Beamline report CH-5234.

Determination of the mechanism and identification of the active sites responsible for selective anaerobic oxidation of methane into methanol over copper-exchanged mordenite

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The experiments within this beam time where successful allowing to observe in situ the changes in copper oxidation state and geometry during the activation, reaction with methane and reactivation with water. This was possible to do due to the smart design of the gas purification system which included three absorbers for the traces of oxygen, water and volatile organic compounds, preliminary testes in our labs. In addition we also proved the usability of novel XAS-cell, designed by Chupas, which allows heating of the samples placed into quartz capillary with the simultaneous control of the temperature in the catalyst bed using thin (0.5mm) thermocouple. Temperature gradient measurements using IR camera confirmed the isothermicity within the catalyst bed.

In terms of the provision of the X-Rays, the BM26A must be considered as an unqualified success on the bases of these results, though only time will tell what structural details may be obtained through modelling and fitting of this data.

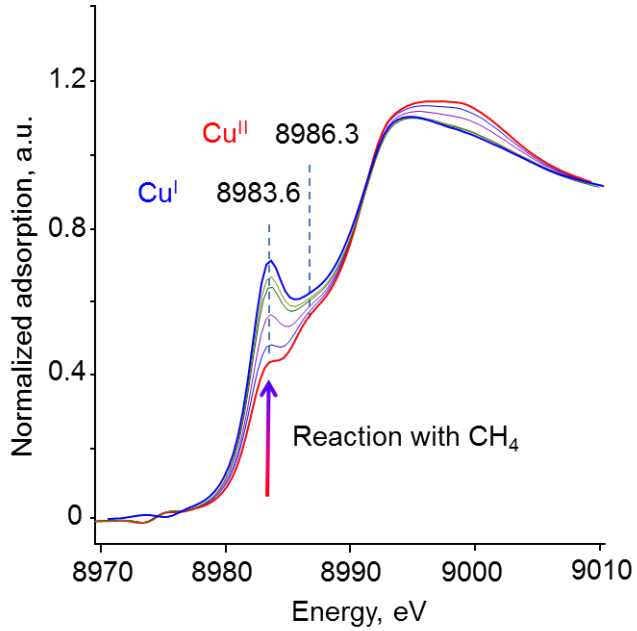

Fig. 1. The XANES spectra obtained during the reaction of copper-exchanged mordenite samples with methane at different temperatures. The development of the peak at 8983.6 eV is clearly observed suggesting the reduction of Cu(II) into Cu(I).

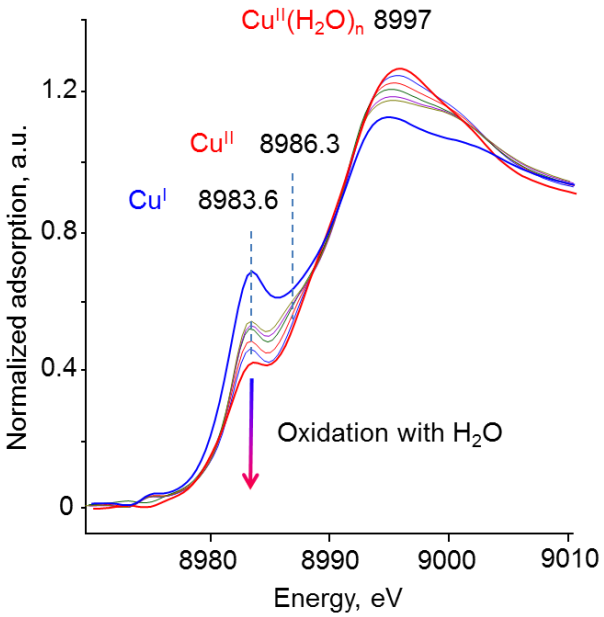

Fig. 2. The XANES spectra obtained during the reaction of methane-reacted CuMOR sample with small doses of water. The decrease of intensity of the peak at 8983.6 eV indicates the oxidation of Cu(I). Simultaneous increase of the intensity of the peak at 8997 eV points to the formation of Cu(II) hydrated species.

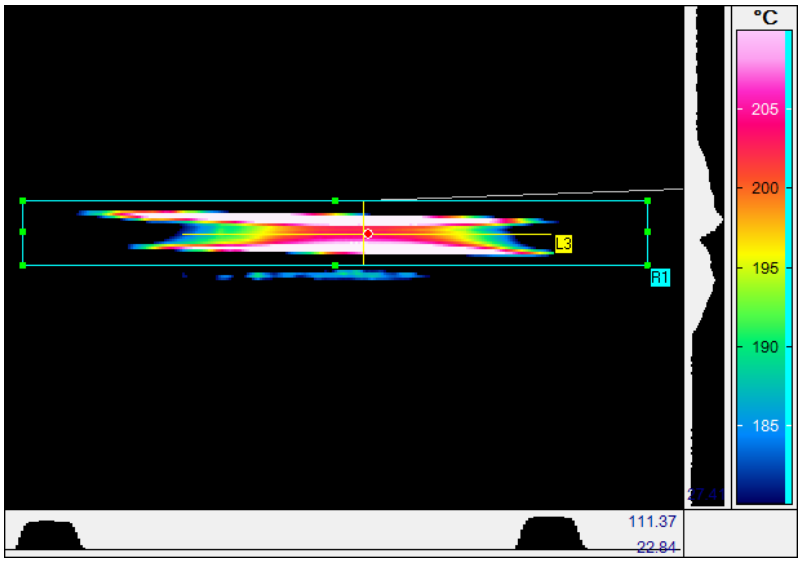

Fig. 3. The IR image of the XAS cell showing the temperature profile along the capillary and catalyst bed.

In further experiments it would be useful to have some opportunities to record on line the temperature values from the cell into the file, containing XAS data or just to the separate file.