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Understanding the mechanism of hydrogen uptake at low pressure in Carbon/Palladium nanostructured composites

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2. Experimental

The amount of PdO present in the composites was determined by hydrogen reduction using a home-made glass volumetric system which is schematically represented below (Fig. S1). The system is equipped with a vacuum pump, a pressure sensor and a mass spectrometer.

Prior to the hydrogen sorption measurements, the total volume of the glass system was determined using hydrogen and a glass cell for which the volume is known. Therefore, the volumes of the sample and the cold trap tubes were determined.

For a typical measurement, first the C/Pd composite was placed in the tube and outgased under vacuum at 150°C for one night using a vertical four. After the sample was cooled down, hydrogen was introduced in the system (the valves 1 and 2 are kept closed) with a pressure around 0.7 10^{-2} bar (slightly superior to the pressure corresponding to the reduction of PdO and inferior to that of PdHy formation). The valves 2 and 1 are then opened and during the reaction of PdO with hydrogen, the water formed is condensed continuously in the cold trap tube using a nitrogen bath. When the reaction is finished (no further variations of the pressure) the final pressure is noted, the valve 2 is closed and the hydrogen is evacuated by vacuum pumping.

After complete evacuation, the nitrogen bath is removed and the cold trap tube is naturally warm up at room temperature. By opening the valve 2, the water condensed can be quantitatively analysed by mass spectrometry. It should be mentioned that all the species present in the system are analysed by mass spectrometry during the different steps of the experiment. The quantity of hydrogen consumed can be calculated and further related to the quantity of PdO in the composite. Bulk palladium oxide was used to calibrate the system and the accuracy was found to be ~95 %.

By this way, we could obtain useful qualitative information's about the species present in the system and also the quantitative evaluation of the hydrogen quantity sorbed by the PdO, carbon and C/Pd materials.

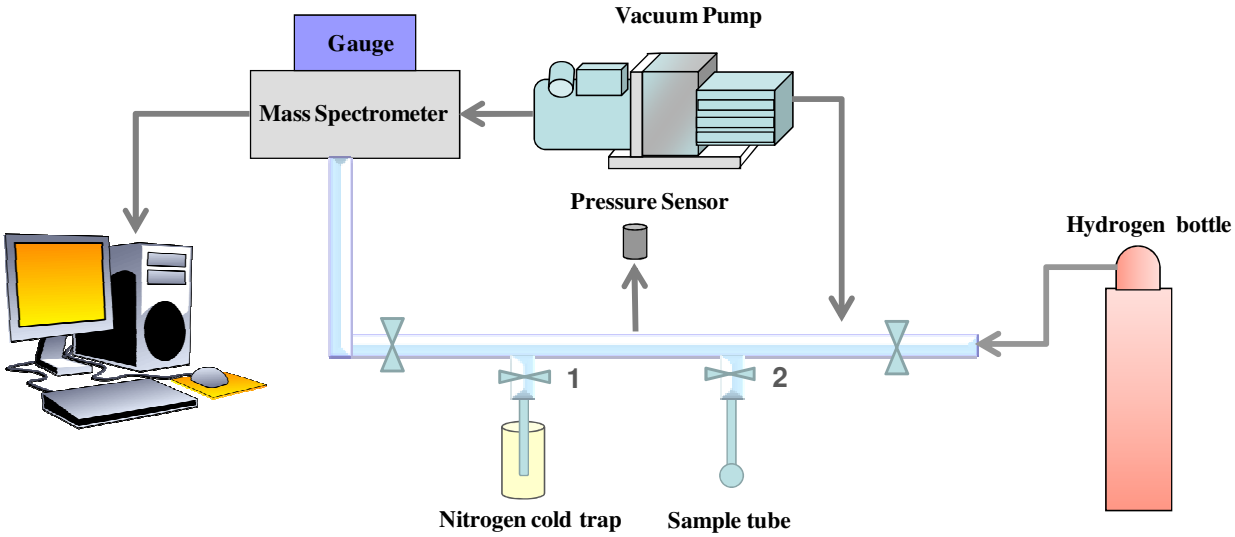

Figure S1. Experimental set-up used to determine the palladium oxide quantity and to monitor the water formed during its reaction with hydrogen

