Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supporting information

TG-MS: Gas flow rate is 25ml/min, nitrogen is protective gas, O_2/N_2 (25% O_2) is reaction gas, the ratio of the two is 2/3. The heating rate was 10 °C/min. No organic ion fragments were found during the test, the external standard method determines the amount of CO_2 produced by the catalyst carbon deposits oxidation reaction, and then determines the carbon mass percentage in catalyst (wt%, carbon elementary substance). Carbon deposits (mmol, carbon elementary substance) in Table S1 is the amount of carbon elementary substance.

The fresh catalysts were used to catalyzed by a toluene combustion reaction. The reactions were continued for 12 hours. The toluene elimination rates were> 99.5% (reaction conditions: toluene concentration is 1000 mg/m^3; GHSV is 5000 h^{-1}; toluene elimination rate> 99.5%; reaction time is 12h). Only residual toluene and CO_2 were detected in the reaction exhaust gas. The carbon deposition of the catalysts after the reaction were measured by TG-MS. Finally, using the CO_2 selectivity were calculated by the results of TG-MS to verify the accuracy of using the CO_2 concentration in the reaction exhaust gas to determine the carbon deposition in the article. The results were shown in Fig. S1 and Table S1. The results show that the carbon balance calculation in the article is correct and the method is feasible.

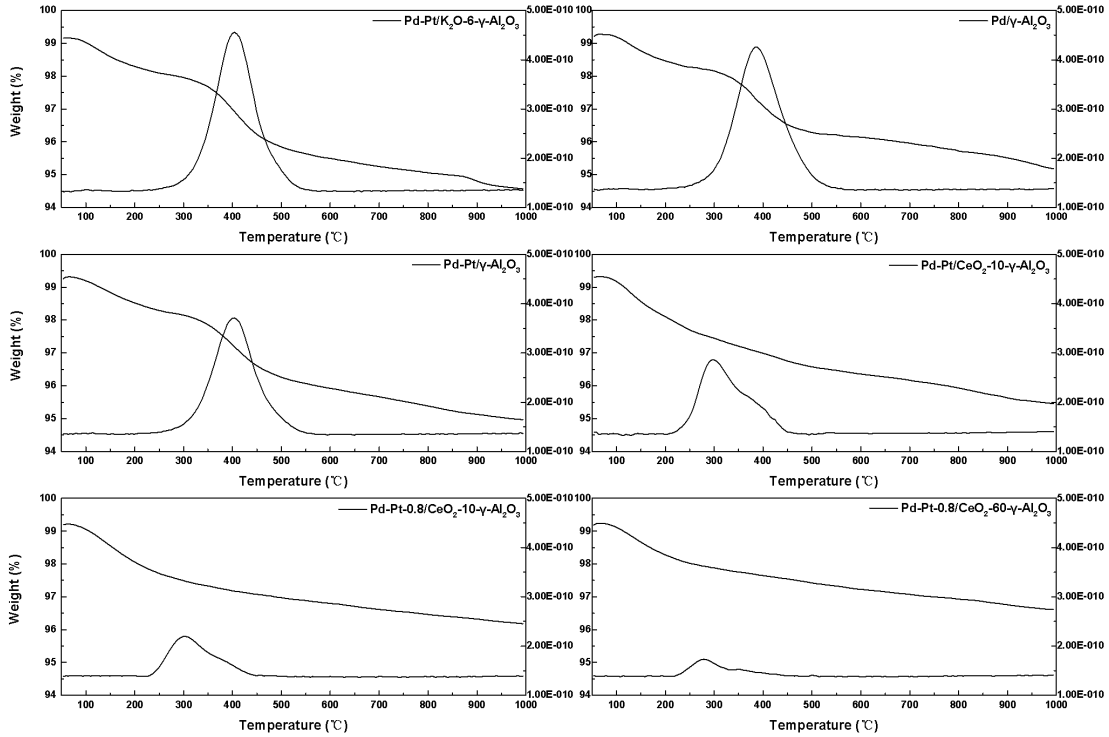


Fig. S1. TG-MS tests of post-reaction catalysts. (reaction conditions: toluene concentration is 1000 mg/m^3; GHSV is 5000 h^{-1}; Toluene elimination rate> 99.5%; reaction time is 12h).

