Advances in the theoretical understanding of electrochemical systems have, over the past decade, led to growing use of periodic Density Functional Theory studies to treat a surprisingly large ensemble of electrocatalytic reactions, ranging from carbon dioxide electroreduction to oxygen evolution. Many such studies have employed simplified models of the electrochemical environment to determine reactivity trends across a broad space of catalytic materials, while other efforts have focused on developing detailed descriptions of electrochemical phenomena, such as the structure of electrochemical double layers, on model catalyst structures. An emerging challenge is to combine these approaches to ultimately enable theoretical design of electrocatalysts for reactions of significantly expanded chemical and materials complexity. In this talk, I will begin by discussing how we have applied strategies from computational heterogeneous catalysis to design enhanced electrocatalysts for the classic oxygen reduction reaction. I will then explore challenges in obtaining more detailed descriptions of the electrocatalytic reaction environment, including the structure of catalysts with solid/solid interfaces, the distribution of charges in electrochemical double layers, and the structure and entropy of solvents near the electrocatalyst surface. I will conclude with some perspectives on how these complexities may be incorporated into traditional computational catalyst screening approaches to identify improved materials for more complex electrocatalytic systems.