Table S-1 Fitting parameters for the three speciation models considered

$\log\beta_{pqr}$	Model 1[a]				Model 2[b]				Model 3[c]			
	Citrate	DL-tartrate	D-tartrate	L-tartrate	Citrate	DL-tartrate	D-tartrate	L-tartrate	Citrate	DL-tartrate	D-tartrate	L-tartrate
LH	5.57	3.87	3.87	3.87	5.57	3.87	3.87	3.87	5.57	3.87	3.87	3.87
LH_2	9.84	6.63	6.65	6.65	9.84	6.63	6.65	6.65	9.84	6.63	6.64	6.64
LH_3	12.71	-	-	-	12.71	-	-	-	12.71	-	-	-
VOLH	10.68				10.68	5.63	5.68	5.60	10.65			
VOL	7.50	4.43	4.48	4.38	7.50	4.43	4.48	4.38	7.85	3.99	4.04	3.80
$VOLH_{-1}$	3.90	1.36	1.40	1.38	3.90	1.36	1.40	1.38				
$VOLH_{-2}$	-5.60				-5.60				-5.7			
$(VO)_2L_2H_{-1}$	15.71	9.01	9.34	9.31	15.71	9.01	9.34	9.31	15.71	9.01	9.34	9.31
$(VO)_2L_2H_{-2}$	10.53	6.04	6.37	6.25	10.53	6.04	6.37	6.25	10.73	6.05	6.37	6.24
$(VO)_2L_2H_{-3}$	1.60	-0.47	-0.49	-0.82	1.60	-0.47	-0.49	-0.82	1.6	-0.48	-0.49	-0.81
$(VO)_2L_2H_{-4}$		-4.90	-5.60	-5.85		-4.90	-5.60	-5.85		-4.86	-5.61	-5.80
VOL_2H_{-2}	-2.89	-2.22	-2.22	-2.28	-2.89	-2.22	-2.22	-2.28	-2.89			
VOL_2H_{-3}		-13.04	-13.00	-13.58		-13.04	-13.00	-13.58				
VOL_2H_{-4}		-24.58	-24.56	-24.66		-24.58	-24.56	-24.66				
σ	1.2	1.1	1.1	1.1	1.2	1.8	1.8	1.8	2.2	3.2	3.3	3.4
X^2	6.1	5.1	5.1	5.1	6.1	7.9	7.9	7.9	9.1	9.2	9.3	9.4

[a] Model 1: final data reported in Table 1 of the manuscript. [b] Model 2: data with VOLH for tartrates. [c] Model 3: data reported in *Inorg. Chim. Acta*, 1995, **239**, 145.

Table S-2 Structural parameters of 59 structures of VO(IV) complexes formed by bidentate ligand

