

Supplementary Material

Insights into reaction mechanism of CO oxidative coupling to dimethyl oxalate over palladium: A combined DFT and IR study

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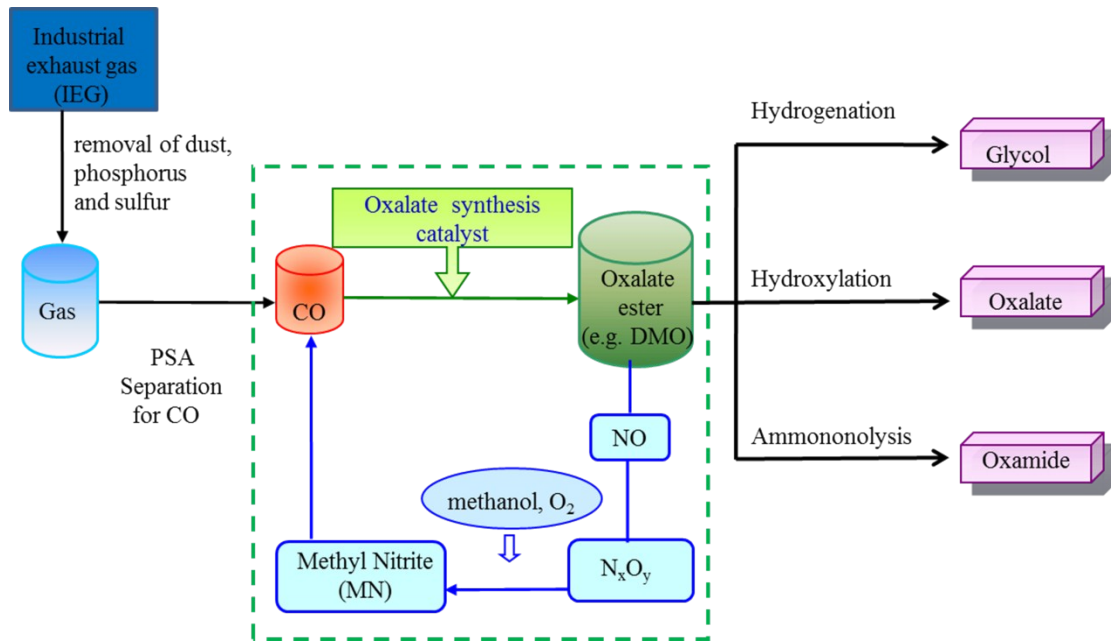
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effectively enhance the yield of DMO. The microscopic elucidation of this important reaction suggests the following improvements in the coal-to-EG (CTEG) production which have been applied in practice, effectively enhancing the yield and reducing the cost. The results may help with the further fine-designing of high-efficient noble metal catalysts.

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Schema 1S. Diagram of industrial exhaust gas to DMO technology

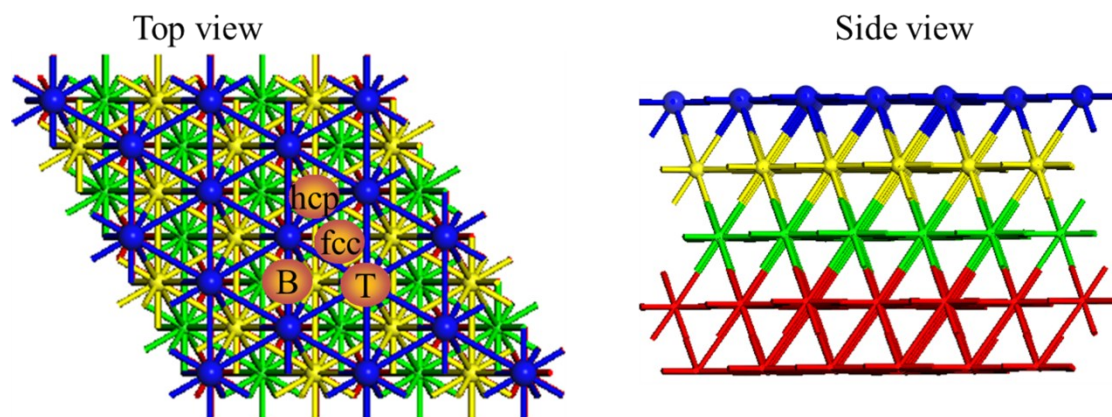


Fig. 1S. Top and side views of the perfect Pd(111) surface used for our calculations. Four adsorptions modes [the $T(top)$, $B(bridge)$, hcp , and fcc modes] on the Pd(111) surface.

