Electronic Supplementary Information

A combined experimental and DFT/TDDFT investigation of structural, electronic, and pH-induced tuning of photophysical and redox properties of osmium(II) mixed-chelate derived from imidazole-4,5-dicarboxylic acid and 2, 2′-bipyridine

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Table S1 Optimized geometry for $\mathbf{1}^{+}$.

	Coordinates (Angstroms)								
	Gas phase/ E(RB3LYP) = -1684.32205691 a.u.			Solution phase/E(RB3LYP) = -1684.41039812 a.u.			Triplet stae in MeCN E(UB3LYP) = -1684.35213481 a.u.		
	X	Y	Z	X	Y	Z	X	Y	Z
Os	0.320944	-0.02072	-0.0433	0.294543	-0.02591	-0.08836	0.281756	-0.04879	-0.10074
N	-3.61627	0.69049	1.563945	-3.60746	0.509895	1.644823	-3.61277	0.360046	1.676327
H	-4.36798	1.009481	2.170513	-4.31407	0.797975	2.315258	-4.32263	0.638196	2.348518
O	-3.03368	-1.41511	-2.20147	-3.09686	-1.4382	-2.23087	-3.0465	-1.63029	-2.17961
N	-1.62078	0.276702	0.68758	-1.63745	0.171769	0.683313	-1.64942	0.066608	0.705737
O	-0.86202	-0.90258	-1.58861	-0.92848	-0.90499	-1.64809	-0.89683	-1.04432	-1.58088
O	-6.19707	0.046596	0.692704	-6.24342	-0.18756	0.730432	-6.23191	-0.44793	0.805164
N	0.245044	1.872172	-0.85351	0.140214	1.908674	-0.79689	-0.07653	1.881255	-0.77723
N	1.312651	1.060209	1.373137	1.266898	1.011758	1.372049	1.217727	1.020083	1.35739
O	-5.39925	-0.98955	-1.19616	-5.37261	-1.09631	-1.17866	-5.35909	-1.34987	-1.10385
H	-4.5322	-1.22535	-1.6976	-4.47167	-1.28122	-1.69123	-4.47455	-1.51158	-1.62403
N	2.027574	-0.66129	-0.99235	2.047979	-0.52885	-1.04243	2.077687	-0.35031	-1.06551
N	0.516409	-1.93998	0.713262	0.602991	-1.95582	0.60513	0.779638	-1.97461	0.532891
C	-2.60569	-0.26101	-0.1556	-2.63824	-0.34092	-0.1495	-2.63455	-0.49758	-0.10846
C	3.875434	-0.42464	-2.54116	3.914837	-0.11289	-2.5293	3.872703	0.305449	-2.54132
H	4.445307	0.221527	-3.20026	4.441921	0.588247	-3.16681	4.307463	1.063548	-3.1823
C	-0.38854	3.482243	-2.5427	-0.57048	3.5946	-2.37877	-1.02537	3.538871	-2.26128
H	-0.84061	3.67299	-3.51018	-1.04825	3.826163	-3.32429	-1.5895	3.756953	-3.16088
C	-3.85597	-0.00718	0.386016	-3.87815	-0.13498	0.445789	-3.87402	-0.31903	0.493261
C	1.501863	-2.71637	0.128979	1.680841	-2.63011	0.05871	1.959232	-2.48543	0.026802
C	-2.26305	0.851614	1.72261	-2.25523	0.685097	1.765209	-2.2696	0.586712	1.780052
H	-1.80271	1.362657	2.55167	-1.77897	1.159184	2.606379	-1.79877	1.095434	2.603886
C	0.701158	4.23206	-0.51199	0.549975	4.25636	-0.33487	0.221492	4.262325	-0.29668
H	1.103633	5.026569	0.106523	0.948556	5.024227	0.317572	0.62402	5.047693	0.333655
C	3.430466	-2.57171	-1.52303	3.645658	-2.29565	-1.52198	3.896954	-1.88803	-1.5083
H	3.664592	-3.62179	-1.39137	3.977764	-3.31683	-1.37842	4.366673	-2.85118	-1.35147
C	2.352513	-1.99619	-0.82601	2.49403	-1.82716	-0.86393	2.682904	-1.57836	-0.87082
C	-5.2583	-0.32159	-0.03827	-5.2572	-0.47331	0.013829	-5.24714	-0.70849	0.081376
C	1.374829	2.436322	1.174857	1.294963	2.393484	1.239731	1.135808	2.432579	1.211722
C	4.200725	-1.78796	-2.3919	4.366204	-1.43791	-2.36458	4.498392	-0.94366	-2.35183
H	5.030441	-2.22433	-2.93856	5.255075	-1.79019	-2.87718	5.434032	-1.17404	-2.85017
C	2.548359	1.33621	3.444653	2.505564	1.212092	3.445853	2.525894	1.277116	3.380449
H	3.001692	0.869056	4.312688	2.969941	0.712485	4.288923	3.056849	0.810374	4.202176
C	1.65114	-4.07447	0.465288	1.949999	-3.96799	0.399336	2.382126	-3.78538	0.356718
H	2.420891	-4.67143	-0.01002	2.796761	-4.48278	-0.03839	3.309012	-4.17802	-0.04289
C	0.761682	2.894362	-0.07714	0.649407	2.899272	0.022529	0.43943	2.893704	0.056773
C	-2.17003	-0.92432	-1.41193	-2.2187	-0.94474	-1.43011	-2.19879	-1.10572	-1.38132
C	-0.30698	-2.51737	1.640161	-0.19901	-2.61408	1.495801	0.01114	-2.75188	1.348924
H	-1.06539	-1.87626	2.071243	-1.02659	-2.05134	1.906592	-0.90429	-2.31107	1.720538
C	-0.31061	2.1697	-2.06818	-0.45203	2.26078	-1.97721	-0.79585	2.218332	-1.89177
H	-0.67861	1.324675	-2.63856	-0.81728	1.444975	-2.58848	-1.16101	1.394465	-2.49479
C	2.004672	3.277682	2.111234	1.914064	3.203557	2.208412	1.760596	3.264367	2.191048
H	2.037426	4.34787	1.93941	1.922197	4.280483	2.08913	1.699943	4.342149	2.086597
C	2.596334	2.734628	3.258761	2.525103	2.616931	3.324717	2.440196	2.707208	3.262417
H	3.085668	3.375716	3.984713	3.005255	3.23355	4.077062	2.910954	3.344623	4.00475
C	2.791777	0.099399	-1.83309	2.762154	0.304264	-1.85877	2.670047	0.57014	-1.88249
H	2.505667	1.137858	-1.93533	2.385951	1.312325	-1.96907	2.158837	1.516127	-1.99798
C	1.90788	0.539248	2.494913	1.875567	0.447356	2.461804	1.919516	0.489835	2.41725
H	1.850889	-0.5362	2.606858	1.840417	-0.63182	2.527326	1.96361	-0.59127	2.470441
C	0.118739	4.534577	-1.75105	-0.06688	4.612673	-1.54287	-0.49899	4.584814	-1.43768
H	0.063584	5.562248	-2.09575	-0.14979	5.65596	-1.82883	-0.66181	5.624339	-1.70611
C	-0.19655	-3.8601	2.010678	0.027957	-3.94213	1.868652	0.387867	-4.04942	1.7079
H	-0.88111	-4.27034	2.745487	-0.6395	-4.41695	2.579206	-0.25143	-4.62851	2.364201
C	0.796896	-4.65939	1.40956	1.119341	-4.63664	1.309603	1.59464	-4.5757	1.205603
H	0.898264	-5.7081	1.669454	1.317271	-5.66907	1.577701	1.911826	-5.57938	1.467903

Table S2 Optimized geometry for **1**.

	Coordinates (Angstroms)								
	Gas phase/E(RB3LYP) = -1683.88366963 a.u.			Solution phase/E(RB3LYP) = -1683.94827221 a.u.			Triplet stae in MeCN E(UB3LYP) = -1683.89552904 a.u.		
	X	Y	Z	X	Y	Z	X	Y	Z
Os	-0.34124	-0.00896	-0.0636	-0.29291	-0.01757	-0.09233	-0.24992	-0.04527	-0.10806
O	0.812539	-0.833	-1.62536	0.925937	-0.86784	-1.65553	0.961642	-0.93371	-1.61964
O	6.22719	0.003874	0.644304	6.278076	-0.18753	0.727311	6.285342	-0.23784	0.826441
O	5.319403	-0.91852	-1.22619	5.348492	-1.04643	-1.15775	5.395835	-1.11117	-1.07002
H	4.452923	-1.11659	-1.71832	4.453359	-1.21961	-1.6669	4.520469	-1.28533	-1.59392
O	2.957324	-1.32674	-2.29828	3.087321	-1.3866	-2.25281	3.126502	-1.45056	-2.20819
N	-0.32152	1.856684	-0.89549	-0.15971	1.908182	-0.80302	-0.02014	1.907736	-0.75035
N	-1.26001	1.08115	1.384282	-1.2526	1.014695	1.375289	-1.24066	0.947652	1.377072
N	-0.42714	-1.90913	0.725795	-0.55799	-1.94445	0.609993	-0.62998	-2.0071	0.481187
N	-2.05118	-0.73752	-0.9355	-2.04363	-0.56187	-1.03198	-2.05201	-0.42751	-1.05741
N	3.582933	0.732965	1.622089	3.621787	0.536417	1.724949	3.616765	0.498392	1.769704
N	1.58628	0.310179	0.620942	1.63552	0.183727	0.669594	1.652012	0.125708	0.695557
C	0.18457	2.135396	-2.13828	0.420816	2.259923	-1.99092	0.693811	2.304298	-1.84909
H	0.529138	1.276615	-2.70314	0.782409	1.441287	-2.60063	1.140011	1.511239	-2.43831
C	0.259606	3.440801	-2.62607	0.534959	3.592479	-2.39536	0.818537	3.638968	-2.21732
H	0.6801	3.615479	-3.6109	1.004004	3.822386	-3.34587	1.38281	3.904127	-3.10423
C	-0.19499	4.511158	-1.82291	0.04091	4.613887	-1.55597	0.18621	4.637214	-1.40843
H	-0.13444	5.534989	-2.17883	0.12169	5.6566	-1.84496	0.263955	5.686595	-1.67714
C	-0.71835	4.230164	-0.55502	-0.56054	4.259624	-0.34024	-0.52397	4.255758	-0.27881
H	-1.06888	5.035937	0.081	-0.94942	5.028492	0.317221	-1.00143	5.005092	0.34315
C	-0.77715	2.897381	-0.10245	-0.65619	2.902828	0.021722	-0.62884	2.874708	0.075187
C	-1.31477	2.459808	1.184498	-1.28221	2.398287	1.246919	-1.28395	2.357362	1.23124
C	-1.78432	0.576564	2.550655	-1.84871	0.450136	2.474011	-1.86091	0.364286	2.455889
H	-1.72719	-0.49851	2.666803	-1.81279	-0.62935	2.537195	-1.807	-0.71641	2.514204
C	-2.34276	1.386976	3.536716	-2.46347	1.213496	3.467508	-2.51621	1.098918	3.432299
H	-2.73491	0.930539	4.439905	-2.91656	0.71283	4.316298	-2.9828	0.589014	4.267533
C	-2.38037	2.78776	3.348605	-2.48205	2.619931	3.350691	-2.56637	2.52767	3.306688
H	-2.7998	3.4403	4.107754	-2.94968	3.235446	4.111902	-3.07671	3.124571	4.056655
C	-1.86303	3.31464	2.160376	-1.88632	3.20712	2.227158	-1.96119	3.137184	2.218586
H	-1.88049	4.385113	1.986189	-1.89314	4.284492	2.109929	-1.99549	4.215846	2.110251
C	0.463647	-2.42668	1.629432	0.264185	-2.57644	1.50312	0.210816	-2.76373	1.245599
H	1.210362	-1.74015	2.006631	1.07871	-1.98663	1.902576	1.122523	-2.28502	1.578192
C	0.433323	-3.76566	2.022879	0.072169	-3.90735	1.883587	-0.09422	-4.08006	1.601433
H	1.172586	-4.12607	2.730503	0.753876	-4.36078	2.594866	0.599569	-4.6409	2.217251
C	-0.54221	-4.62714	1.476437	-1.00212	-4.63488	1.330905	-1.30432	-4.64921	1.151785
H	-0.57791	-5.67503	1.757684	-1.17148	-5.67078	1.605574	-1.56872	-5.66725	1.417819
C	-1.45853	-4.10576	0.555324	-1.85182	-3.99415	0.418648	-2.16155	-3.88326	0.350923
H	-2.21327	-4.7488	0.11616	-2.68542	-4.53354	-0.01517	-3.0906	-4.30911	-0.0083
C	-1.39079	-2.74772	0.188482	-1.6192	-2.65086	0.06944	-1.80756	-2.56253	0.015088
C	-2.3065	-2.08754	-0.74324	-2.45572	-1.87255	-0.85172	-2.59443	-1.68515	-0.85539
C	-3.3829	-2.7217	-1.3935	-3.5983	-2.37131	-1.50499	-3.80449	-2.05211	-1.4747
H	-3.55634	-3.78128	-1.24161	-3.90179	-3.40141	-1.36011	-4.22247	-3.03823	-1.31369
C	-4.22481	-1.98756	-2.23632	-4.34611	-1.53316	-2.34288	-4.46499	-1.13856	-2.30567
H	-5.05289	-2.46974	-2.7464	-5.22764	-1.90878	-2.85177	-5.39667	-1.41332	-2.78892
C	-3.9721	-0.6087	-2.40997	-3.93127	-0.19509	-2.50785	-3.90209	0.141027	-2.5041
H	-4.59844	0.002153	-3.052	-4.48109	0.492033	-3.14173	-4.38386	0.878079	-3.13623
C	-2.88882	-0.02552	-1.75375	-2.78667	0.252132	-1.84405	-2.70399	0.462277	-1.86396
H	-2.65262	1.023838	-1.87735	-2.43665	1.269878	-1.95517	-2.24141	1.432728	-1.98521
C	2.14507	-0.8389	-1.45275	2.225262	-0.8971	-1.42623	2.26885	-0.96427	-1.38757
C	2.588445	-0.20146	-0.20182	2.642741	-0.3104	-0.14794	2.669973	-0.38642	-0.10084
C	3.826558	0.0653	0.428112	3.876081	-0.09015	0.510391	3.889197	-0.15007	0.569495
C	2.236722	0.860082	1.697401	2.270167	0.681528	1.777312	2.268387	0.651884	1.800463
H	1.725766	1.347465	2.514598	1.746385	1.137582	2.60338	1.730784	1.124473	2.607539
C	5.215055	-0.27182	-0.01887	5.24407	-0.43198	0.059514	5.267395	-0.49014	0.142302