Ligand	Donor set	V-L_{eq} [a]	V-L'$_{ax}$ [a]	Δd [b]	L_{eq}-V-L_{eq} [c]	L'$_{ax}$-V-L'$_{ax}$ [c]	τ [d]	Ref.
Acetylacetonate	$VO(O_4)$	1.96, 1.97	1.96, 1.98	-0.0050	149.80	149.50	0.0050	1
Acetylacetonate	$VO(O_4)$	1.962, 1974	1.955, 1.983	-0.0010	149.60	145.50	0.0683	2
Acetylacetonate	$VO(O_4)$	1.967, 1.968	1.969, 1.970	-0.0020	150.13	145.63	0.0750	3
Acetylacetonate	$VO(O_4)$	1.968, 1.969	1.976, 1.977	-0.0080	150.24	146.20	0.0673	4
Acetylacetonate	$VO(O_4)$	1.969, 1.974	1.976, 1.978	-0.0055	150.13	145.63	0.0750	5
Acetylacetonate	$VO(O_4)$	1.965, 1.967	1.968, 1.972	-0.0040	150.24	145.63	0.0768	6
Acetylacetonate	$VO(O_4)$	1.973, 1.975	1.980, 1.983	-0.0075	150.24	146.20	0.0673	7
Acetylacetonate	$VO(O_4)$	1.973, 1.975	1.980, 1.982	-0.0070	150.27	146.29	0.0663	7
Acetylacetonate	$VO(O_4)$	1.970, 1.973	1.978, 1.980	-0.0075	150.23	146.35	0.0647	7
Acetylacetonate	$VO(O_4)$	1.968, 1.972	1.976, 1.979	-0.0075	150.25	146.40	0.0642	7
Acetylacetonate	$VO(O_4)$	1.971, 1.976	1.973, 1.977	-0.0015	150.26	146.42	0.0640	7
Catecholate	$VO(O_4)$	1.958, 1.959	1.933, 1.959	0.0125	149.95	141.08	0.1478	8
2,2,6,6-Tetramethylheptane-3,5-dionate	$VO(O_4)$	1.957, 1.965	1.952, 1.972	-0.0010	147.39	145.89	0.0250	9
2,2,6,6-Tetramethylheptane-3,5-dionate	$VO(O_4)$	1.967, 1.972	1.963, 1.980	-0.0020	147.63	146.04	0.0265	5
N,*N*-Dimethylacetylacetamidate-O,O'	$VO(O_4)$	1.958, 1.966	1.959, 1.976	-0.0035	145.08	144.97	0.0018	10
N-Diphenylphosphoryl-*P*,*P*-diphenylphosphinimidate-O,O'	$VO(O_4)$	1.982, 1.987	1.977, 1.984	0.0040	152.30	143.60	0.1450	11
3-Ethyl-2,4-pentanedionate-O,O'	$VO(O_4)$	1.962, 1.965	1.962, 1.966	-0.0005	147.86	146.56	0.0217	12
3-Methyl-2,4-pentanedionate-O,O'	$VO(O_4)$	1.956, 1.958	1.958, 1.971	-0.0075	149.61	143.45	0.1027	12
(S)-2-Diphenylphosphinoyl-1,1'-binaphathalene-2'-olate	$VO(O_4)$	2.042	1.901	0.1410	156.50	133.80	0.3783	13
Benzilate	$VO(O_4)$	1.970, 1.973	1.900, 1.931	0.0560	151.62	132.93	0.3115	14

Ligand	Donor set	V-L_{eq} [a]	V-L'$_{ax}$ [a]	Δd [b]	L_{eq}-V-L_{eq} [c]	L'$_{ax}$-V-L'$_{ax}$ [c]	τ [d]	Ref.
2-Ethyl-2-hydroxybutanoate	$VO(O_4)$	1.954, 1.977	1.882, 1.986	0.0315	147.80	136.31	0.1915	15
1,3-Dithiolan-2-ylidenemalonate-O,O'	$VO(O_4)$	1.946	1.941	0.0050	157.94	155.04	0.0483	16
1,3-dithiolan-2-ylidenemalonate-O,O'	$VO(O_4)$	1.952, 1.955	1.940, 1.943	0.0120	145.80	142.47	0.0555	16
6-(Phenylamino)hex-5-ene-2,4-dionate-O,O'	$VO(O_4)$	1.972, 1.978	1.944, 1.952	0.0270	151.90	141.90	0.1667	17
1-Phenyl-1,3-butanedionate	$VO(O_4)$	1.946, 1.969	1.953, 1.972	-0.0050	150.10	146.80	0.0550	2
1,3-Diphenyl-1,3-propanedionate	$VO(O_4)$	1.941, 1.944	1.941, 1.957	-0.0065	147.22	145.93	0.0215	18
Maltolate	$VO(O_4)$	1.998, 2.024	1.958, 1.971	0.0465	146.80	140.60	0.1033	19
N-(2-(2-((Methylthio)thiocarbonyl)cyclopenton-1-yl)amino)ethyl)salicylaldiminate	$VO(N_2O_2)$	2.099	1.915	0.1840	163.61	130.99	0.5437	20
N-(Salicylidene)propylamine	$VO(N_2O_2)$	2.107, 2.126	1.898, 1.904	0.2155	161.88	130.42	0.5243	21
2-(4,4-Dimethyl-4,5-dihydro-oxazol-2-yl)phenolate-N,O	$VO(N_2O_2)$	2.102, 2.106	1.909, 1.917	0.1910	165.36	129.64	0.5953	22
2-(5,6-Dihydro-4H-1,3oxazin-2-yl-)phenolate-N,O	$VO(N_2O_2)$	2.079, 2.086	1.883, 1.883	0.1995	165.15	127.67	0.6247	22
N-(4-Chlorophenyl)-salicylideneiminate	$VO(N_2O_2)$	2.145, 2.154	1.899, 1.904	0.2480	159.12	133.13	0.4332	23
2-(2-Oxyphenyl)-2-oxazolinate	$VO(N_2O_2)$	2.061, 2.069	1.926, 1.930	0.1370	151.48	142.77	0.1452	24
2-(2'-Oxyphenyl)-2-thiazolinate	$VO(N_2O_2)$	2.083	1.900, 1.903	0.1815	161.44	129.90	0.5257	24
2-(Dimethylaminomethyl)phenolate-O,N	$VO(N_2O_2)$	2.147, 2.155	1.883, 1.897	0.2610	165.13	130.02	0.5852	25
N-(2-oxido-5-chlorosalicylidene)-(S)-1-phenylethylamine	$VO(N_2O_2)$	2.076, 2.085	1.912, 1.915	0.1670	164.87	125.25	0.6603	26
N-(2-oxido-5-chlorosalicylidene)-(S)-1-phenylethylamine	$VO(N_2O_2)$	2.092, 2.095	1.914, 1.918	0.1775	164.54	127.46	0.6180	26
N-(2-oxidonaphthylmethylidene)-(S)-1-phenylethylamine	$VO(N_2O_2)$	2.065, 2.085	1.900, 1.904	0.1730	161.97	123.18	0.6465	26
N-(2-oxidonaphthylmethylidene)-(S)-1-phenylethylamine	$VO(N_2O_2)$	2.085, 2.100	1.910, 1.913	0.1810	161.45	127.42	0.5672	26
N-(2-oxido-salicylidene)-(R)-1-phenylethylamine	$VO(N_2O_2)$	2.100, 2.112	1.905, 1.910	0.1985	159.65	133.28	0.4395	26
2-Salicylideneaminato-1-hydroxyethane-N,O	$VO(N_2O_2)$	2.096, 2.112	1.892, 1.901	0.2075	159.66	135.29	0.4062	27

