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Supporting information

 Table S1. IR spectra of pristine compounds and salts 1-3.

Components	Cryptand	Bu ₄ PBr	Flavanthrone	$\{Crypt(Na^+)\}$	$(Bu_4P^+)_2$	$(Bu_4P^+)_2$
r r			(Flav)	(Flav [•])	$(Flav^{2-})$	$(Flav^{2-})$
			()		0.5C H Cl	(1 111)
				(1)	$(0.5C_6\Pi_4CI_2)$	(3)
F 1			422	(1)	(2)	(3)
Flav			432W	434W	439W	433W
			530w	529m*	533m	533m
			566m	558w	555w	553w
			589w	587w	588w	586w
			637m	637w*	632w	633w
			669m	668w*	676w*	668w
			686s	685m	706m	704w
			7238	721s 724s* sp	717m*	720m*
			785m	, 210 / 210 op	796w	796w
			800m	806w	7.70W	750%
			812	800w	925*	015
			815W	822W	825W*	815W
			856s	-	-	-
			901w	-	903w*	903w*
			947m	-	-	-
			995w	996w	-	1000m*
			1092w	1089s	1093w*	1115m
			1106w	1103vs	1131s*	1132m
			1183w	1176m	1171s	11728
			1217w	-	1214w*	1214w*
			1231	_	12220*	12320*
			1251w	- 1267m	12525	12325
			12058	120/11	12/98	12/98
			13058	1300m	-	-
			1336w	1345s*	1335w	1337w
			1444w	1441m*	1434vs	1436vs
			1481w	1485m*	1460vs	1458vs*
			-	1509vs	1506s	1506s
			-	-	1519m	1520w
			1554w	1549m	1549m	1549m
			1591m	1571vs	15758	15758
			_	1604w	1602w	1602w
			1660s	1663w	1002.0	1002.
			2052	2057w	2061	2056
			5052W	5057W	5001W	3030W
Cation ⁺	Crypt(Na ⁺)	Bu_4P^+		Crypt(Na ⁺)	Bu_4P^+	Bu_4P^+
	476w	466w		-	-	-
	528w	698w		529m*	-	-
	581w	723s		587w*	717m*	720m*
	735m	747m		724s*	753w	750w*
	922m	787w		927w	776w	778w
	9/8w	812m		,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	-	815w*
	082m	820c		-	825m*	015W
	902III 1029	0.005		-	023W**	*
	1038W	906m		103/5*	903W	903W*
	10/1m	9218		10/8s	921w	922w
	1100s	966w		1103vs*	962w	964w
	1127s	1002w		1134m 1141m	-	-
	1213w	1053w		-	-	-
	1295m	1071w		1290s	1078s*	1078s
	1329m	1091s		1319s	1093w	-
	1360s	1206w		1345s*	1214w*	1214w*
	1446m	1234w		1441m*	1232s*	1232s*
	1462m	1285w		1462s*	1279s*	1279s*
	1/10/11	1316		1/85m*	1315m	1316
	2700	1376m		2810	1392	1394
	2190W	1410		2010W	1303W	1304W
	28//W 2042	1419m		28/0W	-	1416VS
	2943W	14068		2972w	1460vs	1458vs
		2871m			2870m	2850w
		2910m			2927m	2920w
		2934m			-	-
		2959m			2958m	2956w
				668w	676w*	
Solvent				-	753w	
C ₄ H ₄ Cl ₂				1037s*	1030m	
~0· ·4 ~· ·2				10575	11310*	
				1/62**	1/60%	
				1402S**	1400VS*	

Abbreviation: w: weak, m: middle, s- strong, vs-very strong, sp. – split band, *: bands are overlapped.

Table S2. IR spectra of pristine compounds and compounds 4-5.

