Supporting Information for:

Measurements of Hydrogen Spillover in Platinum Doped Superactivated Carbon

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Maxsorb MSC-30

Maxsorb MSC-30 (from Kansai Coke & Chemicals Co. Ltd.) is a suitable support material for catalyst nanoparticles, essentially equivalent to AX-21 (from Anderson Development Co.) previously used in spillover experiments. Both materials are produced by activation of petroleum coke with molten KOH, using a process patented by Standard Oil Company (later, Amoco Corporation). Both have a surface area near 3000 m²g⁻¹, exhibit similar chemical character and textural morphology, and are classified as "superactivated" or "AX-21 type" carbon. Experimentally The oxygen content of superactivated carbon, a characteristic which may be important to the hydrogen spillover mechanism, is proportional to the BET surface area and is expected to be similar for MSC-30 and AX-21.

Adsorption/Desorption Cycle Measurements

High pressure hydrogen adsorption/desorption cycles were performed for multiple gram quantities of both MSC-30 and Pt-MSC-30, and are shown in Figures S1 and S2. Hydrogen uptake and delivery were identical after many cycles for MSC-30. Hydrogen uptake capacity for MSC-30 at room temperature and 7 MPa was 0.64 wt%. Using the same sample and degassing under vacuum at room temperature for 20 min between cycles, the standard deviation in this value was 0.0003 wt%. Complete desorption to 0.00 wt% at 0 MPa was achieved, with a standard deviation of 0.0006 wt%. Reloading the sample and performing the same adsorption/desorption cycles resulted in combined data which had a standard deviation of 0.003 wt%. This suggests that errors in determining the mass of the dried sample after cycling contributed to a small scatter in the data of different samples. This scatter is within the bounds of the overall experimental error.

Hydrogen uptake capacity in Pt-MSC30 was greatest during the first cycle (after degassing at 573 K under vacuum), yielding the reported value of 0.53 wt% at 7 MPa and room temperature. Upon desorption, a hysteresis was observed. The desorption curve was extrapolated to 0 MPa, yielding a value of 0.02 wt% which could not be desorbed at room temperature for the first cycle. Using the same sample and degassing under vacuum at room temperature for 20 min between cycles, the uptake capacity at 7 MPa decreased to 0.52 wt% for subsequent cycles. Desorption under these conditions was possible to 0.02 wt% at 0 MPa in all cycles. If the sample is instead degassed for 8 h between cycles, uptake at 7 MPa is measured to be 0.53 wt%. In all cases, the amount remaining after desorption is approximately equal to the amount chemisorbed by Pt-MSC-30 at low pressure (0-0.1 MPa). This implies that some hydrogen remains chemisorbed on the surface of the Pt nanoparticles during degassing after short times, but can be removed by evacuating the sample under vacuum overnight.

In the low pressure regime hydrogen uptake data are collected by a high resolution pressure manometer that is then blocked off for measurements above 0.1 MPa. It can be seen that the high resolution data are consistent with the high pressure data for large (~3 g) samples in Figure S3. Together, the data are used to interpolate the point of intersection of the two isotherm curves.

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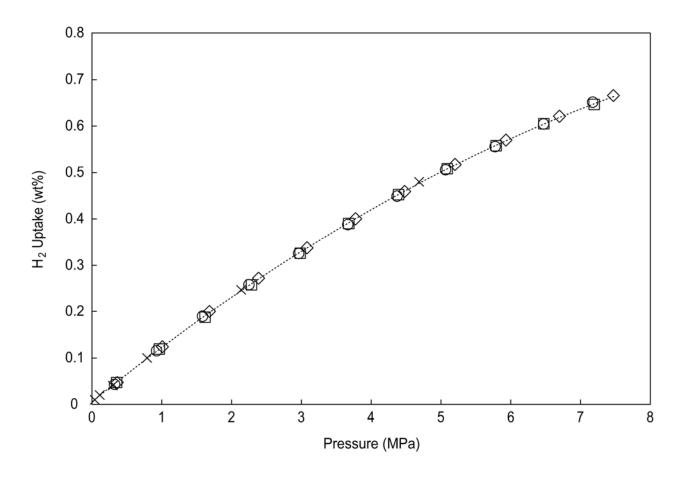


Figure S1. Equilibrium adsorption/desorption isotherms of H_2 on MSC-30 at 296 K (diamonds are first cycle, squares are second cycle, circles are third cycle, and crosses are a typical desorption).

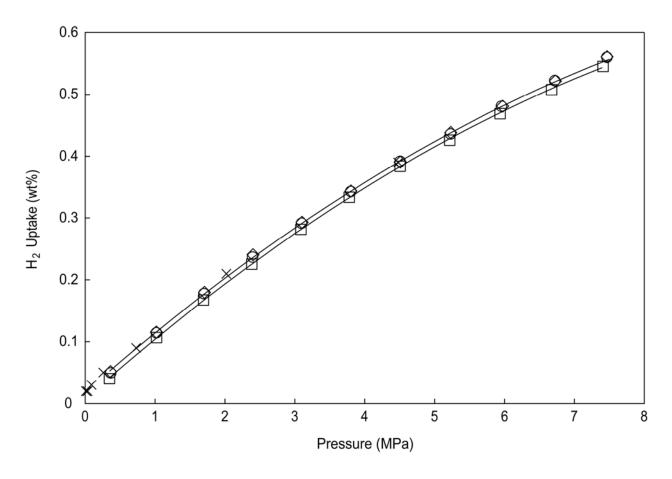


Figure S2. Equilibrium adsorption/desorption isotherms of H_2 on Pt-MSC-30 at 296 K (diamonds are first cycle, squares are second cycle, circles are third cycle, and crosses are a typical desorption). After the first cycle, the sample was degassed under vacuum at 296 K for 20 min. After the second cycle, the sample was degassed under vacuum at 296 K for 8 h.

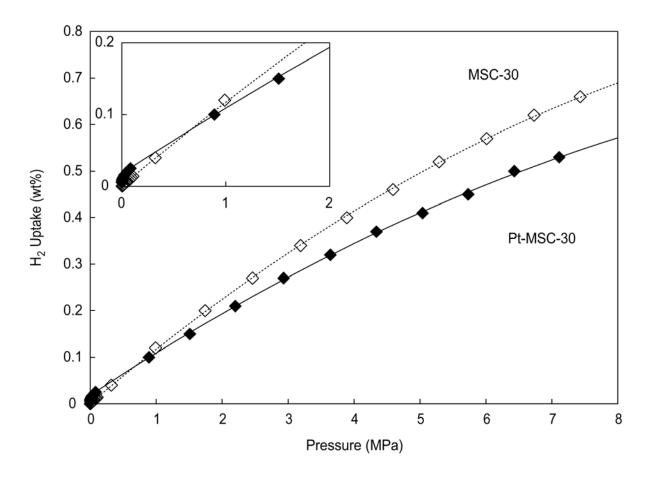


Figure S3. Fitted equilibrium adsorption isotherms for MSC-30 and Pt-MSC-30 at 296 K in mid (inset) and high pressure ranges. The data is interpolated to intersect at 0.7 MPa and 0.08 wt%.