Supplementary Material (ESI) for Chemical Communications

Supporting Information

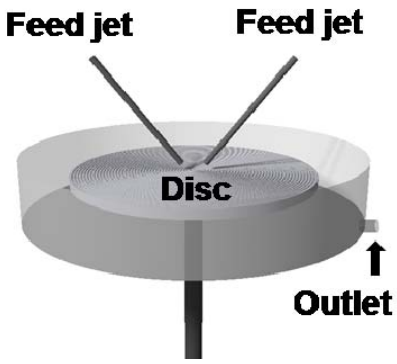
Surface oxygen triggered size change of palladium nano-crystals impedes catalytic efficacy

Jianli Zou, Scott G. Stewart, Colin L. Raston and K. Swaminathan Iyer*

Synthesis and characterization of Pd-PVP nanospheres:

Pd-PVP nanospheres were synthesized by using spinning disc processing (SDP) as a facile one step method with hydrogen gas as the reducing agent at room temperature. SDP (Scheme S1) is a process intensification strategy which offers continuous flowing film (1 to 200 μm) on a rapidly rotating disc surface (usually up to 3000 rpm). The wavy thin film generated on the spinning disc surface has been reported to enhance hydrogen gas uptake in the solution. [1] In a typical experiment, the aqueous solutions of H_2PdCl_4 (0.6mM) were mixed with PVP (polyvinylpyrrolidone) with the molecular ratio of PVP to palladium10, and then fed through a jet feed onto the spinning disc, and hydrogen gas as reducing agent was fed through another jet feed, which resulted in the formation of palladium nano-spheres of uniform size and shape (Figure S1). The resulting mixture was collected from the outlet and washed three times with MilliQ water, then freeze-dried before use in the Heck reaction. A large number of 5 nm palladium particles (Figure S1) spontaneous assembly into nano-spheres within the dynamic thin films on the surface of disc in the presence of PVP, rather than discrete individual palladium nanoparticles. PVP acts as a scaffold entangled with the small palladium nano-particles within the dynamic thin films under intense shearing in the dynamic thin films, as well as stabilising the Pd-PVP

nano-spheres in solution.



Scheme S1. Schematic representation of a spinning disc processor (SDP).

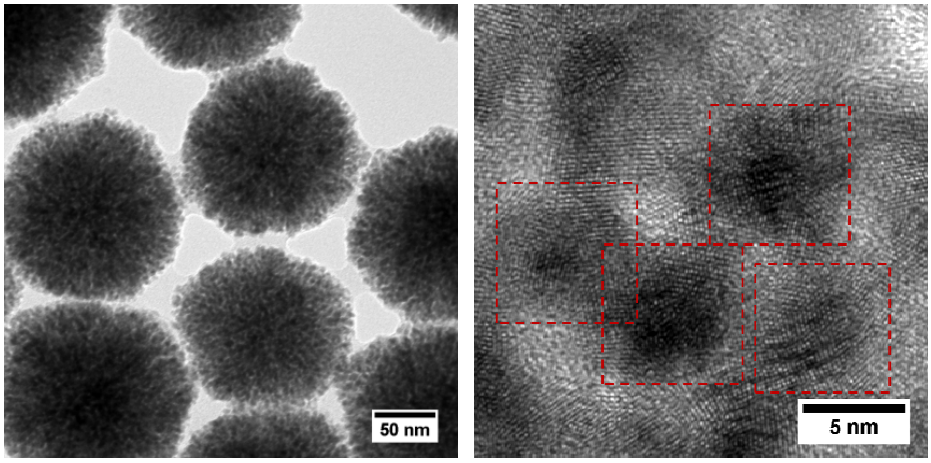


Figure S1.TEM image and high resolution TEM image of Pd-PVP nano-spheres. Quasi-spheroidal 5 nm palladium nano-crystals indicated in dotted box.

