

Supporting information

High-temperature Flame Spray Pyrolysis Induced Stabilization of Pt Single-Atom Catalysts

Shipeng Ding^a, Hsi-An Chen^b, Okorn Mekasuwandumrong^c, Max J. Hülsey^a, Xinpu Fu^a, Qian He^d, Joongjai Panpranot^{e,*}, Chia-Min Yang^{b,f,*}, Ning Yan^{a,*}

^a *Department of Chemical and Biomolecular Engineering, National University of Singapore, 4 Engineering Drive 4, Singapore 117585, Singapore*

^b *Department of Chemistry, National Tsing Hua University, Hsinchu 30013, Taiwan*

^c *Department of Chemical Engineering, Faculty of Engineering and Industrial Technology, Silpakorn University, Nakorn Pathom 73000, Thailand*

^d *Department of Materials Science and Engineering, National University of Singapore, 9 Engineering Drive 1, Singapore 117575, Singapore*

^e *Center of Excellence on Catalysis and Catalytic Reaction, Biorefinery Cluster, Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok, 10330, Thailand*

^f *Frontier Research Center on Fundamental and Applied Sciences of Matters, National Tsing Hua University, Hsinchu 30013, Taiwan*

Corresponding authors: ning.yan@nus.edu.sg, joongjai.p@chula.ac.th, cmyang@mx.nthu.edu.tw.

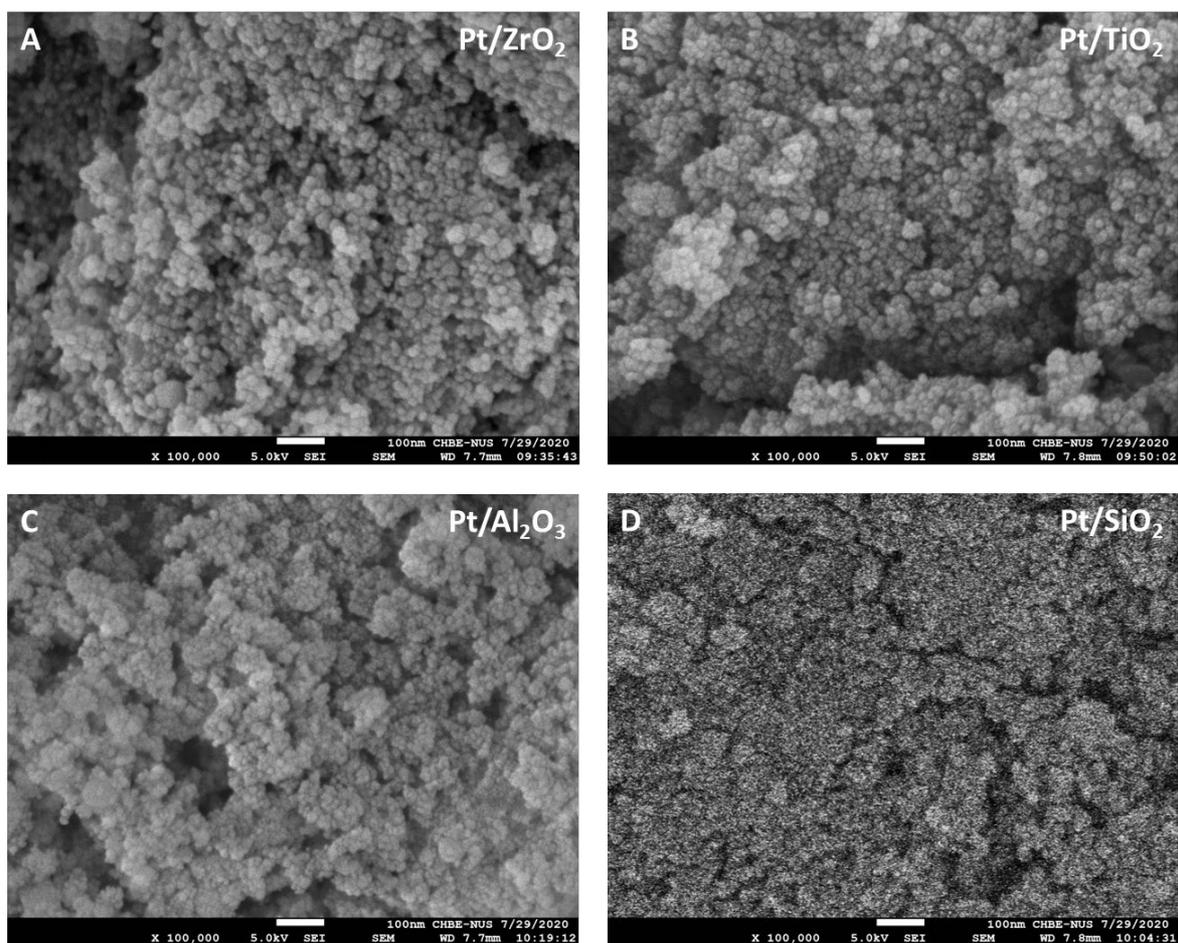


Fig. S1 The SEM images of the freshly prepared (A) 0.2Pt/ZrO₂ (B) 0.2Pt/TiO₂ (C) 0.2Pt/Al₂O₃ (D) 0.2Pt/SiO₂ using flame spray pyrolysis.

