Theoretical understanding of CO_2 electroreduction on atomically precise gold nanoclusters

Efremov Vladimir, 이재민, 유종석†
서울시립대학교
(jsyoo84@uos.ac.kr†)

Electrochemical CO_2 reduction reaction (CO_2RR) is one of the appealing approaches to mitigate the increasing CO_2 level emissions in atmosphere by using electricity from renewable sources. Stabilized by thiolate ligands, ultra-small atomically precise Au nanoclusters appear as an attractive material for catalytic applications due to the unique composition and properties. Periodical density functional theory (DFT) calculations were performed to explore the CO_2 reduction to CO on $Au_{25}(SCH_3)_{18}$ nanocluster (Au_{25} NC). We found that SCH_3 ligands were partially removed under the reducing potential, and this significantly improved the catalytic activity of Au_{25} NC. DFT investigations revealed an active site for CO_2RR as the dethiolated Au atom in staple motifs. Moreover, our results demonstrated that moderate upshift of the d-states and increased charge density of dethiolated Au site facilitates the CO production by significantly stabilizing COOH* compared to CO*.