Atomically dispersed Pd-based catalysts via constructing spatial structure with high performance for lean methane combustion

Supporting Information

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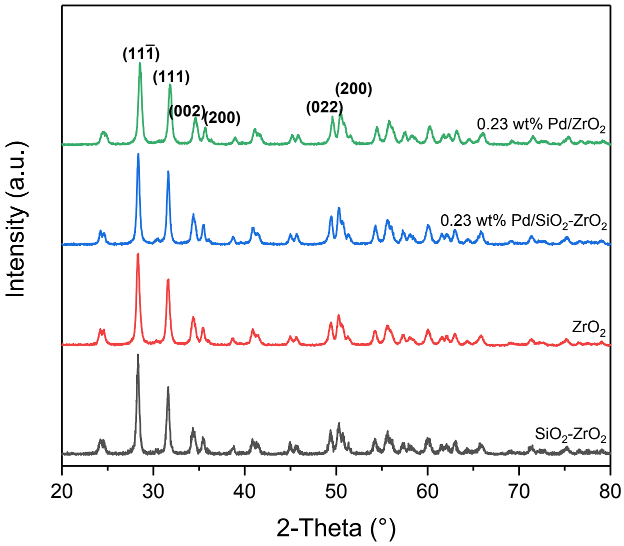
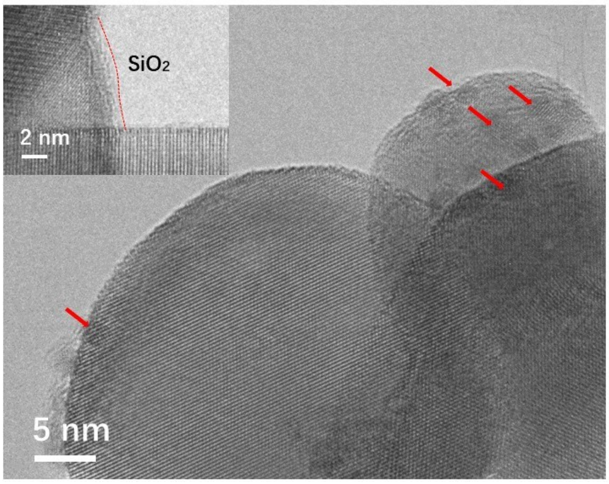
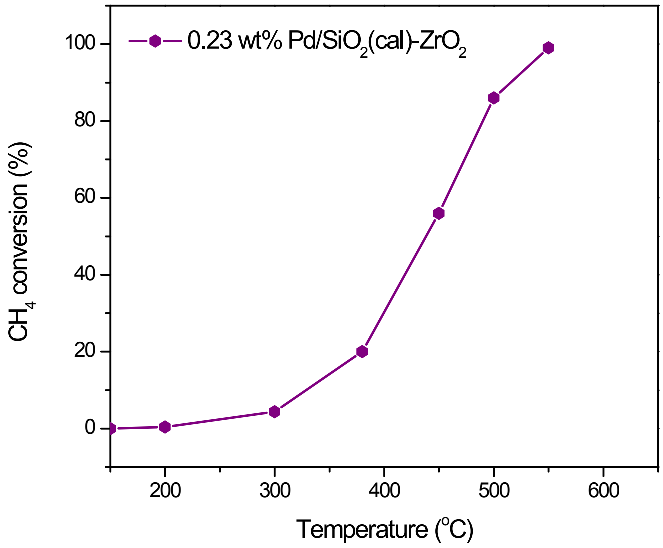


Supplementary Figure 1. XRD patterns of the as-prepared samples

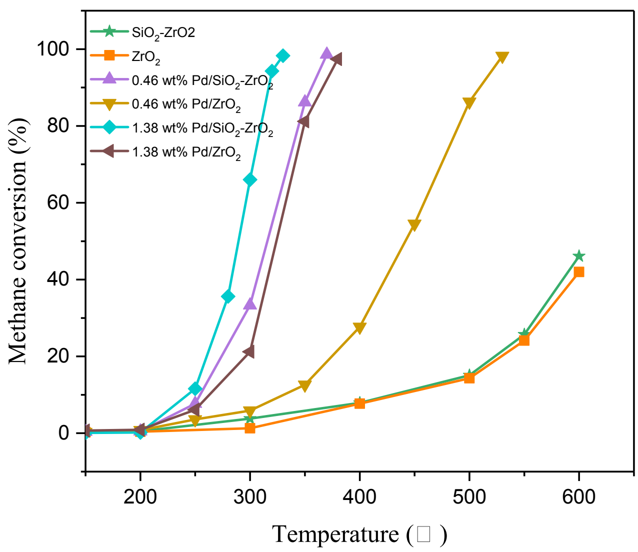


Supplementary Figure 2. TEM image of 0.23 wt% Pd/SiO_2(cal)-ZrO_2 catalyst.

