Studying the formation of Ni_3C from CO and metallic Ni at T=265°C *in situ* using Ni K-edge X-ray absorption spectroscopy.

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Abstract: Metallic Ni nanopowder (Ni^0) was monitored during 23 h of carburization ($2CO+3Ni^0 \rightarrow Ni_3C+CO_2$, T=265°C) using Ni K-edge X-ray absorption spectroscopy. XRD analysis made afterwards at room temperature revealed 28±3% Ni_3C among 72% unreacted Ni^0. The $\chi(k)$ data recorded during carburization showed small changes at low k indicative of carbon backscattering. The identification of carbon was possible with wavelet transform analysis after eliminating the integral contribution from the unreacted Ni^0 phase using experimental $\chi(k)$ data collected during methanation ($CO+3H_2 \rightarrow CH_4+ H_2O$) at T=265°C. The Fourier transformed $\chi(k)$ data recorded during carburization revealed destructive interference between signals from Ni atoms in slightly different (Ni^0, Ni_3C) environments. The interference effect mainly lowered the peak amplitude of the first two Ni-Ni coordination shells compared to metallic Ni at T=265°C and it propagated very slowly with increasing carburization run time. Simulating the amplitude lowering of the first Ni-Ni peak by destructive interference as a function of the carburization run time, it followed that the carbon atoms migrate into the Ni^0 particle lattice according to the diffusion-induced grain boundary motion advocated in the literature.

Keywords: Carburization; Methanation; *in situ* XAS; Ni K-edge; Wavelet transform analysis; Difference file method; Ab initio simulations

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