*Additional comments on materials synthesis and structure*

XRD of material prior to calcination showed no features.

The co-location of ceria and silica was probed using Energy Dispersive Spectroscopy mapping (Figure S1) within a Hitachi HD2000 Scanning Transmission Microscope. The bright field images (top row) are representative of the particles produced, having distinct shells. The dark field image (center left) show the shells to be relatively homogeneous. However, the intensity of the Pt overwhelms the detector, making collection of the much lower atomic number Si, rather challenging. As can be seen from the EDS map, Ce is clearly clustered around the Pt core. The silicon signal is much weaker due to both the low atomic weight and the lower quantity of silicon (Pt:Si:Ce ratios are 4:1:2 by ICP-OES).


Pt La1   Ce La1   Si Ka1
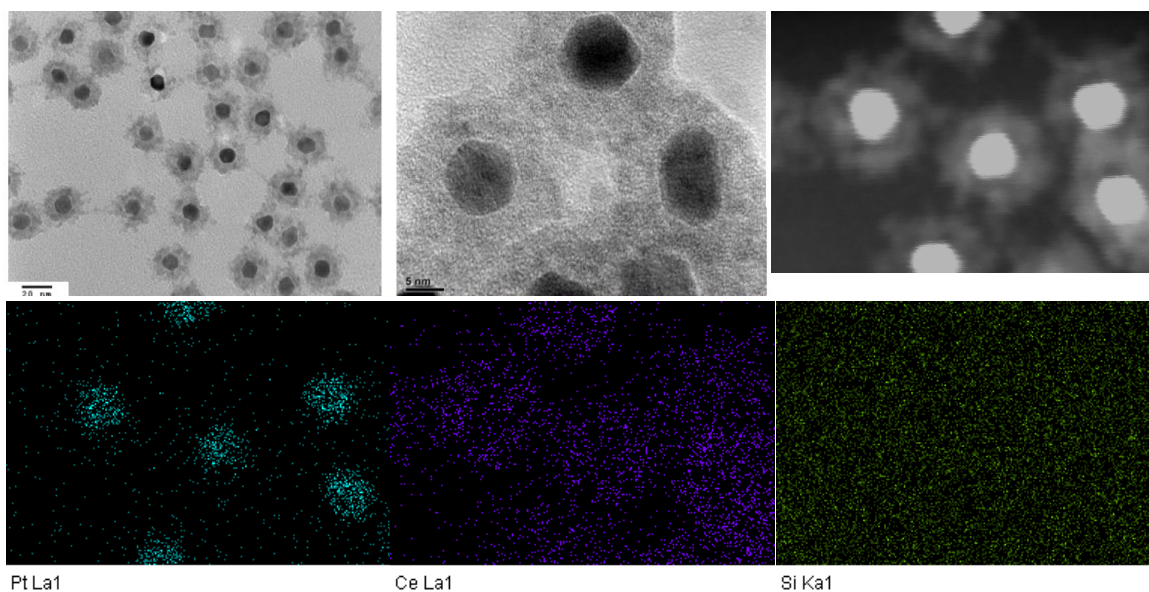
S1. Bright field (top left & center) and dark field (top right) TEM images of Pt@Silica*Ceria. EDS map for Platinum (bottom left), Ceria (bottom center) and Silicon (bottom right) correspond to the dark field image

Calcination of the Pt@silica and Pt@silica*ceria resulted in a fine powder which was easily dispersed due to static charge. To aid in handling, material loss, and most importantly to limit inhalation exposure, the material was deposited onto alumina particles. No effort was made to functionalize the surface of the particles, nor the support to create any sort of bonding between the two. X-ray diffraction of the supported material showed only alumina, and no peaks reflective of the Pt@silica or Pt@silica*ceria particles.

Presence of Surfactant

Thermal Gravitational Analysis of the Pt@silica material showed a mass loss up to the calcination temperature (Figure S2), which is attributed to loss of surfactant. The weight loss is more gradual than the simple decomposition of the surfactant, likely due to the pore structure. Surfactant within the silica pores would prevent access of a probe molecule such as seen in the pulse CO chemisorption of the temperature calcined at 600 °C. The discrepancy between the TEM and Pulse CO measurements can be explained by such pore blockage. The Pt core is measured to be approximately 10 nm by TEM and 36 nm by pulse chemisorption. This would represent a geometric surface area of 314 nm^2 or 4162 nm^2 respectively, suggesting less than 10% of the Pt particle is accessible to the probe CO molecule.
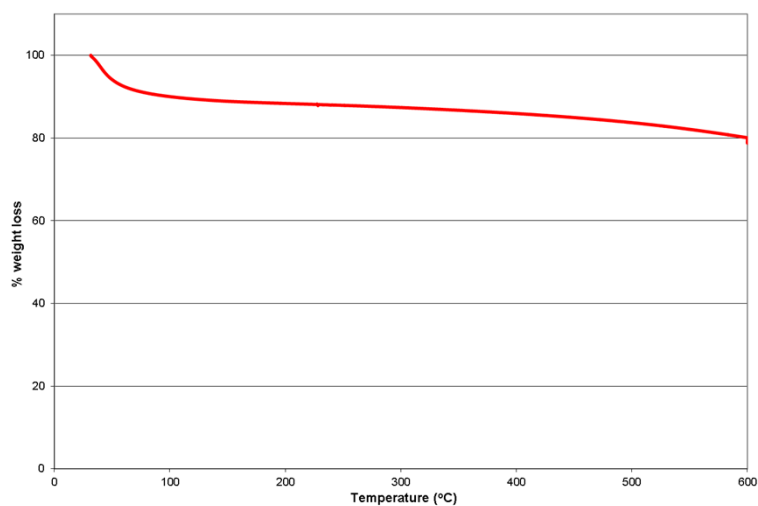
S2. Thermal Gravitational analysis of the unsupported, as-synthesized Pt@silica

Calculation of moles carbon dioxide per mole ceria

The basic reaction for calculation of ceria utilization is $CeO_2 + \frac{1}{2} CO \rightarrow Ce_2O_3 + CO_2$ which would indicate the maximum quantity of carbon dioxide produced from one mole of ceria would be 0.5.