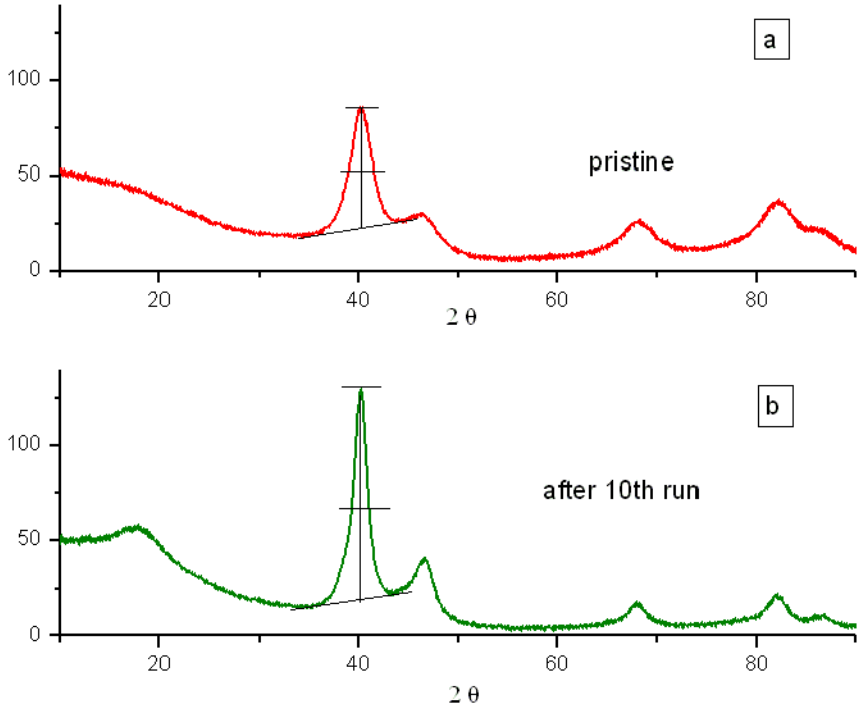


Figure S2. XRD patterns of the nano-catalyst, prior to the first cycle, (a), and after the 10^{th} recycling, (b).

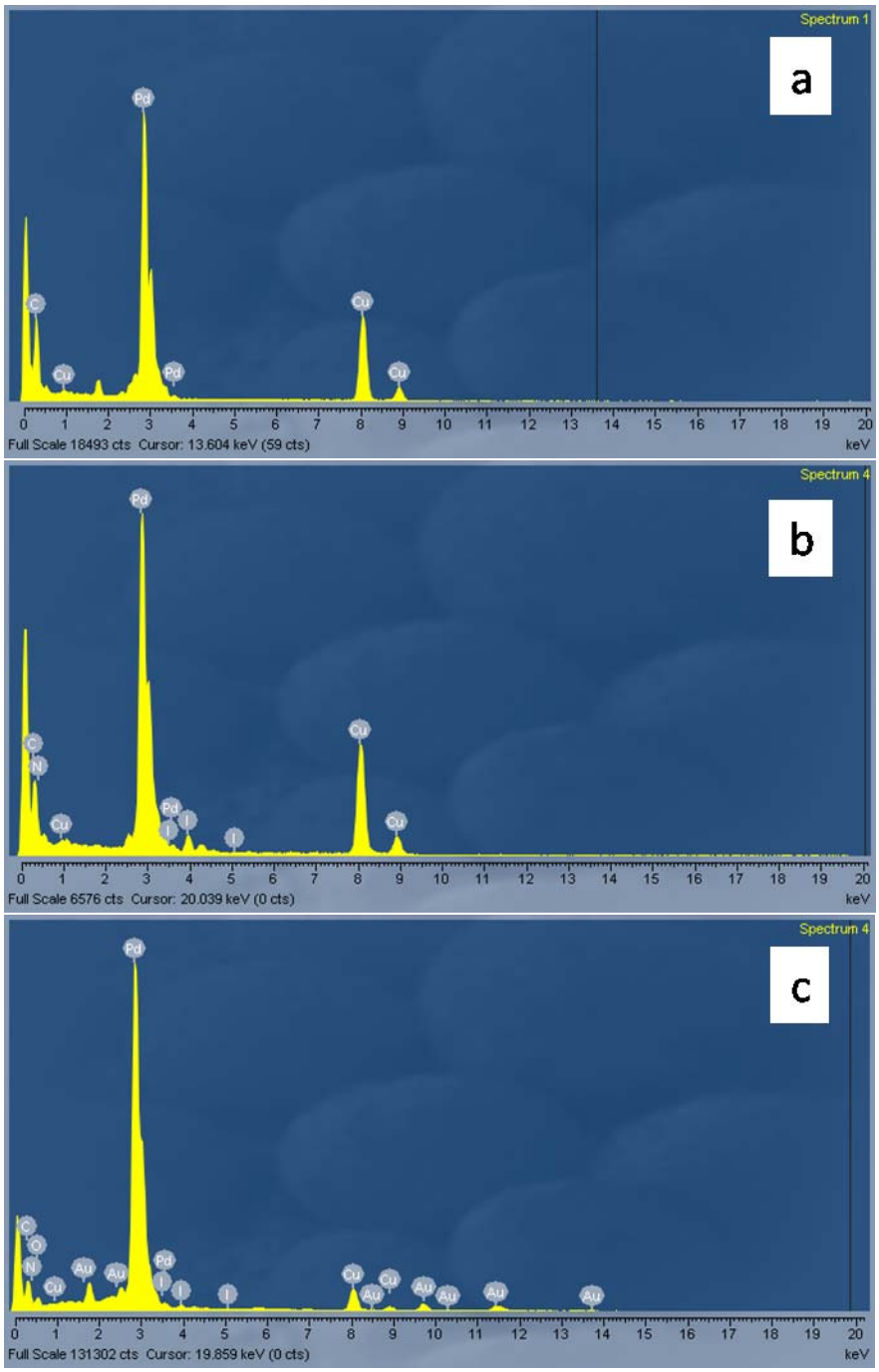


Figure S3. Energy dispersive spectra (EDS) of palladium nanospheres. a) pristine; b) after the 5th recycling in the absence of oxygen; c) after the 10th recycling in the presence of oxygen.

1. Sisoev, G. M.; Matar, O. K.; Lawrence,C. J.; *Chem. Eng. Sci.* ***2005***, 60, 2051-2060.