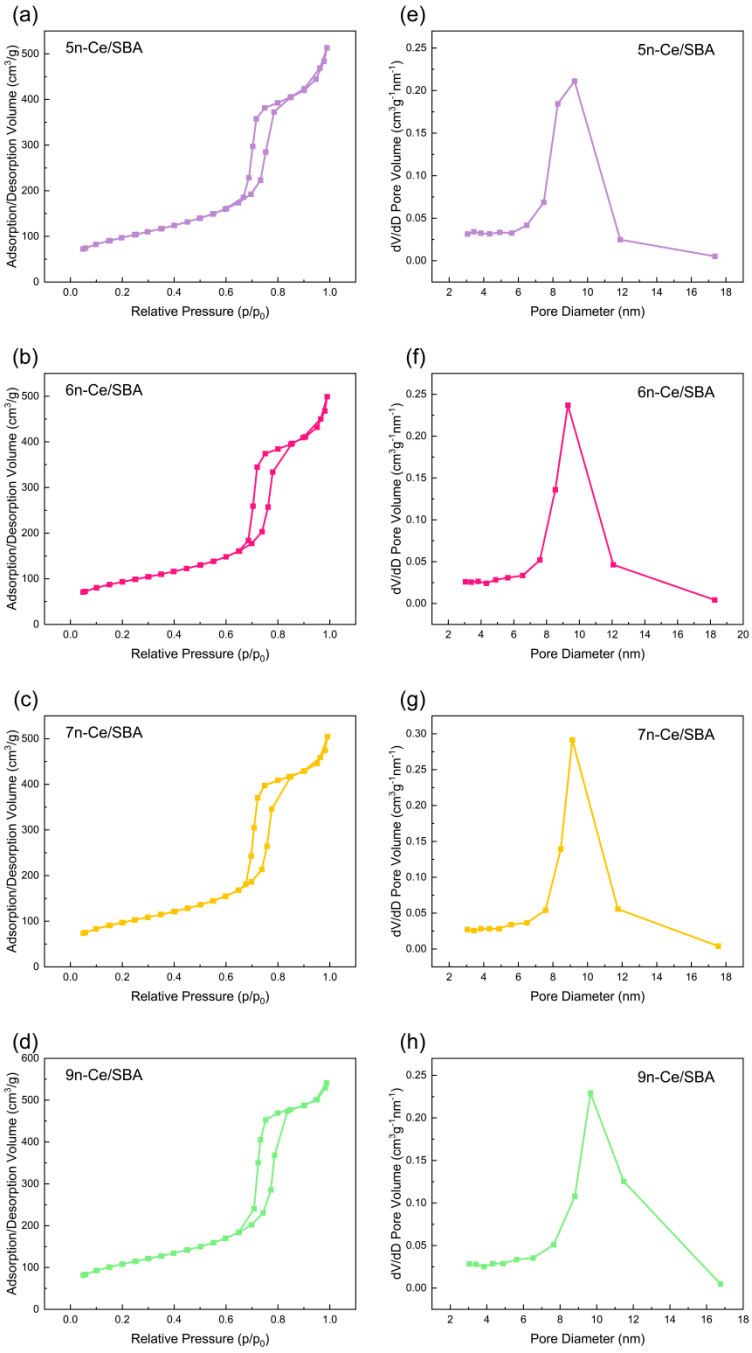

Fig. S6. N_2 adsorption-desorption curve and pore distribution curve of some catalysts.

Table S5. Specific surface area, average pore volume and pore size of catalysts.

Catalyst	BET specific surface area (m^2/g)	Average pore volume (cm^3/g)	Average pore size (nm)	BJH most probable pore size (nm)
5n-Ce/SBA	345	0.812	4.48	9.25
6n-Ce/SBA	324	0.782	4.76	9.30
7n-Ce/SBA	339	0.794	4.59	9.12
9n-Ce/SBA	378	0.845	4.58	9.66