Table S3 Optimized geometry for **1**$^{-}$.

	Coordinates (Angstroms)								
	Gas phase/E(RB3LYP) = -1683.28644505 a.u.			Solution phase/E(RB3LYP) = -1683.45694821 a.u.			Triplet stae in MeCN E(UB3LYP) = -1683.41022077 a.u.		
	X	Y	Z	X	Y	Z	X	Y	Z
Os	0.365403	0.036297	-0.09583	-0.29041	-0.01513	-0.10527	-0.23085	-0.04307	-0.1282
C	-0.94332	2.638032	0.358486	-1.64117	-2.63366	0.050306	-1.79897	-2.55049	-0.01282
C	0.351473	2.967307	-0.22988	-2.49259	-1.83517	-0.83576	-2.59404	-1.66522	-0.85933
C	1.735782	-1.35824	-2.3657	0.405435	2.263664	-1.99697	0.720709	2.364371	-1.77942
H	0.912443	-0.89473	-2.90089	0.728285	1.439233	-2.62074	1.185262	1.588689	-2.377
N	1.757601	-1.09119	-1.01605	-0.14079	1.912044	-0.7913	-0.00387	1.934202	-0.70222
N	-0.96458	-1.51512	0.088624	1.649786	0.128871	0.611692	1.671951	0.084194	0.652831
O	-5.07814	-0.74014	0.48659	5.562341	-1.82579	-0.13334	5.601963	-1.82608	-0.06496
O	-2.83605	-0.22625	-2.74244	2.958922	-1.35295	-2.44481	3.043023	-1.33075	-2.40343
O	-0.77115	0.23316	-1.82829	0.854006	-0.81969	-1.70546	0.922674	-0.84956	-1.66915
N	-2.64097	-2.68933	1.077295	3.652291	0.43175	1.639828	3.643684	0.411468	1.721301
N	1.195238	1.874998	-0.42131	-2.07173	-0.5239	-1.0025	-2.04538	-0.40703	-1.06328
O	-5.40949	-2.69389	-0.72394	6.131336	0.407533	-0.17453	6.17094	0.408311	-0.00052
N	1.554895	-0.48298	1.467268	-1.18121	1.019798	1.407698	-1.2186	0.912438	1.399893
C	-1.99493	3.554125	0.561924	-1.87742	-3.98006	0.38896	-2.15377	-3.87294	0.326448
H	-1.87584	4.589781	0.260124	-2.72696	-4.50554	-0.03132	-3.09801	-4.28708	-0.00691
N	-1.08022	1.307595	0.722009	-0.55996	-1.94162	0.574066	-0.60105	-2.0084	0.426182
C	2.933308	3.381355	-1.19969	-3.99787	-0.11189	-2.41792	-3.90835	0.174212	-2.49206
H	3.945977	3.500514	-1.57321	-4.55887	0.592075	-3.02318	-4.3923	0.915281	-3.11815
C	-2.14045	-1.31756	-0.67496	2.648526	-0.38597	-0.23383	2.695502	-0.42689	-0.1703
C	-1.32766	-2.35957	1.106993	2.295491	0.596829	1.711814	2.288684	0.572891	1.761527
H	-0.63199	-2.71792	1.853318	1.788826	1.047524	2.552649	1.757949	1.025341	2.585782
C	-3.181	-2.00852	-0.02598	3.884824	-0.19195	0.416604	3.912465	-0.21518	0.505575
C	2.690651	-2.1732	-2.96633	0.536883	3.59686	-2.39093	0.832825	3.710161	-2.11545
H	2.626278	-2.36486	-4.03299	0.978091	3.8263	-3.355	1.407524	4.002915	-2.98698
C	3.718293	-2.75359	-2.17744	0.098802	4.622161	-1.5234	0.1746	4.679533	-1.2961
H	4.467933	-3.39661	-2.63006	0.194342	5.665673	-1.80534	0.239374	5.735869	-1.54009
C	3.496009	-1.7772	2.204049	-1.71626	3.217817	2.317737	-1.96187	3.072968	2.289251
H	4.320529	-2.43765	1.953817	-1.69092	4.296886	2.218037	-2.013	4.152829	2.200981
C	0.780339	4.271776	-0.55047	-3.65826	-2.31068	-1.46747	-3.81563	-2.02246	-1.46865
H	0.10034	5.104829	-0.40702	-3.96701	-3.34093	-1.33355	-4.23583	-3.00789	-1.30685
C	-4.69339	-1.82205	-0.13175	5.292756	-0.5658	-0.00457	5.332763	-0.57221	0.108417
C	2.636575	-1.32425	1.179803	-1.17261	2.407126	1.30293	-1.28316	2.320259	1.283797
C	2.187466	-0.53159	3.813381	-2.31601	1.221792	3.543796	-2.46984	1.007776	3.474343
H	1.976975	-0.20613	4.827594	-2.75654	0.721464	4.399599	-2.91904	0.474681	4.30478
C	2.070644	4.493848	-1.04178	-4.42093	-1.45064	-2.26785	-4.47914	-1.10465	-2.28908
H	2.402255	5.496836	-1.29471	-5.31945	-1.80831	-2.75988	-5.41754	-1.37226	-2.76364
C	-2.00436	-0.41079	-1.82842	2.19504	-0.8985	-1.53665	2.275838	-0.90882	-1.49244
C	3.739918	-2.49424	-0.80431	-0.4638	4.26922	-0.28961	-0.5452	4.261087	-0.18395
H	4.506607	-2.93615	-0.17505	-0.80829	5.039771	0.39062	-1.04176	4.989197	0.448212
C	-2.25319	0.898434	1.293068	0.280193	-2.59585	1.436519	0.258594	-2.78248	1.153886
H	-2.33583	-0.13999	1.57367	1.112927	-2.01859	1.816177	1.18568	-2.31251	1.455622
C	2.75187	-1.66596	-0.22793	-0.57904	2.91051	0.063205	-0.63168	2.872406	0.136705
C	-3.21167	3.109814	1.09702	-1.01188	-4.64052	1.270584	-1.27987	-4.65216	1.092828
H	-4.04797	3.795698	1.200075	-1.1836	-5.6784	1.536544	-1.54534	-5.66984	1.36002
C	3.283177	-1.38669	3.528471	-2.29189	2.631296	3.451403	-2.54667	2.435273	3.374079
H	3.937437	-1.73575	4.322104	-2.7122	3.247975	4.238948	-3.05889	3.011256	4.13904
C	2.4665	2.107663	-0.88681	-2.83182	0.312064	-1.77851	-2.70242	0.485806	-1.86444
H	3.089034	1.230803	-1.01971	-2.47415	1.328418	-1.87969	-2.2347	1.453806	-1.98982
C	-3.34781	1.7543	1.460331	0.084749	-3.92911	1.803396	-0.04699	-4.09805	1.507073
H	-4.28211	1.314384	1.789847	0.78217	-4.39933	2.488325	0.661616	-4.67247	2.093018
C	1.358987	-0.11034	2.77833	-1.75988	0.456872	2.518494	-1.81293	0.30163	2.475712
H	0.502877	0.530193	2.953227	-1.75474	-0.62447	2.563151	-1.74149	-0.77915	2.512622

Table S4 Selected molecular orbitals along with their energies and compositions for $\mathbf{1}^{+}$, **1**, and $\mathbf{1}^{-}$

MO	Energy, hartee		(%) Composition					
			Gas			Soln		
$\mathbf{1}^{+}$	Gas	Soln	Os	H_2Imdc^-	bipy	Os	H_2Imdc^-	bipy
HOMO-3	-0.35646	-0.27159	2.41	93.12	3.50	0.20	0.56	99.22
HOMO-2	-0.31348	-0.21320	64.58	8.66	26.73	65.63	7.91	26.20
HOMO-1	-0.30605	-0.20912	61.24	12.52	26.20	64.51	9.92	26.20
HOMO	-0.30085	-0.20277	63.65	10.82	25.51	69.48	7.46	23.05
LUMO	-0.19743	-0.09826	5.05	1.74	93.11	2.73	27.20	70.05
LUMO+1	-0.19015	-0.09258	9.83	1.79	88.36	9.25	7.38	83.36
LUMO+2	-0.17017	-0.09099	1.58	14.29	84.11	4.25	70.10	25.63
LUMO+3	-0.16317	-0.06574	0.75	81.13	18.11	0.811	2.62	96.56
1				$HImdc^{2-}$			$Himdc^{2-}$	
HOMO-3	-0.21494	-0.24974	0.86	98.60	0.52	1.01	96.95	2.03
HOMO-2	-0.20863	-0.20241	51.92	15.89	32.18	62.14	7.92	29.91
HOMO-1	-0.20377	-0.19920	55.81	17.27	26.97	62.54	12.35	25.09
HOMO	-0.19920	-0.19318	57.15	20.8	22.01	68.81	8.67	22.51
LUMO	-0.10674	-0.09236	8.527	1.70	89.76	5.48	3.34	91.17
LUMO+1	-0.10165	-0.08629	11.44	2.10	86.44	10.62	1.64	87.72
LUMO+2	-0.07971	-0.06243	0.77	0.41	98.80	4.09	68.27	27.61
LUMO+3	-0.06840	-0.06121	2.43	0.45	97.10	0.41	23.42	76.16
$\mathbf{1}^{-}$				$Imdc^{3-}$			$Imdc^{3-}$	
HOMO-3	-0.08877	-0.20252	13.12	80.79	6.08	13.12	80.79	6.08
HOMO-2	-0.06101	-0.19107	56.06	13.22	30.73	56.03	13.22	30.73
HOMO-1	-0.05757	-0.18521	56.07	19.16	24.76	56.07	19.16	24.76
HOMO	-0.04362	-0.18152	60.05	19.19	20.75	60.05	19.19	20.75
LUMO	-0.00913	-0.08591	7.30	1.69	90.99	7.30	1.69	90.98
LUMO+1	0.00110	-0.08012	11.48	1.79	86.72	11.48	1.79	86.71
LUMO+2	0.01662	-0.05640	1.10	0.55	98.33	1.10	0.55	98.33
LUMO+3	0.02751	-0.04796	2.56	0.35	97.08	2.56	0.35	97.08

Table S5 Calculated bond distances (Å) and angles (deg) for $\mathbf{1}^{+}$, **1**, and **1** in singlet and triplet States.

