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On the origin of Au charging during green methanol synthesis on Au/ZnO An in situ / operando spectroscopy study

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Some Au catalysts, in particular Au/ZnO, have turned out to be highly active and selective for methanol synthesis from CO₂ and H₂, a key reaction for chemical energy storage. Further improvement of their performance entails a molecular scale understanding of their performance. Here we report on reaction induced changes of structural and electronic properties of Au/ZnO, applying a combination of kinetic and time resolved in situ / operando spectroscopies including high-pressure operando FTIR spectroscopy, near ambient pressure X-ray photoelectron spectroscopy (NAP-XPS) and X-ray absorption spectroscopy (XAS) at the O K-edge, in addition to temporal analysis of products (TAP) measurements. Operando DRIFTS measurements revealed that CO formed as side product during methanol synthesis adsorbs primarily on metallic or negatively charged gold sites. NAP-XPS and XAS measurements point to a close correlation between i) the formation of O-vacancies during reaction, ii) the methanol formation rate, and iii) the CO_{ad} coverage on charged sites. TAP titration of Au/ZnO indicated that CO₂ can reversibly refill the O-vacancies created during reaction by CO or H₂. In total, these results demonstrate a close correlation between the formation/replenishment of O-vacancies, charging of Au nanoparticles and the methanol formation activity.

References

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