Deactivation behavior of an iron-molybdate catalyst during selective oxidation of methanol to formaldehyde - DTU Orbit (18/11/2018)

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An iron molybdate/molybdenum oxide catalyst (Mo/Fe = 2) was synthesized by a hydrothermal method and the catalyst's performance and compositional changes were followed during selective oxidation of methanol to formaldehyde for up to 600 h. The activity was continuously measured for a series of experiments performed in a laboratory fixed-bed reactor with 10, 100, 250 and 600 h on stream under reaction conditions (5% MeOH, 10% O₂ in N₂, Temp. = 384–416 °C, W/F = 1.2 g h mol MeOH⁻¹). The structural and compositional changes of the catalyst were investigated by a number of techniques including: XRD, Raman spectroscopy, XPS, SEM-EDS and STEM-EDS. Methanol forms volatile species with molybdenum at reaction conditions, leading to depletion of Mo from the catalyst. Excess MoO₃ was shown to volatilize and leave the catalyst during the first 10 h on stream, leading to an initial loss in activity of 50%. From 10 to 600 h on stream leaching of molybdenum from the remaining iron molybdate phase (Fe₂(MoO₄)₃, Mo/Fe = 1.5) leads to iron rich phases (FeMoO₄ and Fe₂O₃, Me/Fe < 1.5) and simultaneously an increase in activity to approximately 1.5 times the initial activity. Even at high degrees of molybdenum loss (Mo/Fe = 0.49) the formaldehyde selectivity remained above 92%, and the combined CO/CO₂ selectivity was below 4%. This is likely due to a surface layer of MoOₓ on the catalyst at all times due to segregation and a surface in equilibrium with the gaseous molybdenum compounds. After 600 h on stream formation of β-MoO₃ was observed, indicating that this molybdenum oxide phase is stable to some extent under reaction conditions.

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Contributors: Raun, K. V., Lundegaard, L. F., Chevallier, J., Beato, P., Appel, C. C., Nielsen, K., Thorhauge, M., Jensen, A. D., Hej, M.
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