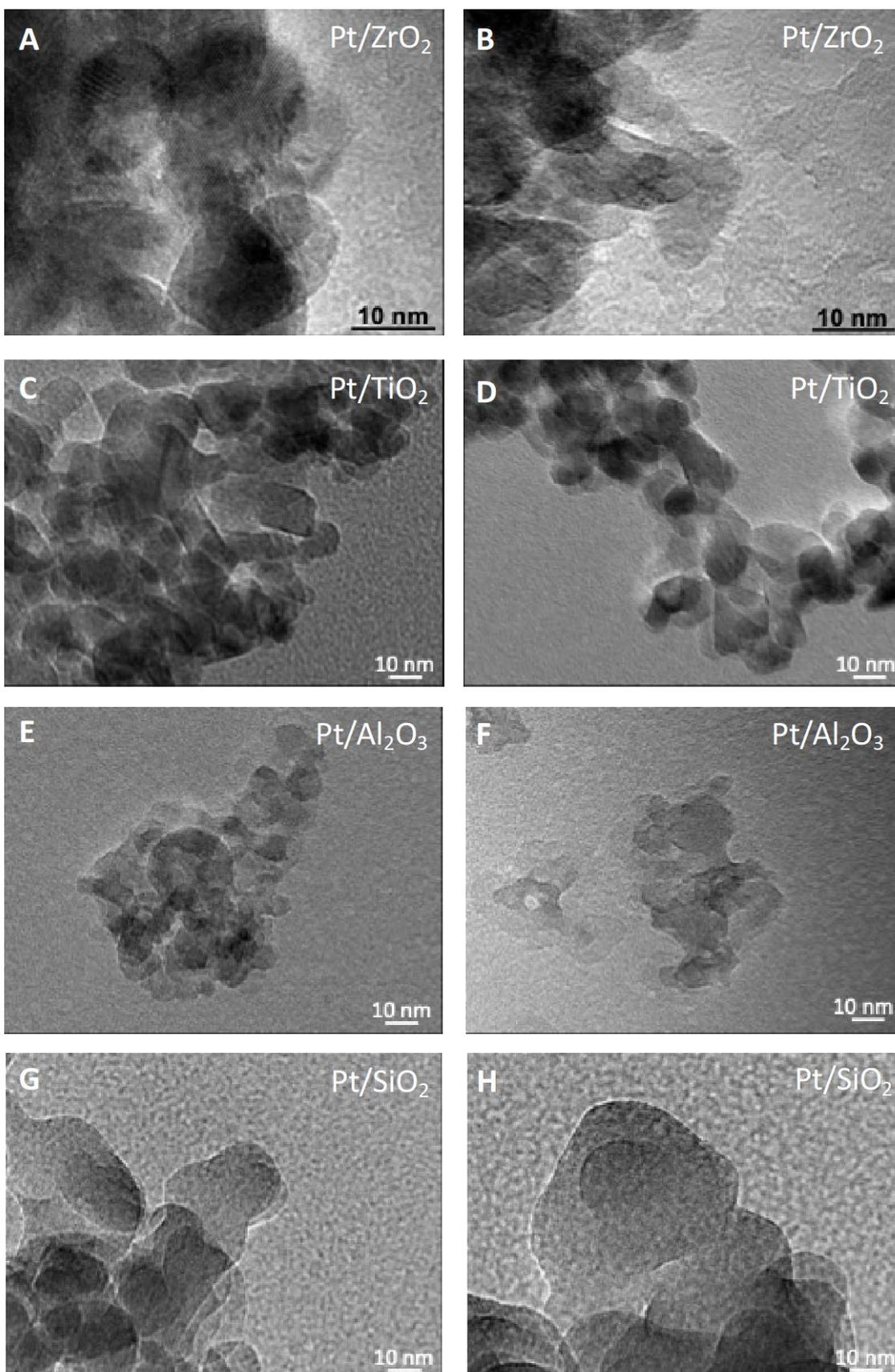


Fig. S2 The TEM images of (A-B) 0.2Pt/ZrO₂ (C-D) 0.2Pt/TiO₂ (E-F) 0.2Pt/Al₂O₃ (G-H) 0.2Pt/SiO₂ catalysts prepared by flame spray pyrolysis technique.