	$\mathbf{1}^{+}$			**1**			$\mathbf{1}^{-}$		
	$^1A'$	$^3A'$ (UKS)	$^3A'$ (TD-DFT)	$^1A'$	$^3A'$ (UKS)	$^3A'$ (TD-DFT)	$^1A'$	$^3A'$ (UKS)	$^3A'$ (TD-DFT)
Os_1-N_1	2.038	2.039	2.012	2.034	2.042	2.019	2.038	2.055	2.061
Os_1-N_2	2.066	2.076	2.090	2.057	2.068	2.087	2.051	2.071	2.062
Os_1-N_3	2.073	2.087	2.084	2.067	2.083	2.077	2.060	2.075	2.131
Os_1-N_4	2.058	2.060	2.061	2.060	2.072	2.072	2.058	2.073	2.087
Os_1-N_5	2.089	2.096	2.100	2.083	2.071	2.084	2.073	2.080	2.082
Os_1-O_4	2.168	2.138	2.136	2.156	2.131	2.121	2.125	2.087	2.070
O_4 -Os_1-N_1	172.0	173.5	172.9	171.5	173.7	173.3	170.5	173.8	170.4
O_4 -Os_1-N_2	94.2	96.4	96.5	94.7	96.3	96.2	93.7	95.9	95.2
O_4 -Os_1-N_3	86.2	84.9	85.0	87.2	84.9	85.1	87.9	85.9	88.1
O_4 -Os_1-N_4	89.8	95.0	93.6	92.6	95.3	93.5	92.5	94.7	92.9
O_4 -Os_1-N_5	78.3	77.5	77.3	77.2	77.7	77.5	77.4	78.0	78.6
N_1-Os_1-N_2	78.7	78.7	80.1	78.7	79.6	79.9	78.6	79.3	77.9
N_1-Os_1-N_3	101.0	99.3	98.6	99.6	99.4	98.8	99.9	98.9	99.2
N_1-Os_1-N_4	94.7	92.8	92.9	93.5	90.0	92.4	94.0	89.7	94.3
N_1-Os_1-N_5	97.6	97.3	96.0	97.1	97.0	96.5	96.7	97.6	94.7
N_2-Os_1-N_3	175.7	175.9	176.6	176.3	175.6	176.9	176.6	175.9	172.9
N_2-Os_1-N_4	97.7	97.6	98.4	98.3	97.3	98.8	98.6	97.9	94.6
N_2-Os_1-N_5	88.4	88.2	84.2	88.6	86.5	84.8	88.9	86.8	92.2
N_3-Os_1-N_4	78.5	78.5	78.5	78.4	78.3	78.4	78.4	78.3	78.9
N_3-Os_1-N_5	95.2	95.8	98.9	94.8	97.7	98.0	94.2	97.0	94.4
N_4-Os_1-N_5	167.0	169.0	170.8	168.2	172.3	170.7	167.8	171.7	169.6

Table S6 Mulliken spin density distribution over different moieties for the three complexes ($\mathbf{1}^{+}$, **1**, and $\mathbf{1}^{-}$).

Spin density	$\mathbf{1}^{+}$			$\mathbf{1}^{0}$			$\mathbf{1}^{-}$		
	Os	H_2Imdc^-	bipy	Os	$HImdc^{2-}$	bipy	Os	$Imdc^{3-}$	bipy
One-e^- oxidized	0.9670	0.0223	0.0107	0.9660	0.0355	-0.0015	0.9439	0.0726	-0.0165
One-e-reduced	0.0766	0.0199	1.0567	0.0856	0.0054	1.0803	0.0951	0.0018	1.0934

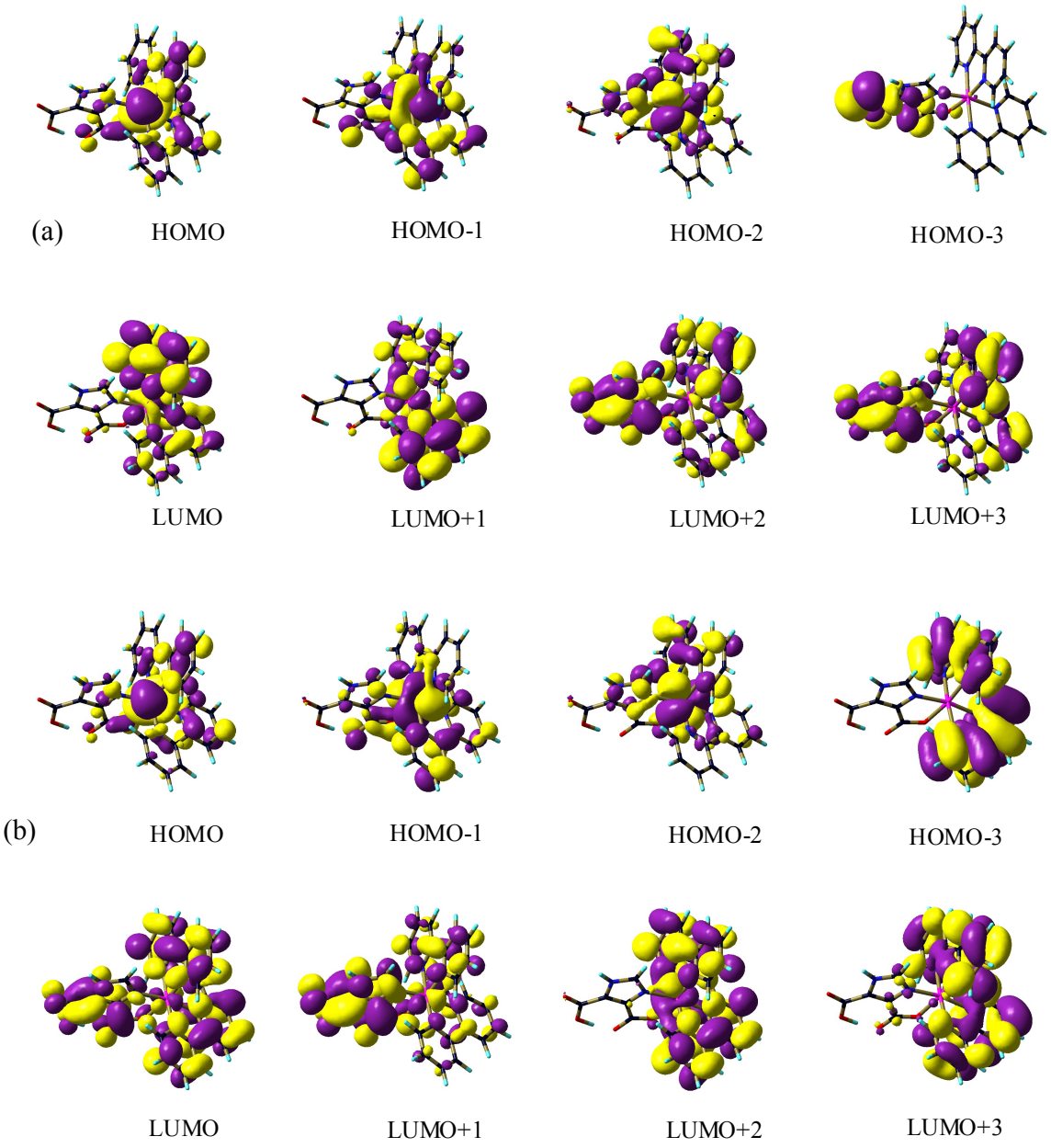


Fig. S1 Schematic drawings of the selective frontier molecular orbitals for $[(bipy)_2Os(H_2Imdc)]^+$ (**1**$^+$) in gas (a) and solution phase (b).

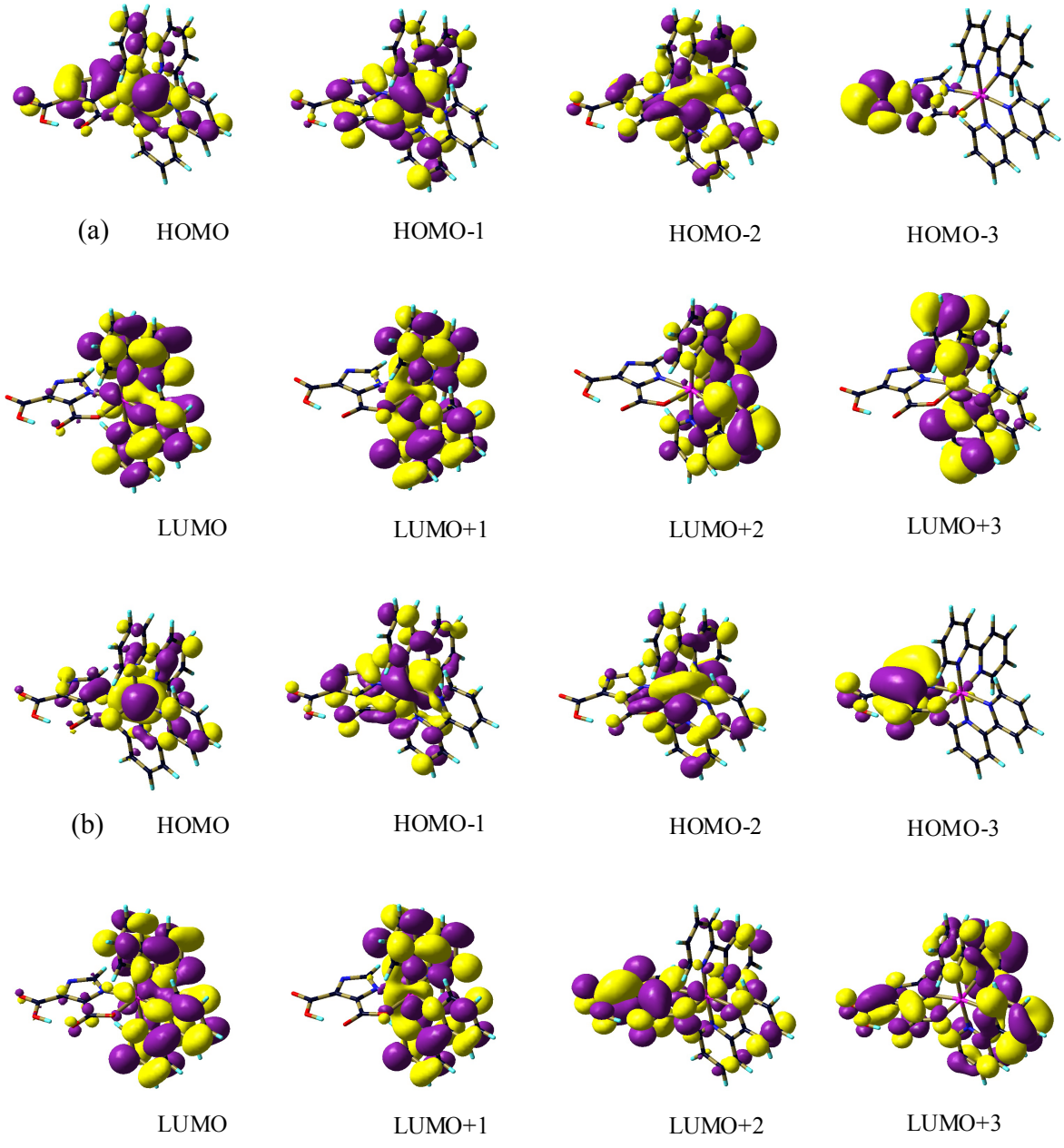


Fig. S2 Schematic drawings of the selective frontier molecular orbitals for [(bipy)$_2$Os (HImdc)] (**1**) in gas (a) and solution phase (b).

