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Redox-Active Metallamacrocycles and Metallacages Constructed From Alkenyl Ruthenium Entities

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Alkenyl ruthenium complexes $[\text{Ru}(\text{CO})\text{Cl}(\text{P}^i\text{Pr}_3)_2(\text{CH}=\text{CHR})]$ and bis(alkenyl)-bridged diruthenium complexes $[\{\text{Ru}(\text{CO})\text{Cl}(\text{P}^i\text{Pr}_3)_2\}_2(\mu\text{-CH}=\text{CH-arylene-CH}=\text{CH})]$ oxidize at low potentials and form stable radical cations and dications. They are characterized by strong ligand participation to the redox orbitals and a high degree of electron delocalization in their mixed-valent states. In a quest for metallamacrocycles as molecule-based (semi)conductive loops, we have followed two different strategies: i) Interconnecting individual divinylarylene-diruthenium building blocks with dicarboxylate linkers, and ii) utilizing self-complimentary, mononuclear alkenyl ruthenium building blocks with a suitable coordinating functionality. It will be demonstrated that metallamacrocycles designed according to method ii) indeed exhibit electronic through-bond coupling in their mixed-valent states, whereas approach i) provides strongly electrochromic compounds with intrinsically delocalized, but mutually insulated diruthenium subunits. Deceptive effects of hydrogen-bonding, the use of such metallamacrocycles as electron-donors in charge-transfer salts and three-dimensional coordination cages based on interlinked diruthenium divinylarylene complexes will also be discussed.

