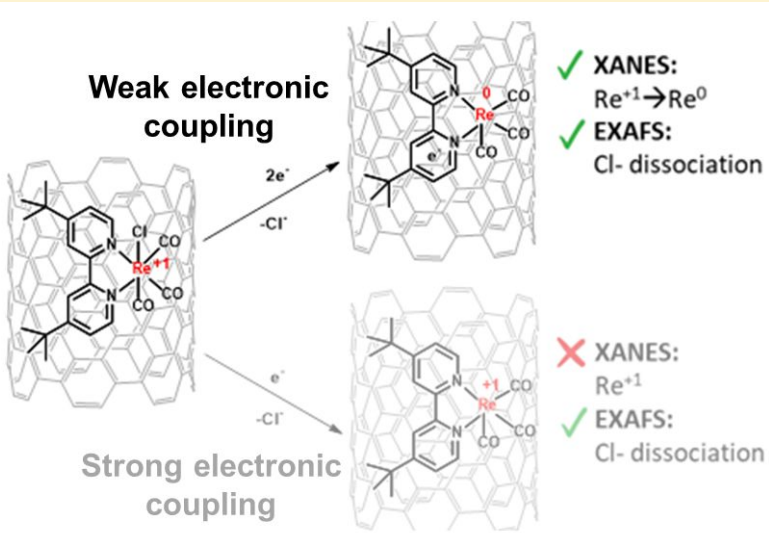
Probing electronic interactions in a highly active heterogenized molecular CO_2 reduction catalyst



The electronic interactions of a molecular catalyst and conductive support were investigated using in situ x-ray absorption spectroscopy. Weak electronic coupling between the initial catalyst species and carbon nanotube support were confirmed from the XANES and EXAFS measured under applied potential.

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Scientific Achievement:

Determined a lack of strong electronic interactions between a molecular CO_2 reduction catalyst and carbon nanotube support, indicating its improved catalytic activity cannot be attributed to strong electronic coupling.

Significance and Impact:

Complicated linkage schemes and strong electronic interactions with the initial catalyst species are not required to improve the activity of heterogenized molecular catalysts.

Research Details:

- Heterogenized Re@CNT electrodes ($Re(bpy)(CO)_3Cl$ catalyst supported by multi-walled carbon nanotubes) were previously found to out-perform the homogeneous catalyst for electrochemical CO_2 reduction.
- X-ray absorption spectroscopy (Re L_3-edge) probed changes in metal oxidation state and ligation under applied cathodic bias to determine if catalyst and support are strongly coupled.

Zoric, M. R.; Chan, T.; Musgrave, C. B.; Goddard, W. A.; Kubiak, C. P.; Cordones, A. A. *In Situ* x-Ray Absorption Investigations of a Heterogenized Molecular Catalyst and Its Interaction with a Carbon Nanotube Support. *J. Chem. Phys.* **2023**, *158* (7), 074703. https://doi.org/10.1063/5.0129724.

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