Supplementary information

1.Preparation procedure of Pt-SAC and Pt-Nano catalysts

The FeO_x supported Pt single-atom catalyst was prepared by co-precipitation method.[S1] Under stirring at 50 ^oC, an aqueous solution of H_2PtCl_6 and $Fe(NO_3)_3$ was added dropwise to a Na_2CO_3 solution with the pH value of the resulting solution controlled at about 8. The recovered solid was dried at 60 ^oC overnight and then calcined at 400 ^oC for 6 h. Prior to being characterized and tested, the sample was reduced in 5% H_2/He at 250 ^oC for 0.5 h. The final sample was denoted as Pt-SAC.

The FeO_x supported Pt nano catalyst was prepared by the colloidal deposition method. The Pt nanoparticles were synthesized by the alkaline ethylene glycol method.[S2] The H_2PtCl_6 was dissolved in ethylene glycol (EG) and mixed with a solution of NaOH in EG at room temperature and then stirred at 140 °C for 3 h under an atmosphere of Ar. After that, a defined amount of the Fe_2O_3 support was added. The solution was stirred further for 3 h. The obtained sample was dried under vacuum overnight and then calcined at 400 ^oC for 6 h. Prior to being characterized and tested, the sample was reduced in 5% H_2/He at 250 ^oC for 0.5 h. The final sample was denoted as Pt-Nano.

2. Measurements of catalytic activities

Catalytic activity measurements were carried out in a fixed-bed reactor with 100 mg of a catalyst. The feed gases for the reaction of NO with H_2 were 2000 ppm NO+ 2000 ppm H_2 balanced with He. Blank experiments showed no activity in the temperature range of 40-300 °C. The gas flow rate was 100 mL min^{-1} which resulted in a space velocity of 60,000 mL h^{-1} g_{cat}^{-1}. Before evaluation, the catalyst sample was reduced in a flow of 20 mL min^{-1} of 5 vol% H_2/He at 250 ^oC for 30 min (for the support, the reduction temperature was chosen at 400 ^oC). The concentrations of NO, N_2 and N_2O in the effluent gas were analyzed by an on-line gas chromatograph (Agilent 6890, Porapak Q column) using He as the carrier gas. For the H_2-SCR detect, the standard feed was composed of 2000 ppm NO, 1% H_2, 1% O_2, balanced with He. In some cases, the 3% H_2O or 10 ppm SO_2 was added. Under these conditions, the GC equipped with a thermal conductivity detector was used for analyzing the products, column A with Porapak Q was chosen for analyzing N_2O, and column B with 13x molecular sieve (40-60 M) was used for separating H_2, N_2, O_2 and NO. The NO concentration was also monitored using a chemiluminescene NO_x analyzer (Infralyst ELD_L). From the mass spectroscopy signals (as shown in Fig. S6), the main products were N_2 and N_2O. Thus, the conversion of NO was calculated with theequation (1):

$$\mathrm{X}_{\mathrm{NO}} = \frac{n_{NO}^{in} - n_{NO}^{out}}{n_{NO}^{in}} \qquad (1)$$

The n_{NO}^{in} is the inlet amount of NO (mol). The n_{NO}^{out} is the outlet amount of NO (mol).

The selectivity toward N_2 was calculated with the equation (2):

$$\mathrm{S}_{\mathrm{N2}} = \frac{n_{\mathrm{N}2}}{n_{N2} + n_{N2O}} \qquad (2)$$

The n_{N2} is the amount of N_2 produced (mol). The n_{N2O} is the amount of N_2O produced (mol).

Specific reaction rates and turnover frequencies (TOF) of Pt-SAC and Pt-Nano were compared at 200 ^oC. For each run, the NO conversions were averaged and used for calculations of the specific rate. The TOF was then calculated based on the specific rate and the dispersion, which was measured by CO chemisorption at 40 ^oC with the assumption of the stoichiometric ratio of adsorbed CO/Pt=1. For Pt-SAC, Pt single atoms were considered as complete dispersion on FeO_x.

3. Characterization techniques

The Pt loadings in the catalyst samples were determined by inductively coupled plasma spectrometer (ICP-AES) on an IRIS Intrepid II XSP instrument (Thermo Electron Corporation).The Cl amounts were detected by XRF characterization. The Cl amounts were smaller than 0.02 wt% and can be neglected.