8 Data of reaction order test

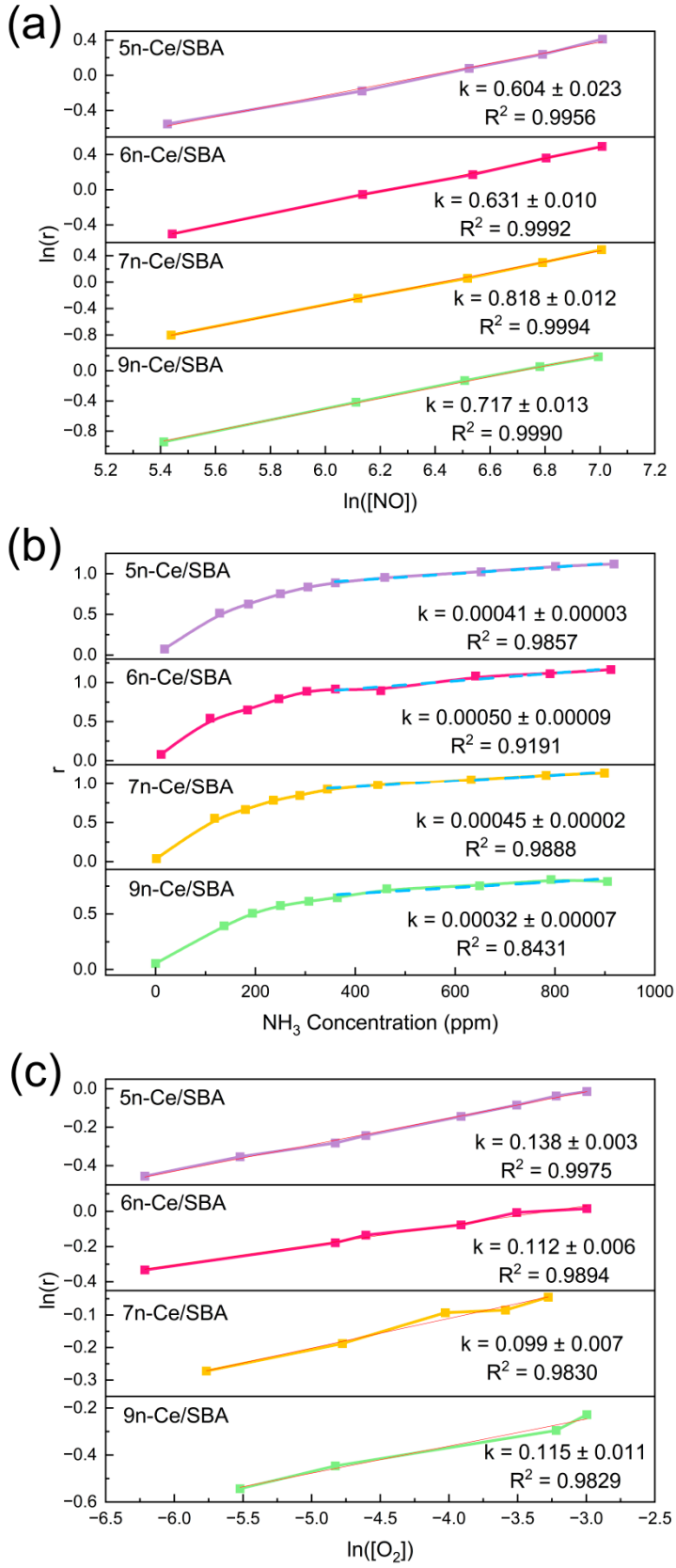

Fig. S7. Dependence of NO conversion rate upon (a) NO, (b) NH_3 and (c) O_2 concentration.

Derivation: How the reaction mechanism is inferred from the rate equation

(1) The Mathematical expression of chemical reaction based on L-H mechanism:

$$r = k\theta_A\theta_B = k\frac{a_A a_B P_A P_B}{1+a_A P_A+a_B p_B} \quad \text{(S1)}$$

(2) When the adsorption of component A is stronger than that of component B, it could be inferred $a_A P_A >> 1+a_B P_B$, so:

$$r = k\theta_A\theta_B \approx k_1\frac{a_B P_B}{a_A P_A} = k_2\frac{P_B}{P_A} \quad \text{(S2)}$$

At this point, the reaction order of A is -1, and the reaction order of B is 1.

(3) When both reactants A and B are weakly adsorbed, it could be inferred $a_A P_A+a_B P_B << 1$, so

$$r = k\theta_A\theta_B \approx k_3 a_A P_A a_B P_B = k_4 P_A P_B \quad \text{(S3)}$$

At this point, the reaction order of A and B are both 1.

(1) The Mathematical expression of chemical reaction based on E-R mechanism:

$$r = k\theta_A P_B = k\frac{a_A P_A P_B}{1+a_A P_A+a_B p_B} \quad \text{(S4)}$$

(2) When the adsorption of component A is stronger than that of component B, it could be inferred $a_A P_A >> 1+a_B P_B$, so:

$$r = k\theta_A P_B \approx k_5 P_B a_B = k_6 P_B \quad \text{(S5)}$$

At this point, the reaction order of A is 0, and the reaction order of B is 1.

