



A.

<b>Experiment title:</b> Time resolved in situ EXAFS study of NO/NO <sub>2</sub> storage and release in Pt/SrO model catalyst	<b>Experiment number:</b> CH-525	
<b>Beamline:</b> ID 24	<b>Date of experiment:</b> from: 27/1 - 99                      to: 30/1 - 99	<b>Date of report:</b> 23/2 99
<b>Shifts:</b> 12	<b>Local contact(s):</b> Sakura Pascarelli	<i>Received at ESRF:</i> <b>01 MAR. 1999</b>
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### Report:

A flow reactor system in combination with time resolved dispersive EXAFS was used to study the evolution of stored NO<sub>x</sub> species on Pt/SrO under transient NO/O<sub>2</sub>/H<sub>2</sub>/He gas compositions, for both the Sr K-edge and The Pt L<sub>III</sub>-edge.

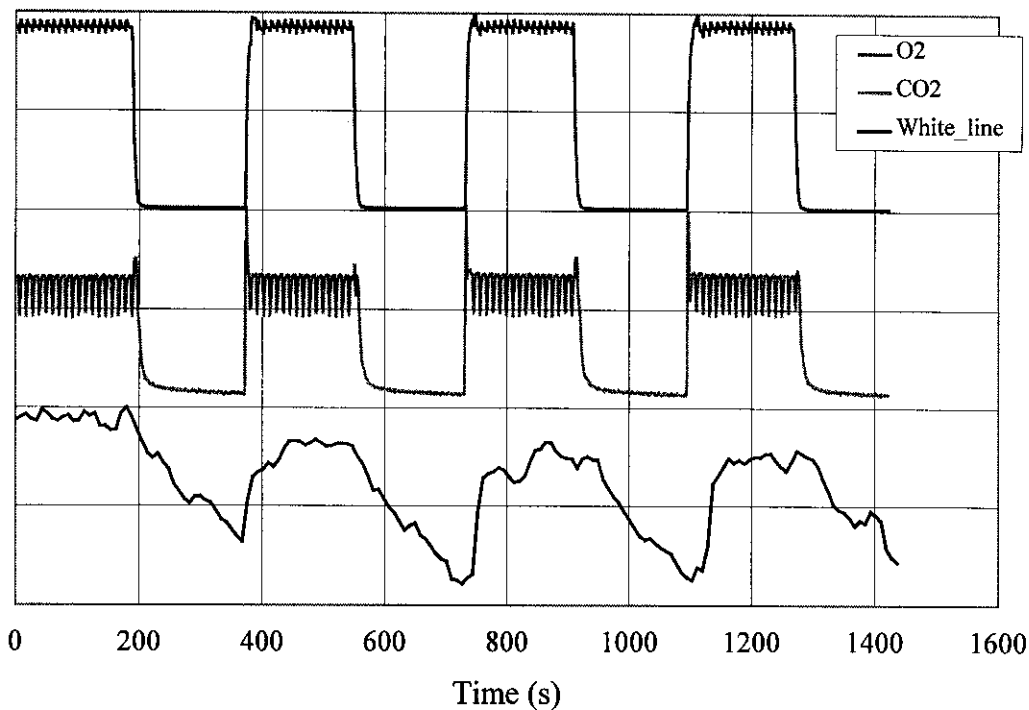
Prior to the experiment we performed some optimisation tests of the catalytic activity for different support materials, like  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> and mixtures thereof. We further optimized the thickness of the active SrO and Pt in order to have a maximum active amount of these compounds. The optimal concentration for SrO 1wt.% and Pt 10 wt.-%, was respectively chosen for the experiment at each edge, and at the same time we were also using 2 wt.-% of Pt when working at the Sr K-edge in order to minimize the absorption in the Pt.

The sample was placed in the reactor cell available at ID24. The experiment were performed at 1 atm, under isothermal condition and at different temperatures in the range of 300-450°C. The different gas mixtures were combinations of: NO (0-2000 vol.-ppm), NO<sub>2</sub> (0-2000 vol.-ppm), O<sub>2</sub> (0-10vol.-%), H<sub>2</sub> (0-3vol.-%), C<sub>3</sub>H<sub>6</sub> (0-2000vol.-ppm), CO (0-3vol.-%), He carrier gas. The resulting gas flow after the catalyst was measured with a Mass-Spectrometer.

As the experiment was carried out in the last days of January of 1999, we have only time to make a brief evaluation of the results.

### Sr K-edge measurements:

The sample was a Pt/SrO on a  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> pressed to pellets. By nature the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> is a porous material with a high surface area in comparison to its weight.



A typical value of 200 m<sup>2</sup>/g is normal and with a particle size of about 100 μm. We tried different ways of pressing the pellet and grinding down the particle size together with a very low focusing of the Laue monochromator. The quality of the EXAFS data was not sufficient to make any normal data treatment. However the data were compared to standard spectra of SrCO<sub>3</sub> and Sr(NO<sub>3</sub>)<sub>2</sub>, both are stable a room temperature, and a clear change can be observed, the data has not yet been examined in respect to this. Further more the near edge structure of Sr did not show a significant change when exposing the material to oxidising conditions, reducing conditions or different NO<sub>x</sub> mixtures.

#### Pt L<sub>III</sub>-edge measurements:

As in the case of Sr K-edge the EXAFS pattern changed slowly with time, probably due to a very slow and a small change of the primary beam in combination with the inhomogenities of the sample. The effect at the Pt edge was smaller than at the Sr edge.

The Pt edge was studied for different catalysts and different gas mixtures. In the figure an example of a synthetic lean burn gas mixture is shown. The catalyst is a Pt/SrO on γ-Al<sub>2</sub>O<sub>3</sub> and the gas mixture is NO (600ppm), C<sub>3</sub>H<sub>6</sub> (600ppm) in He and O<sub>2</sub> is being switched off and on (8%) every 3 minutes. During the reducing part of the period one can see a consumption of C<sub>3</sub>H<sub>6</sub> and NO and during the oxidising part a production of CO<sub>2</sub>, H<sub>2</sub>O and NO<sub>2</sub>. In the figure only CO<sub>2</sub> production is shown.

Note the O<sub>2</sub> signal is changing from 0 % – 8 % (top), CO<sub>2</sub> from 0 % to 0.8 % (middle) and the white line of the Pt L<sub>III</sub>-edge is shown at the bottom

The Pt signal shows a slow decrease during the reducing part of each pulse. The high part of the Pt signal shows a oxidised Pt and the lower parts show a partly reduced Pt.

Furthermore the reduction of the Pt was investigated with different gases CO, C<sub>3</sub>H<sub>6</sub> and H<sub>2</sub> and in combination with NO which reduced the reduction speed.

Samples like Pt on γ-Al<sub>2</sub>O<sub>3</sub> were also tested.