X-ray Diffraction (XRD) patterns were recorded on a PW3040/60 X' Pert PRO (PANalytical) diffractometer equipped with a Cu Kα radiation source (λ=0.15432 nm), operating at 40 kV and 40 mA. A continuous modewas used for collecting data in the 2θ range from 20° to 80° at a scanning speed of 10° min^{-1}.

High-angle annual dark-filed scanning transmission electron microscopy (HAADF-STEM) images were obtained on a JEOL JEM-ARM200F equipped with a CEOS probe corrector, with a guaranteed resolution of0.08 nm. Before microscopy examination, the samples after H_2 treatment were suspended in ethanol with an ultrasonic dispersionfor 5-10 minutes and then a drop of the resulting solution was dropped on a holey carbon film supported by a copper TEM grid.

H_2 temperature-programmed reduction (H_2-TPR) was performed on an Auto Chem II 2920 automatic catalyst characterization system. First, 50 mg of a catalyst was loaded into a U-shape quartz reactor and purged with He at 120 ^oC for 2 h to remove adsorbed carbonates and hydrates. Then, after cooling to room temperature, the flowing gas was switched to a 10 vol% H_2/Ar, and the catalyst was heated to 900 ^oC at a ramping rate of 10^oC min^{-1}.

The NO temperature-programmed desorption (NO-TPD) experiment with on-line mass spectroscopy (MS) analysis were also carried out on Auto Chem II 2920 automatic catalyst characterization system. The Pt-SAC and Pt-Nano samples were reduced in situ with H_2 and then submitted to pulses of NO at 50 ^oC until adsorption saturation. Then the catalyst was heated to 900 ^oC under He gas flow at a ramping rate of 10 ^oC min^{-1}. The effluent gases of NO, N_2 and N_2O were analyzed by MS.

A Temperature-programmed surface reaction (TPSR) experiment with on-line mass spectroscopy (MS) analysis was carried out as following: a 100 mg catalyst was reduced in situ with 5% H_2/He at 200 ^oC for 0.5 h, and purged with He for 1 h. Then the reaction gas at a flow rate of 30 mL min^{-1} was introduced and the effluent gases of NO, N_2 and N_2O were analyzed by MS. The gate time for MS analysis was 0.1 s for each detected component (NO, N_2, N_2O, NH_3), equivalent to the acquisition of 2 data point per second.

References:

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Table S1 Specific rates and turnover frequencies (TOF) of Pt-SAC and Pt-Nano catalysts compared with other catalyst in the reduction of NO by H_2.

	Pt loadings (wt%)	Reaction conditions	T (^oC)	Specific rate ($mol_{NO}\ g_{Pt}^{-1}\ h^{-1}$)	TOF_{NO} (s^{-1})	Note
Pt-SAC	0.06	2000 ppm NO+2000 ppm H_2	200	9.46	0.51	This work
Pt-Nano	1.22	2000 ppm NO+2000 ppm H_2	200	1.56	0.23	This work
Pt-SAC	0.06	2000 ppm NO+1% H_2+1% O_2	200	4.15	0.22	This work
Pt/SiO_2	1.0	500 ppm NO+2000 ppm H_2 +6% O_2	140	0.055	0.032	Ref. S3
Pt/TiO_2	1.0	1000 ppm NO+3000 ppm H_2 +5% O_2+10% H_2O	140	0.016	0.004	Ref. S3
Pt/MnO_x	2.0	480 ppm NO+8000 ppm H_2+5% O_2	200	0.024	-	Ref. S4
$Pt/MgO-CeO_2$	0.1	2500 ppm NO+1% H_2	200	4.32	0.28	Ref. S5
$Pt/MgO-CeO_2$	0.1	2500 ppm NO+1% H_2+5% O_2	200	3.24	0.21	Ref. S5

Table S2 The NO conversion and N_2 selectivity over Pt-SAC and the reported Pt or other transition metal based catalysts.

