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NAP-XPS study of Mo/HZSM-5 under Methane Dehydroaromatization conditions

Content

The direct conversion of methane to value-added chemicals such as methanol, ethers and aromatics is a key-factor in the efficient use of natural gas resources. Methane can be transformed to hydrogen, ethylene, and aromatics by employing molybdenum-modified zeolite catalysts at elevated temperatures [1]. Rapid catalyst deactivation makes this reaction highly dynamic, while in situ reduced Mo oxycarbides and carbides are discussed to be the active species [2]. In this study a laboratory based near ambient pressure X-ray photoelectron spectrometer (NAP-XPS) is used to investigate Mo/HZSM-5 catalysts with a nominal Mo loading of 6 wt.% under operando non-oxidative methane dehydroaromatization conditions. At a reaction temperature of 900 K the evolution of the electronic properties of the catalyst is followed with time on stream under continuous flow of reaction gas (90% CH_4/10% N_2; 2 mbar) revealing a pronounced change of the molybdenum oxidation state. The Mo 3d signal is deconvoluted assuming contributions of Mo^{2+}, Mo^{4+}, Mo^{5+}, and Mo^{6+} using similar peak shapes / FWMH as described by Murugappan et al. [3]. The deconvolution shows a stepwise reduction of Mo^{6+} to Mo^{5+} and Mo^{4+} before a further reduction to Mo^{2+} starts. The obtained results are discussed in connection with activity measurements under ambient pressure conditions and corresponding *ex situ* and *pseudo in situ* XPS results.

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