Contribution ID: **180** Type: **Talk**

Probing the active site during methane conversion over Cu-MOR with XAS

Friday, 16 September 2011 11:30 (25 minutes)

In the last decade, Groothaert et al. showed that methane can be converted to a methanol-like species at mild conditions using Cu-exchanged zeolites [1]. However, the product strongly sorbed to the surface and the process was not catalytic. To successfully activate methane by partially oxidation to a methanol derivative one must understand the reaction mechanism. This necessitates knowing the structure of the Cu sites that participate in the reaction by determining them during the different reaction conditions. This poses a challenge, since there might be different Cu sites present and not all might participate in the reaction. High energy resolved fluorescence detected X-ray absorption spectroscopy (HERFD XAS) and time resolved XAS (QuickXAS) were explored to monitor the structure of the Cu atoms under different gas feeds.
In situ XAS showed the changes from Cu2+ after oxygen activation to a mixture of Cu2+ and Cu+ states of the copper particles in Cu-MOR upon methane conversion, similar to the reduction of Cu after methanol interaction.

[1] Groothaert, M.H., Smeets, P.J., Sels, B.F., Jacobs, P.A., Schoonheydt, R., Selective oxidation of methane by the bis(u-oxo)dicopper core stabilized on ZSM-5 and mordenite zeolites, Journal of the American Chemical Society, 127 (2005), 1394-1395.

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Heterogeneous catalysis

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Session Classification: Heterogeneous Catalysis

Track Classification: Heterogeneous Catalysis