	Metal loadings (wt%)	Reaction conditions	Space velocity (mL g_{cat}^{-1} h^{-1})	T (^oC)	NO reduction efficiency		Note
					NO conversion (%)	N_2 selectivity (%)	
Pt-SAC	0.06	2000 ppm NO+2000 ppm H_2	60000	200	96	71	This work
Pt-SAC	0.06	2000 ppm NO+2000 ppm H_2	60000	280	100	100	This work
Pt-SAC	0.06	2000 ppm NO+1% H_2 +1% O_2	60000	240	50	58	This work
Pt/MnO_x	2.0	480 ppm NO+0.8% H_2	100000	200	95	8	Ref. S4
Pt/MgO-CeO_2	0.1	150 ppm NO+0.7% H_2	33000[a]	180	100	92	Ref. S6
Pt/MFI	1.0	1000 ppm NO+5000 ppm H_2 +6.7% O_2	120000	75	37	45	Ref. S7
Pt/Al_2O_3	1.0	500 ppm NO+0.2% H_2+6% O_2	120000	140	50	50	Ref. S8
Pt/CeO_2	1.0	1000 ppm NO+0.4% H_2+10% O_2	12000	100	80	40	Ref. S9
Pt/MgO-CeO_2	0.1	2500 ppm NO+1% H_2+5% O_2	80000[a]	150	94	80	Ref. S10
Pt/$LaCoO_3$	1.0	10000 ppm NO+10000 ppm H_2	100000	205	~90	-	Ref. S11
Pd/K_2O-TiO_2	1.0	1000 ppm NO+0.6% H_2+5% O_2	60000	164	41	76	Ref. S12
Ir/SiO_2	0.5	1000 ppm NO+0.3% H_2	134831	500	98	98	Ref. S13
Ir/SiO_2	0.5	1000 ppm NO+0.3% H_2+0.65% O_2+20 ppm SO_2+10% H_2O	134831	300	73	61	Ref. S13
Rh-Zn/SiO_2	5.0	1000 ppm NO+0.6% H_2+5% O_2+20 ppm SO_2+6% H_2O	134831	300	55	86	Ref. S14

[a]the unit is h^{-1}

Table S3 Reducibility of Pt-SAC and Pt-Nano by H_2-TPR

	Metal loading (wt%)	H_2 consumed (μmol g_{cat}^{-1})	
		Peak (T/°C)[a]	FeO_x/Pt[b]
FeO_x	-	1421(361)	-
Pt-SAC	0.06	1700(253)	275
Pt-Nano	1.22	1426(227)	30

[a] The central reduction temperatures and H_2 consumption amounts.

[b] The ratio of H_2 consumption amount for surface FeO_x to surface Pt species reduction. It can reflect the reducibility of support promoted by Pt atoms.

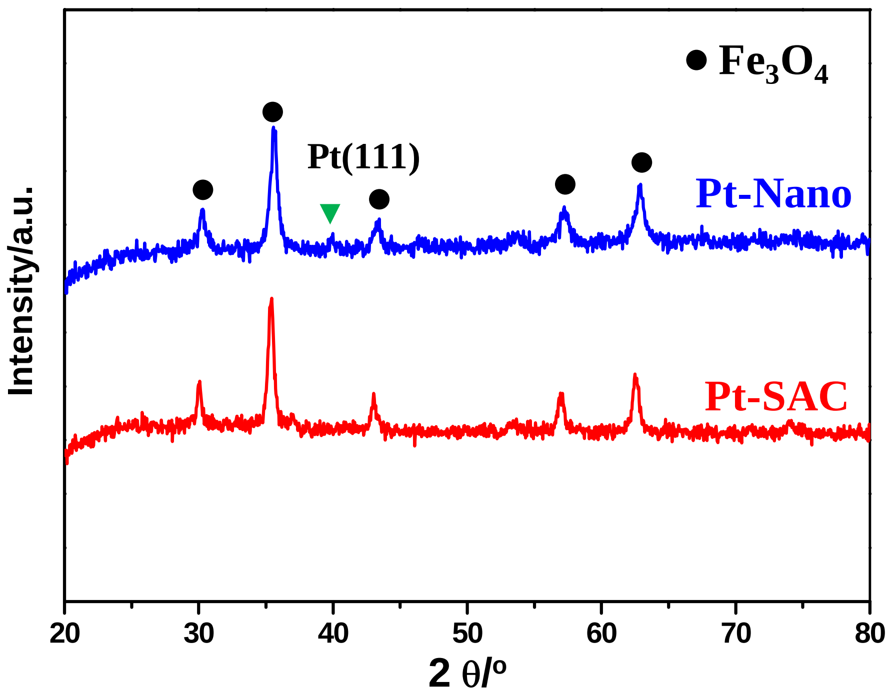

Fig. S1 XRD patterns of the Pt-SAC and Pt-Nano catalysts.