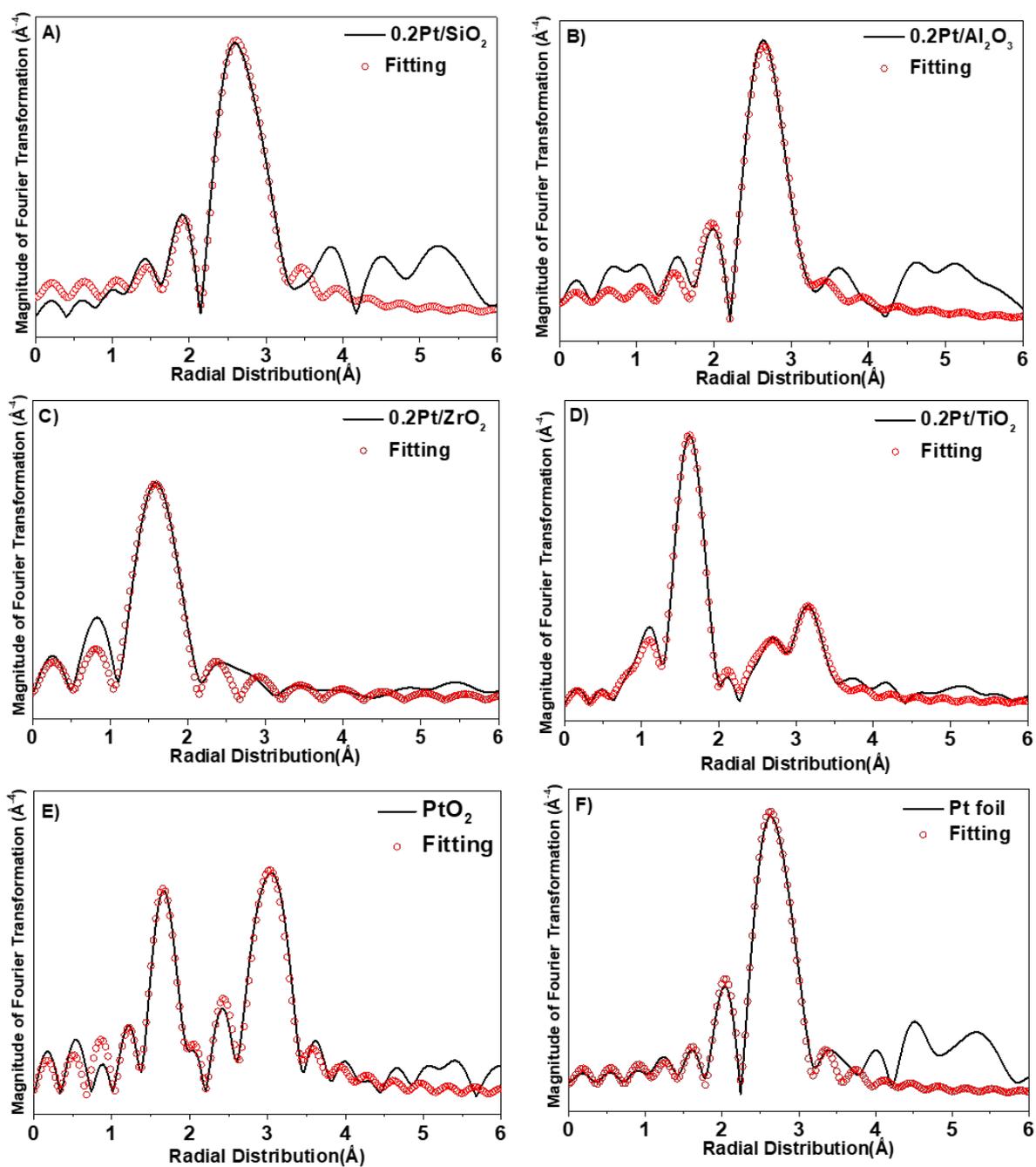


Fig. S3 The FT profiles of Pt L₃-edge k^3 -weighted EXAFS and the fitting results of the samples prepared by flame spray pyrolysis and reference samples.