Table S1. Carbon balance calculation (to verify the accuracy of using CO_2 to measure carbon deposits)

Catalyst	Catalyst mass (g)	12h reaction toluene total feed (g)	Total carbon (mmol)	Carbon mass percentage in catalyst (wt%, TG-MS)	Carbon deposits (mmol,ele-mentary substance)	CO_2 selectivity (%)
Pd-Pt/K_2O-6-γ-Al_2O_3	145.010	8.478	644.085	1.20	145.010	77.49
Pd/γ-Al_2O_3	144.427	8.478	644.085	1.03	123.967	80.75
Pd-Pt/γ-Al_2O_3	144.256	8.478	644.085	0.92	110.596	82.83
Pd-Pt/CeO_2-10-γ-Al_2O_3	144.473	8.478	644.085	0.60	72.237	88.78
Pd-Pt-0.8/CeO_2-10-γ-Al_2O_3	143.983	8.478	644.085	0.31	37.196	94.22
Pd-Pt-0.8/CeO_2-60-γ-Al_2O_3	147.150	8.478	644.085	0.14	17.168	97.33

The γ-Al_2O_3 support was impregnated with the aluminum nitrate solution, and then dried at 120 °C for 3 h and calcined in a muffle furnace at 600 °C for 2 h. This process was repeated 5 times to obtain γ-Al_2O_3-R support, after which CeO_2 (10wt%) and Pd-Pt (2wt%) were loaded. Preparation of monolithic catalyst: powder catalyst was mixed with water to form a slurry. Then, cordierite honeycomb ceramics (∅ 60 mm, height = 50 mm, 400 holes per square inch, 136 g) were impregnated in the slurry. The suspension remaining in the hole of the cordierite honeycomb ceramics was then blown off by compressed air, dried at 120 °C for 3 h and calcined in a muffle furnace at 500 °C for 2 h (γ-Al_2O_3 in the powder catalysts accounts for 5 wt% of cordierite honeycomb ceramics).

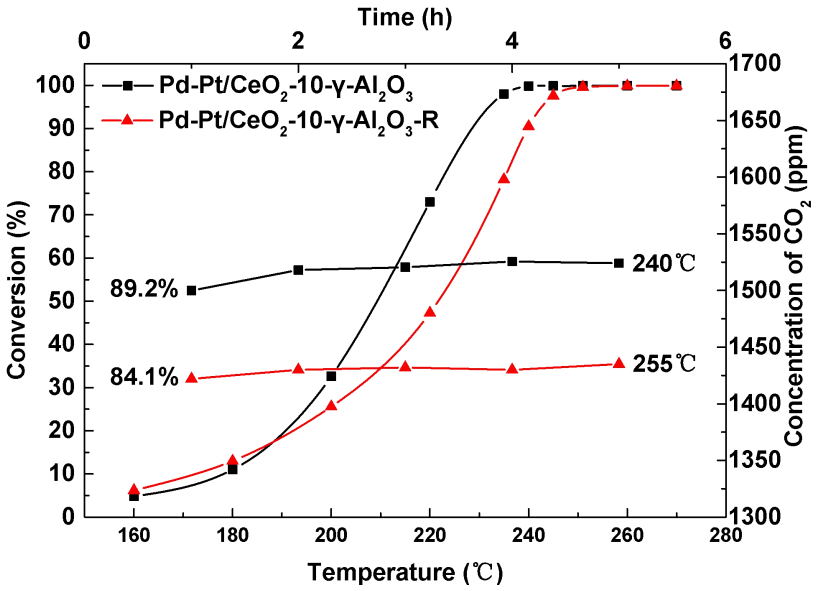
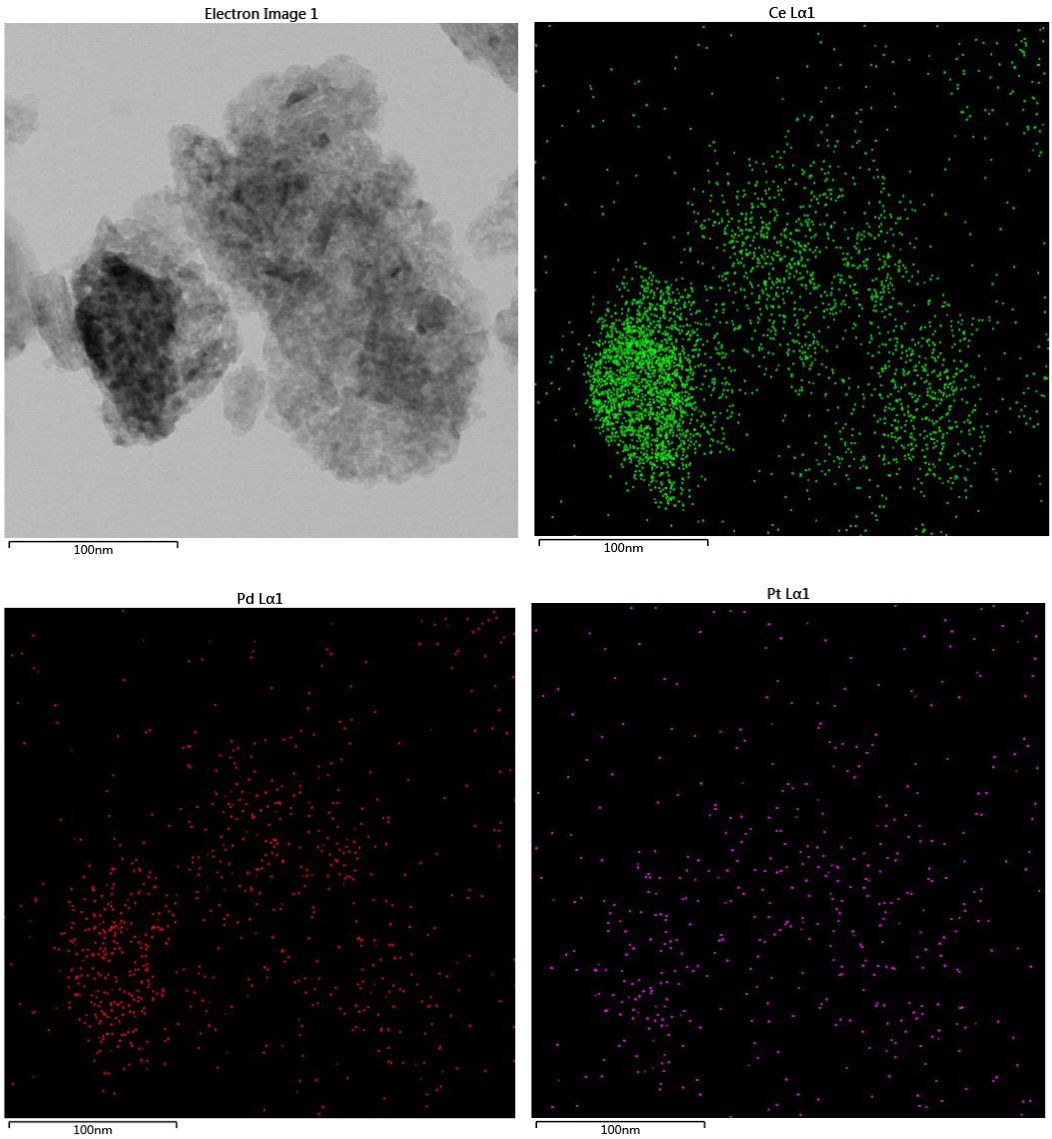


