



Experiment title:

In-situ XAS studies on oscillatory behaviour in the CO oxidation on a supported Pt catalyst

Experiment number:
CH 146

Beamline:
8 (ID24A)

Date of experiment:

from: 19-03-1996 to: 25-03-1996

Date of report:
18-04-1996

shifts:
21

Local contact(s):

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Received at ESRF:

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Report:

We have studied bistabilities as well as oscillatory behaviour in the CO oxidation on a 14 wt% Pt Y zeolite model catalyst using the energy dispersive spectrometer on beamline ID24 at ESRF. XAS spectra were recorded at the Pt L_{III} edge using a Si(111) monochromator in transmission geometry.

The catalyst was prepared by ion-exchange of zeolite NaY with Pt(NH₃)₄(NO₃)₂. CO combustion was carried out in a flow reactor provided by ID24. Reaction temperature and exhaust gas composition were continuously monitored via thermocouples directly attached to the catalyst and a quadruple mass spectrometer, respectively.

Temperature programmed resorption (TPD) in a temperature range up to 650 K revealed a multiple-step reaction mechanism for the platinum oxidation, resulting in strongly dispersed oxidized Pt clusters. Figure 1 and 2 show the behaviour of the Pt 'whiteline' together with the reactant partial pressures and the evolution of the Fourier transformed $\chi(k)$ during the activation.

Afterwards, the clusters were reduced with CO at 490 K and the reaction was started by adding an understoichiometric amount of air (T = 520 K). The low active phase of the catalyst

could be determined to consist of Pt clusters blocked by adsorbed CO molecules. The high active phase could be recovered by increasing the oxygen partial pressure, which resulted in an oxidized surface. The evolution of Pt L_{III} edge spectra during several activation/deactivation cycles is shown in Figure 3.

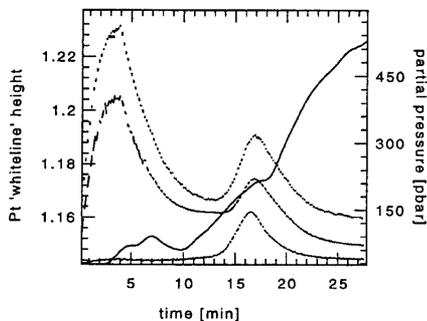


Fig. 1: Change in Pt L_{III} edge 'whiteline' height (solid) during activation of Pt Y zeolite catalyst together with reactant partial pressures (dotted, bottom : CO_2 , center : NH_3 , top : H_2O).

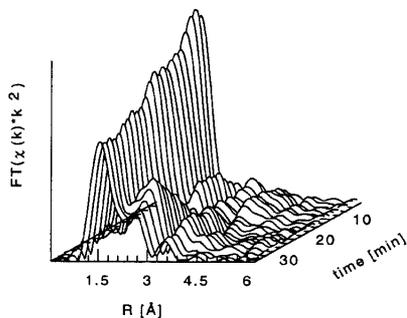


Fig. 2: Evolution of Fourier transformed Pt L_{III} $\chi(k)$ during activation of Pt Y zeolite in air.

Almost no difference could be observed in the Pt L_{III} edge spectra of the catalyst under reaction conditions and under flowing oxygen. Small temperature oscillations (1 K) during reaction (Figure 4) are accompanied by oscillations of the prominent peak position of the Fourier transformed XAFS. Furthermore, an oscillating variation of the degree of oxidation of the Pt clusters could be deduced. Further data analysis is under way.

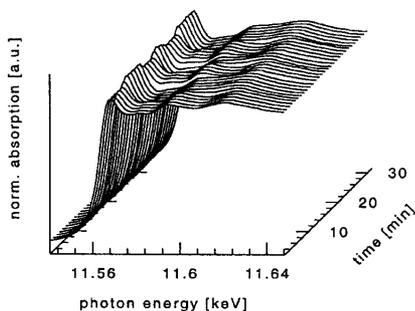


Fig. 3: Evolution of the Pt L_{III} edge spectra during several activation/deactivation cycles.

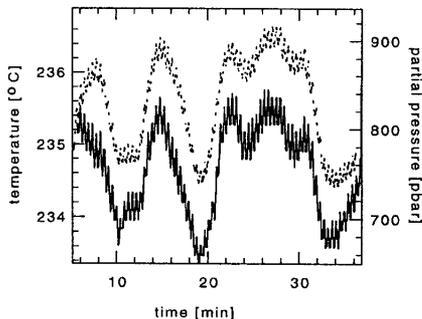


Fig. 4: Temperature (solid) and CO_2 partial pressure (dotted) oscillations during CO oxidation on Pt Y zeolite.