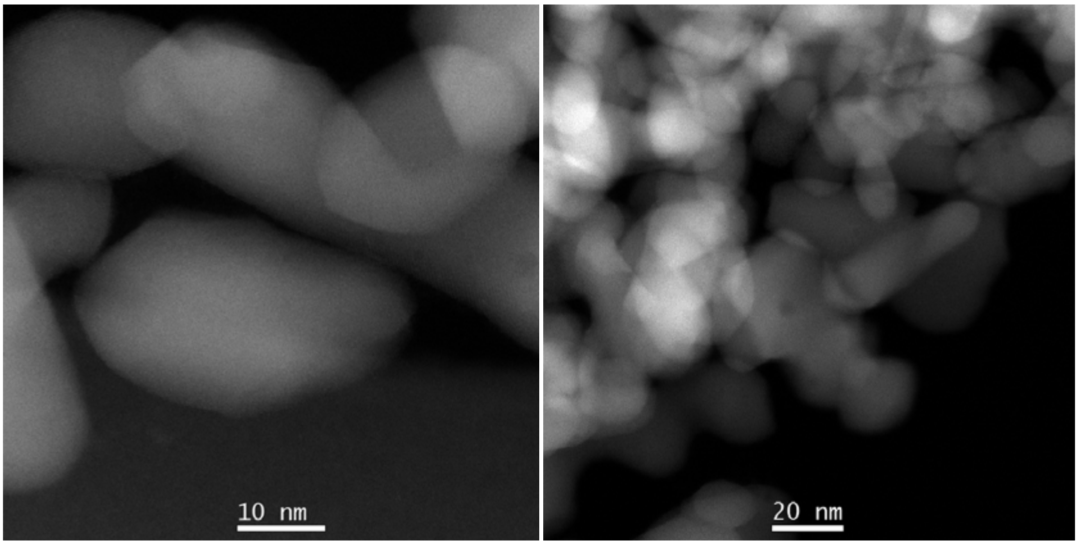

Fig. S2 HAADF-STEM images of Pt-SAC with different magnifications.

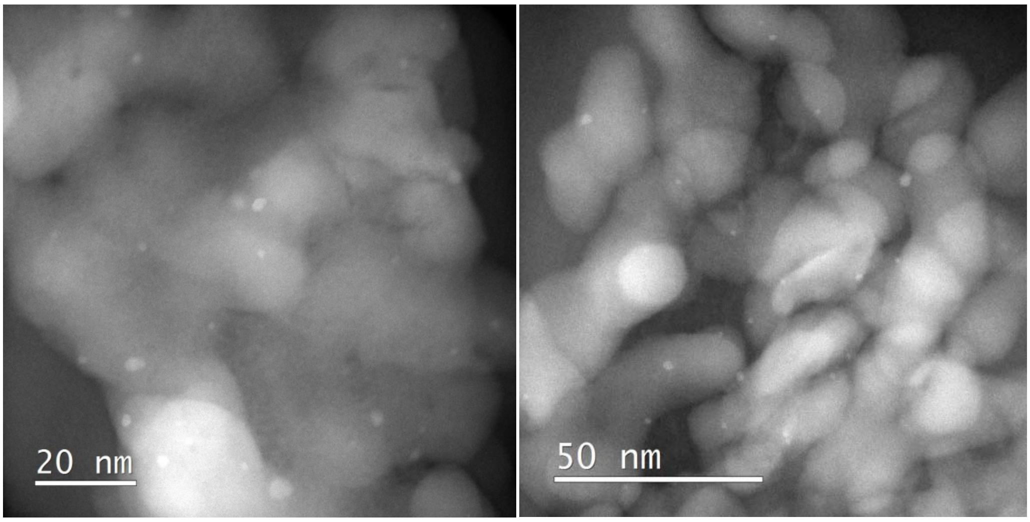

Fig. S3 HAADF-STEM images of Pt-Nano with different magnifications.