(4) When both reactants A and B are weakly adsorbed, it could be inferred $a_A P_A+a_B P_B << 1$, so

$$r = k\theta_A P_B \approx k_6 P_A a_B P_B = k_7 P_A P_B \quad \text{(S6)}$$

At this point, the reaction order of A and B are both 1.

For the Ce/SBA catalysts in this work, part of NO participates in the reaction by L-H mechanism, so its reaction order is less than 0, while the rest of NO participates in the reaction by E-R mechanism, so its reaction order is 1. After weighted average, the resulting apparent reaction order of NO is between 0 and 1.

9 In-situ DRIFT spectra of the adsorption of NH_3 at different temperatures of 9n-Ce/SBA and 9n-Ce/SBA-S

Table S6. Materials corresponding to different wavenumbers and references.

Wavenumber (cm^{-1})	Species	Ref.
3300 peak groups	N-H bonds of Lewis Acid-NH_3 species	[1, 2]
1419, 1439, 1435	Bronsted Acid-NH^{4+} species	
1574	Symmetric and asymmetric bending vibrations of N−H bonds in the NH_3 coordinated to Lewis acid sites	[3]
1338, 1327	Oxidized species of NH_3	[4]
1284, 1277	Symmetric deformation of coordinated NH_3	
1103	NH_3 on Lewis acid sites over pure CeO_2	[5]
1535, 1539	Monodentate nitrate species	[6]
1578	Stretching vibration (v_3) of bidentate nitrate Species	
1273, 1277	Asymmetric vibration of NO_2^- species	
1817	Stretching vibration (v_3) of N-O bonds	
1381, 1385	v_3 vibration of free NO_3^- ion	
1315, 1304	Stretching vibration of M-NO_2 (nitro compound)	
1107, 1103	Cis-$N_2O_2^{2-}$ species on lanthanide oxide	

The catalyst was pretreated at 400 °C in 40 mL/min of Argon atmosphere for 30 min. Then 500 ppm NH_3-Ar was passed into the sample cell at 200 °C and the changes of IR spectrum were recorded for 15 min. Then sample cell was heated to 250 °C, 300 °C, 350 °C and 400 °C, during which time the changes in the IR spectra were continuously recorded. The IR spectra of the catalysts are plotted as **Fig. S8**.

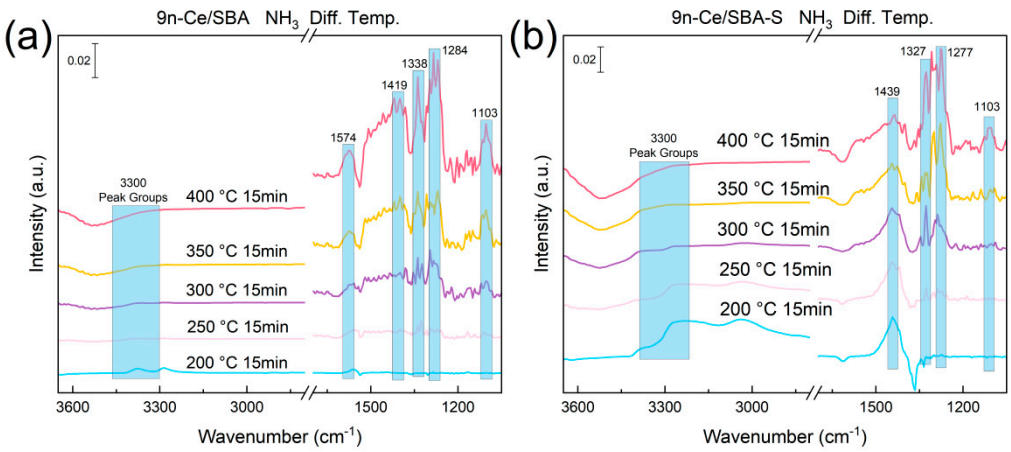

Fig. S8. In-situ DRIFT spectra of the adsorption of NH_3 at different temperatures of (a) 9n-Ce/SBA and (b) 9n-Ce/SBA-S.

10 In-situ DRIFT spectra of the adsorption of NO + O_2 at different temperatures of 9n-Ce/SBA and 9n-Ce/SBA-S

The catalyst was pretreated at 400 °C in 40 mL/min of Argon atmosphere for 30 min. Then 500 ppm NO-Ar and 5% O_2 were passed into the sample cell at 200 °C and the changes of IR spectrum were recorded for 15 min. Then sample cell was heated to 250 °C, 300 °C, 350 °C and 400 °C, during which time the changes in the IR spectra were continuously recorded. The IR spectra of the catalysts are plotted as **Fig. S9**.

