**Shaped Platinum Nanoparticles Directly Synthesized Inside Mesoporous Silica Supports**

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**Figure S1.** $^{29}$Si CP MAS NMR results measured for bare SBA-15, SBA-15 treated with the same synthesis recipe as cubic Pt/SBA-15 except the absence of Pt precursor (SBA-15+MPTMS), cubic Pt/SBA-15. All samples were washed thoroughly before the measurements. ($T^m$ peaks: –C–Si–(OSi)$_m$(OH)$_{3-m}$)

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Figure S2. TEM images of the samples prepared with (a) APTMS+Pt precursor+SBA-15 or (b) hexanethiol+Pt precursor+SBA-15. MPTMS was not added in these cases.

Figure S3. SEM images of the samples prepared using (a) porous SBA-15 after calcination and (b) as-made SBA-15 without calcination.
Figure S4. TEM images of cubic Pt/SBA-15 with different platinum contents of (a) 6.1 wt%, (b) 9.1 wt%, (c) 13.3 wt%, and (d) 17.8 wt%. The amounts of the reactants (SBA-15(mg)/Pt(acac)₂(mg)/MPTMS(ml)) were (a) 50/10/4, (b) 50/15/7.5, (c) 50/25/7.5, and (d) 50/35/7.5, respectively.
Figure S5. TEM images of the samples prepared with different amounts of MPTMS. The volumes of MPTMS added for the synthesis were (a) 2 μL, (b) 3 μL and (c) 6 μL. SBA-15 20 mg and Pt(acac)\(_2\) 4 mg were used in all cases.

Figure S6. BET isotherms and pore size distributions of the samples prepared with (a), (b) SBA-15, and (c), (d) SBA-16.
Figure S7. TEM image of platinum nanocubes directly overgrown onto fumed silica. Fumed silica 50 mg, Pt(acac)₂ 12 mg, and MPTMS 5 μl were used for the synthesis.

Figure S8. Selectivity for pyrrole hydrogenation of cubic Pt/fumed silica at different temperatures. ◆ pyrrolidine; ◇ butylamine.
Figure S9. TEM images of the samples after pyrrole hydrogenation; (a) cubic Pt/SBA-15 and (b) cubic Pt/SBA-16.