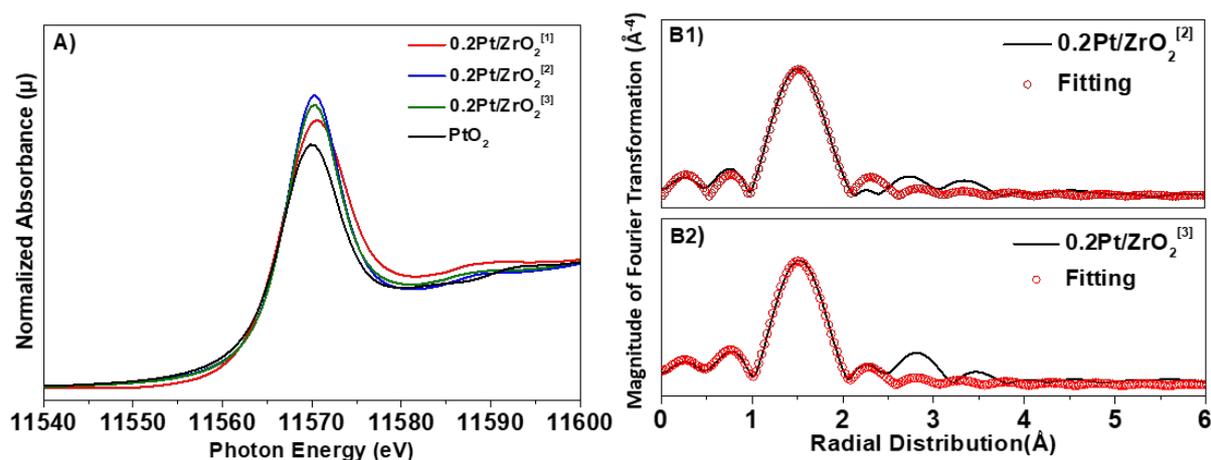


Fig. S4 (A) The normalized Pt L_3 -edge XANES and (B1-B2) The FT profiles of k^3 -weighted EXAFS of the 0.2Pt/ZrO₂. ^[2] and ^[3] represent 0.2Pt/ZrO₂ prepared under synthetic conditions 2) concentration 0.3 M, flow rate 3 mL/min; and 3) concentration 0.3 M, flow rate 2 mL/min, respectively.

Tab. S1. EXAFS parameters of Pt samples, Pt foil and PtO₂.

Samples	Shell	CN	R(Å)	$\Delta\sigma^2 \times 10^3(\text{Å}^2)$	$\Delta E_0(\text{eV})$	r-Factor(%)
Pt foil	Pt-Pt	12(fixed)	2.77	5.45	6.82	0.9
PtO ₂	Pt-O	4.9	2.02	2.74	8.72	1.1
	Pt-Pt	5.0	3.10	2.63	8.95	
0.2Pt/ZrO ₂ ^[1]	Pt-O	4.3	1.99	0.10	7.90	0.7
0.2Pt/ZrO ₂ ^[2]	Pt-O	5.2	1.95	2.06	4.19	0.1
0.2Pt/ZrO ₂ ^[3]	Pt-O	4.4	1.96	0.12	4.18	0.04
0.2Pt/Al ₂ O ₃	Pt-Pt	7.4	2.76	4.17	7.76	0.2
0.2Pt/SiO ₂	Pt-Pt	8.1	2.76	3.75	7.06	0.7
0.2Pt/TiO ₂	Pt-O	4.5	1.99	6.23	9.92	0.6

CN, coordination number; R, interatomic distance; σ^2 , the Debye–Waller factor; ΔE_0 , inner potential correction to account for the difference in the inner potential between the sample and each simulated path. ^[1], ^[2] and ^[3] represent 0.2Pt/ZrO₂ prepared under different preparation conditions (total metal concentration and flow rate of precursor solution): 1) concentration 0.5 M, flow rate 3 mL/min; 2) concentration 0.3 M, flow rate 3 mL/min; and 3) concentration 0.3 M, flow rate 2 mL/min.

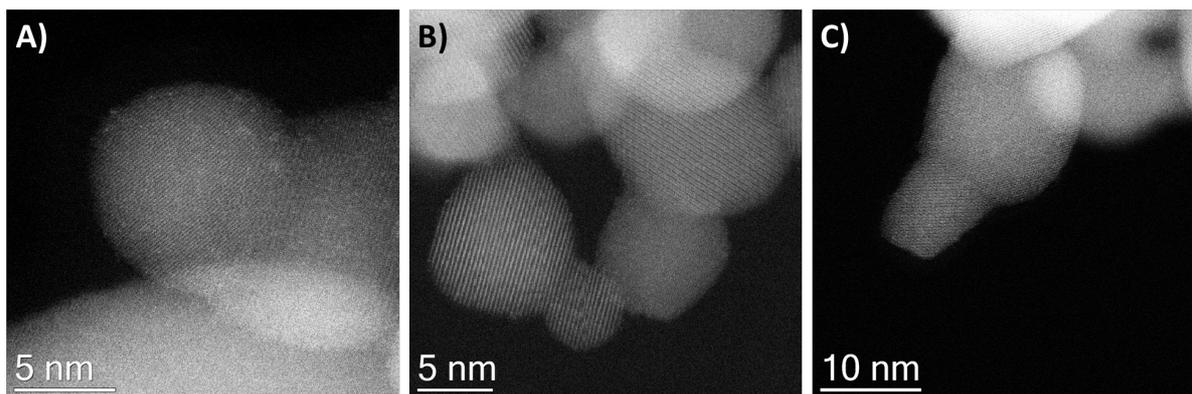


Fig. S5 HAADF-STEM images of the 0.1Pt₁/ZrO₂-flame catalyst.