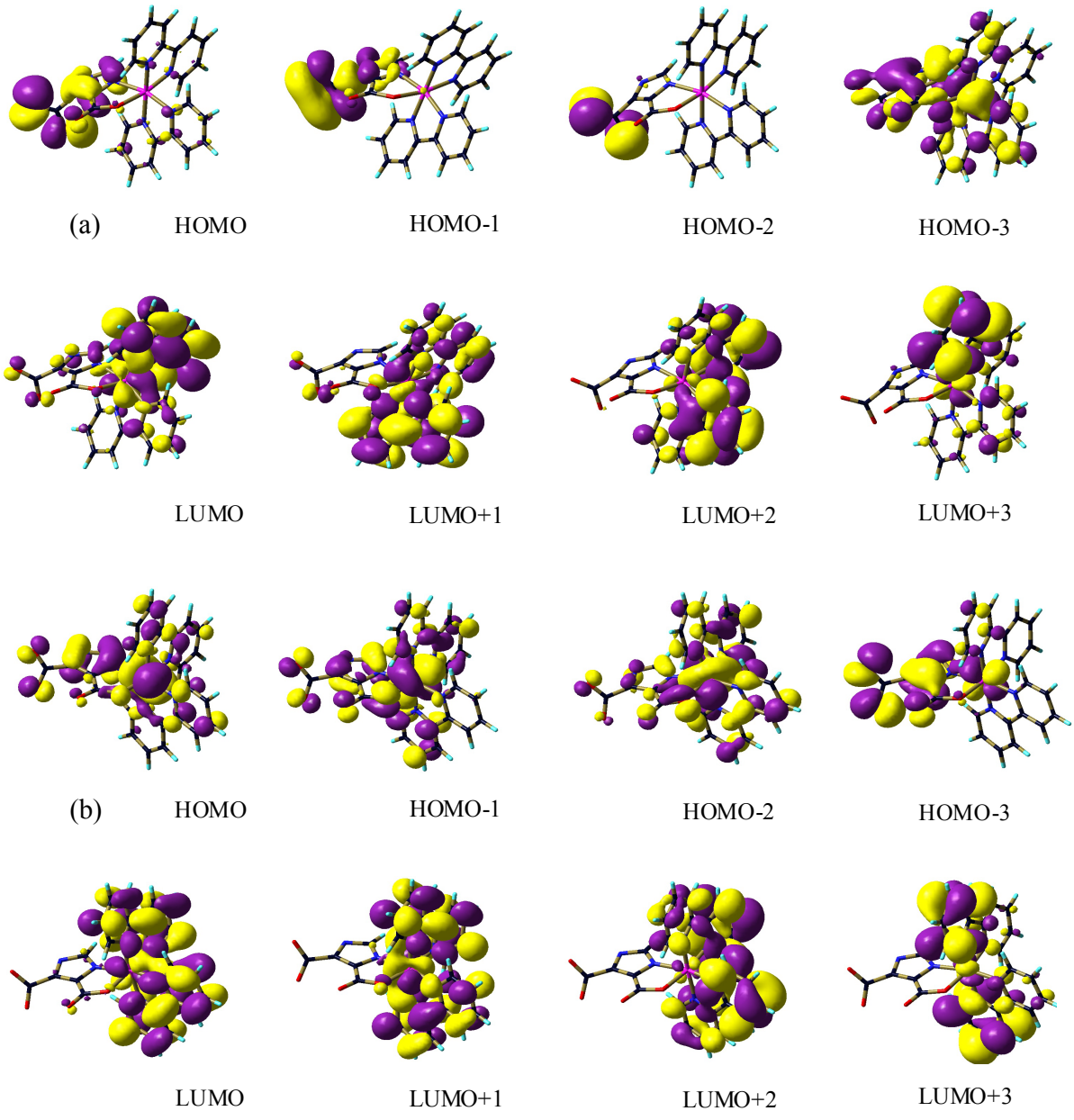


Fig. S3 Schematic drawings of the selective frontier molecular orbitals for $[(bipy)_2Os(Imdc)]^-$ (**1**$^-$) in gas (a) and solution phase (b).

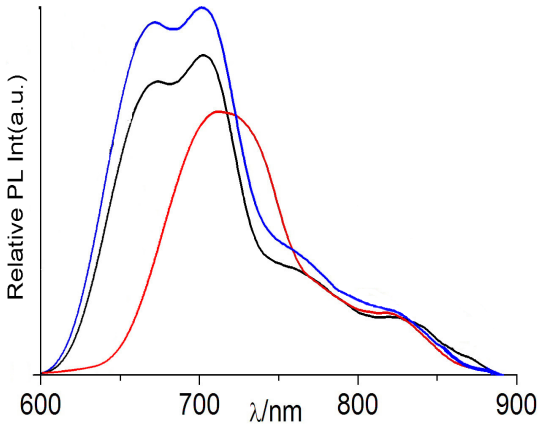
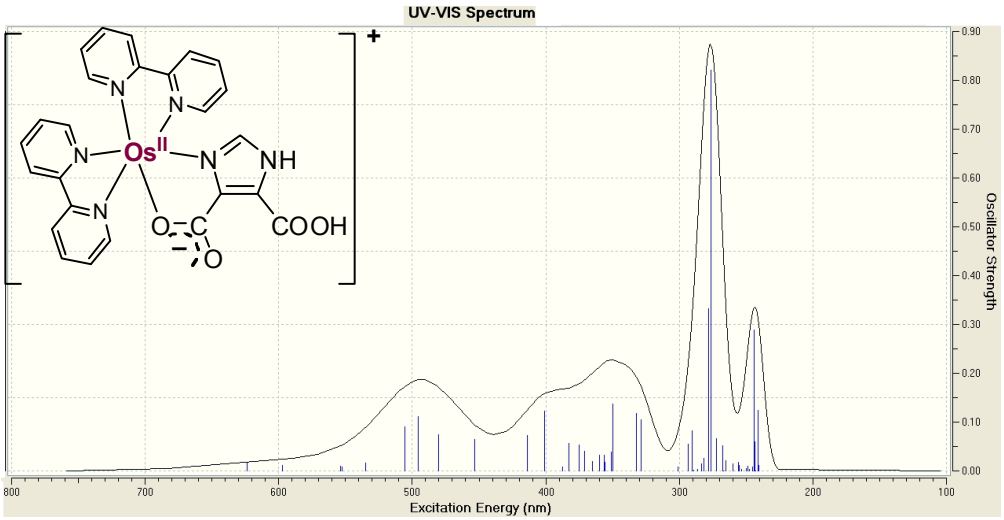


Fig. S4 Emission spectra of $[(bipy)_2Os(H_2Imdc)]^+$ (**1**$^+$), $[(bipy)_2Os(HImdc)]$ (**1**), and $[(bipy)_2Os(Imdc)]^-$ (**1**$^-$) in ethanol-methanol (1:4) glass at 77 K.

Fig. S5 TD-DFT calculated electronic transitions for $[(bipy)_2Os(H_2Imdc)]^+$ (**1**$^+$)



Excitation energies and oscillator strengths:

Excited State 1:	Singlet-A	1.9887 eV 623.45 nm	f= 0.0079<S**2>=0.000
HOMO-2 → LUMO		0.10162	
HOMO → LUMO		0.63917	
HOMO → LUMO+1		-0.26071	
Excited State 2:	Singlet-A	2.0760 eV 597.24 nm	f= 0.0013<S**2>=0.000
HOMO-2 → LUMO+2		0.13309	
HOMO-1 → LUMO+2		-0.10173	
HOMO → LUMO+2		0.67602	
Excited State 3:	Singlet-A	2.2382 eV 553.95 nm	f= 0.0083<S**2>=0.000
HOMO-2 → LUMO		-0.10534	
HOMO-1 → LUMO		0.46572	
HOMO-1 → LUMO+1		-0.20572	
HOMO-1 → LUMO+2		-0.36102	
HOMO → LUMO		0.16891	
HOMO → LUMO+1		0.22833	
Excited State 4:	Singlet-A	2.2434 eV 552.66 nm	f= 0.0021<S**2>=0.000
HOMO-1→LUMO		-0.20543	
HOMO-1→LUMO+2		0.16773	
HOMO→LUMO		0.22151	
HOMO→LUMO+1		0.60140	
Excited State 5:	Singlet-A	2.3179 eV 534.90 nm	f= 0.0133<S**2>=0.000
HOMO-2 → LUMO		0.48002	
HOMO-2 → LUMO+1		-0.13960	
HOMO-2 → LUMO+2		0.33987	
HOMO-1 → LUMO		0.29990	
HOMO-1 → LUMO+2		0.18830	
Excited State 6:	Singlet-A	2.4538 eV 505.28 nm	f=0.0902 <S**2>=0.000

HOMO-2 → LUMO 0.39416
HOMO-2 → LUMO+1 0.14198
HOMO-1 → LUMO -0.32193
HOMO-1 → LUMO+1 -0.30729
HOMO-1 → LUMO+2 -0.32705
Excited State 7: Singlet-A 2.5031 eV 495.32 nm f=0.1115 <S**2>=0.000
HOMO-2 → LUMO 0.27596
HOMO-2 → LUMO+1 0.12051
HOMO-2 → LUMO+2 -0.37618
HOMO-1 → LUMO 0.14146
HOMO-1 → LUMO+1 0.46650
HOMO-1 → LUMO+2 -0.11437
Excited State 8: Singlet-A 2.5821 eV 480.16 nm f=0.0739 <S**2>=0.000
HOMO-2 → LUMO+1 0.57628
HOMO-1 → LUMO+1 -0.16568
HOMO-1 → LUMO+2 0.32409
Excited State 9: Singlet-A 2.7341 eV 453.48 nm f=0.0639 <S**2>=0.000
HOMO-2 → LUMO+1 0.29230
HOMO-2 → LUMO+2 0.40991
HOMO-1 → LUMO+1 0.30155
HOMO-1 → LUMO+2 -0.20888
HOMO → LUMO+2 -0.13085
HOMO → LUMO+4 -0.19162
HOMO → LUMO+6 0.17431
Excited State 10: Singlet-A 2.9974 eV 413.64 nm f=0.0721 <S**2>=0.000
HOMO → LUMO+3 0.68791
Excited State 11: Singlet-A 3.0945 eV 400.66 nm f=0.1224 <S**2>=0.000
HOMO-1 → LUMO+3 0.51025
HOMO → LUMO+4 -0.43511
HOMO → LUMO+5 0.15623
Excited State 12: Singlet-A 3.2012 eV 387.30 nm f=0.0083 <S**2>=0.000
HOMO-2 → LUMO+3 -0.11336
HOMO-1 → LUMO+3 0.36102
HOMO-1 → LUMO+5 -0.13855
HOMO → LUMO+4 0.45251
HOMO → LUMO+5 0.28581
HOMO → LUMO+6 0.15256
Excited State 13: Singlet-A 3.2377 eV 382.94 nm f=0.0562 <S**2>=0.000
HOMO-2 → LUMO+3 -0.26702
HOMO-1 → LUMO+3 -0.28747
HOMO-1 → LUMO+4 -0.14502
HOMO → LUMO+4 -0.13266
HOMO → LUMO+5 0.50804
HOMO → LUMO+6 -0.17161

Excited State 14: Singlet-A 3.3049 eV 375.15 nm f=0.0525 <S**2>=0.000

HOMO-2 → LUMO+3 0.61287
HOMO-1 → LUMO+4 -0.21160
HOMO → LUMO+5 0.20721
Excited State 15: Singlet-A 3.3429 eV 370.89 nm f=0.0408 <S**2>=0.000
HOMO-2 → LUMO+6 0.17186
HOMO-1→ LUMO+4 0.20631
HOMO-1 → LUMO+5 0.29520
HOMO-1 → LUMO+6 -0.23692
HOMO → LUMO+5 0.17079
HOMO → LUMO+6 0.46832
Excited State 16: Singlet-A 3.3967 eV 365.01 nm f=0.0187 <S**2>=0.000
HOMO-2 → LUMO+3 0.14700
HOMO-2 → LUMO+4 -0.14243
HOMO-2 → LUMO+6 -0.11029
HOMO-1 → LUMO+4 0.56227
HOMO → LUMO+5 0.17701
HOMO → LUMO+6 -0.24415
Excited State 17: Singlet-A 3.4487 eV 359.51 nm f=0.0319 <S**2>=0.000
HOMO-2 → LUMO+4 0.48315
HOMO-2 → LUMO+5 -0.18076
HOMO-2 → LUMO+6 -0.11722
HOMO-1 → LUMO+5 -0.20304
HOMO-1 → LUMO+6 -0.13914
HOMO → LUMO+7 0.36872
Excited State 18: Singlet-A 3.4809 eV 356.18 nm f=0.0329 <S**2>=0.000
HOMO-2 → LUMO+4 -0.19006
HOMO-1 → LUMO+5 0.25882
HOMO-1 → LUMO+6 0.27367
HOMO → LUMO+7 0.53745
Excited State 19: Singlet-A 3.4885 eV 355.41 nm f=0.0180 <S**2>=0.000
HOMO-2 → LUMO+4 0.30768
HOMO-2 → LUMO+5 -0.21272
HOMO-2 → LUMO+6 -0.14713
HOMO-1 → LUMO+5 0.40439
HOMO-1 → LUMO+6 0.29853
HOMO → LUMO+7 -0.21943
Excited State 20: Singlet-A 3.5306 eV 351.17 nm f=0.0392 <S**2>=0.000
HOMO-2 → LUMO+4 0.13771
HOMO-2 → LUMO+5 0.12625
HOMO-2 → LUMO+6 0.37437
HOMO-1 → LUMO+4 0.19046
HOMO-1 → LUMO+5 -0.22441
HOMO-1 → LUMO+6 0.44102
HOMO-1 → LUMO+7 -0.10211
HOMO → LUMO+6 0.10317
Excited State 21: Singlet-A 3.5470 eV 349.55 nm f=0.1376 <S**2>=0.000