Components	Flavanthrone	QCl ₄	Cp ₂ V	${(Cp_2V^+)_2}$	$\{(Cp_2V^{\scriptscriptstyle +})_2$
	(Flav)		1 -	$(Flav^{2-})$	(QCl_4^{2-})
				$\cdot C_6 H_4 Cl_2$ (4)	(5)
Flav	432w	470w		447w	-
(QCl ₄)	530w	714m		-	706w
	566m	752m		556w	-
	589w	905w		592w	902m
	637m	1112s		631w	1103w
	669m	1212w		6/0m*	-
	080S	1254W 1250w		089W	- 1264w
	7253 785m	1319w		7103 7203 777m	1204w
	800w	1491w		800s*	
	813w	1570m		808s*	
	856s	1680s sp		856w	1420vs
	901w	1691s sp		903w	1631w
	947m			944w	
	995w			1008m	
	1092w			1080s1096w	
	1183w			1121w 1180m*	
	1217w			-	
	1231w			1227s	
	1265s			1281s	
	1305s			1305m	
	1336w			1333w	
	1444w			1416vs	
	1481w			1462s*	
	-			-	
	1554w			1550m	
	1591m			1587s	
	-			-	
	1660s			1657m	
	3052w			3058w	
Cp_2V^+			121	1.17	110
			424w	44'/w	448W
			-	507W 670m	508W 673w
			769s	800s	804s
			-	808s	-
			-	917w	922m
			1005m	1020m	1020m
			1106w	1180m*	1176w 1185w
			1424w	-	-
			2854W 2924m	2850W	2851W 2020w
			2924III 2955w	2916w	2920w 2958w
			3101 w	3125w	3116w
			0101	670m*	0110
C.H.Ch				-	
~ ₀ ,14~12				1032m	
				-	
	1	1	1	1462s*	1

Abbreviation: w: weak, m: middle, s- strong, vs-very strong, sp. – split band, *: bands are overlapped

IR spectra of starting metal phthalocyanines and their anion-radical salts



Figure S1. IR spectrum of pristine flavanthrone in KBr pellet.



Figure S2. IR spectrum of radical-anion $\{Crypt(Na^+)\}(Flav^{\bullet-})\cdot 2C_6H_4Cl_2$ (1) salt in KBr pellet prepared in anaerobic conditions.



Figure S3. IR spectrum of dianion salt $(Bu_4P^+)_2(Flav^{2-})\cdot 0.5C_6H_4Cl_2(2)$ in KBr pellet prepared in anaerobic conditions.



Figure S4. IR spectrum of dianion salt $(Bu_4P^+)_2(Flav^{2-})$ (**3**) in KBr pellet prepared in anaerobic conditions.



Figure S5. IR spectrum of $\{(Cp_2V^+)_2(Flav^{2-})\}\cdot C_6H_4Cl_2$ (4) in KBr pellet prepared in anaerobic conditions.



Figure S6. IR spectra of pristine chloranil (QCl₄) and compound $\{(Cp_2V^+)(QCl_4^{2-})_2\}$ (5) in KBr pellet prepared in anaerobic conditions.



Figure S7. View on the packing of the salt $(Bu_4P^+)_2(Flav^{2-})\cdot 0.5C_6H_4Cl_2$ (2). Solvent *o*-dichlorobenzene molecules are not shown. On the figure carbon atoms are brown, oxygen atoms are red, nitrogen atoms are blue and phosphorus atoms are is orange.



Figure S8. View on the packing of the $(Bu_4P^+)_2(Flav^{2-})$ (3) salt. On the figure carbon atoms are brown, oxygen atoms are red, nitrogen atoms are blue and phosphorus atoms are is orange.

UV-visible-NIR spectra of starting metal phthalocyanines and thier anion-radical salts.



Figure S9. Spectrum of salt **3** in the UV-visible-NIR ranges in KBr pellet prepared in anaerobic conditions.



Data of magnetic measurements for obtained compounds.

Figure S10. Temperature dependence of effective magnetic moment of polycrystalline **1** in the 1.9-300 K range.



Figure S11. Temperature dependence of effective magnetic moment of polycrystalline **4** in the 1.9-300 K range.



Figure S12. Temperature dependence of reciprocal molar magnetic susceptibility of polycrystalline **4** in the 1.9-300 K range.



Figure S13. Temperature dependence of effective magnetic moment of polycrystalline **5** in the 1.9-300 K range.



Figure S14. Temperature dependence of reciprocal molar magnetic susceptibility of polycrystalline **5** in the 1.9-300 K range.



Figure S15. Calculated geometry of neutral flavanthrone.



Figure S16. Calculated geometry of flavanthrone^{•-} radical anion



Figure S17. Calculated geometry of flavanthrone^{2–} dianion.



Figure S18. Calculated geometry of {(Cp₂V)₂Flavanthrone} coordination unit.

DFT calculations were performed using the PBE density functional theory¹ with the extended basis set V: [9s9p8d/5s5p4d], O, N, C: [5s5p2d/3s3p2d], H: [5s1p/3s1p] for the valence electrons and the SBK pseudopotential². The Hirschfeld method was applied to determine the charge on the atoms.³ Electronic spectra are estimated using results of TDDFT calculations and fixed linewidth 30 nm of Gaussian peaks. All calculations were carried out using the PRIRODA program⁴ at Joint Supercomputer Center of Russian Academy of Sciences.