3. Results and discussion

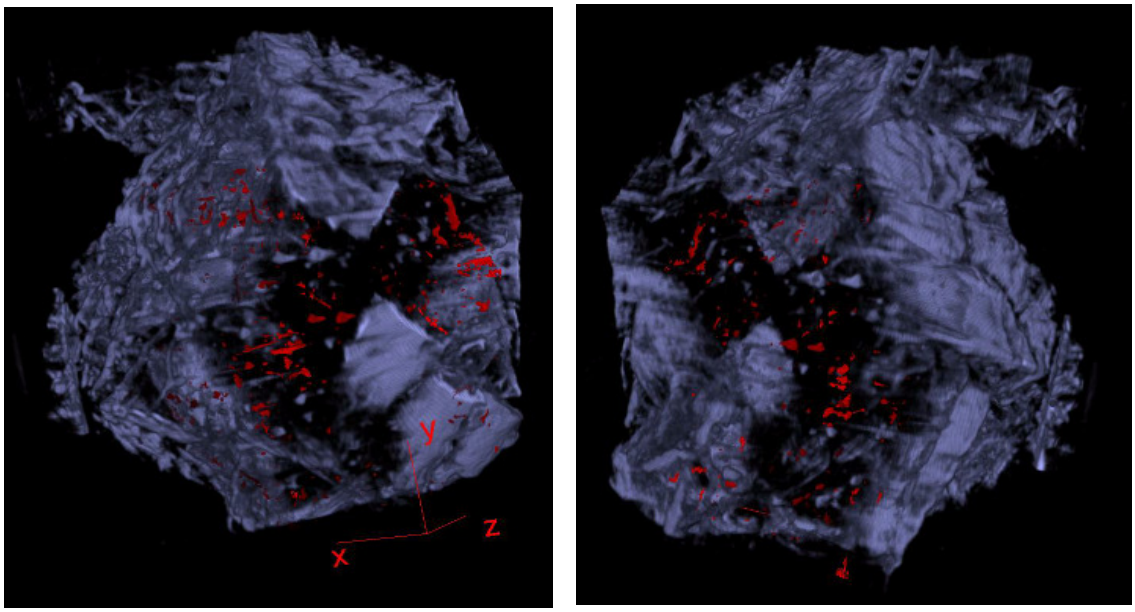

Figure S2: 3D TEM tomography reconstruction pictures of C/Pd1.4 composite

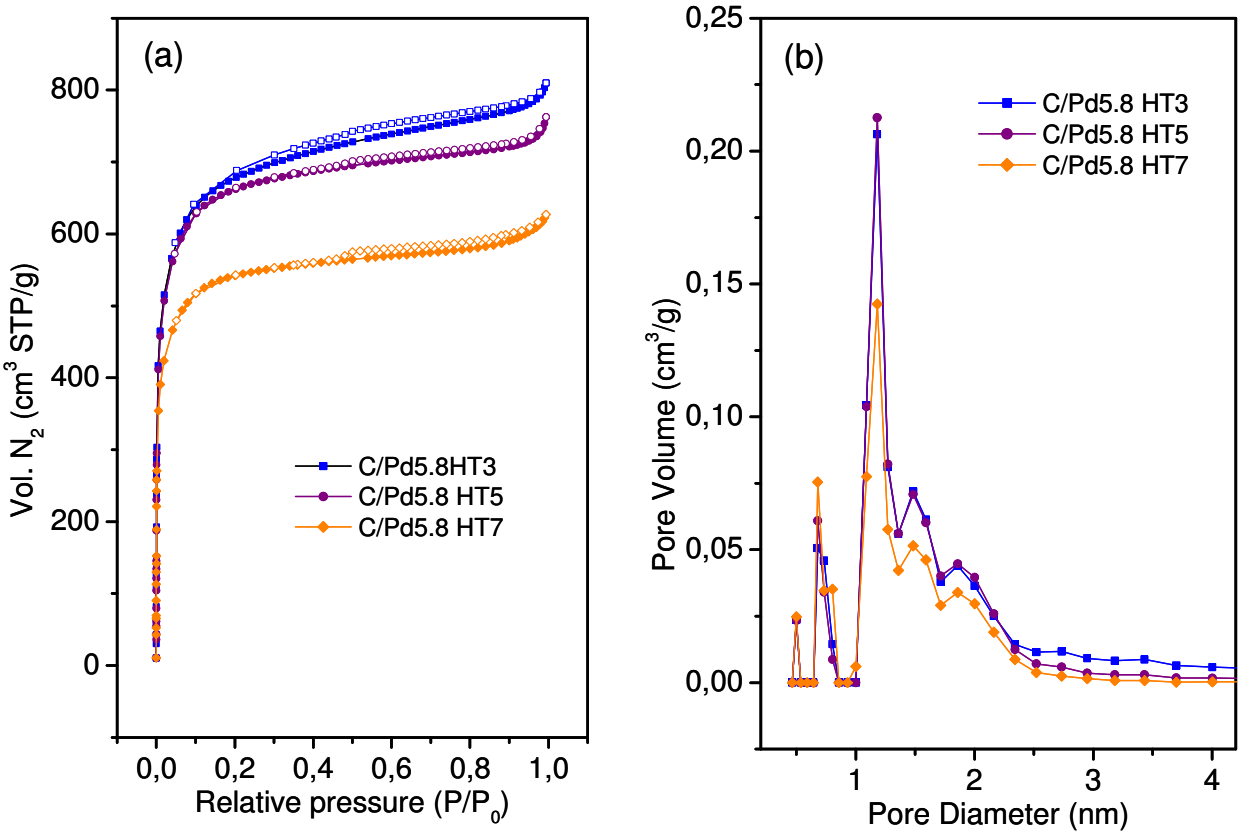

Figure S3. (a) Adsorption-desorption isotherms of N_2 at 77K for C/Pd5.8 composites prepared at 300 °C, 500 °C and 700 °C and (b) the corresponding DFT pore size distribution