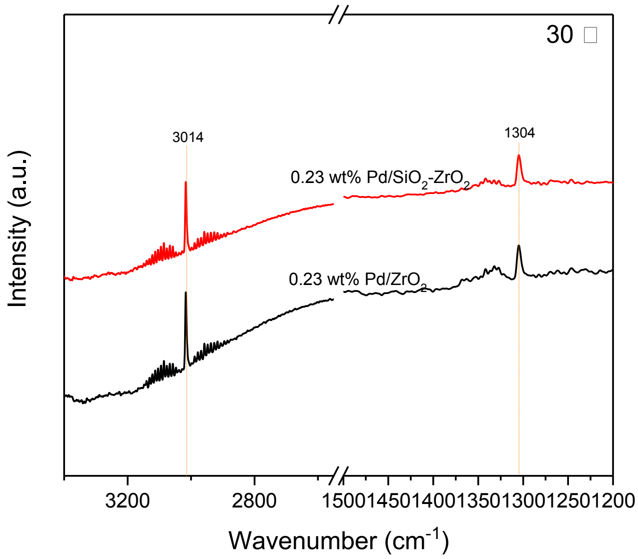
The modified support was calcined at 500 ° C in air for 3 hours, and then loaded with 0.23 wt% palladium. The catalyst was marked as 0.23 wt% Pd/SiO_2(cal)-ZrO_2 catalyst. Amorphous silica was coated on the surface of zirconia. It was obvious that palladium species were in the form of nanoparticles.



Supplementary Figure 3. Methane conversion as the function as temperature over 0.23 wt% Pd/SiO_2(cal)-ZrO_2 catalyst. Reaction gas: 1 vol% CH_4+99 vol% Air; GHSV: 30,000 ml/h $^{-1}$. g $^{-1}$.



Supplementary Figure 4. Methane conversion as the function as temperature over other catalysts



Supplementary Figure 5. In situ DRIFTS spectra of 0.23 wt% Pd/SiO_2-ZrO_2 catalyst and 0.23 wt% Pd/ZrO_2 catalyst in the presence of 1% CH_4/Air at room temperature.

Supplementary Table 1. Summarized nitrogen adsorption/desorption data and actual palladium and silicon loadings of various catalysts.

Pd loading amount (wt %)	SiO_2-ZrO_2 supported				ZrO_2 supported		
	Si[a] loading (wt %)	Pd[a] loading (wt %)	S_{BET} (m^2/g)	V_p[b] (cm^3/g)	Pd[a] loading (wt %)	S_{BET} (m^2/g)	V_p[b] (cm^3/g)
0	4.512	0	27.53	0.247	0	27.41	0.189
0.23	4.492	0.213	29.78	0.250	0.215	28.42	0.243
0.46	4.498	0.441	30.05	0.243	0.439	28.70	0.235
1.38	4.489	1.355	31.60	0.264	1.362	27.33	0.226

[a] Data were determined by the ICP-AES technique.

[b] Data were obtained from BJH Adsorption cumulative volume of pores between 17.000 Å and 3,000.000 Å diameter.

Supplementary Table 2. Performance comparison of various Palladium-based catalysts for lean methane combustion.

Sample	WHSV (ml/g-1.h-1)	T50 (℃)	T90 (℃)	T100 (℃)	Reactant gas	References
Pd5/$MgAlO_2$	20,000		477		CH_4(1%)/Air	[1]
Pd2/$LaFeO_3$	18,400		460		CH_4(1%)/O_2(4.4%)/He	[2]
Pd@CeO_2/H-Al_2O_3	200,000			400	CH_4(0.5%)/O_2(2%)/Ar	[3]
Pd0.5/g-Al2O_3	120,000		563		CH_4(1%)/O_2(20%)/N_2	[4]
Pd1/CeO_2/PG151	30,000			350	CH_4(1%)/O_2(18%)/N_2	[5]
Pd0.8/Co0.2	40,000	300	370		CH_4(0.5%)/O_2(2%)/Ar	[6]
LaAlPd-H-900	15,000		368		CH_4(1%)/O_2(20%)/Ar	[7]
Pd/Na-MOR	70,000			450	CH_4(1%)/O_2(4%)/N_2	[8]
Pd/o-CeO_2	30,000		348		CH_4(1%)/Air	[9]
Pd/NA-Al_2O_3	15,000			370	CH_4(1%)/O_2(20%)/Ar	[10]
Pd0.8Ni0.2@S-1	24,000		360		CH_4(1%)/ O_2(20%) / N_2	[11]
Pd6/CeO_2-0.1/Co_3O_4	60,000		350		CH_4(0.5%)/O_2(2%)/He	[12]
Pd@CeO_2/Si-Al_2O_3	50,000	368	416		CH_4(1%)/ O_2(20%) / N_2	[13]
6LSCPd	24,000		580		CH_4(1%)/ O_2(6%) /CO(6)/ He	[14]
Pd/6P-OMA	30,000			345	CH_4(1%)/ O_2(5%) / N_2	[15]
Pd-$NiCo_2O_4$/SiO_2	30,000			378	CH_4(1%)/Air	[16]
Pd/CeZr5	60,000			500	CH_4(1%)/O_2(4%)/He	[17]
Pd/15TA	50,000		340		CH_4(1%)/ O_2(10%) / N_2	[18]
Pd/ADP-OMA	50,000		420		CH_4(1%)/ O_2(5%) / N_2	[19]
PdCo/Hal	72,000			420	CH_4(1%)/ O_2(20%) / Ar	[20]
Pd-CeO_2CASs/Al_2O_3	60,000			410	CH_4(1%)/ O_2(4%) / N_2	[21]
Pd1.38/SiO_2-ZrO_2	**30,000**			**330**	**CH_4(1%)/Air**	This work

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