The flat flavanthrone molecule has C_{2h} symmetry. When the first and second electrons are added, its symmetry is conserved and the charge on the heteroatoms O, -0.222, and N, -0.130, does not change much -0.287, -0.157 and -0.360, -0.183, respectively. This indicates that approximately 80% of the electron density is accepted by the conjugated π -electron system. A slight elongation of the C – O bond in the calculated structures by 0.017 A for each additional electron corresponds to this picture (see Figs. S15-S17).

The theoretical values of the C – O bond lengths 1.235 (1.21(3)) and C-N 1.327 (1.30(3)), 1.374 (1.37(3)) Å in the neutral flavanthrone molecule; C-O 1.252 (1.284(4)), C-N 1.344 (1.342(4)), 1.360 (1.362(4)) Å in the flavanthrone^{•–} radical anion; C-O 1.269 (1.271(3)), C-N 1.349 (1.348(3)) 1.360 (1.360(3)) Å in the flavanthrone^{2–} dianion are in good agreement with experimental data (in brackets).

The calculated bond lengths for the $(Cp_2V)_2$ (Flavanthrone) complex: V-O 1.986, O – C 1.318, C-N 1.343 and 1.357 Å, and the C-O-V angle is 140.2° are also in good agreement with the

experimental data 1.971(1), 1.312(2) 1.339(3), 1.363(3) Å and 140.6°, respectively The average bond lengths of the V-C(Cp) is 2.312 and 2.284(3) Å for calculated and experimental structures, respectively.

In the flavanthrone molecule, the HOMO (a_g) and HOMO-2 (b_u) consist mainly of lone pairs of oxygen atoms. HOMO-3 (a_u), HOMO-1 (b_g), LUMO (a_u), LUMO + 1 (b_g), LUMO + 2 (a_u) LUMO + 3 (b_g) are π -orbitals. HOMO (a_g) - LUMO (a_u) (679 nm) transition does not appear in the spectrum due to the small moment of transition between σ - and π -orbitals. The most intense transition in the visible region, the band at 528 nm, is due to the HOMO-1 (b_g) - LUMO (a_u) transition. The next π - π transition, which has a noticeable intensity in the visible region is at 441 nm, it can be attributed to the HOMO-7 (b_g) - LUMO (a_u) transition

Long-wave transitions SOMO (a_u) LUMO (b_g) at 934 nm and SOMO (a_u) LUMO + 1 (b_g) at 802 nm appear in the flavanthrone^{•-} radical anion. The most intense transition at 609 nm is the HOMO (b_g) - SOMO (a_u) , analogue of the HOMO - LUMO transition in the neutral system at 679 nm. The following π - π transitions, which have a noticeable intensity in the visible region, are the HOMO-5 (b_g) - LUMO (a_u) transition at 512 nm and the SOMO (a_u) - LUMO + 4 (b_g) transition at 484 nm.

LUMO and LUMO + 1 have the same b_g symmetry in the flavanthrone²⁻ dianion and close energies. Therefore, excited terms are a superposition of configurations corresponding to various one-electron excitations. This leads to a redistribution of intensities in the spectrum — a less intense transition at 934 nm and a very intense transition at 680 nm. The transition - HOMO (a_u) - LUMO + 2 (a_u) is forbidden in parity and the next transition, which has a noticeable intensity in the visible region is the HOMO (a_u) - LUMO + 6 (a_u) transition at 473 nm.

Upon coordination of Cp_2V^+ fragments on the dianion of the flavanthrone, a significant transfer of electron density to vanadium atoms occurs. As a result, the charge of the Cp_2V^+ fragments decreases to +0.341 (the charge on the V atom itself is +0.275, the spin density is 1.781). The negative charge on the ligand is concentrated mainly on oxygen (-0.202) and nitrogen (-0.151) heteroatoms. The elongation of CO bonds to 1.318 Å in calculated structure (Fig. S18) indicates a two-electron reduction of the ligand to diol.

