## **Electronic Supplementary Information**

## Luminescent bis-tridentate ruthenium(II) and osmium(II) complexes based on terpyridyl-imidazole ligand: synthesis, structural characterization, photophysical, electrochemical, and solvent dependence studies

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Compounds				
Protons	1	2	3	4
H(3)	7.62-7.43, m, 4H	7.68-7.43, m, 2H	7.59-7.43, m, 4H	7.61-7.28, m, 4H
H(4)	8.09-8.03, m, 4H	7.96, t (7.7), 2H	8.08, t (7.8), 4H	7.94, t (7.7), 4H
H(5)	7.38-7.26, m, 4H	7.30-7.23, m, 2H	7.29, t (6.6), 4H	7.23, t (6.6), 4H
H(6)	9.11, t (8.0), 4H	9.05, d (8.0), 2H	9.14, d (7.8), 4H	9.11, d (8.2), 4H
H(7)	8.45, d (8.3), 2H	8.50, d (8.2), 2H	8.45, d (8.2), 4H	8.45, d (8.3), 4H
H(7')	7.62-7.43, m, 2H	-	-	-
H(8)	8.58, d (8.4), 2H	8.78, d (7.9), 2H	8.60, d (7.7), 4H	8.55, d (8.4), 4H
H(8')	8.36, d (8.0), 2H	-	-	-
H(9)	-	8.63, t (8.0), 1H	-	-
H(10)	-	8.73, d (8.3), 2H	-	-
H(11)	-	7.68-7.43, m, 2H	-	-
H(12)	-	7.30-7.23, m, 2H	-	-
H(13)	-	7.03, t (7.6), 2H	-	-
H(14)	-	6.09, d (8.2), 2H	-	-
H(3')	9.53, s, 2H	9.65, s, 2H	9.55, s, 4H	9.56, s, 4H
H(3")	9.45, s, 2H	-	-	-
H(Ph)	7.62-7.43, m,	7.68-7.43, m,	7.59-7.43, m,	7.61-7.28, m,
	10H	10H	20H	20H
H(-	2.53, s, 3H	-	-	-
CH <sub>2</sub> )				

**Table S1** <sup>1</sup>H NMR (300 MHz) spectral data<sup>*a,b*</sup> of the complexes **1-4** in DMSO- $d_6$ 

 $a^{a}$  For <sup>1</sup>H NMR data respectively: chemical shift (ppm), multiplicity, *J* (Hz) in parentheses, number of protons. <sup>*b*</sup> Proton numbering shown in Fig. 3



**Fig. S1** ESI-MS (positive) for the complex cations (a)  $[Ru(tpy-HImzPh_3)_2]^{2+}$  (m/z = 577.92) and (b)  $[Os(tpy-HImzPh_3)_2]^{2+}$  (m/z = 623.19) in acetonitrile showing the observed and simulated isotopic distribution patterns.



**Fig. S2** <sup>1</sup>H-<sup>1</sup>H COSY spectra of (a)  $[(tpy-PhCH_3)Ru(tpy-HImzPh_3)](ClO_4)_2$  (1) and (b)  $[(H_2pbbzim)Ru(tpy-HImzPh_3)](ClO4)_2$  (2) in DMSO-*d*<sub>6</sub>.



Fig. S3 UV-vis (a) and luminescence (b) spectra of 3 in different solvents.



**Fig. S4** Square wave voltammograms of **tpy-HImzPh**<sub>3</sub> and complexes **1-4** in acetonitrile showing oxidation in the positive potential window.





**Fig. S5** Square wave voltammograms of **tpy-HImzPh**<sub>3</sub> and complexes **1-4** in acetonitrile showing reduction processes in the negative potential window.