Ligand	Donor set	V-L_{eq} [a]	V-L'$_{ax}$ [a]	Δd [b]	L_{eq}-V-L_{eq} [c]	L'$_{ax}$-V-L'$_{ax}$ [c]	τ [d]	Ref.
2-Methyl-8-quinolinolate	$VO(N_2O_2)$	2.136	1.921	0.2150	160.98	127.16	0.5637	28
N-Methylsalicylaldiminate	$VO(N_2O_2)$	2.105	1.907	0.1980	162.47	129.56	0.5485	29
N-Isopropyl-*o*-methylsalicylaldiminate	$VO(N_2O_2)$	2.096	1.912, 1.915	0.1825	164.17	129.32	0.5808	29
N-Methyl-*o*-*t*-butyl-*p*-methylsalicylaldiminate	$VO(N_2O_2)$	2.076, 2.089	1.900, 1.903	0.1810	165.75	123.99	0.6960	29
2-Methyl-5-methylthio-8-hydroxyquinolate	$VO(N_2O_2)$	2.104, 2.106	1.934, 1.970	0.1530	157.89	127.91	0.4997	30
2-Methyl-5-methylthio-8-hydroxyquinolate	$VO(N_2O_2)$	2.111, 2.122	1.923, 1.948	0.1810	158.71	127.48	0.5205	30
2-((1S,2S,5R)(-)-Menthol)-pyridine	$VO(N_2O_2)$	2.078	1.8770	0.2010	150.22	132.93	0.2882	31
4-Trifluoromethoxysalicylideneaminato-N,O	$VO(N_2O_2)$	2.110, 2.121	1.891, 1.904	0.2180	157.40	132.19	0.4202	32
2-(5-Ethyl-oxazolinyl)phenylate	$VO(N_2O_2)$	2.050, 2.087	1.921, 1.923	0.1465	153.33	139.82	0.2252	33
2-(5-Isopropyl-oxazolinyl)phenylate	$VO(N_2O_2)$	2.068, 2.075	1.915, 1.916	0.1560	155.83	133.24	0.3765	33
Salicylideneiminate	$VO(N_2O_2)$	2.056, 2.058	1.917, 1.920	0.1385	152.29	139.21	0.2180	34
2-(Piperid-1-ylmethyl)-4-methylphenolate-N,O	$VO(N_2O_2)$	2.165, 2.165	1.883, 1.899	0.2740	161.17	131.20	0.4995	35
2-((Dimethylamino)methyl)-6-*t*-butylphenolate-N,O	$VO(N_2O_2)$	2.150, 2.151	1.904, 1.912	0.2425	173.25	125.33	0.7987	35
2-((S)-1-(Dimethylamino)ethyl)phenolate-N,O	$VO(N_2O_2)$	2.164, 2.169	1.900 1.904	0.2645	160.15	130.78	0.4895	35
2-((R)-1-(Dimethylamino)ethyl)phenolate-N,O	$VO(N_2O_2)$	2.154, 2.164	1.898, 1.901	0.2595	160.37	130.99	0.4897	35
Ethane-1,2-dithiolate-S,S'	$VO(S_4)$	2.376, 2.388	2.370, 2.377	0.0085	151.92	142.75	0.1528	36
Ethane-1,2-dithiolate-S,S'	$VO(S_4)$	2.376, 2.388	2.370, 2.377	0.0085	151.89	142.72	0.1528	37
Diethyldithiocarbamate	$VO(S_4)$	2.404, 2.406	2.387, 2.410	0.0065	145.15	141.46	0.0615	38