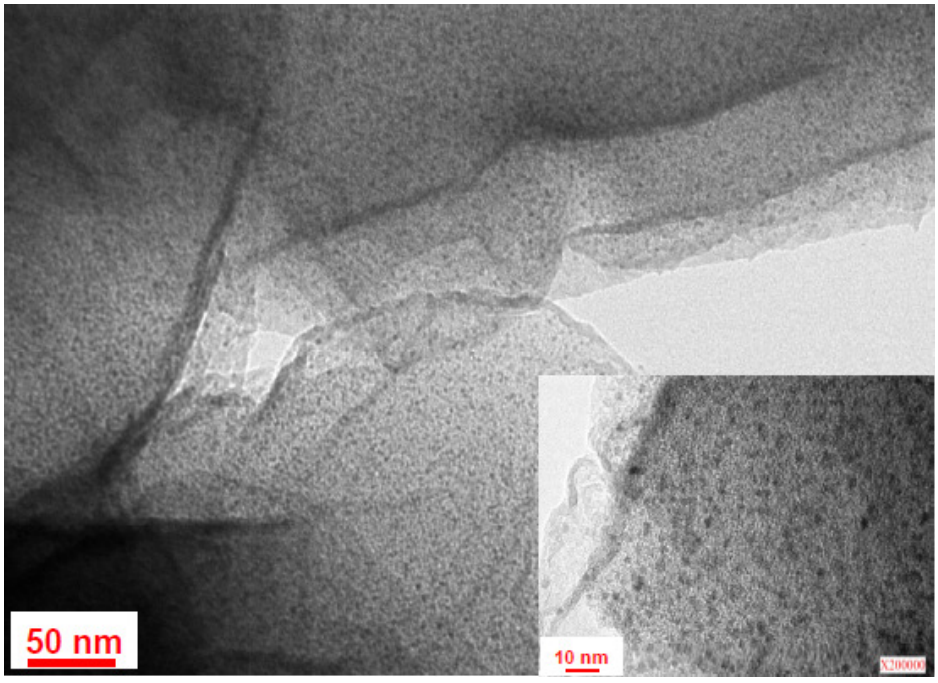

Figure S4: 3D TEM tomography reconstruction pictures of C/Pd1.4 composite

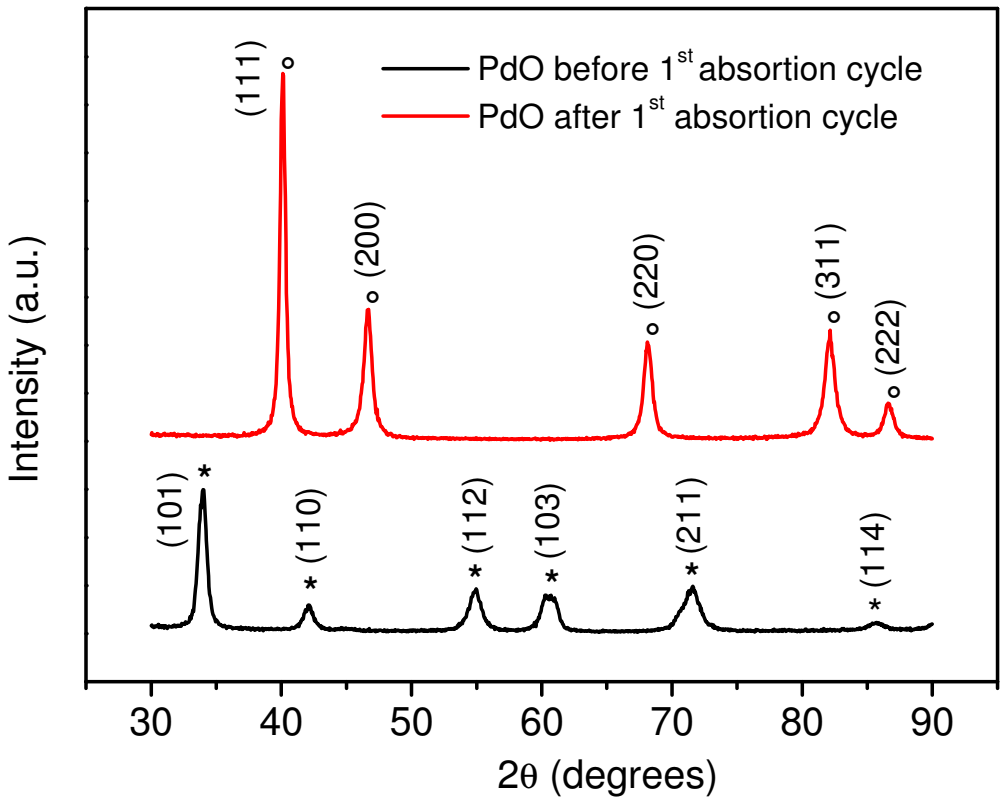

Figure S5: XRD patterns of PdO powder before and after the first hydrogen sorption cycle (* -PdO and °-Pd)