

Experiment

number:

CH-5639



Experiment title: Understanding the role of the noble metal particle size in Pt/CeO₂ catalysts on the CO oxidation mechanism at low temperature by using HERFD-XANES and XES investigations

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18	Mauro Rovezzi		

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Report:

The aim of this experiment was to gain new insights into the mechanism of low temperature CO-oxidation over Pt/CeO₂ catalysts and its boost after pulses of reductive agents. Two approaches were used to tackle this issue: (1) Monitoring the Pt-CO interaction during CO oxidation light-off for 1%Pt/CeO₂ compared to a 1%Pt/Al₂O₃ reference catalyst, including identification of inhibition effects and (2) investigation of the influence of noble metal particle size and Pt-CeO₂ interface on the Pt-adsorbate interaction and corresponding catalytic activity. In this regard, High Energy Resolved Fluorescence Detected X-ray Absorption Near Edge Structure (HERFD-XANES) is a powerful tool, since it allows the identification of structural changes influencing the gas phase interaction.

For our study, a Si(220) double crystal monochromator was used to tune the incident energy (Pt L3 edge) on the sample and three Ge(660) crystals were used to select the energy of the emitted fluorescence radiation (L α 1 emission line). The beam size on the sample was set to 150 x 300 µm. Spectra were recorded in step scanning mode with an energy resolution of 1-2 eV. A capillary micro reactor heated by a hot air gas blower (Oxford) was used as *in situ* cell. Gases were dosed by mass flow controllers (Bronkhorst). The gas concentration in the

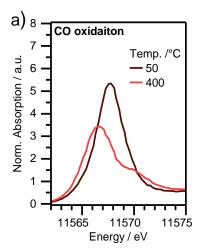


Figure 1: HERFD-XANES spectra of a 1% Pt/CeO₂ with highly dispersed Pt species at 50 °C and 400 °C in reaction mixture containing 1000 ppm CO, 10% O₂ / He

product flow was monitored on-line by a mass spectrometer (Omnistar, Pfeiffer Vacuum) and a Fourier transformed infrared spectrometer (MultiGas[™] 2030 FTIR Continuous Gas Analyzer, MKS Instruments).

Oxidative and reductive *in situ* pre-treatments of catalysts revealed striking differences of the structural response and catalytic activity of Pt/CeO₂ and on the Pt/Al₂O₃. Similarly, the two catalysts behave differently under CO oxidation conditions. Under reaction conditions and oxygen excess (Figure 1a), the structure of platinum behaves similarly to the oxidizing conditions, with a pronounced decrease of the white-line intesity (Fig.1). In general, the results of the performed HERFD-XAS study allowed us to develop a more detailed understanding of the reaction pathway and catalytic activity

of different Pt species on CeO₂ and Al₂O₃. The study is expected to be published soon.