The race to solve the sustainable energy puzzle is on. First-principles based computational methods, which balance accuracy and computational cost, are at the forefront for solving this puzzle. These computational approaches allow one to describe and understand chemistries of already known materials, and, importantly, they can be used to predict new materials. In this talk, I will first demonstrate how we have been able to computationally predict several new complex catalyst materials including metal oxides, which have been experimentally synthesized, characterized and tested. I then share recent insights on the stability and activity of computationally predicted catalyst systems and conclude with our new efforts to understand the dynamic behavior of these complex materials. Together, this research agenda aims to replace trial-and-error, time-consuming approaches as the main source of development for new materials and chemical routes with a systematic computational materials modeling strategy.