Electronic supplementary information (ESI)

# A highly active Pd/H-ZSM-5 catalyst in lean methane combustion prepared through sol-gel method and treated by reduction-oxidation

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As the Electronic supplementary information (ESI) of the manuscript "*A highly active Pd/H-ZSM-5 catalyst in lean methane combustion prepared through sol-gel method and treated by reduction-oxidation*", following materials are provided:

A comparison of various zeolites supported palladium catalysts in their activity for lean methane combustion; TGA profiles of the precursor Pd/H-ZSM-5 catalyst; light-off profiles for methane combustion and Pd 3d XPS spectra of the Pd/H-ZSM-5 catalysts reduced with hydrogen at different temperatures; light-off tests of methane combustion over the Pd/H-ZSM-5-R catalyst at different space velocities; repeated 10 cycle light-off tests of lean methane combustion over the Pd/H-ZSM-5-R catalyst; TGA profiles of the spent Pd/H-ZSM-5-R catalyst after the 10 cycle repeated light-off tests.

**Table S1** A comparison of various zeolites supported palladium catalysts in their activity for lean methane combustion

| Catalyst | Loading method | Reaction conditions | Pd loading (wt. %) | $T_{90\%}$ (°C) | Ref. |
|---|---|---|---|---|---|
| Pd/H-ZSM-5 | impregnation | 2% $CH_4$ + 8% $O_2$; GHSV = 48,000 $h^{-1}$ | 1.00 | 400 | [1] |
| Pd/H-ZSM-5 | deposition | 1% $CH_4$ + 20% $O_2$; GHSV = 15,000 mL $g^{-1}$ $h^{-1}$ | 0.77 | 311 | [2] |
| Pd-SSZ-13 | ion exchange | 0.15% $CH_4$ + 5% $O_2$; GHSV = 100,000 $h^{-1}$ | 1.1 | 362 | [3] |
| Pd-ZSM-5 | ibid | ibid | 1.30 | 384 | [3] |
| Pd-H-Mordenite | ion-exchange | 1% $CH_4$ + 99% Air; GHSV = 100,000 $h^{-1}$ | 0.70 | 495 | [4] |
| Pd-H-Y | ibid | ibid | 1.00 | 475 | [4] |
| Pd-H-SAPO-5 | ibid | ibid | 0.96 | 480 | [4] |
| Pd/H-MCM-41 | wet impregnation | $O_2/CH_4$ = 4; GHSV = 15,000 mL $g^{-1}$ $h^{-1}$ | 0.98 | 454 | [5] |
| Pd/MCM-48 | ibid | ibid | 1.05 | 483 | [5] |
| Pd/H-ZSM-5 | sol-gel | 1% $CH_4$ + 99% Air; GHSV = 10,000 mL $g^{-1}$ $h^{-1}$ | 0.92 | 293 | this work |
| | | GHSV = 30,000 mL $g^{-1}$ $h^{-1}$ | | 298 | |
| | | GHSV = 60,000 mL $g^{-1}$ $h^{-1}$ | | 308 | |
| | | GHSV = 100,000 mL $g^{-1}$ $h^{-1}$ | | 324 | |

**Note:** $T_{90\%}$ denotes the temperature for lean methane combustion at which a methane conversion of 90% can be achieved.

