

Morphology Evolution of AuCl₃ during Its Impregnation and the Activation of Au/Al₂O₃ Catalyst -- EXAFS Analysis

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The morphology of a supported metal catalyst is directly related to its catalytic activity. Among the many currently used tools for morphology characterization, EXAFS analysis provides a good insight via detailed modeling and with in-situ experiment when needed. This is especially true when the active species has been well dispersed within the catalyst. Furthermore, the morphology evolution of the active species during catalyst pretreatment and reaction can also be probed by EXAFS. This provides information on further improvement of catalyst design. In this study, we discuss the morphology evolution of AuCl₃ precursor during the Au/Al₂O₃ catalyst preparation and activation using EXAFS analysis at Au L_{III} edge.

The aqueous AuCl₃ solution used for impregnation has a pH value of 2.4 and is known to contain neutral species AuCl₃(H₂O) and ionic species AuCl_n(OH)_{4-n}⁻ (n = 4, 3, and 2). However, EXAFS analysis shows that Au center is coordinated to three O neighbors after the catalyst is vacuum dried at 298 K. No Cl ligand is presented in the freshly prepared catalyst but the Au-Cl coordination is observed again while heating the catalyst under He in in-situ EXAFS analysis. The Au precursor species obviously interact with surface hydroxyl and shifting the Cl to nearby Al₂O₃ surface. If the freshly prepared catalyst is heated under air or H₂, the appearance of reduced state Au (by the presence of Au-Au coordination at ca. 2.86 Å) occurs at lower temperature than that under He.

Though only reduced Au state presents after the heating under H₂, He or air, the catalyst shows relatively low activity. The activation can be achieved via soaking the catalyst in NH_{3(aq)}, before or after the heating. The extraction of Cl residue from Al₂O₃

surface is considered as the reason of such activation. The activated catalyst can be deactivated again by the addition of trace amount of HCl or HNO₃. The acidity of Al₂O₃ support is considered an important parameter in the catalytic activity. Near-edge XAFS analysis indicates that the Au electron density is lower when the support is acidic. Results in this study suggest that the activity of Au/Al₂O₃ is affected by the electron charge density of the reduced state Au.