HOMO-2 → LUMO+4 0.18538
HOMO-2→ LUMO+5 0.58971
HOMO-2 → LUMO+6 -0.26703
Excited State 22: Singlet-A 3.7355 eV 331.91 nm f=0.1173 <S**2>=0.000
HOMO-2 → LUMO+4 0.11269
HOMO-2 → LUMO+6 0.31678
HOMO-2 → LUMO+7 -0.11181
HOMO-1→ LUMO+7 0.55225
HOMO → LUMO+6 -0.14290
Excited State 23: Singlet-A 3.7750 eV 328.43 nm f=0.1043 <S**2>=0.000
HOMO-2 → LUMO+4 0.11215
HOMO-2 → LUMO+6 0.23550
HOMO-2 → LUMO+7 0.54330
HOMO-1 → LUMO+5 0.12392
HOMO-1→ LUMO+6 -0.14961
HOMO-1 → LUMO+7 -0.17268
HOMO → LUMO+6 -0.16605
Excited State 24: Singlet-A 4.1208 eV 300.87 nm f=0.0077 <S**2>=0.000
HOMO-2 → LUMO+2 -0.10343
HOMO-2 → LUMO+4 -0.11053
HOMO-2 → LUMO+6 -0.18540
HOMO-2 → LUMO+7 0.39991
HOMO-1 → LUMO+5 -0.11235
HOMO-1 → LUMO+6 0.11031
HOMO-1 → LUMO+7 0.33743
HOMO → LUMO+4 -0.10617
HOMO → LUMO+6 0.18973
HOMO → LUMO+8 0.18382
Excited State 25: Singlet-A 4.2304 eV 293.08 nm f=0.0545 <S**2>=0.000
HOMO-4 → LUMO+2 0.15681
HOMO-3 → LUMO 0.66074
HOMO-3 → LUMO+2 -0.10376
Excited State 26: Singlet-A 4.2671 eV 290.56 nm f=0.0829 <S**2>=0.000
HOMO-4 → LUMO 0.62345
HOMO-3 → LUMO+2 0.28816
Excited State 27: Singlet-A 4.2883 eV 289.12 nm f=0.0014 <S**2>=0.000
HOMO-7 → LUMO+1 -0.10212
HOMO-6 → LUMO 0.40969
HOMO-6 → LUMO+1 0.53773
Excited State 28: Singlet-A 4.3325 eV 286.17 nm f=0.0031 <S**2>=0.000
HOMO-1 → LUMO+15 -0.11389
HOMO → LUMO+11 0.61894
HOMO → LUMO+14 -0.22056

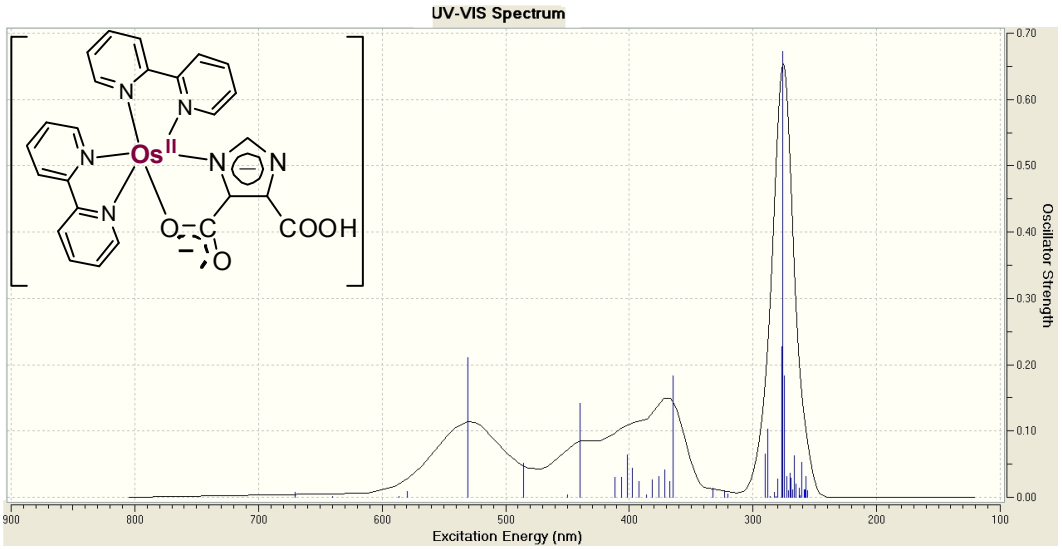
Excited State 29: Singlet-A 4.3747 eV 283.41 nm f=0.0144 <S**2>=0.000

HOMO-4 → LUMO+2 -0.14481
HOMO-3 → LUMO+1 0.67571
Excited State 30: Singlet-A 4.4071 eV 281.33 nm f=0.0264 <S**2>=0.000
HOMO-4 → LUMO 0.17057
HOMO-4 → LUMO+1 0.61686
HOMO-3 → LUMO+2 -0.27581
Excited State 31: Singlet-A 4.4550 eV 278.30 nm f=0.3320 <S**2>=0.000
HOMO-4 → LUMO+2 0.58200
HOMO-3 → LUMO -0.13082
HOMO-3 → LUMO+1 0.14153
HOMO-3 → LUMO+2 -0.25823
Excited State 32: Singlet-A 4.4900 eV 276.13 nm f=0.8214 <S**2>=0.000
HOMO-4 → LUMO -0.21017
HOMO-4 → LUMO+1 0.29899
HOMO-4 → LUMO+2 0.23913
HOMO-3 → LUMO+2 0.46517
HOMO-2 → LUMO+5 -0.10178
HOMO-1 → LUMO+11 -0.12758
Excited State 33: Singlet-A 4.5510 eV 272.43 nm f=0.0655 <S**2>=0.000
HOMO-2 → LUMO+11 -0.21532
HOMO-1 → LUMO+11 0.51813
HOMO-1 → LUMO+14 -0.22605
HOMO → LUMO+15 -0.19257
Excited State 34: Singlet-A 4.6347 eV 267.51 nm f=0.0515 <S**2>=0.000
HOMO-1 → LUMO+8 -0.10033
HOMO → LUMO+8 0.63780
Excited State 35: Singlet-A 4.6464 eV 266.84 nm f=0.0004 <S**2>=0.000
HOMO-7 → LUMO 0.42863
HOMO-7 → LUMO+1 0.51614
HOMO-6 → LUMO 0.11987
Excited State 36: Singlet-A 4.6801 eV 264.92 nm f=0.0203 <S**2>=0.000
HOMO → LUMO+9 0.68524
Excited State 37: Singlet-A 4.7699 eV 259.93 nm f=0.0145 <S**2>=0.000
HOMO-2 → LUMO+11 0.10595
HOMO-1 → LUMO+8 0.67191
Excited State 38: Singlet-A 4.8433 eV 255.99 nm f=0.0180 <S**2>=0.000
HOMO-5 → LUMO -0.44003
HOMO-5 → LUMO+1 0.14536
HOMO-2 → LUMO+8 0.48838
Excited State 39: Singlet-A 4.8473 eV 255.78 nm f=0.0042 <S**2>=0.000
HOMO-5 → LUMO 0.40222
HOMO-5 → LUMO+1 -0.11890
HOMO-2 → LUMO+8 0.30639
HOMO-2 → LUMO+11 -0.35208
HOMO-2 → LUMO+15 0.17345
Excited State 40: Singlet-A 4.8547 eV 255.39 nm f=0.0107 <S**2>=0.000

HOMO-5 → LUMO	0.25635	
HOMO-2 → LUMO+8	0.33800	
HOMO-2 → LUMO+11	0.42670	
HOMO-2 → LUMO+15	-0.18343	
HOMO-1 → LUMO+9	0.12898	
HOMO-1 → LUMO+11	0.12552	
HOMO → LUMO+15	-0.10457	
Excited State 41: Singlet-A	4.8882 eV 253.64 nm	f=0.0033 <S**2>=0.000
HOMO-1 → LUMO+9	0.64768	
Excited State 42: Singlet-A	4.9572 eV 250.11 nm	f=0.0041 <S**2>=0.000
HOMO-5 → LUMO+2	0.12044	
HOMO-4 → LUMO+3	-0.17381	
HOMO-2 → LUMO+9	0.62459	
Excited State 43: Singlet-A	4.9845 eV 248.74 nm	f=0.0090 <S**2>=0.000
HOMO-5→ LUMO	0.13151	
HOMO-5→ LUMO+1	0.10588	
HOMO-5→ LUMO+2	0.66439	
HOMO-2→ LUMO+9	-0.11464	
Excited State 44: Singlet-A	5.0050 eV 247.72 nm	f=0.0029 <S**2>=0.000
HOMO-2 → LUMO+9	0.14279	
HOMO-2 → LUMO+11	0.18477	
HOMO-2 → LUMO+12	-0.15052	
HOMO-2 → LUMO+14	0.15134	
HOMO-2 → LUMO+15	0.27592	
HOMO-1 → LUMO+11	0.23278	
HOMO-1 → LUMO+14	-0.10275	
HOMO → LUMO+10	-0.14745	
HOMO → LUMO+9	-0.20643	
HOMO → LUMO+14	-0.14031	
HOMO → LUMO+15	0.35980	
Excited State 45: Singlet-A	5.0581 eV 245.12 nm	f=0.0077 <S**2>=0.000
HOMO-7 → LUMO+1	-0.11600	
HOMO-6 → LUMO	0.50133	
HOMO-6 → LUMO+1	-0.38654	
HOMO-3 → LUMO+3	0.19724	
Excited State 46: Singlet-A	5.0809 eV 244.02 nm	f=0.2893 <S**2>=0.000
HOMO-5 → LUMO	0.16306	
HOMO-5 → LUMO+1	0.61586	
HOMO-5 → LUMO+2	-0.14696	
HOMO-1→ LUMO+10	0.15101	
Excited State 47: Singlet-A	5.0930 eV 243.44 nm	f=0.0598 <S**2>=0.000
HOMO-9 → LUMO	-0.13175	
HOMO-8 → LUMO	-0.11599	
HOMO-6 → LUMO	-0.16722	
HOMO-6 → LUMO+1	0.13252	
HOMO-5 → LUMO+1	0.10015	

HOMO-3 → LUMO+3	0.57996			
HOMO-2 → LUMO+8	0.11125			
HOMO-1 → LUMO+9	0.10104			
Excited State 48: Singlet-A	5.1014 eV	243.04 nm	f=0.0004	<S**2>=0.000
HOMO-3 → LUMO+3	-0.10600			
HOMO → LUMO+10	0.64041			
Excited State 49: Singlet-A	5.1414 eV	241.15 nm	f=0.1240	<S**2>=0.000
HOMO-9 → LUMO+2	0.10625			
HOMO-8 → LUMO	0.13439			
HOMO-4 → LUMO+3	0.58675			
HOMO-2 → LUMO+9	0.18318			
Excited State 50: Singlet-A	5.1606 eV	240.25 nm	f=0.0113	<S**2>=0.000
HOMO-8 → LUMO	0.24113			
HOMO-8 → LUMO+2	0.14901			
HOMO-7 → LUMO+2	0.12178			
HOMO-6 → LUMO	-0.10506			
HOMO-6 → LUMO+1	0.11677			
HOMO-6 → LUMO+2	0.39632			
HOMO-4 → LUMO+3	-0.11939			
HOMO-4 → LUMO+4	-0.11325			
HOMO-4 → LUMO+5	0.13824			
HOMO-3 → LUMO+4	0.24638			
HOMO-3 → LUMO+5	-0.22727			

