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Supplementary Information for

Promotional effects of Ascorbic acid on the low-temperature catalytic activity of selective catalytic oxidation of ammonia over Pt/SA: Effect of Pt^0 Content

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Experimental

Preparation of catalysts

A series of platinum-based catalysts were prepared with commercial SiO_2-Al_2O_3 (SiO_2 content of 30 wt. %) as support material and H_2PtCl_6 solution as precursor solution by equal water pore volume impregnation method. Specific operation steps are shown in Supplementary material.

For comparison, a monolithic catalyst labeled Pt/SA without ascorbic acid was prepared by the same method as described above.

Performance evaluation of catalysts

The reactions of NH_3-SCO over catalysts were evaluated in a self-assembled fixed-bed quartz-glass reactor at atmospheric pressure. The compositions of the inlet gases of the simulated diesel exhaust were as follow: 200 ppm NH_3, 10 % O_2, 8 % CO_2, N_2 as equilibrium gas, and the space velocity was 100,000 h^{-1}. The mixture gases flow rate was controlled by the mass flow controller. FT-IR (Antaris IGS-6700, Nicolet Thermo Fisher Scientific) gas analyzer was used to quantitatively analyze the gas composition before and after the reaction. Typically, 2.2mL monolithic catalyst was put into the self-assembled fixed-bed quartz-glass reactor and the reactor was performed in the temperature range of 150–500°C. Before the activity test, the catalyst was pretreated for 1 hour under 500°C reaction atmosphere to stabilize the catalyst, and then the transient temperature reduction test was carried out. The exhaust tailpipe is wrapped with a heating belt to prevent NH_3 adsorption and is controlled at 100 °C with a temperature controller. The NH_3 conversion and N_2 selectivity were

were calculated according to the following Eqs：

NH_3 conversion = $(NH_{3[in]}-NH_{3[out]})/NH_{3[in]}\times100\%$

N_2 selectivity=$(NH_{3[in]}-NH_{3[out]}-NO_{[out]}-NO_{2[out]}-2N_2O_{[out]})/(NH_{3[in]}-NH_{3[out]})\times100\%$

Characterization of catalysts

The texture properties of the samples were analyzed by N_2 adsorption-desorption at 77 K using QUANTACHROME SI automatic surface area and pore size analyzer. Before measurement, the catalyst was pretreated at 300°C vacuum for 3h, and the specific surface area and average pore diameter of the catalyst powder were calculated using Brunauer-Emmett-Teller (BET) and Barrett-Joyner-halenda (BJH) methods, respectively.

The H_2 temperature programmed reduction (H_2-TPR) of the catalyst was carried out in a self-assembled quartz tube reactor. The 0.10 g sample was first pretreated in a 25 ml/min N_2 gas stream at 450°C for 60 minutes, and then cooled to room temperature in the N_2 gas stream. Programmed reduction was performed at a heating rate of 8°C/min, a flow rate of 20 ml/min, and a mixture of 5vol.% H_2-N_2. Meanwhile, signal changes were recorded with a thermal conductivity detector (TCD).

The powder X-ray diffraction (XRD) patterns were collected on a Rigaku D/max-RA Diffractometer using Cu $K\alpha$ radiation (40 kV, 100 mA, λ = 0.15406 nm), and the sample was scanned in the range of 10-80°. The compound was identified by comparison with reference data from the International Centre for Diffraction Data (ICDD).

The X-ray photoelectron spectrum of the sample was carried out on an AXIS Ultra DLD (KRATOS) spectrometer using Al *K* α (1486.6 eV) as the radiation source. The experiment was carried out at 150 W by setting up an electronic energy analyzer.

The catalyst was tested by Tecnai G2 F20 S-TWIN transmission electron microscope (TEM, FEI Company, USA, and 200 kV) and high-resolution transmission electron microscope (HR-TEM). Before the test, the milled powder sample was placed in ethanol and dispersed by ultrasonic wave, and a small amount of liquid was dropped on the porous carbon film. The results were analyzed by digital micrograph software.

CO chemisorption was carried out in a quartz tube reactor. Prior to analysis, the sample (0.10 g) was pretreated with a hydrogen stream (H_2, 99.999%) at 500°C for 1 hour. The sample was then cooled to room temperature in the same reduction stream. Then, the Pt dispersibility of different catalysts was measured by pulsed CO. Due to the existence of linear and bridge adsorption, the calculated coefficient of exposed Pt atoms is 0.8 ×CO/Pt，according to the equations for D (dispersion) $=\frac{0.8 V_{co} M_{Pt}}{22.414 W P}$. Where V_{CO} was the amounts of CO assumption, and M_{Pt} was the Pt atomic weight (195 g/mol). W was the weight of catalyst, P was the metal content of Pt. Moreover, the average particle size was about 1.1/ D.

The In situ DRIFTS experiments were tested by a Fourier transform infrared spectrometer (Thermo Nicole 6700) using an MCT detector. The catalysts were pretreated with N_2 for 60 min at 450°C to remove the undesired species, and then collected the background spectra in flowing N_2 atmosphere. When cooled to the target temperature of 50°C, 200ppm NH_3 and excess O_2 were simultaneously introduced into the reaction chamber and the in situ DRIFT spectra were collected with the increasing temperature from 50-400°C.