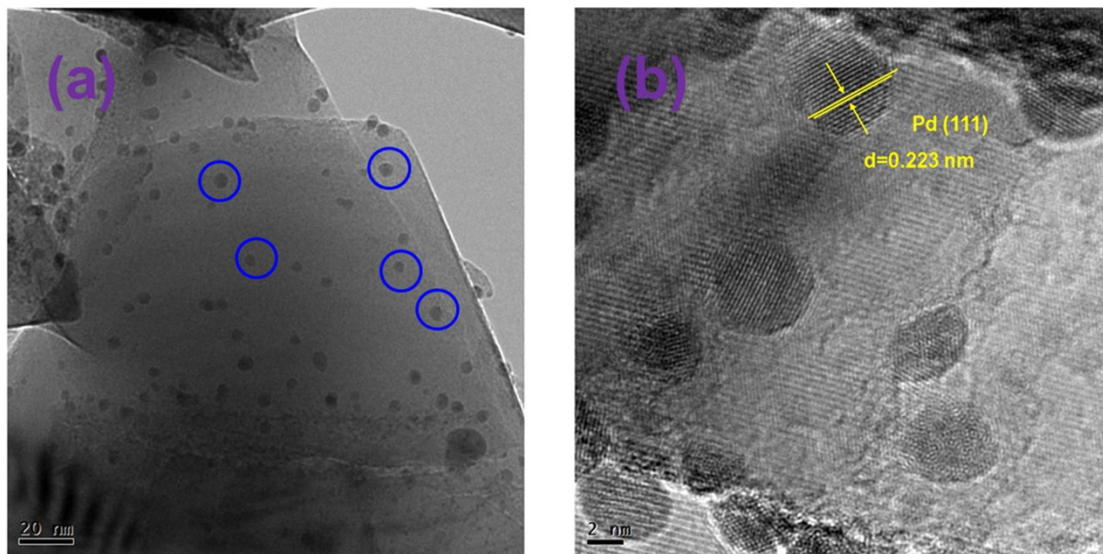


Fig. 2S. TEM (a) and HRTEM (b) images fresh of Pd/ α -Al₂O₃ catalysts after H₂ reduction.

The transmission electron microscope (TEM) image presented in Fig. 2S(a) reveals that the Pd nanoparticles (NPs) of catalyst is highly dispersed on the α -Al₂O₃ support with a size of 2-5nm. The high resolution TEM (HRTEM) image of catalyst in Fig. 2(b) shows that the interval between two lattice fringes of Pd NPs is about 0.223 nm, corresponding to the interplanar distance of the (111) plane of the *fcc* Pd[1, 2].