Fig. S6 TD-DFT calculated electronic transitions for [(bipy)$_2$Os(HImdc)] (**1**)



Excitation energies and oscillator strengths:

Excited State 1:	Singlet-A	1.8502 eV	670.10 nm	f= 0.0075<S**2>=0.000
HOMO-2 → LUMO			-0.12620	
HOMO → LUMO			0.68449	
Excited State 2:	Singlet-A	1.9364 eV	640.28 nm	f= 0.0010<S**2>=0.000
HOMO-2 → LUMO+1			-0.14296	
HOMO → LUMO+1			0.68113	
Excited State 3:	Singlet-A	2.1154 eV	586.11 nm	f= 0.0021<S**2>=0.000
HOMO-2 → LUMO			0.43059	
HOMO-1 → LUMO			0.14221	
HOMO-1 → LUMO+1			0.51990	
Excited State 4:	Singlet-A	2.1394 eV	579.52 nm	f=0.0096 <S**2>=0.000
HOMO-2 → LUMO+1			0.29121	
HOMO-1 → LUMO			0.62597	
Excited State 5:	Singlet-A	2.3358 eV	530.79 nm	f=0.2107 <S**2>=0.000
HOMO-2 → LUMO			0.52001	
HOMO-1→ LUMO+1			-0.43417	
HOMO → LUMO			0.10071	
HOMO → LUMO+5			-0.10563	
Excited State 6:	Singlet-A	2.5542 eV	485.41 nm	f=0.0520 <S**2>=0.000
HOMO-2→ LUMO+1			0.56509	
HOMO-1 → LUMO			-0.23305	
HOMO-1 → LUMO+1			0.13069	
HOMO-1 → LUMO+2			-0.10940	
HOMO → LUMO+1			0.13040	
HOMO → LUMO+4			0.16963	
HOMO→ LUMO+6			-0.15584	
Excited State 7:	Singlet-A	2.7569 eV	449.72 nm	f=0.0043 <S**2>=0.000
HOMO-1→ LUMO+2			0.10247	
HOMO → LUMO+2			0.67652	
Excited State 8:	Singlet-A	2.8178 eV	440.00 nm	f=0.1414 <S**2>=0.000
HOMO→ LUMO+3			0.68314	
Excited State 9:	Singlet-A	3.0122 eV	411.60 nm	f=0.0305 <S**2>=0.000
HOMO-1→ LUMO+2			0.43684	
HOMO-1→ LUMO+3			0.30832	
HOMO→ LUMO+4			0.43928	
Excited State 10:	Singlet-A	3.0506 eV	406.42 nm	f=0.0295 <S**2>=0.000
HOMO-1→ LUMO+3			0.52420	
HOMO-1 → LUMO+5			-0.11574	
HOMO → LUMO+4			-0.39588	
HOMO → LUMO+5			0.14950	
HOMO→ LUMO+6			-0.12569	
Excited State 11:	Singlet-A	3.0540 eV	405.97 nm	f=0.0038 <S**2>=0.000
HOMO-2→ LUMO+2			0.60974	
HOMO-1 → LUMO+2			0.10955	
HOMO → LUMO+5			0.30531	

Excited State 12:	Singlet-A	3.0859 eV	401.77 nm	f=0.0640 <S**2>=0.000
HOMO-2→ LUMO+2		-0.20460		
HOMO-2 → LUMO+3		0.40520		
HOMO-1 → LUMO+3		-0.14374		
HOMO-1 → LUMO+4		0.18913		
HOMO → LUMO+5		0.44514		
Excited State 13:	Singlet-A	3.1188 eV	397.54 nm	f=0.0443 <S**2>=0.000
HOMO-2→ LUMO+3		-0.10638		
HOMO-1 → LUMO+2		0.39446		
HOMO-1 → LUMO+3		-0.29429		
HOMO→ LUMO+4		-0.23286		
HOMO→ LUMO+6		-0.39026		
Excited State 14:	Singlet-A	3.1612 eV	392.20 nm	f=0.0239 <S**2>=0.000
HOMO-2→ LUMO+2		0.18950		
HOMO-2→ LUMO+3		0.53508		
HOMO-1→ LUMO+4		-0.24625		
HOMO → LUMO+5		-0.24479		
HOMO→ LUMO+6		-0.11609		
Excited State 15:	Singlet-A	3.2086 eV	386.41 nm	f=0.0033 <S**2>=0.000
HOMO-2 → LUMO+5		-0.10012		
HOMO-2 → LUMO+6		0.25907		
HOMO-1→ LUMO+2		-0.23825		
HOMO-1 → LUMO+5		0.40040		
HOMO → LUMO+6		-0.39664		
Excited State 16:	Singlet-A	3.2501 eV	381.48 nm	f=0.0261 <S**2>=0.000
HOMO-2 → LUMO+2		0.10240		
HOMO-2 → LUMO+4		-0.13031		
HOMO-2 → LUMO+5		-0.14853		
HOMO-1 → LUMO+4		0.50278		
HOMO-1 → LUMO+6		-0.28980		
HOMO → LUMO+5		-0.28202		
Excited State 17:	Singlet-A	3.2973 eV	376.02 nm	f=0.0310 <S**2>=0.000
HOMO-2 → LUMO+4		0.60846		
HOMO-1 → LUMO+4		0.20286		
HOMO-1 → LUMO+5		-0.17278		
HOMO → LUMO+6		-0.19397		
Excited State 18:	Singlet-A	3.3371 eV	371.53 nm	f=0.0408 <S**2>=0.000
HOMO-2 → LUMO+2		0.10967		
HOMO-2 → LUMO+4		-0.13848		
HOMO-2 → LUMO+5		-0.10376		
HOMO-1 → LUMO+4		0.22187		
HOMO-1 → LUMO+6		0.61277		
Excited State 19:	Singlet-A	3.3753 eV	367.33 nm	f=0.0243 <S**2>=0.000
HOMO-2 → LUMO+4		-0.17255		
HOMO-2 → LUMO+6		0.47003		
HOMO-1 → LUMO+5		-0.44413		

HOMO → LUMO+4		0.10211		
Excited State 20:	Singlet-A	3.4010 eV	364.55 nm	f=0.1834 <S**2>=0.000
HOMO-5 → LUMO		-0.10313		
HOMO-2 → LUMO+5		0.64442		
HOMO-1 → LUMO+4		0.20180		
Excited State 21:	Singlet-A	3.7310 eV	332.31 nm	f=0.0141 <S**2>=0.000
HOMO-3 → LUMO		0.65931		
HOMO-3 → LUMO+1		-0.10570		
HOMO-2 → LUMO+6		-0.13292		
Excited State 22:	Singlet-A	3.8401 eV	322.87 nm	f=0.0087 <S**2>=0.000
HOMO-3 → LUMO		0.20215		
HOMO-3 → LUMO+1		-0.13487		
HOMO-2 → LUMO+1		0.15735		
HOMO-2 → LUMO+4		0.16004		
HOMO-2 → LUMO+6		0.37800		
HOMO-1 → LUMO		-0.10928		
HOMO-1 → LUMO+2		0.17170		
HOMO-1 → LUMO+5		0.21035		
HOMO-1→ LUMO+6		0.11884		
HOMO → LUMO+4		-0.14778		
HOMO → LUMO+6		0.25186		
HOMO → LUMO+7		0.14452		
Excited State 23:	Singlet-A	3.8698 eV	320.39 nm	f=0.0048 <S**2>=0.000
HOMO-3 → LUMO		0.14219		
HOMO-3 → LUMO+1		0.68390		
Excited State 24:	Singlet-A	4.2691 eV	290.42 nm	f=0.0655 <S**2>=0.000
HOMO-5 → LUMO		0.15488		
HOMO-5 → LUMO+1		0.18029		
HOMO-4 → LUMO		0.64739		
Excited State 25:	Singlet-A	4.3008 eV	288.28 nm	f=0.1025 <S**2>=0.000
HOMO-5 → LUMO		0.59778		
HOMO-4 → LUMO		-0.15103		
HOMO-4 → LUMO+1		0.31512		
Excited State 26:	Singlet-A	4.3319 eV	286.21 nm	f=0.0018 <S**2>=0.000
HOMO-6 → LUMO		0.50323		
HOMO-6 → LUMO+2		0.40954		
HOMO-6 → LUMO+3		-0.22857		
Excited State 27:	Singlet-A	4.3883 eV	282.53 nm	f=0.0073 <S**2>=0.000
HOMO-1 → LUMO+10		0.17240		
HOMO-1 → LUMO+13		-0.11021		
HOMO→ LUMO+7		0.19480		
HOMO → LUMO+8		-0.25307		
HOMO → LUMO+10		0.42808		
HOMO → LUMO+11		0.15287		
HOMO → LUMO+13		-0.25522		

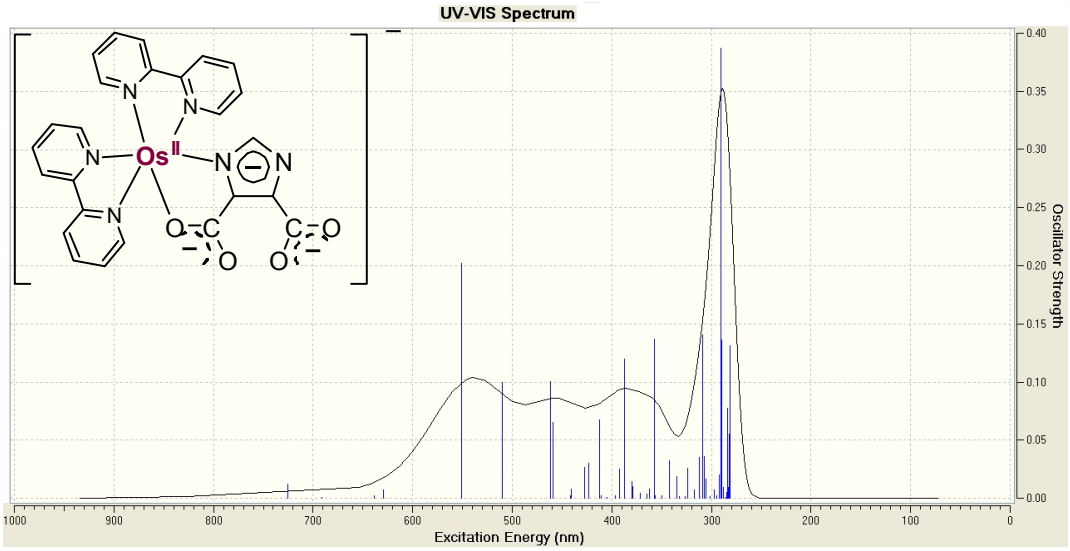
Excited State 28:	Singlet-A	4.4103 eV	281.12 nm	f=0.0010	<S**2>=0.000
HOMO-7 → LUMO			0.66722		
HOMO-7→ LUMO+2			0.13697		
Excited State 29:	Singlet-A	4.4289 eV	279.94 nm	f=0.0276	<S**2>=0.000
HOMO-5 → LUMO+1			0.28310		
HOMO-4 → LUMO			-0.10673		
HOMO-3 → LUMO+2			-0.10396		
HOMO → LUMO+7			0.54415		
HOMO → LUMO+10			-0.15676		
Excited State 30:	Singlet-A	4.4798 eV	276.76 nm	f=0.2269	<S**2>=0.000
HOMO-5 → LUMO+1			0.54667		
HOMO-4 → LUMO			-0.11819		
HOMO-4 → LUMO+1			-0.12343		
HOMO → LUMO+7			-0.25011		
HOMO → LUMO+8			0.14902		
Excited State 31:	Singlet-A	4.4943 eV	275.87 nm	f=0.6719	<S**2>=0.000
HOMO-5 → LUMO			-0.26076		
HOMO-5 → LUMO+1			0.15864		
HOMO-4 → LUMO+1			0.56103		
HOMO → LUMO+9			0.13675		
Excited State 32:	Singlet-A	4.5154 eV	274.58 nm	f=0.1831	<S**2>=0.000
HOMO-7 → LUMO+1			-0.17382		
HOMO-3 → LUMO+2			0.35067		
HOMO-3 → LUMO+3			-0.10271		
HOMO → LUMO+7			0.20150		
HOMO → LUMO+8			0.45708		
HOMO → LUMO+10			0.14155		
Excited State 33:	Singlet-A	4.5424 eV	272.95 nm	f=0.0311	<S**2>=0.000
HOMO-3 → LUMO+2			0.12587		
HOMO → LUMO+8			-0.20444		
HOMO → LUMO+9			0.62654		
Excited State 34:	Singlet-A	4.5646 eV	271.62 nm	f=0.0100	<S**2>=0.000
HOMO-7 → LUMO+1			-0.31770		
HOMO-6 → LUMO			0.35521		
HOMO-6 → LUMO+1			0.23934		
HOMO-6 → LUMO+2			-0.28853		
HOMO-6 → LUMO+3			0.15084		
HOMO-3 → LUMO+2			0.11238		
HOMO → LUMO+8			-0.18764		
HOMO → LUMO+9			-0.12038		
Excited State 35:	Singlet-A	4.5713 eV	271.22 nm	f=0.0044	<S**2>=0.000
HOMO-7 → LUMO			0.11040		
HOMO-7 → LUMO+1			0.37174		
HOMO-6 → LUMO			0.31763		
HOMO-6 → LUMO+1			-0.12328		