Four single filled d - orbitals of metals are located below the HOMO - π -ligand orbital. The parity-allowed π - π transitions: HOMO(u)-LUMO(g), HOMO(u)-LUMO + 2(g), and HOMO(u)-LUMO + 4(g) transitions correspond to the absorption bands at 1207, 884, and 807 nm. The metal – ligand transitions are mixed-type transitions, since the corresponding excited states are the superpositions that also include configurations of π - π excitations. In these superpositions, configurations of u \rightarrow g and g \rightarrow u excitations are simultaneously present. These transitions lead

to the absorption in the calculated spectra at 967, 780, 679, 632 and 609 nm with the participation of LUMO (u), LUMO + 3 (g), LUMO + 5 (u) + LUMO + 4 (g), LUMO + 5 (u) + LUMO + 6 (g) and LUMO + 2 (g), respectively. The difference in the most intense transition at 609 nm is the largest amplitude of the contribution of π - π excitation HOMO(u)-LUMO + 2 (g). Interestingly, there is a proximity of intense transitions at 609 nm in the radical anion and 609 nm in the complex, there is small total charge on the π system in the both systems -0.112 and +0.025, respectively.

In general there is a reasonable similarity of calculated and observed spectra.

References:

- 1. J. P. Perdew, K. Burke, M. Ernzerhof, Phys. Rev. Lett. 1996, 77, 3865–3868.
- 2. W. J. Stevens, H. Basch, M. Krauss, J. Chem. Phys. 1984, 81, 6026–6033.
- 3. F. L. Hirshfeld, *Theor. Chim. Acta*, 1977, **44**, 129–138.
- 4. D. N. Laikov, Chem. Phys. Lett. 2005, 416, 116–120.

Cartesian coordinates of the optimized structure of flavanthrone molecule.

Jui v	containa	tes of the opti-	millea stractar
8	-2.18312755	6.58468525	-2.80348011
7	0.61879620	4.69430643	-7.15807762
8	4.51869768	-0.39652582	-7.68975478
7	1.71834886	1.49500468	-3.33464663
6	-0.58332414	4.24669938	-2.45500941
6	-0.98234119	7.01402644	-7.45934799
6	-0.88703405	6.33712956	-6.22851638
6	-0.04706286	5.13263283	-6.09769335
6	0.00986829	4.48109142	-4.80551989
6	-0.69727864	4.94445108	-3.67538938
6	-1.55309828	6.15654686	-3.77520306
6	-1.61013015	6.82700406	-5.10980291
6	-2.40798866	7.97611733	-5.24557260
6	-1.77846684	8.15304748	-7.57793185
6	0.81616316	3.32541773	-4.68200424
6	0.21225883	3.10765715	-2.33589233
6	-2.49380828	8.63719551	-6.47033142
6	0.93412456	2.61679831	-3.44937482
1	-1.14189442	4.62892284	-1.59902796
1	0.29387175	2.57493289	-1.38741983
1	-3.11561826	9.52863537	-6.56705925
1	-1.84412405	8.66890854	-8.53746117
1	-0.42174271	6.62688150	-8.30998393
6	2.92008846	1.94233541	-8.03783663
6	3.31943949	-0.82474929	-3.03339003
6	3.22399034	-0.14795364	-4.26426881
6	2.38423795	1.05670195	-4.39502034
6	2.32735222	1.70828027	-5.68717786
6	3.03419500	1.24469528	-6.81740673
6	3.88938948	0.03214561	-6.71779750
6	3.94670157	-0.63811081	-5.38310729
6	4.74433856	-1.78738809	-5.24740957
6	4.11538830	-1.96389854	-2.91486387
6	1.52119343	2.86405126	-5.81064630
6	2.12471552	3.08153220	-8.15688397
6	4.83034204	-2.44832967	-4.02259092
6	1.40311065	3.57257691	-7.04331668
1	3.47830858	1.55986210	-8.89393457
1	2.04290525	3.61410947	-9.10542192
1	5.45196337	-3.33990762	-3.92592139
1	4.18113998	-2.47969597	-1.95530671
1	2.75909830	-0.43741212	-2.18267253
1	-2.95178732	8.32839422	-4.36798541
1	5.28775342	-2.13994431	-6.12512187

Cartesian coordinates of the optimized structure of flavanthrone^{•-} radical anion.

- 6 -0.63682657 4.36413122 -2.50425223
- 6 -0.96823837 6.98223958 -7.58891451
- 6 -0.88317325 6.33253041 -6.33630315
- 6 -0.05275287 5.13932495 -6.16532138
- 6 -0.01584190 4.53342470 -4.86226518