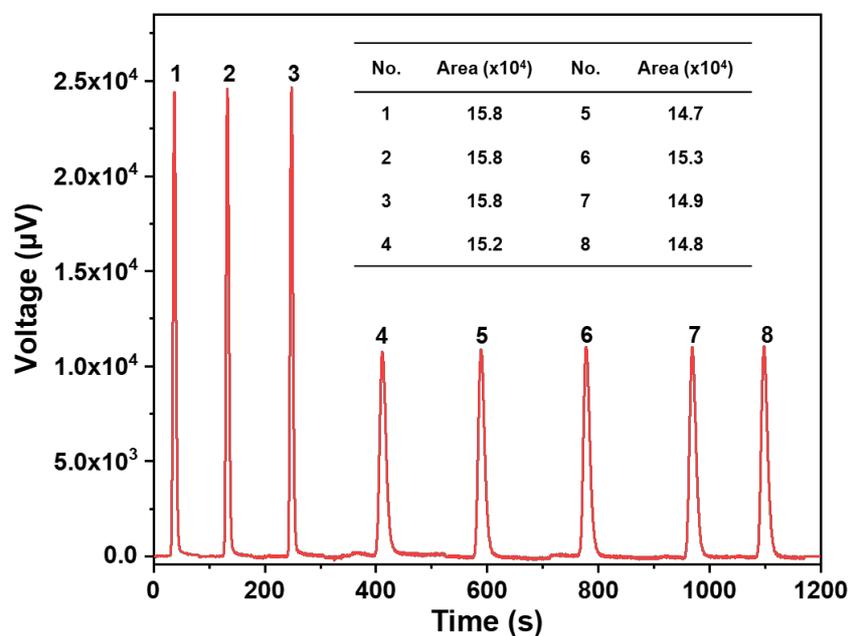


Fig. S6 H₂ titration file of 0.1Pt₁/ZrO₂-flame. The inset is the integrated area of each peak. The first three peaks with the integrated area of 15.8x10⁴ indicate the control injections (no H₂ adsorption). No obvious H₂ uptake was observed when H₂ passed through the catalyst (from peak 4 to 8). Titration conditions: N₂ carrier gas, 100 °C, 0.2 mL 5% H₂/pulse, 62 mg catalyst.

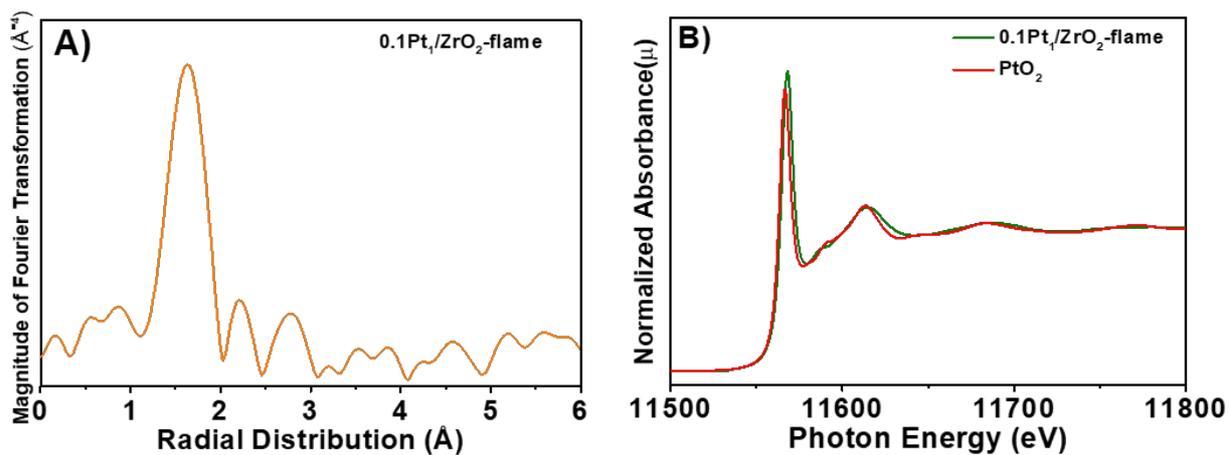


Fig. S7 (A) The FT profiles of k^3 -weighted EXAFS and (B) the normalized XANES spectra at the Pt L_3 -edge for 0.1Pt₁/ZrO₂-flame and PtO₂.

