Insights into the redox mechanism of methane-to-methanol conversion on copper zeolites

<u>A. Knorpp¹</u>, M. Newton¹, A. B. Pinar², J. A. van Bokhoven^{1,2} (1) ETH Zurich, (2) Paul Scherrer Institut <u>amy.knorpp@chem.ethz.ch</u>

Methane is often combusted and flared to the atmosphere when there is no cost-effective alternative, and especially in large quantities at rural petroleum extraction sites¹. One proposed alternative to flaring is to utilize this methane by converting it to more easily transportable products like methanol². Copper exchanged zeolites are able to convert this methane to methanol by a stepwise process³. However, there is debate in the scientific community on the copper speciation and mechanism that is responsible for the partial oxidization of methanol^{4,5,6}.

In this work, in-situ XAS was conducted on a series of copper exchanged zeolites (mordenite and mazzite) during the methane-to-methanol conversion. Changes in the XANES spectra at the copper K-edge were monitored throughout the activation stage and reaction stage for two different stepwise methane-to-methanol treatment procedures (isothermal activation⁷ and high temperature activation³). By monitoring the reduction of copper(II) to copper(I) and comparing it to the methanol production, we are able to quantify the electrons involved in the reduction mechanism for the conversion of methane to methanol.

By this method we conclude that the methane-to-methanol conversion on copper zeolites (mazzite and mordenite) is a two-electron transfer mechanism. The two stepwise procedures (isothermal and high activation temperature) show a similar two electron transfer mechanism.

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