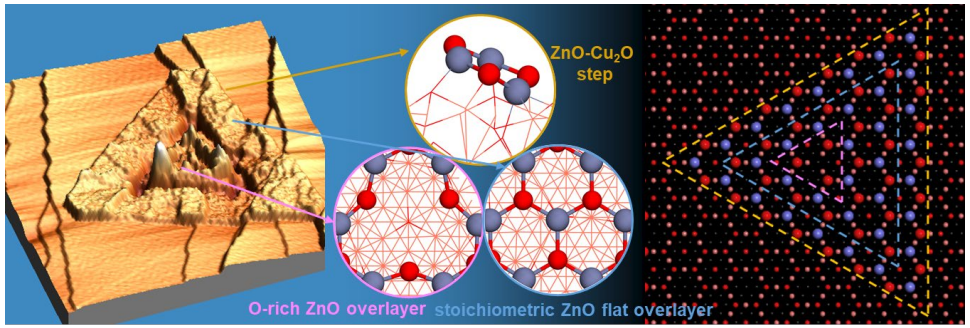
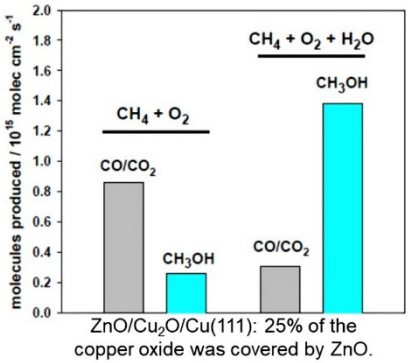
Site-dependent Methane to Methanol Conversion on $ZnO/Cu_2O/Cu(111)$ Catalysts

Multiple sites on $ZnO/Cu_2O/Cu(111)$ catalysts were observed by STM (top left) and simulated by DFT (top middle) and KMC (top right) to identify the active sites during methane oxidation.



Varying methanol selectivity (right) is attributed to different reactive sites that become important in different gas mixtures.



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Scientific Achievement:

The model catalyst $ZnO/Cu_2O/Cu(111)$ is found to enable direct methane-to-methanol conversion at 450 K. The product distributions depend on multiple reactive sites whose roles change in different reaction conditions.

Significance and Impact:

Conversion of abundant natural gas methane into value-added methanol is desirable but challenging. This work shows that $ZnO/Cu_2O/Cu(111)$, a model Cu-ZnO catalyst, is selective for CH_4 to CH_3OH conversion under several reaction conditions. The results reveal new functionality for Cu-ZnO catalysts, which are widely used for CO_2 hydrogenation. The site-dependent behavior suggests design strategies for high methanol selectivity.

Research Details:

- Scanning tunneling microscopy (STM) showed multiple potential reaction sites;
- Reactor testing and ambient-pressure x-ray photoemission spectroscopy (XPS) demonstrated methane dissociation at room temperature and conversion to methanol under methane and oxygen, or methane, oxygen and water;
- Density functional theory (DFT) and kinetic Monte Carlo (kMC) simulation identified the ZnO-Cu_2O step and O-rich ZnO layer as active sites for methanol synthesis from methane oxidation by oxygen and a mixture of oxygen and water, respectively.

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