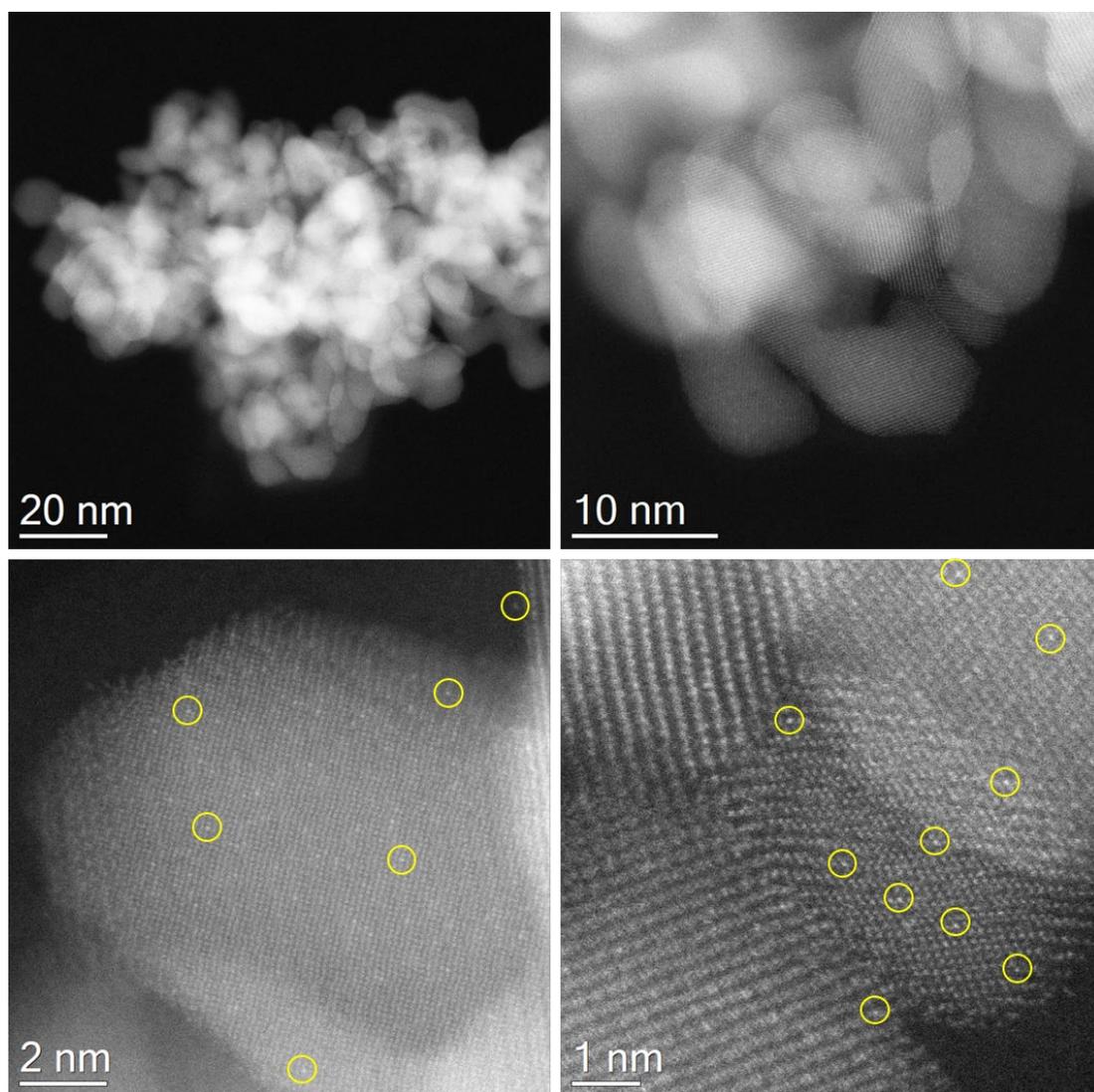


Fig. S8 Representative STEM-HAADF images of the 0.1Pt₁/ZrO₂(m)-wet catalyst. Atomically dispersed Pt species are highlighted with yellow circles.

