Electrochemistry is an exceptionally powerful tool in the development of cleaner energy technologies. We have found, though, that by harnessing the inherent activity of biomolecules and microbes, we can improve upon conventional electrochemical systems. Specifically, by combining biomolecular assembly with conventional electrocatalysis, we have improved the specificity and efficiency of electrocatalytic CO2 reduction. Additionally, we have engineered microbial systems for efficient electron transfer to anodes in microbial fuel cells. By combining these microbes with engineered enzymes, we have developed a platform to degrade and electrochemically detect harmful pesticides. The combination of electrochemistry and biomolecular engineering affords advantages beyond the capabilities of either technology alone.