HOMO-6 → LUMO+2	-0.30419			
HOMO-6 → LUMO+3	0.16037			
HOMO-3 → LUMO+2	-0.14125			
HOMO → LUMO+8	0.18701			
HOMO → LUMO+9	0.11502			
Excited State 36:	Singlet-A	4.5937 eV	269.90 nm	f=0.0361 <S**2>=0.000
HOMO-7 → LUMO+1	0.18876			
HOMO-6 → LUMO+1	-0.21550			
HOMO-3 → LUMO+2	0.25893			
HOMO-1 → LUMO+7	0.54717			
Excited State 37:	Singlet-A	4.6060 eV	269.18 nm	f=0.0286 <S**2>=0.000
HOMO-7 → LUMO+1	0.12382			
HOMO-6 → LUMO+1	- 0.21669			
HOMO-3 → LUMO+2	0.35791			
HOMO-3 → LUMO+3	0.26132			
HOMO-2 → LUMO+7	0.15771			
HOMO-1 → LUMO+7	-0.32360			
HOMO-1 → LUMO+8	-0.16774			
HOMO → LUMO+8	-0.14796			
Excited State 38:	Singlet-A	4.6270 eV	267.96 nm	f=0.0029 <S**2>=0.000
HOMO-7 → LUMO+1	0.32467			
HOMO-6 → LUMO+1	0.54356			
HOMO-3 → LUMO+3	0.21388			
Excited State 39:	Singlet-A	4.6315 eV	267.70 nm	f=0.0115 <S**2>=0.000
HOMO-3 → LUMO+3	0.20634			
HOMO-2 → LUMO+10	0.36682			
HOMO-2 → LUMO+11	0.12326			
HOMO-2 → LUMO+12	0.11266			
HOMO-2 → LUMO+13	-0.21712			
HOMO-2 → LUMO+14	-0.11497			
HOMO-1 → LUMO+8	0.12040			
HOMO-1 → LUMO+10	-0.22690			
HOMO-1 → LUMO+13	0.13552			
HOMO → LUMO+10	0.15915			
HOMO → LUMO+15	0.19360			
Excited State 40:	Singlet-A	4.6514 eV	266.55 nm	f=0.0624 <S**2>=0.000
HOMO-7 → LUMO+1	-0.20759			
HOMO-6 → LUMO+1	-0.11815			
HOMO-3 → LUMO+3	0.49931			
HOMO-2 → LUMO+7	-0.27449			
HOMO-2 → LUMO+8	0.10734			
HOMO-1 → LUMO+8	0.12785			
HOMO → LUMO+8	0.14743			
Excited State 41:	Singlet-A	4.6802 eV	264.91 nm	f=0.0198 <S**2>=0.000
HOMO-8 → LUMO	-0.11669			
HOMO-3→ LUMO+2	-0.14616			

HOMO-3 → LUMO+3 0.12950
HOMO-2 → LUMO+7 0.59696
HOMO-2→ LUMO+8 0.11979
HOMO-1 → LUMO+9 0.12802
Excited State 42: Singlet-A 4.7230 eV 262.51 nm f=0.0144 <S**2>=0.000
HOMO-1 → LUMO+8 -0.46776
HOMO-1 → LUMO+9 0.48085
Excited State 43: Singlet-A 4.7370 eV 261.74 nm f=0.0026 <S**2>=0.000
HOMO-8 → LUMO 0.22597
HOMO-7 → LUMO -0.14710
HOMO-7 → LUMO+2 0.52300
HOMO-7 → LUMO+3 -0.26991
HOMO-2 → LUMO+8 0.15475
Excited State 44: Singlet-A 4.7531 eV 260.85 nm f=0.0161 <S**2>=0.000
HOMO-8 → LUMO 0.45832
HOMO-7 → LUMO+2 -0.21620
HOMO-7 → LUMO+3 0.11027
HOMO-3 → LUMO+4 -0.32842
HOMO-1 → LUMO+8 0.19147
HOMO-1 → LUMO+9 0.16038
Excited State 45: Singlet-A 4.7626 eV 260.33 nm f=0.0521 <S**2>=0.000
HOMO-8 → LUMO 0.24085
HOMO-3 → LUMO+4 0.57071
HOMO-1 → LUMO+8 0.18458
HOMO-1 → LUMO+9 0.18701
Excited State 46: Singlet-A 4.7909 eV 258.79 nm f=0.0119 <S**2>=0.000
HOMO-9 → LUMO 0.36187
HOMO-9 → LUMO+2 0.20249
HOMO-9 → LUMO+3 -0.12049
HOMO-8 → LUMO -0.23234
HOMO-3 → LUMO+4 -0.12949
HOMO-2 → LUMO+9 -0.14884
HOMO-1 → LUMO+7 -0.10652
HOMO-1 → LUMO+8 0.22816
HOMO-1 → LUMO+9 0.26748
Excited State 47: Singlet-A 4.7998 eV 258.31 nm f=0.0064 <S**2>=0.000
HOMO-9 → LUMO -0.13506
HOMO-5 → LUMO+3 -0.10295
HOMO-3 → LUMO+4 -0.13961
HOMO-2 → LUMO+8 -0.34538
HOMO-2 → LUMO+9 0.49824
HOMO-1 → LUMO+9 0.15382
Excited State 48: Singlet-A 4.8045 eV 258.06 nm f=0.0107 <S**2>=0.000
HOMO-9 → LUMO 0.40891
HOMO-9 → LUMO+2 0.20708
HOMO-9 → LUMO+3 -0.12252

HOMO-8 → LUMO		0.18451
HOMO-7 → LUMO+2		-0.11992
HOMO-3 → LUMO+3		0.10142
HOMO-3 → LUMO+5		-0.18529
HOMO-2 → LUMO+9		0.28171
HOMO-1 → LUMO+8		-0.15804
HOMO-1 → LUMO+9		-0.13248
Excited State 49:	Singlet-A	4.8151 eV 257.49 nm f=0.0311 <S**2>=0.000
HOMO-7 → LUMO+2		-0.13261
HOMO-3 → LUMO+2		0.11860
HOMO-3 → LUMO+3		-0.14700
HOMO-3 → LUMO+5		0.18629
HOMO-2 → LUMO+8		0.51487
HOMO-2 → LUMO+9		0.31574
Excited State 50:	Singlet-A	4.8465 eV 255.82 nm f=0.0097 <S**2>=0.000
HOMO-8 → LUMO		0.13752
HOMO-8 → LUMO+1		0.18896
HOMO-3 → LUMO+2		-0.13792
HOMO-3 → LUMO+5		0.59266
HOMO-2 → LUMO+8		-0.11999
HOMO-1 → LUMO+9		-0.10933

Fig. S7 TD-DFT calculated electronic transitions for $[(bipy)_2Os(Imdc)]^-$ (**1**$^-$)



Excitation energies and oscillator strengths:

Excited State 1:	Singlet-A	1.7093 eV 725.34 nm f= 0.0062<S**2>=0.000
HOMO-1 → LUMO		0.27410
HOMO → LUMO		0.63675

Excited State 2:	Singlet-A	1.7945 eV	690.90 nm	f= 0.0007<S**2>=0.000
HOMO-1 → LUMO+1		0.25388		
HOMO → LUMO+1		0.63950		
Excited State 3:	Singlet-A	1.9420 eV	638.45 nm	f= 0.0022<S**2>=0.000
HOMO-2 → LUMO		-0.37956		
HOMO-2 → LUMO+1		-0.16113		
HOMO-1 → LUMO		0.12684		
HOMO-1 → LUMO+1		0.50325		
HOMO → LUMO+1		-0.18764		
Excited State 4:	Singlet-A	1.9702 eV	629.30 nm	f=0.0078 <S**2>=0.000
HOMO-2 → LUMO+1		-0.22568		
HOMO-1 → LUMO		0.58134		
HOMO-1 → LUMO+1		-0.15233		
HOMO → LUMO		-0.23408		
Excited State 5:	Singlet-A	2.2506 eV	550.88 nm	f=0.2019 <S**2>=0.000
HOMO-2 → LUMO		0.56020		
HOMO-1 → LUMO+1		0.35204		
HOMO → LUMO+1		-0.16287		
Excited State 6:	Singlet-A	2.4302 eV	510.18 nm	f=0.0996 <S**2>=0.000
HOMO-2 → LUMO+1		0.58275		
HOMO-1 → LUMO		0.20444		
HOMO-1 → LUMO+1		0.10916		
HOMO → LUMO		-0.10703		
HOMO → LUMO+3		0.16194		
HOMO → LUMO+5		-0.13583		
Excited State 7:	Singlet-A	2.6839 eV	461.95 nm	f=0.1004<S**2>=0.000
HOMO-3 → LUMO		0.47714		
HOMO-1 → LUMO+2		-0.12985		
HOMO → LUMO+2		-0.45942		
Excited State 8:	Singlet-A	2.7006 eV	459.10 nm	f=0.0650 <S**2>=0.000
HOMO-3 → LUMO		0.48208		
HOMO-1 → LUMO+2		0.11576		
HOMO → LUMO+2		0.47690		
Excited State 9:	Singlet-A	2.8056 eV	441.92 nm	f=0.0023 <S**2>=0.000
HOMO-1 → LUMO+2		0.65411		
HOMO → LUMO+2		-0.17303		
HOMO → LUMO+3		0.11470		
Excited State 10:	Singlet-A	2.8126 eV	440.82 nm	f=0.0078 <S**2>=0.000
HOMO-3 → LUMO+1		0.67340		
Excited State 11:	Singlet-A	2.9022 eV	427.21 nm	f=0.0264 <S**2>=0.000
HOMO-2 → LUMO+2		-0.16102		
HOMO → LUMO+3		0.51722		
HOMO → LUMO+4		0.40163		
HOMO → LUMO+5		0.10074		
Excited State 12:	Singlet-A	2.9318 eV	422.89 nm	f=0.0303 <S**2>=0.000
HOMO-2 → LUMO+2		-0.30419		