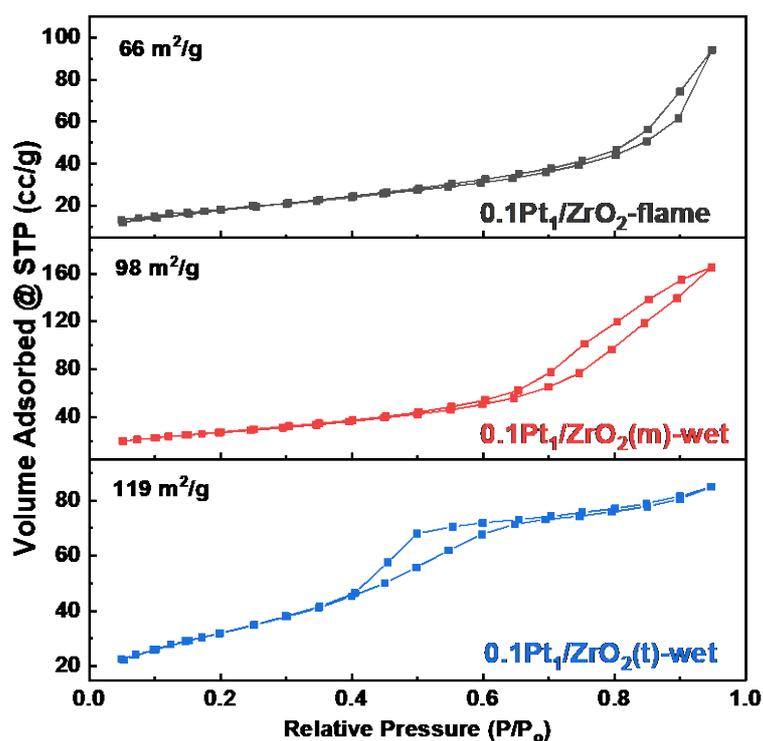


Fig. S9 N₂ adsorption/desorption isotherms of 0.1Pt₁/ZrO₂-flame, 0.1Pt₁/ZrO₂(m)-wet and 0.1Pt₁/ZrO₂(t)-wet.

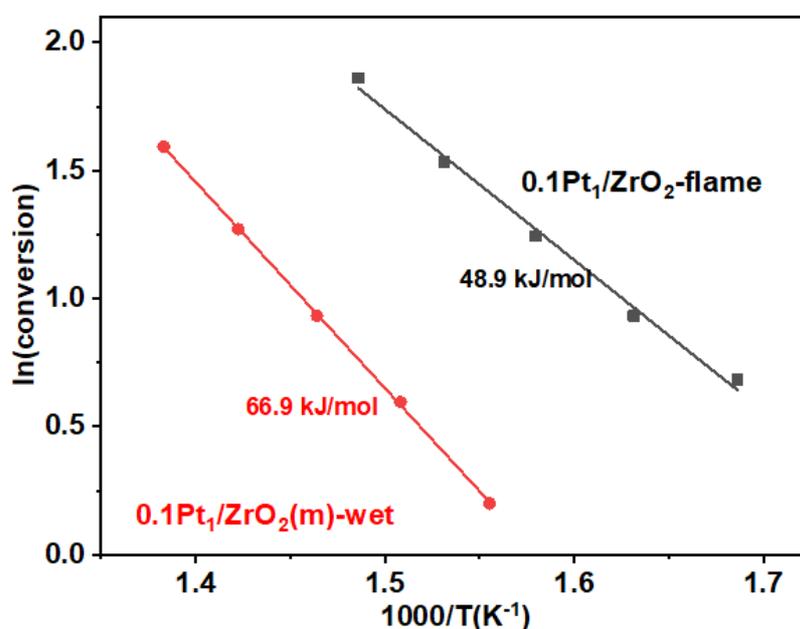


Fig. S10 Arrhenius plots of methane combustion reactions over 0.1Pt₁/ZrO₂-flame and 0.1Pt₁/ZrO₂(m)-flame.

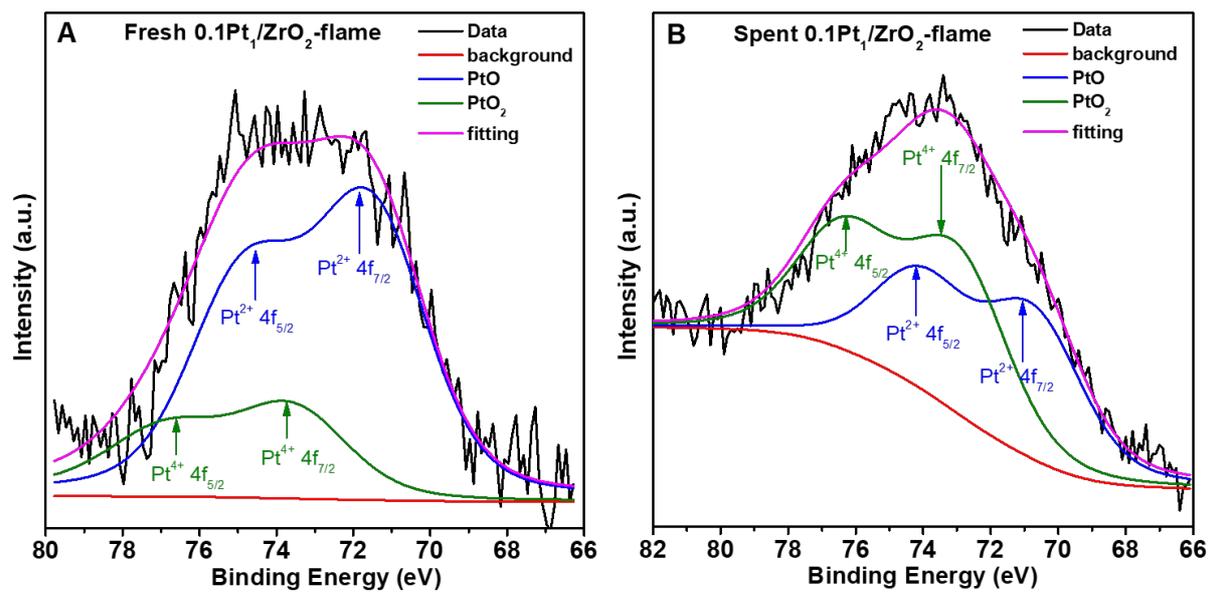


Fig. S11 Pt 4f XPS spectra of (A) freshly prepared and (B) spent 0.1Pt₁/ZrO₂-flame in methane combustion at 700 °C.

