

## Nanocrystal Bilayer for Tandem Catalysis

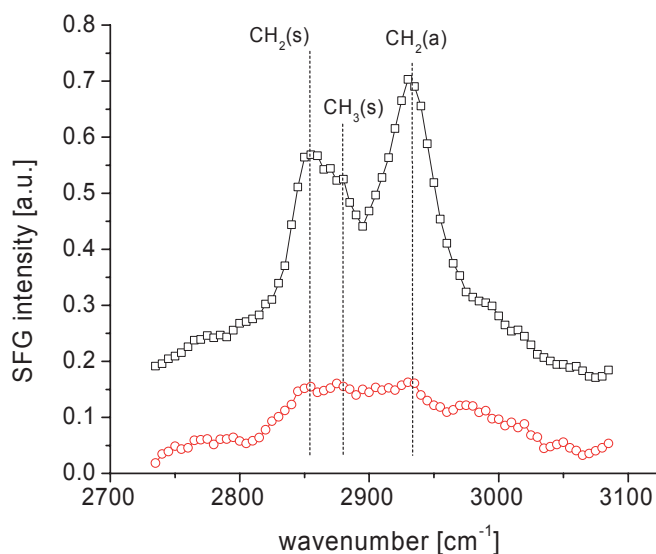
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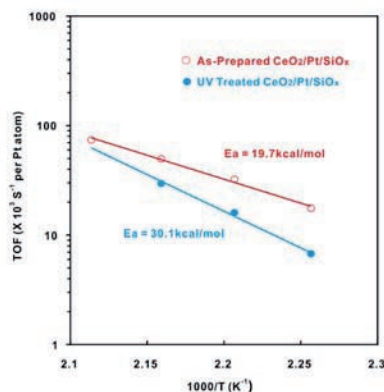
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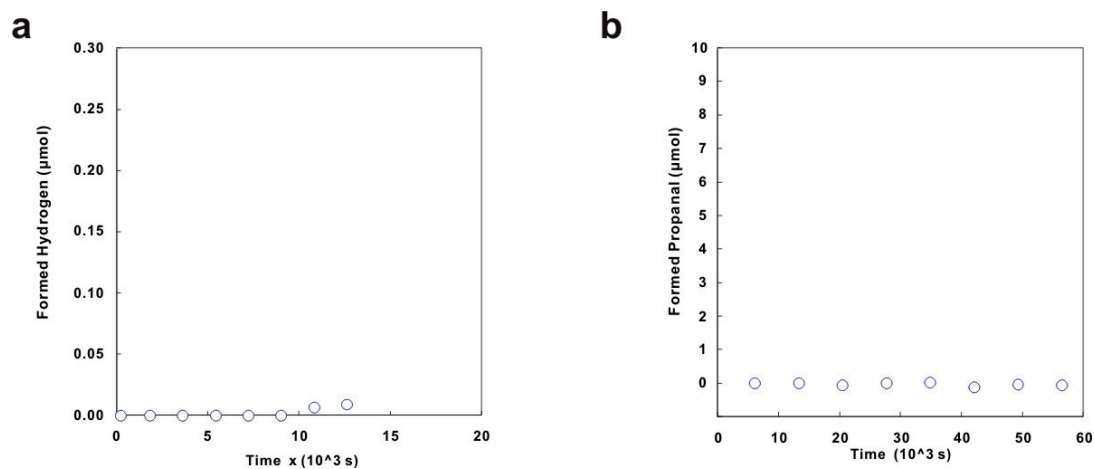
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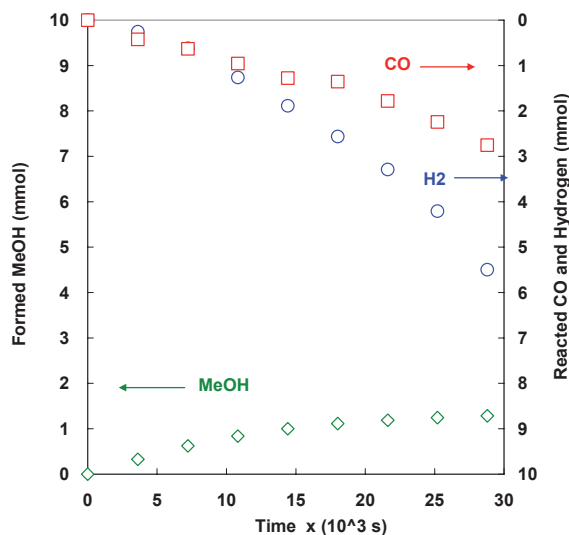
**Figure S1.** Sum frequency generation spectra of an LB film of oleylamine-coated Pt nanoparticles after UV/ozone treatment for 200 min (black line: sample as prepared, red line: UV/ozone treated sample)



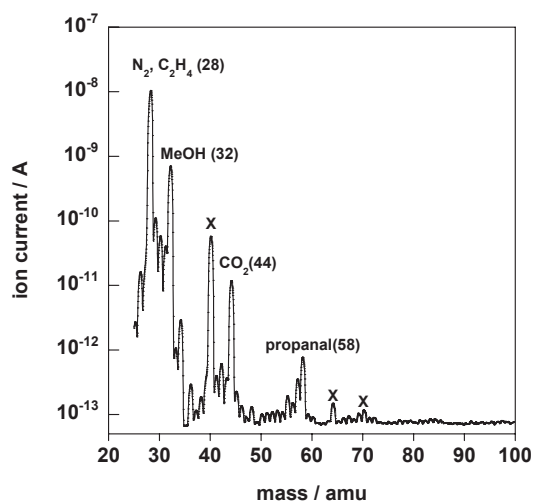
**Figure S2.** Turn-over frequency on CO oxidation over CeO<sub>2</sub> on Pt nanoparticles (open circle; as prepared sample, closed circle; plasma treated sample).



