

METALS, MACROCYCLES, AND SMALL MOLECULES: A TALE OF BOND BREAKING AND FORMATION

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Metal-ion redox cooperativity is an underlying presupposed principal for the activation of small molecules by multimetallic cofactors in proteins and by metal surfaces. To delineate the principles that govern this cooperativity requires organic ligands to template specific metal binding sites and enforce metal-metal separations. In particular, there is a limited understanding of the reactivity of metal clusters coordinated by weak-field ligands, which is typical for metal cluster cofactors in biological systems. To that end, we have targeted cyclophanes bearing β -diketiminato arms as our preferred ligand type because of the constraints imposed by the macrocycle defined the inter-metallic distance while simultaneously affording a predictable coordination environment. Results from our ongoing reactivity studies of these multimetallic complexes with dinitrogen, dioxygen, carbon dioxide, and related substrates will be presented.