High-valent Metal-oxo and Imido Cores in Chemistry and Biology

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Although terminal Co^{IV} -O, Ni^{III} -O and Cu^{III} -O intermediates have been implicated as active intermediates in a number of important chemical transformations, no spectroscopic evidences for the species are available, leaving the pathway uncertain. Evidences of the presence of terminal M-O units (M = Cu(III), Ni(III) or Co(IV)) are to date limited to mass spectrometric studies in the gas phase. Theory suggests that they should be powerful oxidants, perhaps even more reactive than the related $[Fe^{IV}=O]^{2+}$ units that have been extensively studied. In this presentation, we will summarize some of our recent efforts to stabilize the elusive metal-oxo and isoelectronic metal-imido units of Cu(III), Co(IV) and Ni(III)) in solution phase at low temperatures. The high-valent metal-oxo or metal-imido assignments are made on the basis of a variety of spectroscopic methods. The reactivity of the intermediates in hydrogen atom abstraction, oxo transfer reactions and dioxygen reduction reactions are also discussed.