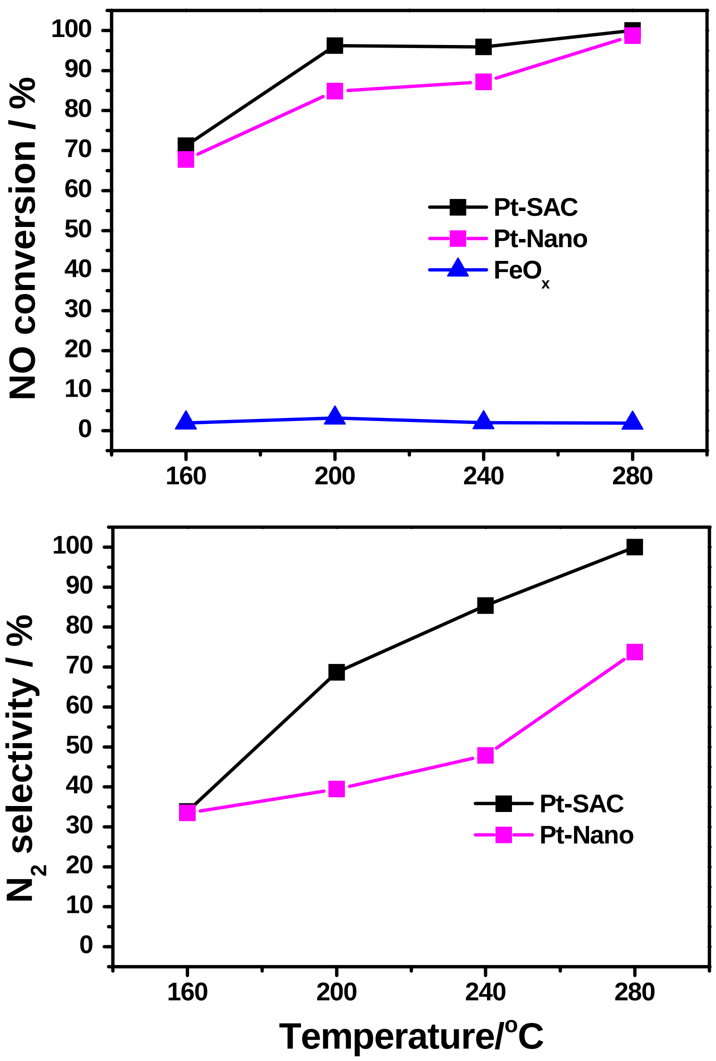

Fig. S4 NO conversion and N_2 selectivity with the temperatures over different catalysts. Reaction condition: 2000 ppm NO, 2000 ppm H_2, and balance He. Weight hourly space velocity (WHSV): 60,000 mL h^{-1} g_{cat}^{-1}.

