Contribution ID: **46** Type: **Poster contribution**

Mechanism of oxygen storage capacity of nano-ceria: Correlation of in situ XAS, RIXS, XRD and Raman spectroscopy

Thursday, 5 July 2012 10:30 (1h 30m)

Oxygen storage capacity (OSC) is a unique property of ceria (CeO2) allow-ing this oxide to store oxygen in its structure and provide it for catalytic processes under reducing conditions [1]. This property is extensively used in exhaust catalysis. The surface oxygen in ceria is much more active com-pared to the bulk one and its activity can be further enhanced by addition of noble metals (Pt, Pd, Rh). However, the structural reasons for higher activity is still not clear as well as the structure of Ce3+ defects forming in reducing atmospheres. It the present work we studied the structure of well-defined ceria nanoparticles of different size and shape promoted by Pt under differ-ent redox conditions. OSC was determined in pulse experiments and corre-lated to the structural changes observed in situ by Ce K-edge XANES, RIXS across Ce L3-edge, XRD and Raman spectroscopy under identical reaction conditions. The changes in the Ce K-edge XANES indicate reversible change in the oxidation state of cerium (from 4+ to 3+) while RIXS shows unexpectedly large variations in its charge state that can be related to un-usual local geometry of Ce3+ defects. XRD and Raman spectra also show reversible changes suggesting that bulk structure of ceria nanoparticles is af-fected by formation of Ce3+ defects on the surface.

[1] A. Trovarelli, Catalysis by Ceria and Related Materials, Imperial Col-lege Press, London, 2002.

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Session Classification: Poster Session

Track Classification: Catalysis