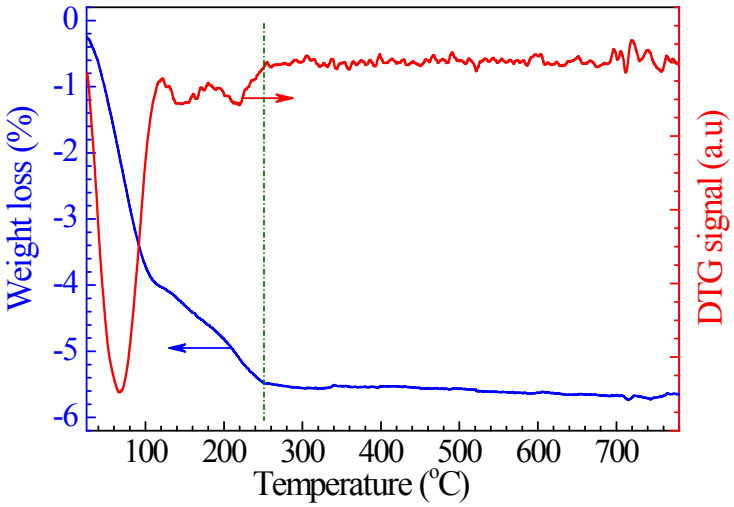


**Fig. S1** Weight loss and DTG curves for the thermogravimetric analysis of the precursor Pd/H-ZSM-5 catalyst.

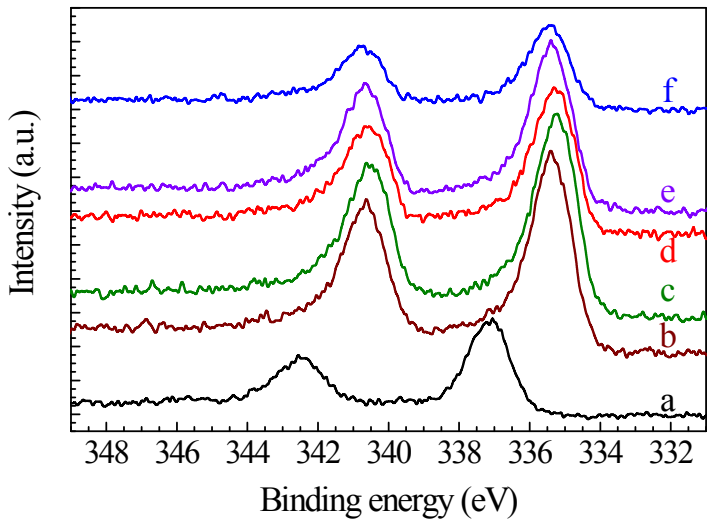


**Fig. S2** Pd 3d XPS spectra of the Pd/H-ZSM-5 catalysts subjected to reduction with hydrogen at different temperatures: (a) calcined without reduction; (b) 100 °C; (c) 200 °C; (d) 300 °C; (e) 400 °C; (f) 500 °C.

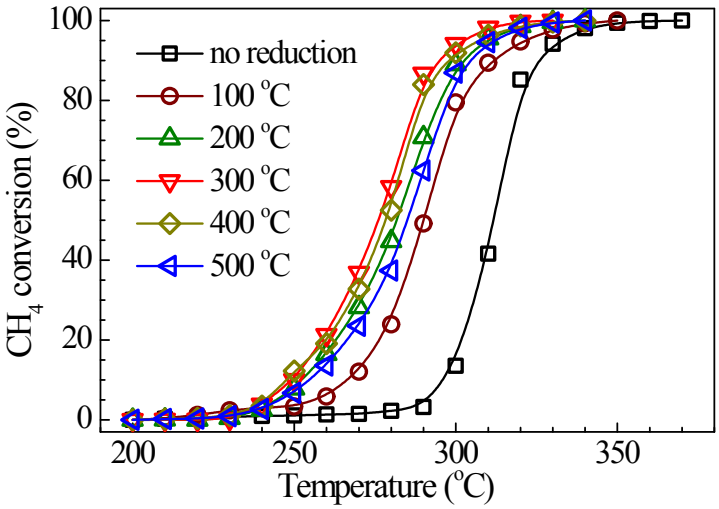


**Fig. S3** Effect of reduction temperature (marked in the legend) on the catalytic activity of Pd/H-ZSM-5 in lean methane combustion, represented by the light-off profiles (1.0 vol.% $CH_4$, GHSV = 30,000 mL $g^{-1}$ $h^{-1}$).