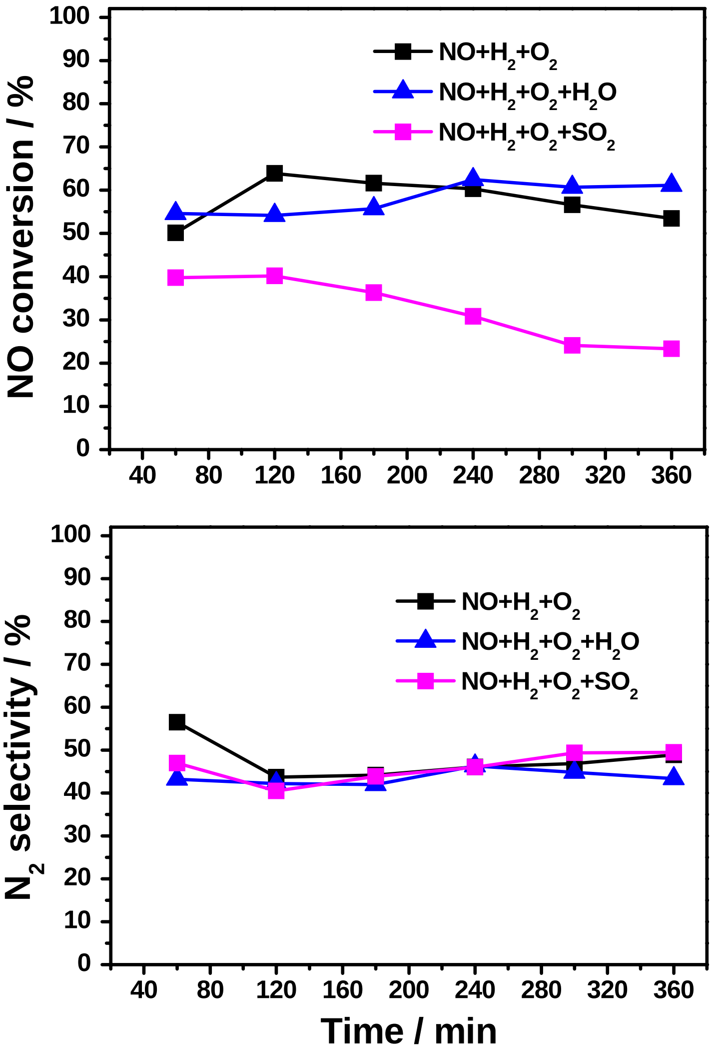

Fig. S5 NO conversion and N_2 selectivity of Pt-SAC under different reaction conditions at 240 °C. Reaction conditions: 2000 ppm NO + 1% H_2+ 1% O_2, with or without 3% H_2O or 10 ppm SO_2, balance He, Weight hourly space velocity (WHSV) = 60,000 mL g_{cat}^{-1} h^{-1}.

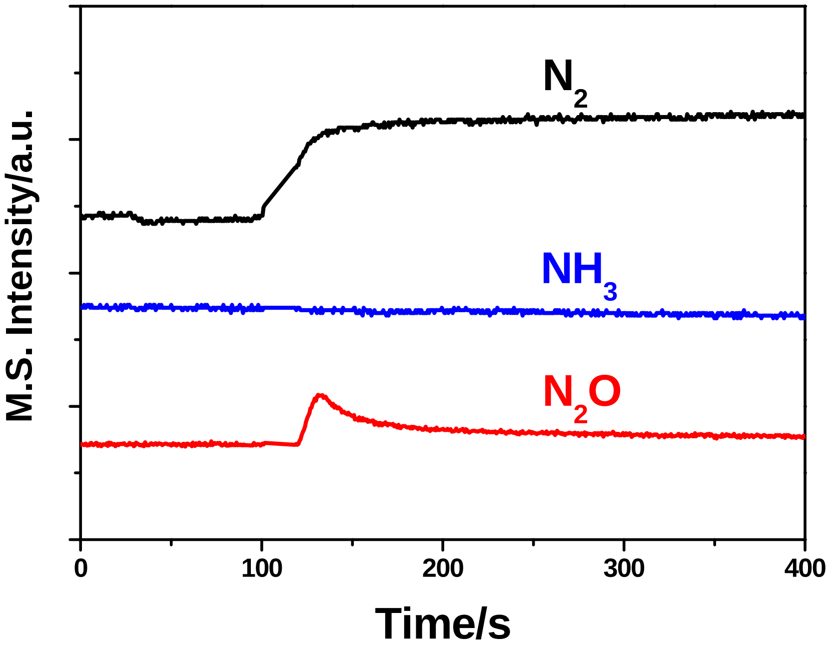

Fig. S6 MS analysis of product after introducing NO+H_2 onto the Pt-SAC catalyst.

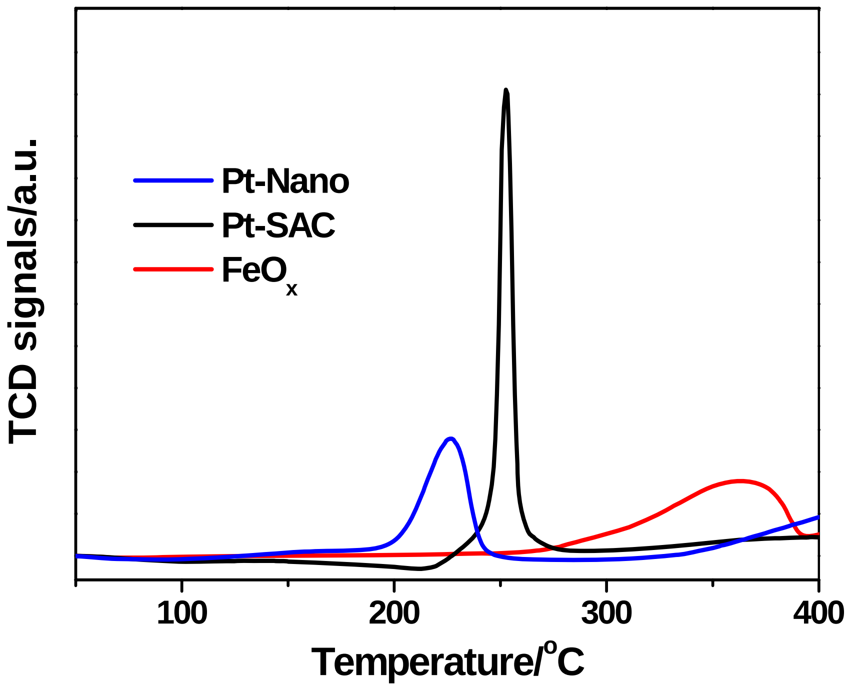

Fig. S7 H_2-TPR results of FeO$_x$ support, Pt-SAC and Pt-Nano samples.

