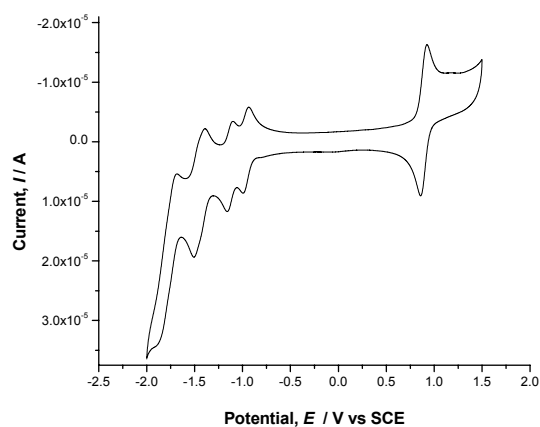
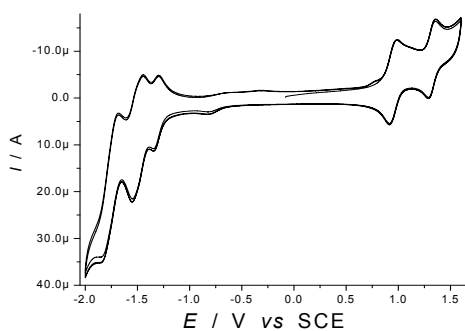


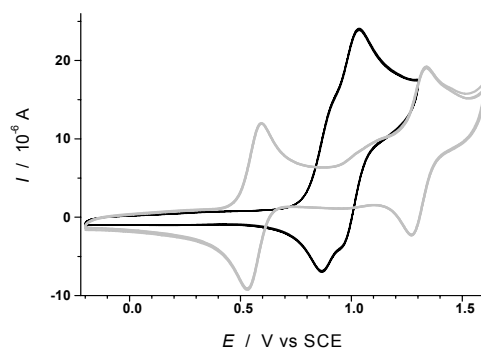
## ELECTRONIC SUPPORTING INFORMATION



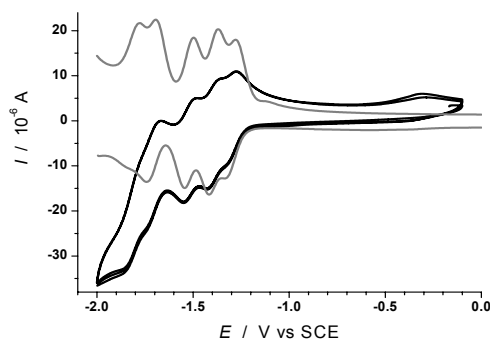
**Fig. S11.** Cyclic voltammogram (CV) of **2** (1mM) in 0.1 M TBAPF<sub>6</sub> CH<sub>3</sub>CN:CH<sub>2</sub>Cl<sub>2</sub> (1:1) at 0.1 V s<sup>-1</sup>.



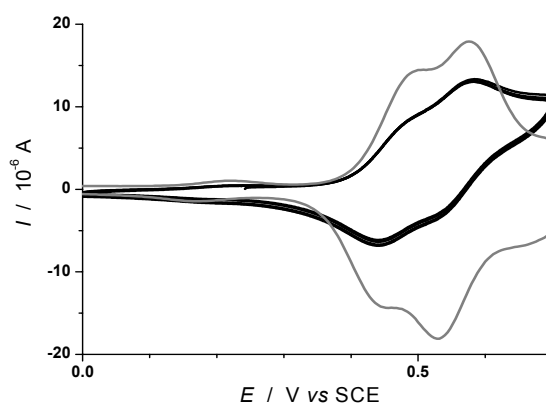
**Fig. S11.** Cyclic voltammogram (CV) of **3** (1mM) in 0.1 M TBAPF<sub>6</sub> CH<sub>3</sub>CN:CH<sub>2</sub>Cl<sub>2</sub> (1:1) at 0.1 V s<sup>-1</sup>.



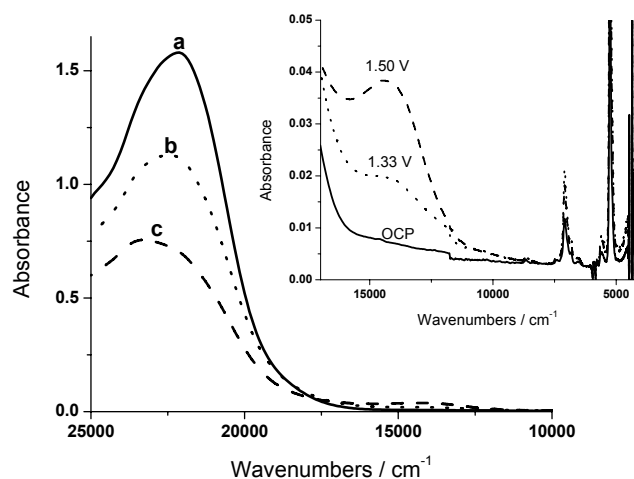
**Fig. SI2.** Cyclic voltammogram (CV) of  $[(\text{bipy})_2\text{Ru}(\text{pytr-bipy})\text{Os}(\text{bipy})_2]^{3+}$  (**4**) (1 mM, black trace) and  $[(\text{bipy})_2\text{Os}(\text{pytr-bipy})\text{Ru}(\text{bipy})_2]^{3+}$  (**5**) (1 mM, grey trace) in 0.1 M  $\text{TBAPF}_6 \text{CH}_3\text{CN}:\text{CH}_2\text{Cl}_2$  (1:1) at  $0.1 \text{ V s}^{-1}$ .



