# Artificial Metalloenzymes: Reaction Scope and Optimization Strategies

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The incorporation of a synthetic, catalytically competent metallocofactor into a protein scaffold to generate an artificial metalloenzyme (ArM) has been explored since the late 1970's. Progress in the ensuing years was limited by the tools available for both organometallic synthesis and protein engineering. Advances in both of these areas, combined with increased appreciation of the potential benefits of combining attractive features of both homogeneous catalysis and enzymatic catalysis, led to a resurgence of interest in ArMs starting in the early 2000's. Perhaps the most intriguing of potential ArM properties is their ability to endow homogeneous catalysts with a genetic memory. Indeed, incorporating a homogeneous catalyst into a genetically encoded scaffold offers the opportunity to improve ArM performance by directed evolution. This capability could, in turn, lead to improvements in ArM efficiency similar to those obtained for natural enzymes, providing systems suitable for practical applications and greater insight into the role of second coordination sphere interactions in organometallic catalysis. Since its renaissance in the early 2000's, different aspects of artificial metalloenzymes have been extensively reviewed and highlighted. Our intent is to provide a comprehensive overview of all work in the field up to December 2016, organized according to reaction class. Because of the wide range of non-natural reactions catalyzed by ArMs, this was done using a functional-group transformation classification. The review begins with a summary of the proteins and the anchoring strategies used to date for the creation of ArMs, followed by a historical perspective. Then follows a summary of the reactions catalyzed by ArMs and a concluding critical outlook. This analysis allows for comparison of similar reactions catalyzed by ArMs constructed using different metallocofactor anchoring strategies, cofactors, protein scaffolds, and mutagenesis strategies. These data will be used to construct a searchable Web site on ArMs that will be updated regularly by the authors.
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