Graphite-Conjugated Catalysis

The interconversion of electrical and chemical energy requires the coupling of electron transfer with substrate bond rearrangement. This can be achieved at surface exposed active sites of heterogeneous electrocatalysts or via redox mediation facilitated by a homogeneous molecular electrocatalyst. Molecular electrocatalysts yield readily to synthetic alternative of their redox properties permitting systematic tuning of catalyst activity and selectivity. Similar control is difficult to achieve with heterogeneous electrocatalysts because they typically exhibit a distribution of active site geometries and local electronic structures, which are recalcitrant to molecular-level synthetic modification. However, heterogeneous electrocatalysts typically exhibit greater durability and are more readily integrated into functional energy conversion devices such as fuel cells and electrolyzers. We have developed a simple synthetic strategy for conjugating well-defined molecular catalyst active sites with the extended states of graphitic solids. Our latest efforts to develop this new class of catalysts will be discussed.