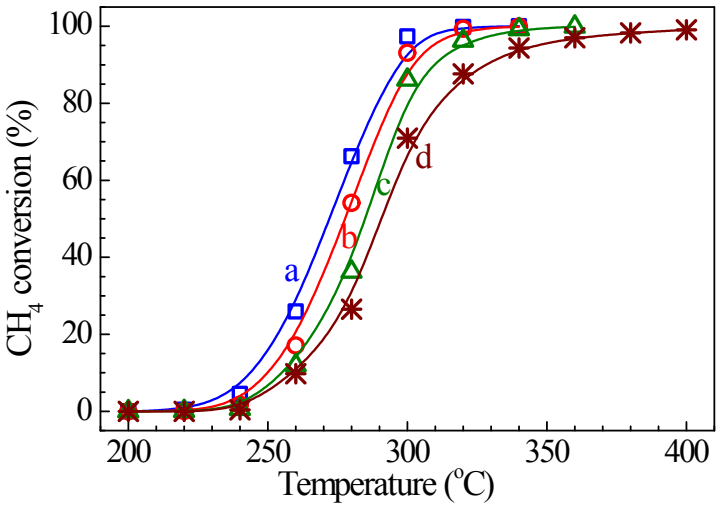


**Fig. S4** Light-off tests of lean methane combustion over the Pd/H-ZSM-5-R catalyst at different gas hourly space velocities (GHSV, mL $g^{-1}$ $h^{-1}$): (a) 10,000; (b) 30,000; (c) 60,000; (d) 100,000.

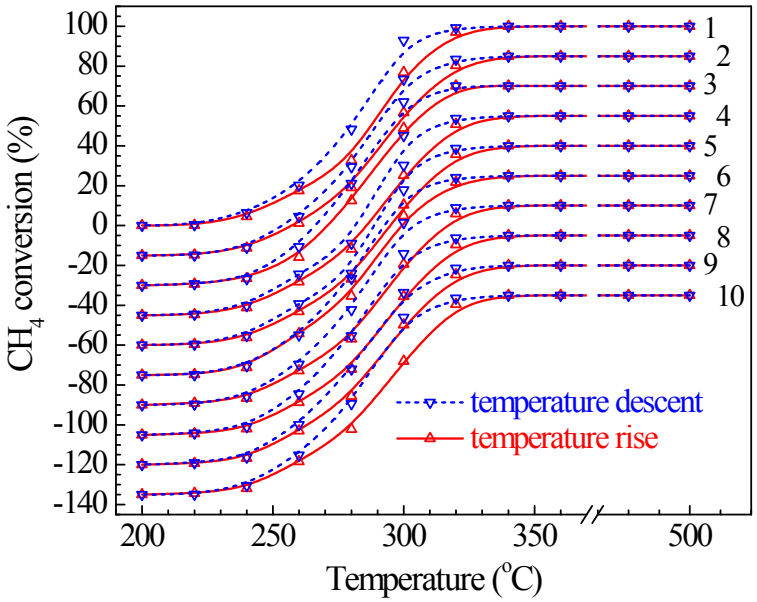


**Fig. S5** Repeated 10 cycle light-off tests of lean methane combustion over the Pd/H-ZSM-5-R catalyst (1.0 vol.% $CH_4$, GHSV = 30,000 mL $g^{-1}$ $h^{-1}$). From cycle 1 to cycle 10, the conversion of methane is downshifted by 15% sequentially in the graph.

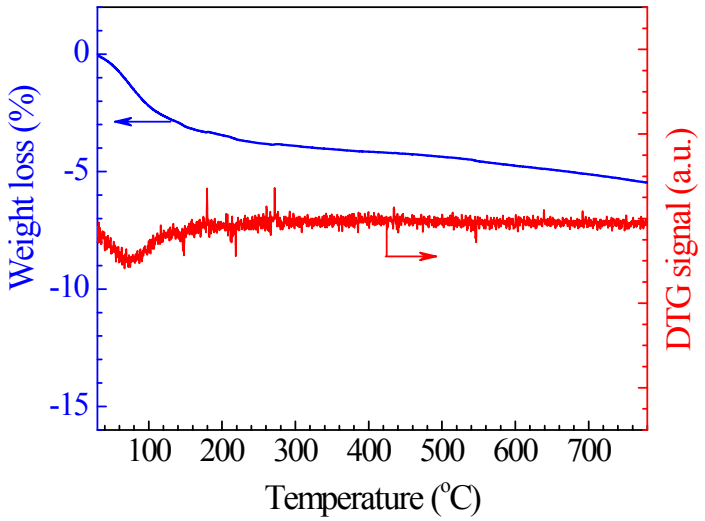


**Fig. S6** Weight loss and DTG curves for the thermogravimetric analysis of the spent Pd/H-ZSM-5-R catalyst after the 10 cycle repeated light-off tests.

## References


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