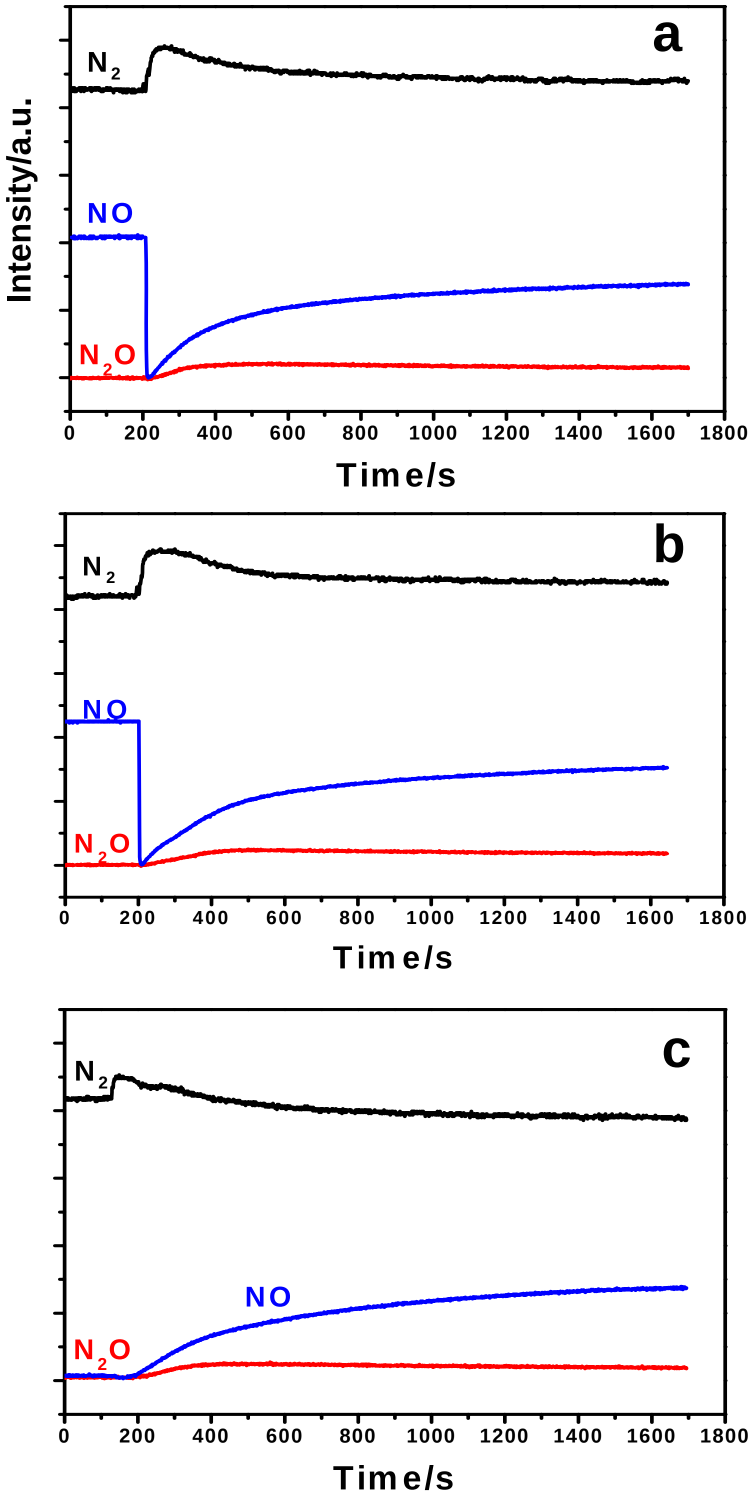

Fig. S8 TPSR results of NO+H_2 reaction on the Pt-SAC catalyst at 200 °C. (a) the freshly reduced Pt-SAC under 2000 ppm NO with a flow rate of 30 mL min^{-1}; (b) The Pt-SAC after reaction in (a) was reduced at 200 °C for 30 min and then resubmitted to NO; (c) The Pt-SAC after reaction in (b) was submitted to NO+H_2 for 0.5 h and then H_2 was removed.