Electronic Supplementary Information (ESI)

Quinone tailored selective oxidation of methane over palladium catalyst with molecular oxygen as an oxidant

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SI-1: Activated carbon purification

Activated carbon (sieved into <100 mesh) was purchased from Tianjin Kermal Chemical Reagent Company. It was purified in 1 M HNO_3 at room temperature for 12 h, followed by filtration and washing with deionized water till the filtrate was neutral. Then it was dried at 100 °C overnight. Subsequently it was further treated in 1 M HCl and the same washing and drying procedure were carried out. Thus treated activated carbon was used as support to prepare Pd/C.

SI-2: Catalyst preparation and characterization

Pd/C was prepared by impregnation. The nominal loading of palladium was 1 wt. %. The sample was dried at 60 °C for 12 h, followed by reduction in H_2 at 400 °C for 10 h. Transmission electron microscopy (TEM) was used to characterize the morphology of the catalysts before and after reaction, which was carried out on a FEI Tecnai G2 microscope operated at an accelerating voltage of 120 kV.

SI-3: Catalytic reaction test

The reaction was carried out in an autoclave made of titanium-nickel alloy with a glass-liner. The standard reaction conditions were: 0.01 g Pd/C, 100 μmol TCQ, 3 ml

† Y. Fan and Z. An made the same contribution to this paper.

CF_3COOH, 1 ml H_2O, 2.5 MPa CH_4 (with 10% N_2), 1.0 MPa CO, 0.5 MPa O_2, 140 ºC and 3 h unless otherwise stated. GC-MS (Agilent 6890-5973N) and NMR (Bruker DRX-400) was used to analyze the products. The quantity of CF_3COOCH_3 was obtained by GC analysis. After removal of solid by centrifugation and distillation, the liquid product was analyzed by ^1H NMR to determine the yield of HCOOH. A known amount of CH_3CN was added to the reaction solution as an internal standard and a capillary tube containing 60 μl of D_2O was used as an external lock and reference. The amount of H_2O_2 generated in-situ was determined by titration with a standard solution of $KMnO_4$. The concentration of Pd^{2+} was determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, TJA IRIS Advantage).

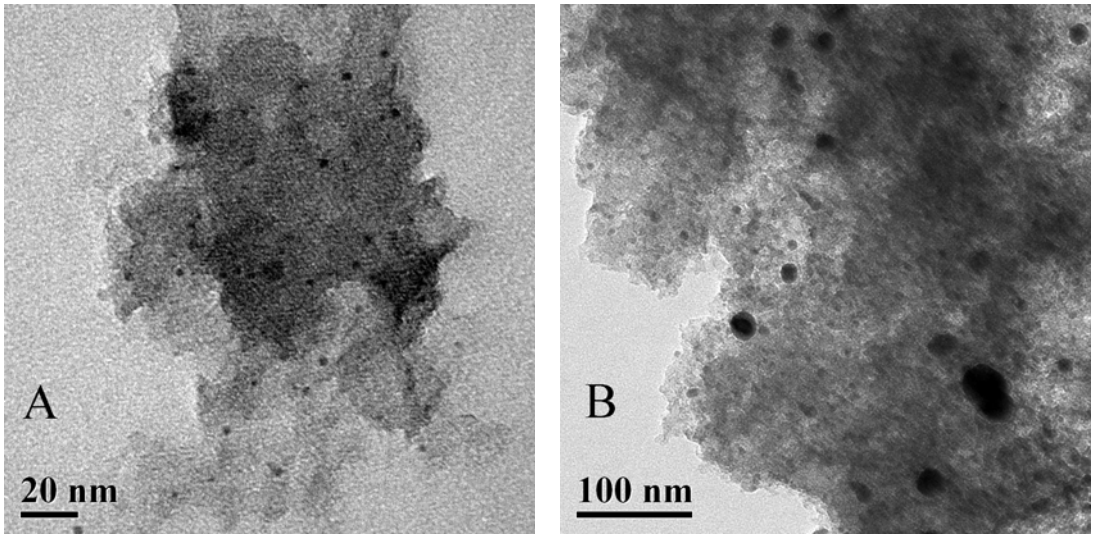

Figure S1. TEM images of the fresh (A) and used (B) Pd/C catalyst. The average size of Pd particles in the fresh catalyst is 3.1 ± 0.5 nm and it grows to 15.0 ± 4.4 nm after reaction.