Deactivating Carbon Formation on a Ni/Al2O3 Catalyst under Methanation Conditions -
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The carbon formation causing deactivation during CO methanation was studied for a Ni/Al2O3 catalyst. Sulfur-free methanation at low temperature (573 K) for various lengths of time was followed by temperature-programmed hydrogenation (TPH) providing information on carbon types involved in the deactivation of the catalyst. Three main carbon hydrogenation peaks were evident from TPHs following methanation: ~460, ~650, and ~775 K. It is suggested that the ~460 K TPH peak was composed of two peaks: a surface carbide peak at 445–460 K, and a peak due to carbon dissolved into the nickel at 485 K based on CO and CH4 adsorption measurements and XRD analysis. The 650 and 775 K temperature peaks are assigned to polymerized carbon structures and the ~775K peak was found to be the primary cause of deactivation as judged by a linear correlation between its amount and the degree of catalyst deactivation. The longer the duration of the methanation test, the more carbon was built up on the Ni surfaces and the highest observed amount was quantified to be as much as eight carbon atoms per Ni surface atom (8 C/Nsurf), which would roughly correspond to an average coverage of four monolayers of graphene. From H2 desorption measurements after reaction the 650 K TPH peak carbon structure is proposed to be partially hydrogenated, possibly resembling polycyclic aromatic-like carbon. The 775 K peak carbon species are likely more graphene-like. Results indicate that although carbon deposition nucleation may be initiated at the most active methanation sites, i.e., the Ni step sites, subsequent growth takes place over Ni terracesites. A strongly inhomogeneous carbon growth distribution over the Ni nanoparticle surfaces could also account for our findings. Similar to suggestions regarding catalyst deactivation in Fischer–Tropsch synthesis, a surface CH* coupling mechanism is likely taking place, and our results suggest these polymeric hydrocarbon species become more ordered, aromatic, and eventually graphene-like over time.

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