Superoxide reductases (SORs) are non–heme iron enzymes that reduce superoxide ($O_2^-$) to $H_2O_2$ in anaerobic microbes. The iron is redox active, and ligated by four $N^{\text{his}}$ and an $S^{\text{cys}}$. Activity requires that the iron be in the reduced Fe$^{2+}$ state. The mechanism by which SOR reduces superoxide has been controversial, particularly with regard to the number of intermediates involved, and the nature of these intermediates. We have been attempting to sort out the molecular-level details of this mechanism using synthetic models. Superoxide reduction by thiolate–ligated [Fe$^{III}$(SMe$_2$N$_4$(tren))]$^+$ (1) involves two proton–dependent steps and a single peroxide intermediate [Fe$^{III}$(SMe$_2$N$_4$(tren))(OOH)]$^+$ (2) — the first reported example of a thiolate–ligated iron–peroxide. Intermediate 2 is low-spin (S=1/2), displays a $\nu_{O-O}$ at 784 cm$^{-1}$ (that shifts to 753 cm$^{-1}$ upon isotopic labeling with $^{18}$O$^-$), and a coordinated diatomic oxygen ligand with one short, and one long Fe–O distance at 1.86(3) Å, and 2.78(3) Å, respectively, as determined by EXAFS. An external proton donor is required for the formation of 2, ruling out mechanisms involving H$^+$ or H–atom abstraction from the ligand N–H. The initial protonation step affording 2 occurs with fairly basic proton donors (EtOH, MeOH, NH$_4^+$) in THF. More acidic proton donors are required to cleave the Fe–O(peroxide) bond in MeOH, and this occurs via a dissociative mechanism. Reaction rates are dependent on the $pK_a$ of the proton donor, and a common solvent–bound intermediate [Fe$^{III}$(SMe$_2$N$_4$(tren))(MeOH)]$^{2+}$ (3) is involved. Acetic acid releases $H_2O_2$ from 2 under pseudo first-order conditions ([H$\text{HOAc}$]= 138 mM, [2]= 0.49 mM) with a rate constant of 8.2 x $10^{-1}$ sec$^{-1}$ at $-78$ °C in MeOH. Reduction of 3 with Cp$_2$Co regenerates the active catalyst 1. Thus far, 8 turnovers have been achieved.