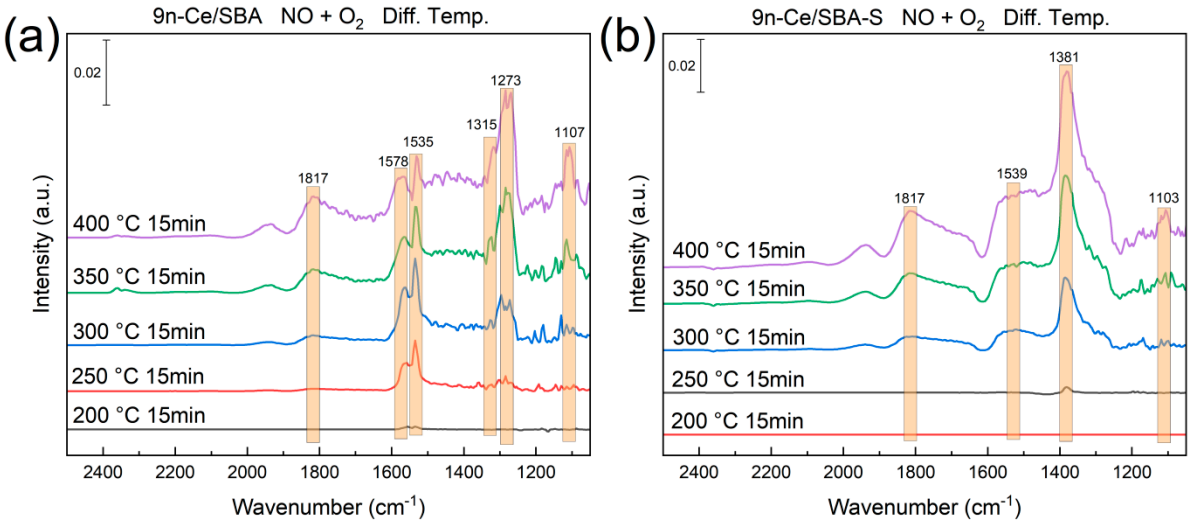

Fig. S9. In-situ DRIFT spectra of the adsorption of NO + O_2 at different temperatures of (a) 9n-Ce/SBA and (b) 9n-Ce/SBA-S.

11 In-situ DRIFT spectra of the reaction of NH_3, NO and O_2 at different temperatures of 9n-Ce/SBA and 9n-Ce/SBA-S

The catalyst was pretreated at 400 °C in 40 mL/min of Argon atmosphere for 30 min. Then 500 ppm NO-Ar, 500 ppm NH_3-Ar and 5% O_2 were then passed into the sample cell at 200 °C and the changes of IR spectrum were recorded for 15 min. Then sample cell was heated to 250 °C, 300 °C, 350 °C and 400 °C, during which time the changes in the IR spectra were continuously recorded. The IR spectra of the catalysts are plotted as **Fig. S10**.

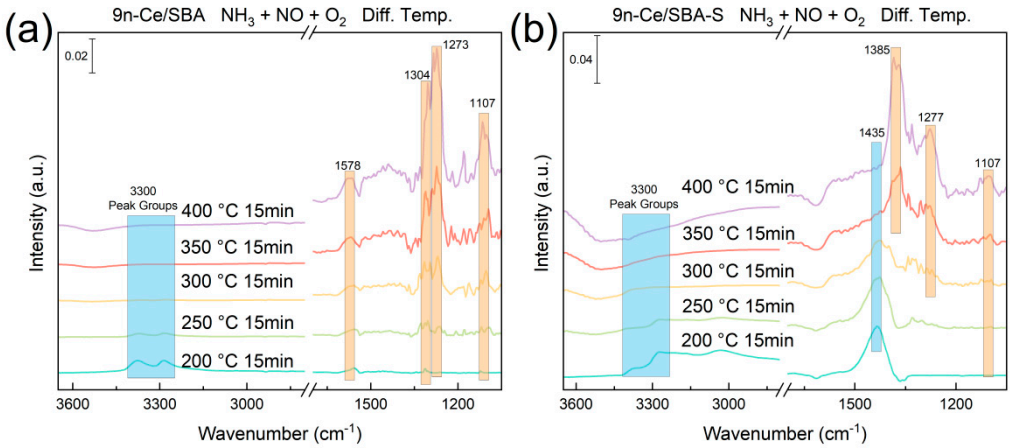

Fig. S10. In-situ DRIFT spectra of the reaction of NO, NH_3 and O_2 at different temperatures of (a) 9n-Ce/SBA and (b) 9n-Ce/SBA-S.