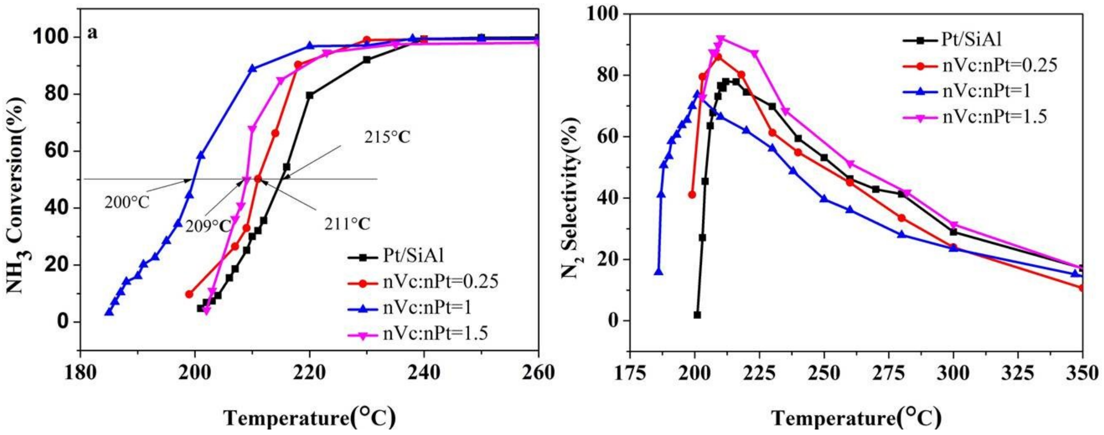

Fig.S1 (a) NH_3 conversion ;(b) N_2 selectivity over Pt/SA and Pt/SA-x samples. Experimental condition: 200 ppm NH_3, 10% O_2, 8% CO_2, 5%H_2O and balanced with N_2. The space velocity was 100,000 h^{-1}

Table.S1 textural parameters of catalysts

Sample	Surface area (m^2/g)	Pore volume(cm^3/g)	Average pore diameter(nm)
SiO_2-Al_2O_3	148	0.49	6.4
Pt/SA	146	0.51	6.8
Pt/SA-0.25	146	0.51	6.9
Pt/SA-1	143	0.50	6.8
Pt/SA-1.5	139	0.50	7.3

Table.S1 was the textural parameters of catalysts. The results of the textural properties of the catalyst were summarized in Table .S1. It could be seen from the table that all samples have similar and higher specific surface area. The specific surface area of the catalyst did not change much after the Pt species were impregnated on the support material, indicating that the Pt species can be well and uniformly dispersed on the surface of the support material without clogging the support pores. However, there was no obvious difference between all catalysts. Therefore, the texture properties could not be one of the main factors affecting the catalytic reaction.

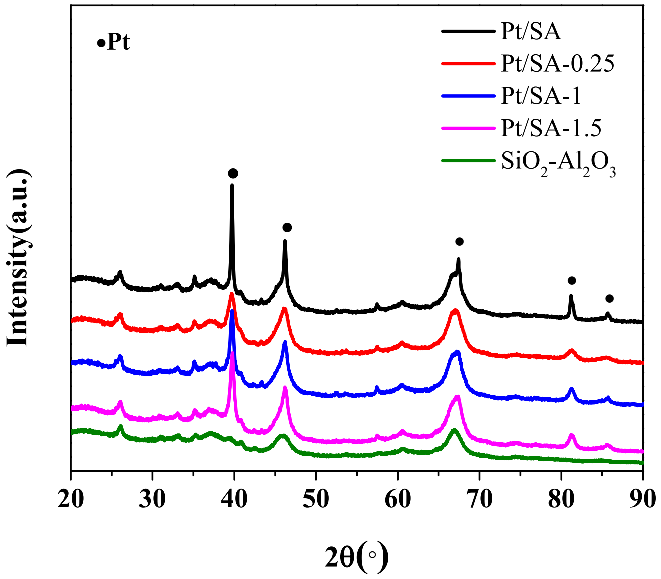

Fig.S2 XRD patterns of Pt/SA and Pt/SA-x catalysts with Cu Kα radiation (40kV, 25mA, λ=0.15406).XRD was performed to identify the crystalline phases presented in the Pt-based catalysts. Fig. S2 was the XRD results of the catalysts Pt/SA and Pt/SA-x with the test range of 20-90°. The four catalysts in the figure could all detected strong diffraction peaks of the support material, indicating that the addition of active species Pt and ascorbic acid did not damage the structure of the support material and maintains the integrity of the support material structure. Although some diffraction peaks of Pt coincide with SiO_2–Al_2O_3, the characteristic diffraction peaks attributed to Pt (111), Pt (311) and Pt (222) at 39.7°, 81.3°, 85.7° could still be clearly observed, indicating that all catalysts have highly active Pt^0 species. Compared with Pt/SA catalyst, the diffraction peak intensity of Pt^0 species in the catalyst with different content of ascorbic acid was decreased and the corresponding half-peak width increased, indicating that the addition of ascorbic acid can reduce the crystallization degree of the active species in the catalyst to a certain extent, so that there are more defect sites in the catalyst structure, thus providing more active sites and improving the reaction activity. However, ascorbic acid with different contents does not show any significant difference in lowering the crystallization degree of Pt^0 species, which may be due to the detection limit of the instrument.