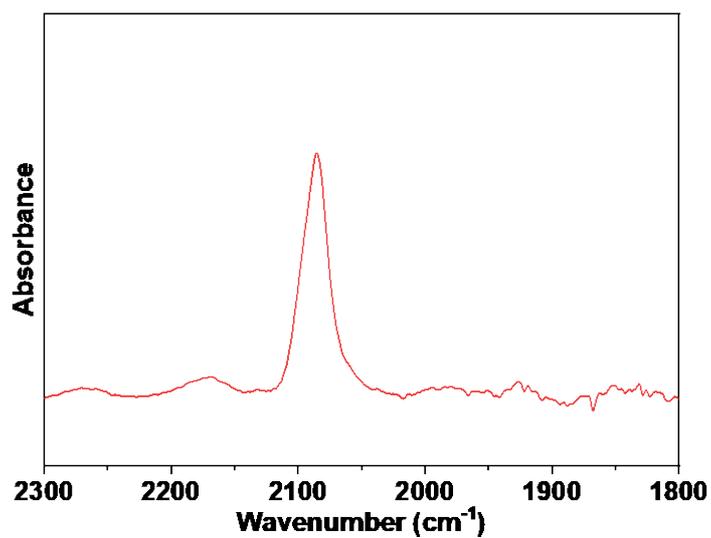


Fig. S12 CO adsorption on the 0.1Pt₁/ZrO₂-flame after long term stability test in methane combustion at 700 °C.

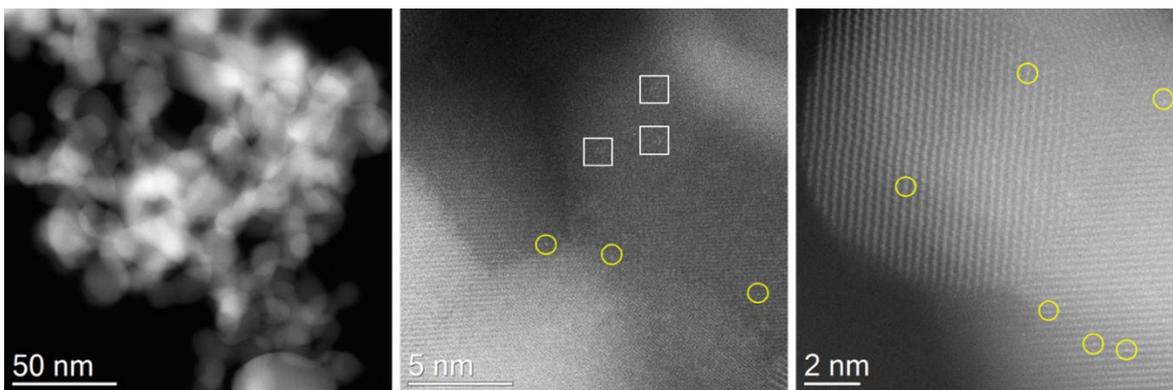


Fig. S13 Representative STEM-HAADF images of 0.1Pt₁/ZrO₂-flame after long term stability test in methane combustion. Pt clusters are highlighted by white squares while single Pt atoms are highlighted with yellow circles.

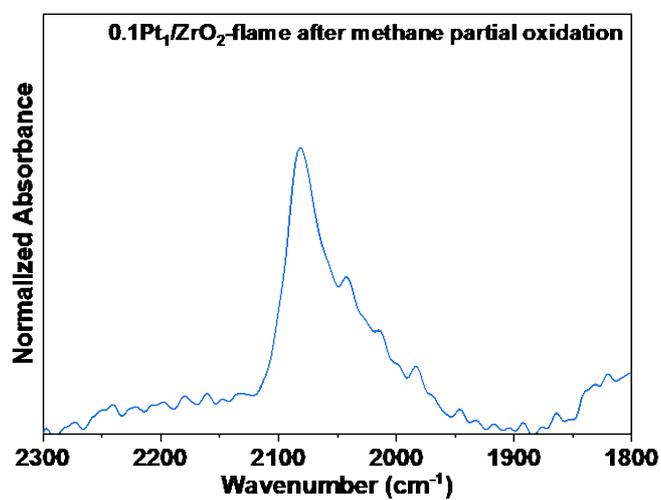


Fig. S14 CO adsorption on the 0.1Pt₁/ZrO₂-flame after long term stability test in methane combustion at 700 °C