In situ XAFS study of nitric oxide oxidation over Pt-based catalysts

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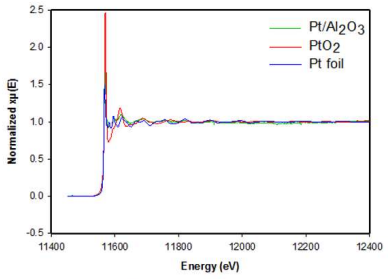
The oxidation of nitric oxide to nitrogen dioxide plays an important role in nitric acid and fertilizer production[1]. The usually homogeneous reaction has been investigated in pertinent literature for heterogeneous catalysts based on noble metals supported on metal oxides.[2,3] The most important deactivation mechanism for such catalysts is reported to be the oxidation of the noble metal e.g. platinum due to excess oxygen and the formation of strongly oxidizing and strongly adsorbed NO_2. It is suggested that the acidity of the catalyst support influences the oxidation resistance of platinum.[5]
We have therefore studied Pt supported on alumina, zirconia and silica, synthesized by impregnation and characterized by nitrogen adsorption, CO chemisorption, TPR and XRF. Catalyst activity has been investigated at elevated nitric oxide concentrations (1 % NO, 6 % O_2), close to industrially relevant conditions for nitric acid and fertilizer production. *In situ* XAS-XRD characterization has been carried out at atmospheric pressure between 300 – 450 °C in an experimental setup built with focus on minimizing the contributions from homogeneous gas phase reactions by mixing the reactants at the capillary reactor inlet. *In situ* X-ray powder diffraction, time and temperature resolved X-ray absorption spectroscopy were used to elucidate the composition and active phase during activation, reaction and deactivation with simultaneous mass spectrometry for monitoring the product stream.
The characterization results will be discussed in connection with catalytic activity data showing the importance of surface acidity, and the corresponding degree of oxidation of the Pt nanoparticles. While the platinum nanoparticles where roughly to 50 wt% in metallic state at ambient conditions, they transitioned to almost complete metal form during reduction before the reaction. During reaction conditions, the platinum was oxidized to roughly 25 wt% PtO_2.



Normalized EXAFS spectra at room temperature

References

1. Honti GD. **1976**. The nitrogen industry, Akademiai Kiado, Budapest.
2. Bourges P, Lunati S, Mabilon G. Proceedings of the Fourth International Congress on Catalysis and Automotive Pollution Control, vol. 1, Brussels, April **1997**, 81.
3. Xue E, Seshan K, Ross JRH. *Appl. Catal. B-Environ*. **1996**, 11, 65-79.
4. Després J, Elsener M, Koebel M, Kröcher O, Schnyder B, Wokaun A. *Appl. Catal. B-Environ.* **2004**, 50, 73-82.
5. Dawody J, Skoglundh M, Fridell E. *J. Mol. Catal. A-Chem.* **2004**, 209, 215-225.

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