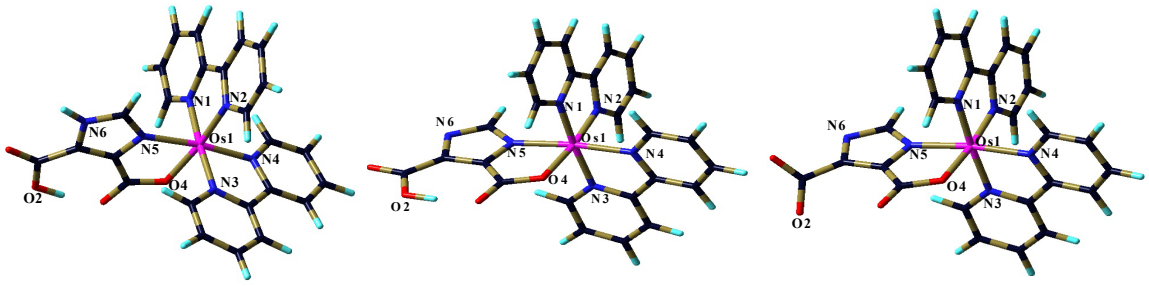
HOMO-1 → LUMO+3	-0.29505			
HOMO-1 → LUMO+4	0.31117			
HOMO → LUMO+3	-0.27985			
HOMO → LUMO+4	0.30579			
HOMO → LUMO+5	-0.18350			
Excited State 13: Singlet-A	3.0074 eV	412.27 nm	f=0.0677	<S**2>=0.000
HOMO-2 → LUMO+2	0.51851			
HOMO-2 → LUMO+4	-0.13288			
HOMO-1 → LUMO+3	-0.37453			
HOMO → LUMO+3	0.10127			
HOMO → LUMO+4	0.10509			
HOMO → LUMO+5	-0.13996			
Excited State 14: Singlet-A	3.0215 eV	410.33 nm	f=0.0023	<S**2>=0.000
HOMO-2 → LUMO+3	0.12803			
HOMO-2 → LUMO+5	-0.19597			
HOMO-1 → LUMO+3	-0.12324			
HOMO-1 → LUMO+4	0.32800			
HOMO-1 → LUMO+5	0.17682			
HOMO → LUMO+4	-0.14428			
HOMO → LUMO+5	0.48552			
Excited State 15: Singlet-A	3.0655 eV	404.45 nm	f=0.0008	<S**2>=0.000
HOMO-3 → LUMO+1	0.10991			
HOMO-2 → LUMO+2	0.20693			
HOMO-1 → LUMO+3	0.31208			
HOMO-1 → LUMO+4	0.10219			
HOMO-1 → LUMO+5	-0.32056			
HOMO → LUMO+3	-0.17995			
HOMO → LUMO+4	0.34648			
HOMO → LUMO+5	0.20853			
Excited State 16: Singlet-A	3.1241 eV	396.87 nm	f=0.0023	<S**2>=0.000
HOMO-2 → LUMO+2	0.18821			
HOMO-2 → LUMO+5	-0.10994			
HOMO-1 → LUMO+3	0.28296			
HOMO-1 → LUMO+5	0.53230			
HOMO → LUMO+4	0.13988			
HOMO → LUMO+5	-0.17912			
Excited State 17: Singlet-A	3.1599 eV	392.37 nm	f=0.0253	<S**2>=0.000
HOMO-2 → LUMO+1	-0.10085			
HOMO-2 → LUMO+3	0.53240			
HOMO-2 → LUMO+5	0.19496			
HOMO-1 → LUMO+3	0.16170			
HOMO-1 → LUMO+4	0.27521			
HOMO-1 → LUMO+5	-0.13040			
HOMO → LUMO+5	-0.16393			
Excited State 18: Singlet-A	3.2037 eV	387.00 nm	f=0.1198	<S**2>=0.000
HOMO-2 → LUMO+3	0.40363			

HOMO-2 → LUMO+5		-0.36899			
HOMO-1 → LUMO+4		-0.36969			
HOMO → LUMO+4		0.13010			
Excited State 19:	Singlet-A	3.2628 eV	379.99 nm	f=0.0144	<S**2>=0.000
HOMO-2 → LUMO+4		0.66506			
HOMO-1 → LUMO+3		-0.13023			
Excited State 20:	Singlet-A	3.2725 eV	378.86 nm	f=0.0101	<S**2>=0.000
HOMO-4 → LUMO		0.68662			
Excited State 21:	Singlet-A	3.3368 eV	371.56 nm	f=0.0043	<S**2>=0.000
HOMO-6 → LUMO		0.24575			
HOMO-5 → LUMO		0.64674			
HOMO-4 → LUMO		0.11357			
Excited State 22:	Singlet-A	3.4000 eV	364.66 nm	f=0.0039	<S**2>=0.000
HOMO-6 → LUMO		0.61605			
HOMO-5 → LUMO		-0.26408			
HOMO-4 → LUMO+1		-0.17764			
Excited State 23:	Singlet-A	3.4210 eV	362.43 nm	f=0.0081	<S**2>=0.000
HOMO-6 → LUMO		0.19753			
HOMO-4 → LUMO+1		0.66239			
Excited State 24:	Singlet-A	3.4799 eV	356.28 nm	f=0.0023	<S**2>=0.000
HOMO-6 → LUMO+1		0.29837			
HOMO-5 → LUMO+1		0.61369			
HOMO-4 → LUMO+1		0.13070			
Excited State 25:	Singlet-A	3.4705 eV	357.25 nm	f=0.1369	<S**2>=0.000
HOMO-3 → LUMO+2		0.61815			
HOMO-3 → LUMO+4		-0.10975			
HOMO-2 → LUMO+5		-0.20161			
Excited State 26:	Singlet-A	3.5431 eV	349.93 nm	f=0.0019	<S**2>=0.000
HOMO-6 → LUMO+1		0.62159			
HOMO-5 → LUMO+1		-0.31774			
Excited State 27:	Singlet-A	3.6226 eV	342.25 nm	f=0.0319	<S**2>=0.000
HOMO-3 → LUMO+2		0.29602			
HOMO-3 → LUMO+3		-0.15746			
HOMO-3 → LUMO+4		0.15212			
HOMO-2 → LUMO+1		0.11932			
HOMO-2 → LUMO+3		0.11942			
HOMO-2 → LUMO+5		0.38209			
HOMO-1 → LUMO+4		-0.16759			
HOMO-1 → LUMO+5		0.13135			
HOMO-1 → LUMO+6		-0.13681			
HOMO → LUMO+3		-0.12359			
HOMO → LUMO+4		0.10204			
HOMO → LUMO+5		0.15961			
HOMO → LUMO+6		0.16193			
Excited State 28:	Singlet-A	3.7046 eV	334.68 nm	f=0.0183	<S**2>=0.000
HOMO-3 → LUMO+3		0.65137			

HOMO-3 → LUMO+4		0.10454			
HOMO → LUMO+6		0.18204			
Excited State 29:	Singlet-A	3.7280 eV	332.57 nm	f=0.0016	<S**2>=0.000
HOMO-3 → LUMO+3		-0.14069			
HOMO-3 → LUMO+4		-0.10363			
HOMO-1 → LUMO+6		0.29755			
HOMO → LUMO+6		0.59402			
Excited State 30:	Singlet-A	3.7945 eV	326.75 nm	f=0.0016	<S**2>=0.000
HOMO-3 → LUMO+4		0.64044			
HOMO-2 → LUMO+5		-0.12190			
HOMO-1 → LUMO+6		0.16686			
Excited State 31:	Singlet-A	3.8290 eV	323.80 nm	f=0.0261	<S**2>=0.000
HOMO-7 → LUMO		0.69187			
Excited State 32:	Singlet-A	3.9065 eV	317.38 nm	f=0.0070	<S**2>=0.000
HOMO-3→ LUMO+5		0.68495			
Excited State 33:	Singlet-A	3.9750 eV	311.91 nm	f=0.0354	<S**2>=0.000
HOMO-7 → LUMO+1		0.66009			
HOMO-1 → LUMO+6		0.18053			
Excited State 34:	Singlet-A	4.0105 eV	309.15 nm	f=0.1408	<S**2>=0.000
HOMO-7 → LUMO+1		0.19544			
HOMO-2 → LUMO+5		0.10233			
HOMO-2 → LUMO+6		0.30951			
HOMO-1 → LUMO+6		0.48401			
HOMO → LUMO+6		-0.21476			
HOMO → LUMO+7		0.13175			
Excited State 35:	Singlet-A	4.0324 eV	307.47 nm	f=0.0362	<S**2>=0.000
HOMO-8 → LUMO		0.10459			
HOMO-2 → LUMO+6		0.61402			
HOMO-1 → LUMO+6		-0.24245			
HOMO → LUMO+6		0.10131			
Excited State 36:	Singlet-A	4.0625 eV	305.19 nm	f=0.0167	<S**2>=0.000
HOMO-8 → LUMO		0.68072			
HOMO-7 → LUMO+1		-0.10715			
Excited State 37:	Singlet-A	4.1119 eV	301.52 nm	f=0.0012	<S**2>=0.000
HOMO-4 → LUMO+2		0.69442			
Excited State 38:	Singlet-A	4.1758 eV	296.91 nm	f=0.0074	<S**2>=0.000
HOMO-6 → LUMO+2		0.19695			
HOMO-5 → LUMO+2		0.66676			
Excited State 39:	Singlet-A	4.2046 eV	294.88 nm	f=0.0019	<S**2>=0.000
HOMO-8 → LUMO+1		0.69155			
Excited State 40:	Singlet-A	4.2484 eV	291.84 nm	f=0.0202	<S**2>=0.000
HOMO-6 → LUMO+2		0.65461			
HOMO-5 → LUMO+2		-0.20736			
Excited State 41:	Singlet-A	4.2732 eV	290.14 nm	f=0.3874	<S**2>=0.000
HOMO-11 → LUMO		-0.25012			
HOMO-10 → LUMO		0.59944			

HOMO-9 → LUMO+1 -0.19353
Excited State 42: Singlet-A 4.2806 eV 289.64 nm f=0.1365 <S**2>=0.000
HOMO-11 → LUMO+1 -0.16711
HOMO-9 → LUMO 0.48601
HOMO-4 → LUMO+3 -0.43224
HOMO → LUMO+7 -0.13805
Excited State 43: Singlet-A 4.2837 eV 289.43 nm f=0.0759 <S**2>=0.000
HOMO-10 → LUMO+1 -0.16875
HOMO-9 → LUMO 0.34990
HOMO-4 → LUMO+3 0.52656
HOMO → LUMO+7 -0.20024
Excited State 44: Singlet-A 4.3084 eV 287.77 nm f=0.0093 <S**2>=0.000
HOMO-12 → LUMO -0.10447
HOMO-11 → LUMO+1 -0.13280
HOMO-9 → LUMO 0.28049
HOMO → LUMO+7 0.55784
Excited State 45: Singlet-A 4.3436 eV 285.44 nm f=0.0008 <S**2>=0.000
HOMO-6 → LUMO+3 0.24233
HOMO-5 → LUMO+3 0.55910
HOMO-4 → LUMO+3 0.11707
HOMO-4 → LUMO+4 0.29448
Excited State 46: Singlet-A 4.3567 eV 284.58 nm f=0.0047 <S**2>=0.000
HOMO-5 → LUMO+3 -0.31591
HOMO-4 → LUMO+4 0.61420
Excited State 47: Singlet-A 4.3633 eV 284.15 nm f=0.0774 <S**2>=0.000
HOMO-11 → LUMO 0.44636
HOMO-10 → LUMO 0.17797
HOMO-9 → LUMO+1 -0.25699
HOMO-3 → LUMO+6 -0.35222
HOMO-1 → LUMO+7 0.11069
HOMO → LUMO+8 -0.14796
Excited State 48: Singlet-A 4.3830 eV 282.87 nm f=0.0093 <S**2>=0.000
HOMO-11 → LUMO -0.10990
HOMO-10 → LUMO+1 0.10769
HOMO-9 → LUMO+1 0.13420
HOMO-3 → LUMO+6 -0.12364
HOMO-1 → LUMO+7 0.54337
HOMO-1 → LUMO+8 0.13923
HOMO → LUMO+7 -0.10395
HOMO → LUMO+8 0.27376
Excited State 49: Singlet-A 4.3978 eV 281.92 nm f=0.0552 <S**2>=0.000
HOMO-11 → LUMO -0.14298
HOMO-3 → LUMO+6 -0.38226
HOMO-1 → LUMO+7 -0.31807
HOMO-1 → LUMO+8 0.11476
HOMO → LUMO+7 0.10275

HOMO → LUMO+8			0.38843	
Excited State 50:	Singlet-A	4.4041 eV	281.52 nm	f=0.1311 <S**2>=0.000
HOMO-11 → LUMO			0.32021	
HOMO-6 → LUMO+3			-0.30760	
HOMO-5 → LUMO+3			0.12886	
HOMO-3 → LUMO+6			0.30620	
HOMO → LUMO+8			0.36642	



1^{+} **1** **1**$^{-}$

Fig. S8 Optimized geometries and labeling schemes of $[(bipy)_2Os(H_2Imdc)]^+$ (**1**$^+$), $[(bipy)_2Os(HImdc)]$ (**1**), and $[(bipy)_2Os(Imdc)]^-$ (**1**$^-$) in triplet state in a solution phase.