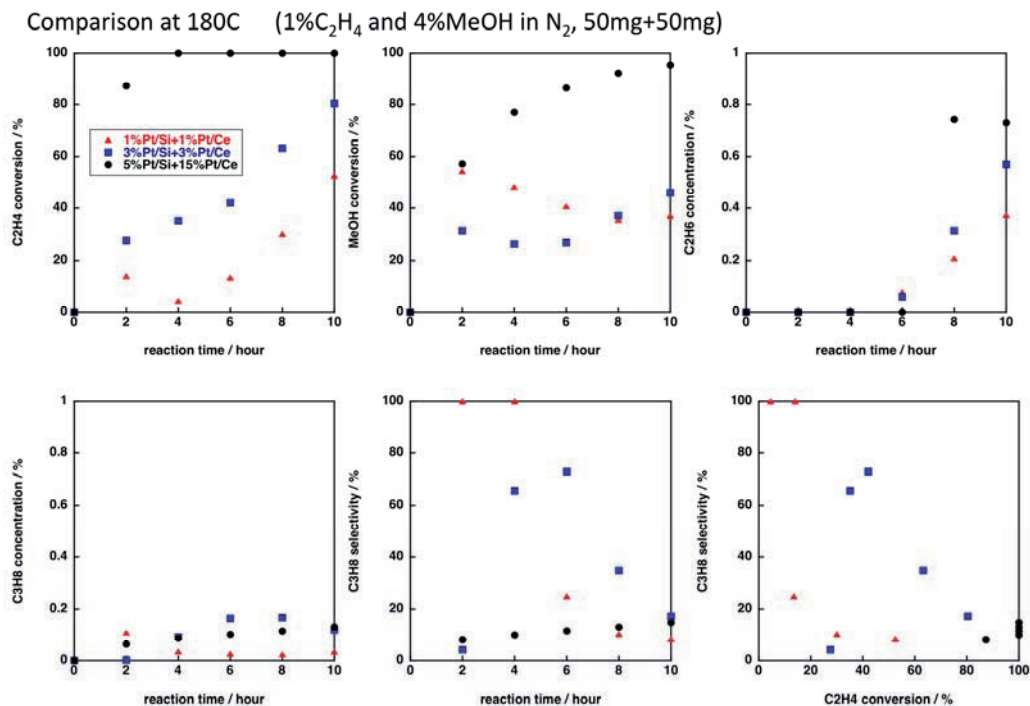
**Figure S3.** The as-prepared catalyst showed no catalytic activity for the reactions due to the lack of clean metal-metal oxide interfaces. (a) Hydrogen produced as a function of reaction time over Pt/CeO<sub>2</sub> interface without UV treatment at 190 °C by catalytic thermal decomposition of methanol. (b) Propanal produced as a function of reaction time over CeO<sub>2</sub>-Pt-SiO<sub>2</sub> without UV treatment at 190°C from ethylene and MeOH.



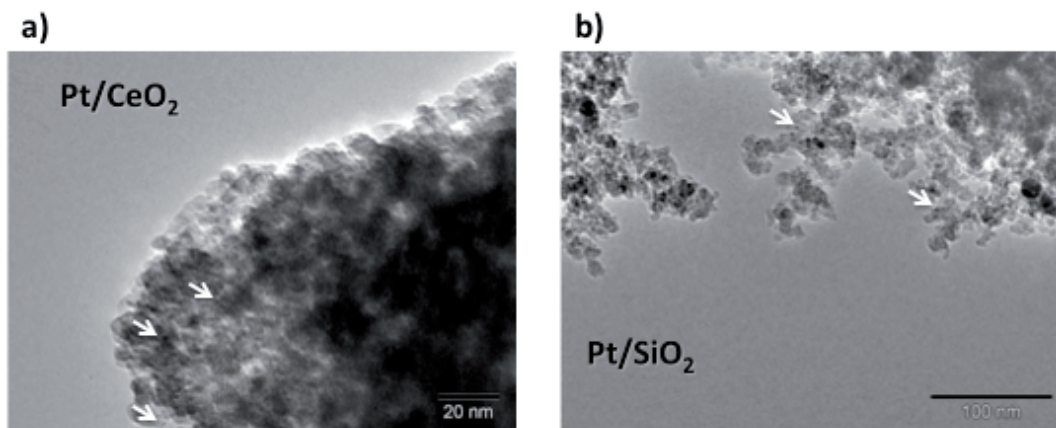
**Figure S4.** CO hydrogenation with CO and H<sub>2</sub> (without ethylene) as a function of reaction time over the Pt nanocubes on SiO<sub>2</sub>. (open square; reacted CO, open circle; reacted H<sub>2</sub>, open diamond; formed MeOH)



**Figure S5.** Mass spectrogram of the outlet gas after 24 hours reaction. (the peaks designated “x” are background peaks.)



**Figure S6.** Time courses of ethylene and MeOH conversions and ethane and propane concentrations in an effluent over the mixture of Pt/SiO<sub>2</sub> and Pt/CeO<sub>2</sub> in a cyclic reactor. (50 mg each, 180°C).



**Figure S7.** TEM images of 15%Pt/CeO<sub>2</sub> and of 10%Pt/SiO<sub>2</sub>.