Supporting Information.

1. **Figure S1.** Fit of absorbance changes for the MMCT(A) band during the oxidation of \( \text{trans-Ru(py)}_4(\text{CNRu(NH}_3)_3)_2^{4+} \). Solid squares are experimental data; open circles are calculated from equation 1 based on the best fit values of absorptivity and comproportionation constants.

2. **Figure S2.** Fit of absorbance changes for the MMCT(A) band during the oxidation of \( \text{trans-Cr([14]aneN}_6)(\text{CNRu(NH}_3)_3)_2^{5+} \). Solid squares are experimental data; open circles are calculated from equation 1 based on the best fit values of absorptivity and comproportionation constants.

3. **Figure S3.** FTIR spectra of \( \text{trans-Ru(py)}_4(\text{CN})_2 \) (upper, light line) and \( \text{trans-Ru(py)}_4(^{13}\text{CN})_2 \) (dark line).

4. **Figure S4.** Correlation of \( RTlnK_c \) with the oscillator strength of the MMCT(A) absorption band.

5. **Figure S5.** Comparison uv-vis-NIR of parent \( \text{LM(CN)}_2 \) complexes (dark line) and their bis-ruthenates (Ru(III)_2; light line): upper, \( \text{cis-Os(bpy)}_2(\text{CN})_2 \); lower, \( \text{trans-Ru(py)}_4(\text{CN})_2 \).
Fit of titration data for $\text{trans-}[\text{py}_2\text{Ru(CNRu(NH}_3)_3\text{H}^+]$ to Sutton/Taube equation for $K_c$. 

![Graph showing absorbance vs. equivalents of Ce(IV)]
Fit of titration data for $\text{trans-}[[\text{[14]aneN}_4]\text{Cr(CNRu(NH}_3)_3\text{)}_2]^{+5}$ to Sutton/Taube equation for $K_c$. 
FT-IR trans-Ru(py)$_4$(CN)$_2$ and trans-Ru(py)$_4$(\(^{13}\)CN)$_2$ (KBr pellets).

FT-IR trans-Ru(py)$_4$(CN)$_2$ and trans-Ru(py)$_4$(\(^{13}\)CN)$_2$ (KBr pellets) / CN-Stretching Region.
The diagram shows absorption spectra for two compounds:

1. **Os^{II} (bpy)_2(CN)_2**
   - Absorptions at 330 (sh), 439 (sh), 358, 476, 394, 579, and 664 (sh) nm.
   - Peak at 827 nm.

2. **[bpy)_2Os^{II} (CNRuA)_{2}] (PF_6)_2**
   - Absorption at 579 nm.

3. **Ru^{II} (bpy)_2(CN)_2**
   - Absorptions at 476 and 685 nm.

4. **[(bpy)_2Ru^{II} (CNRuA)_{2}] (PF_6)_2**
   - Absorption at 685 nm.