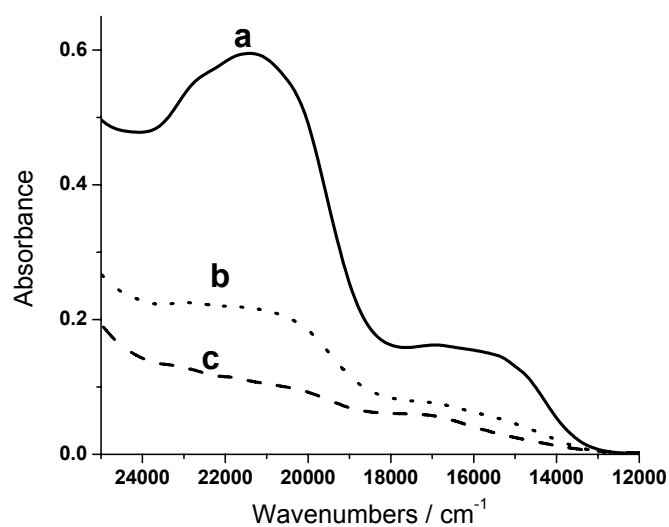
**Fig. SI4.** Cyclic (black trace) and differential pulse (grey trace) voltammograms of **5** (1mM) in 0.1 M  $\text{TBAPF}_6 \text{CH}_3\text{CN}:\text{CH}_2\text{Cl}_2$  (1:1) at  $0.1 \text{ V s}^{-1}$ .



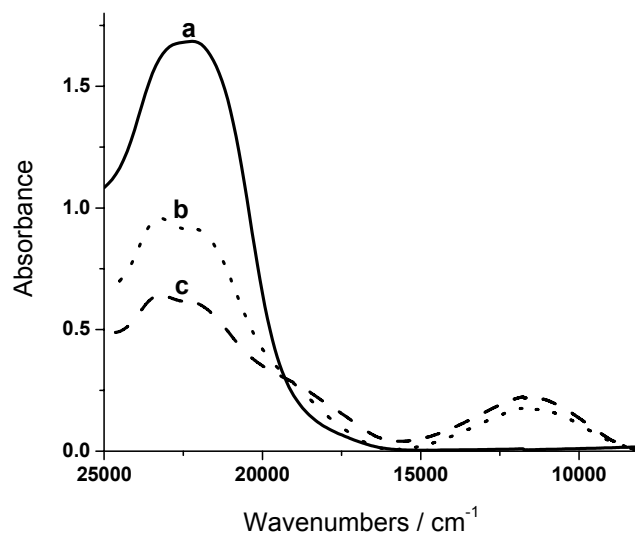
**Fig. SI5.** Cyclic (black line) and differential pulse (grey line) voltammograms of **6** (1mM) in 0.1 M  $\text{TBAPF}_6 \text{CH}_3\text{CN}:\text{CH}_2\text{Cl}_2$  (1:1) at  $0.1 \text{ V s}^{-1}$ .



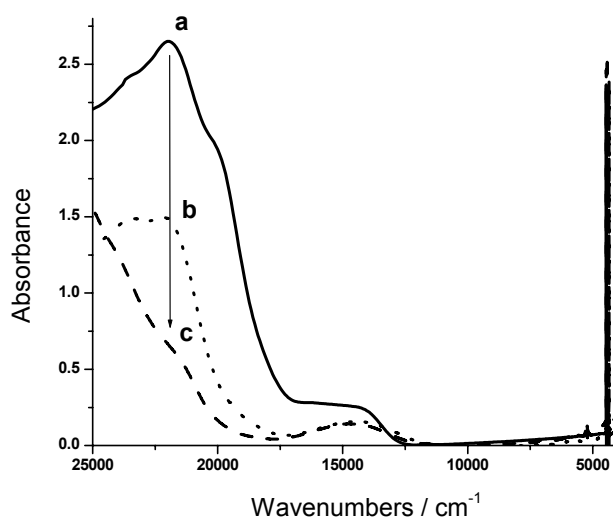
**Fig. SI6.** UV/Vis-NIR absorption spectra of  $[(bipy)_2Ru(qpy)Ru(bipy)_2]^{4+}$  (**1**) in 0.1 M TBAPF<sub>6</sub>/CH<sub>3</sub>CN at (a) E = OCP (open circuit potential) (b) E = 1.33 V [Ru(III)-Ru(II)] and (c) E = 1.50 V [Ru(III)-Ru(III)]. vs SCE. Inset: expansion of NIR region.



**Fig. SI7.** UV/Vis-NIR absorption spectra of  $[(bipy)_2Os(qpy)Os(bipy)_2]^{4+}$  (**2**) in 0.1 M TBAPF<sub>6</sub>/CH<sub>3</sub>CN at (a): E = OCP; (b) E = 0.89 V [Os(III)-Os(II)] and (c) E = 1.20 V [Os(III)-Os(III)] vs SCE.



**Fig. SI8.** UV/Vis-NIR absorption spectra of  $[(\text{bipy})_2\text{Ru}(\text{pytr-bipy})\text{Ru}(\text{bipy})_2]^{3+}$  (**3**) in 0.1 M TBAPF<sub>6</sub>/CH<sub>3</sub>CN at (a)  $E = \text{OCP}$ ; (b)  $E = 1.10$  V [Ru(III)-Ru(II)]; (c)  $E = 1.40$  V [Ru(III)-Ru(III)] vs SCE.



**Fig. SI9.** UV/Vis-NIR absorption spectra of  $[(\text{bipy})_2\text{Os}(\text{pytr-bipy})\text{Ru}(\text{bipy})_2]^{3+}$  (**5**) in 0.1 M TBAPF<sub>6</sub>/CH<sub>3</sub>CN at (a)  $E = \text{OCP}$ , (b)  $E = 0.90$  V [Os(III)-Ru(II)] and (c)  $E = 1.50$  V [Os(III)-Ru(III)] vs SCE.