Fig. S2. Conversion of toluene on catalysts and CO_2 concentration at the reactor outlet and the selectivity of CO_2. Monolithic catalyst: ∅ 60 mm, height = 50 mm, 400 holes per square inch; toluene concentration: 1000 mg/m^3; GHSV: 5000 h^{-1}. (The catalyst with reduced pore diameter after the γ-Al_2O_3 support is treated is indicated by Pd-Pt/CeO_2-10-γ-Al_2O_3-R)

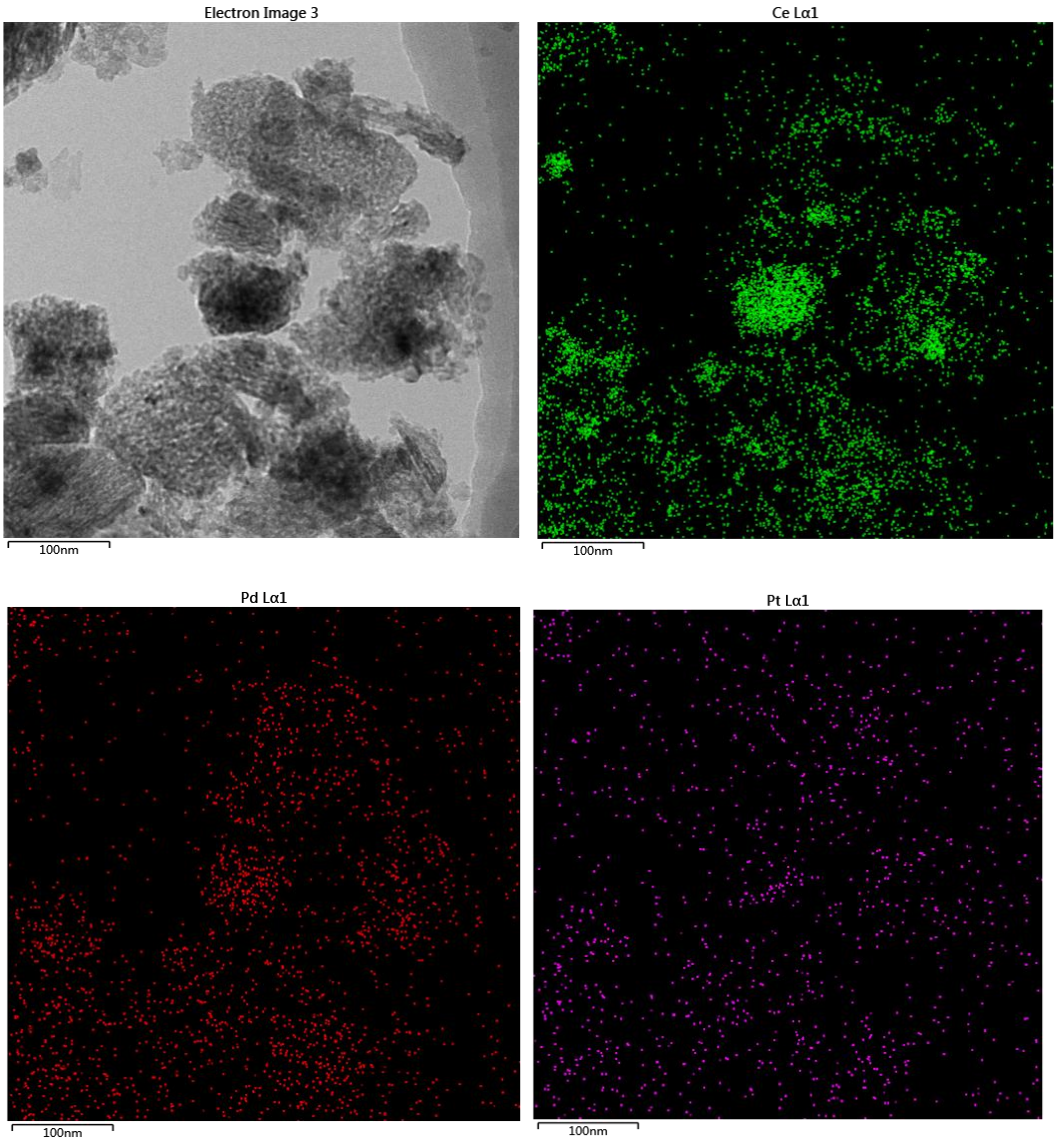
Table S2. Nitrogen adsorption-desorption experiment data.

Catalyst	Surface area (m^2/g)	Pore volume (cm^3/g)	Pore diameter (nm)
Pd-Pt/CeO_2-10-γ-Al_2O_3	130.09	0.1911	5.75
Pd-Pt/CeO_2-10-γ-Al_2O_3-R	70.39	0.0494	2.81

The TEM test results and EDS test results of Pd-Pt-0.8/CeO_2-10-γ-Al_2O_3 and Pd-Pt-0.8/CeO_2-60-γ-Al_2O_3 were shown in Fig. S3. The results show that when CeO_2 loading increases, the easier the CeO_2 agglomerates, the larger the particles. From the EDS results, we can see that Pd tends to be supported on CeO_2 (EDS results show that red dots are also dense where green dots are dense), while Pt is evenly dispersed on CeO_2 and Al_2O_3.



TEM and EDS test of Pd-Pt-0.8/CeO_2-10-γ-Al_2O_3

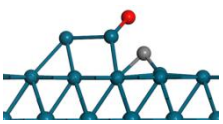
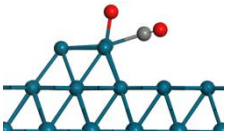
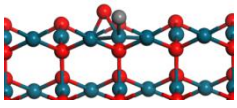
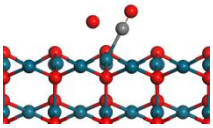
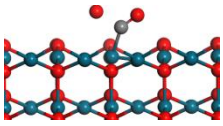


TEM and EDS test of Pd-Pt-0.8/CeO_2-60-γ-Al_2O_3

Fig. S3. TEM and EDS tests of catalysts.

The frequency calculations were performed to verify the transition state. The results show that every transition state has only one imaginary frequency. The transition states are correct, and the result were listed in Table S3.

Table S3. The imaginary frequency of transition state.

	Disproportionation of CO on Pd^0	Oxidation of CO on Pd^0	Disproportionation of CO on PdO	Oxidation of CO on PdO (dissociated O)	Oxidation of CO on PdO (lattice oxygen)
Transition state					
Imaginary frequency	-321.57 cm^{-1}	-349.70 cm^{-1}	-394.69 cm^{-1}	-539.73 cm^{-1}	-178.22 cm^{-1}