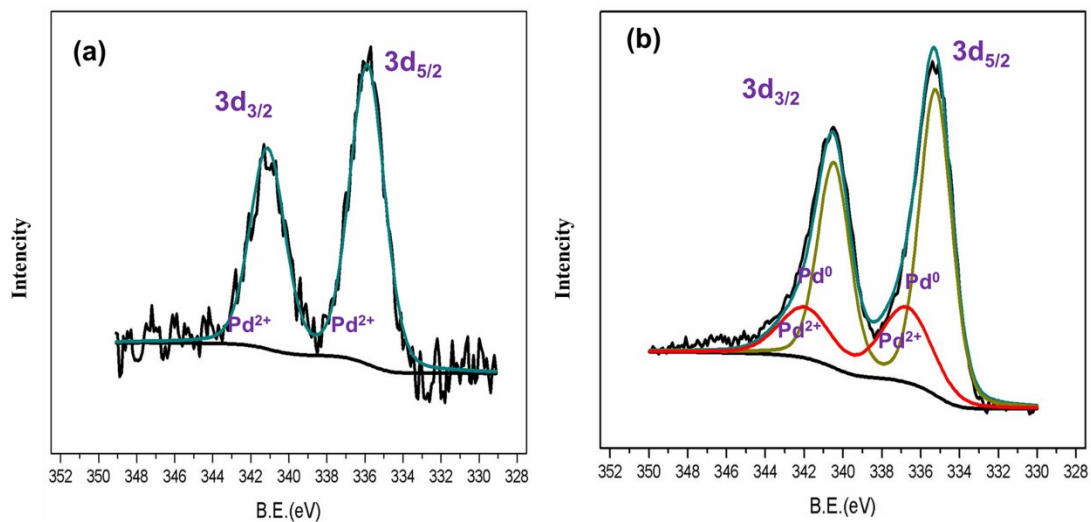


Fig. 3S. Curvefit of Pd 3d for XPS profiles of Pd/ α - Al_2O_3 catalyst (a) fresh catalyst and (b) catalyst after H_2 reduction.

The Pd-peaks at 337 and 342 eV are assigned to Pd^{2+} , and these at 335 and 340 eV are assigned to Pd^0 [3], respectively. Thus the Pd on the surface of the fresh catalyst is in Pd^{2+} state, whereas it on the catalyst after H_2 reducing was mainly in Pd^0 state.

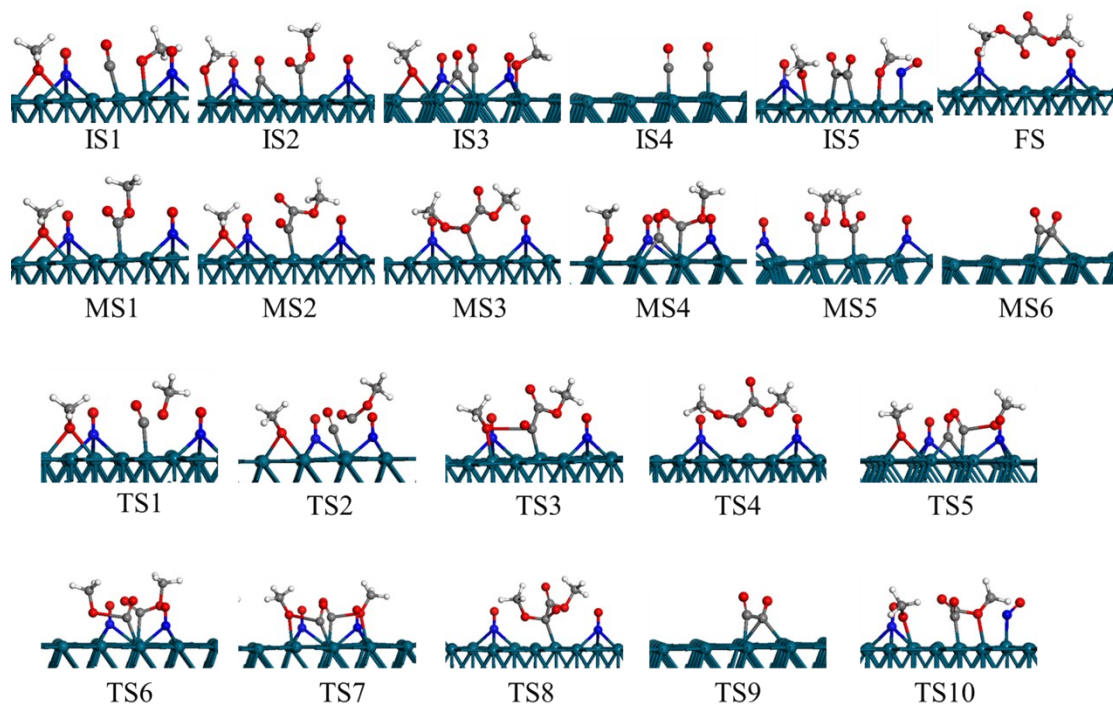


Fig.4S all the initial states (IS1-IS5), intermediate states (MS1-MS6), transition states (TS1-TS10) in three energy profiles for the reaction of CO and MN.

References

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