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In situ studies on the structure of copper oxide/zinc oxide catalysts

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Abstract:
Cu/ZnO supported on alumina is a well-known catalyst for steam reforming of methanol. In this work it is attempted to assess the influence of elemental composition on the resulting active copper phase. XAFS measurements of calcined precursors were carried out at the Cu K edge and the Zn K edge. Corresponding RDF show that both copper oxide and zinc oxide exhibit considerable deviations from a linear dependence of their structure on the composition coinciding with changes in phase composition of hydroxycarbonate precursor. From time-resolved in situ experiments at the Cu K edge the degree of reduction can be monitored using a combination of factor analysis (PCA) and least-squares XANES fitting with suitable reference spectra (e.g. Cu metal, Cu2O, and CuO). It is shown that Cu2O forms prior to Cu. The extent of reduction to Cu exhibited a typical nucleation growth behavior with an enhanced reaction rate for more diluted samples. Adding oxygen to the feed gas leads to the formation of mixed Cu2+ and Cu+ phases accompanied by a complete loss of activity in methanol steam reforming. After switching back to steam reforming conditions the activity is regained.