6	-0.74350014	5.03970968	-3.74929991
6	-1.58419304	6.23044373	-3.88358804
6	-1.62018187	6.85639798	-5.23514296
6	-2.41421207	8.00682393	-5.42099106
6	-1.75775030	8.11620097	-7.75039194
6	0.78926996	3.37570512	-4.69465529
6	0.14913675	3.23501559	-2.34239433
6	-2.48683051	8.63423497	-6.66063269
6	0.89262753	2.69926267	-3.43690367
8	-2.24023851	6.70698301	-2.92936125
7	0.63321521	4.66329488	-7.21830496
1	-1.20619000	4.77439403	-1.66774796
1	0.21971260	2.72787136	-1.37820747
1	-3.10635637	9.52486482	-6.78929769
1	-1.81158006	8.60542822	-8.72590114
1	-0.39680016	6.56584410	-8.41906692
6	2.93372224	1.88976292	-7.99358035
6	3.26498919	-0.72846412	-2.90896477
6	3.17994801	-0.07873311	-4.16156814
6	2.34991793	1.11476574	-4.33242237
6	2.31309337	1.72073275	-5.63545043
6	3.04039240	1.21418889	-6.74852744
6	3.88042049	0.02296644	-6.61445870
6	3.91643923	-0.60297483	-5.26289514
6	4.70983675	-1.75386839	-5.07725199
6	4.05386133	-1.86289959	-2.74769486
6	1.50820050	2.87861525	-5.80299506
6	2.14811111	3.01913403	-8.15533534
6	4.78235264	-2.38136027	-3.83764521
6	1.40478186	3.55500779	-7.06076747
8	4.53577190	-0.45409254	-7.56890264
7	1.66404000	1.59086107	-3.27941330
1	3.50265402	1.47910304	-8.83019265
1	2.07745829	3.52623411	-9.11953930
1	5.40131635	-3.27240752	-3.70916514
1	4.10762211	-2.35218635	-1.77221183
1	2.69388572	-0.31180890	-2.07871193
1	-2.96480574	8.37972314	-4.55531829
1	5.25997625	-2.12710689	-5.94306712

Cartesian coordinates of the optimized structure of flavanthrone^{2–} dianion.

6	-1.76488182	1.31798234	1.99292286
6	-2.08998418	3.93040901	-3.09201515
6	-1.99826939	3.27107400	-1.83877476
6	-1.17328084	2.08783248	-1.67279508
6	-1.14113318	1.48815762	-0.37184722
6	-1.87726791	2.00403555	0.74375905
6	-2.71186678	3.18233648	0.62011464
6	-2.74081431	3.79786134	-0.72919766
6	-3.53448471	4.95186361	-0.92605327
6	-2.87795325	5.06296164	-3.25685086
6	-0.33322661	0.32529109	-0.20174425

6	-0.98411124	0.19381142	2.15856736
6	-3.61050011	5.58259726	-2.16230529
6	-0.23054656	-0.35403456	1.06188268
8	-3.38116434	3.67491512	1.57916403
7	-0.47656265	1.60270897	-2.73533293
1	-2.33675152	1.73261931	2.82719605
1	-0.91392614	-0.31401929	3.12413651
1	-4.23179914	6.47461223	-2.28995657
1	-2.93097545	5.55224844	-4.23446897
1	-1.51656981	3.51170069	-3.92097660
6	1.82159626	-1.16900096	-3.50467173
6	2.14459377	-3.78277239	1.57964328
6	2.05348951	-3.12312799	0.32653942
6	1.22941608	-1.93921830	0.16082141
6	1.19763069	-1.33929654	-1.14000036
6	1.93372855	-1.85524544	-2.25558759
6	2.76763773	-3.03407271	-2.13213666
6	2.79577740	-3.65018408	-0.78306284
6	3.58853605	-4.80485372	-0.58636496
6	2.93145280	-4.91612046	1.74426670
6	0.38990770	-0.17628631	-1.31001968
6	1.04108640	-0.04464130	-3.67022866
6	3.66375176	-5.43605754	0.64971382
6	0.28725077	0.50304917	-2.57362997
8	3.43674910	-3.52681369	-3.09123335
7	0.53304818	-1.45388671	1.22344628
1	2.39348617	-1.58363504	-4.33893245
1	0.97108957	0.46328867	-4.63575783
1	4.28442079	-6.32852533	0.77726090
1	2.98392577	-5.40571491	2.72176134
1	1.57154408	-3.36366158	2.40865435
1	-4.08279638	5.31860107	-0.05464339
1	4.13676835	-5.17175403	-1.45775642

Cartesian coordinates of the optimized structure of $\{(Cp_2V)_2Flavanthrone\}$ complex

23	3.91933058	1.00062116	11.39925719
8	4.63655664	-0.04017489	9.86733775
7	4.84336152	-0.02430726	4.28969135
6	4.08706847	0.27881861	13.56687408
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