Structures of bioinorganic catalysts can often uniquely rationalize important aspects of chemical and biological reaction control. My research group studies the structural differences between members of large metalloenzyme superfamilies that share common characteristics but trigger different reactions or use distinct cofactors. We have initially focused on systems unified in their ability to activate strong C-H, N-H, or O-H bonds. Key objectives include identification of the outcome-dictating structural features of a given catalyst and structure-guided reprogramming for new function. To achieve these ends, we determine stable reactant and product complexes, with an increasing focus on development and implementation of crystallographic approaches to study metalloenzyme reaction intermediates. These experiments are challenging due to the fleeting and reactive nature of these states but uniquely informative because of the fully contextualized view they provide at critical points in the catalytic cycle.