[a] Measured in Å; [b] Δd is the difference between the mean axial and the mean equatorial bond distance: $\Delta d = [d_{mean}(V\text{-}L'_{ax}) - d_{mean}(V\text{-}L_{eq})]$; [c] Measured in °; [d] Distortion towards the trigonal bipyramid: $\tau = \{[L'_{ax}\text{-}V\text{-}L'_{ax}] - [L_{eq}\text{-}V\text{-}L_{eq}]\}/60$.

Table S-3 Structural parameters of the dinuclear VO(IV) complexes formed by tartrates and their derivatives

Complex[a]	V-O(hydr)$_{eq}$[b]	V-O(carb)$_{ax}$[b]	O-V-O(hydr)$_{eq}$[c]	O-V-O(carb)$_{ax}$[c]	τ[d]	Ref.
$(NH_4)_4[(VO)_2(L\text{-}tartH_{-1})_2]\cdot 2H_2O$	1.787, 1.932	2.008, 2.028	123.19	161.94	0.646	39
$Rb_4[(VO)_2(D\text{-}tartH_{-1})_2]\cdot 2H_2O$	1.884, 1.910	1.999, 2.057	124.5	159.8	0.588	40
$Cs_4[(VO)_2(D\text{-}tartH_{-1})_2]\cdot 3H_2O$	1.893, 1.914	2.006, 2.024	125.0	160.3	0.588	40
$(Na)_4[(VO)_2(L\text{-}tartH_{-1})_2]\cdot 6H_2O$	1.890, 1.894	2.012, 2.016	124.98	163.04	0.634	41
$Na_4[(VO)_2(DL\text{-}tartH_{-1})_2]\cdot 12H_2O$	1.902, 1.917	1.994, 2.004	143.7	150.5	0.113	42
$(NEt_4)_4[(VO)_2(DL\text{-}tartH_{-1})_2]\cdot 8H_2O$	1.913, 1.955	2.021, 2.026	145.7	154.1	0.140	43
$Rb_4[(VO)_2(DL\text{-}tartH_{-1})_2]\cdot 2H_2O$	1.916, 1.918	1.989, 2.002	145.1	145.2	0.002	40
$Cs_4[(VO)_2(DL\text{-}tartH_{-1})_2]\cdot 2H_2O$	1.908, 1.920	1.997, 2.001	145.5	146.5	0.017	40
$\{(Ba)_2[(VO)_2(DL\text{-}tartH_{-1})_2]\cdot 2H_2O\}_n$	1.913, 1.927	1.940, 1.995	144.5	146.4	0.032	44
$(Na)_4[(VO)_2(DL\text{-}dmtH_{-1})_2]$	1.964, 1.974	1.981, 2.015	151.4	158.7	0.122	45
$(Na)_4[(VO)_2(DL\text{-}mmtH_{-1})_2]\cdot 14H_2O$	1.904, 1.912	1.979, 1.991	144.5	149.2	0.078	46
$(Na)_4[(VO)_2(DL\text{-}dmtH_{-1})_2]\cdot 6H_2O$	1.946, 1.956	1.969, 2.030	151.2	160.3	0.152	47

[a] dmt = dimethyltartrato(2-); mmt = *threo*-monomethyltartrato(2-); [b] Measured in Å; [c] Measured in °; [d] Distortion towards the trigonal bipyramid: $\tau = \{[\text{O-V-O (carb)}_{ax}] - [\text{O-V-O(hydr)}_{eq})]\}/60$.

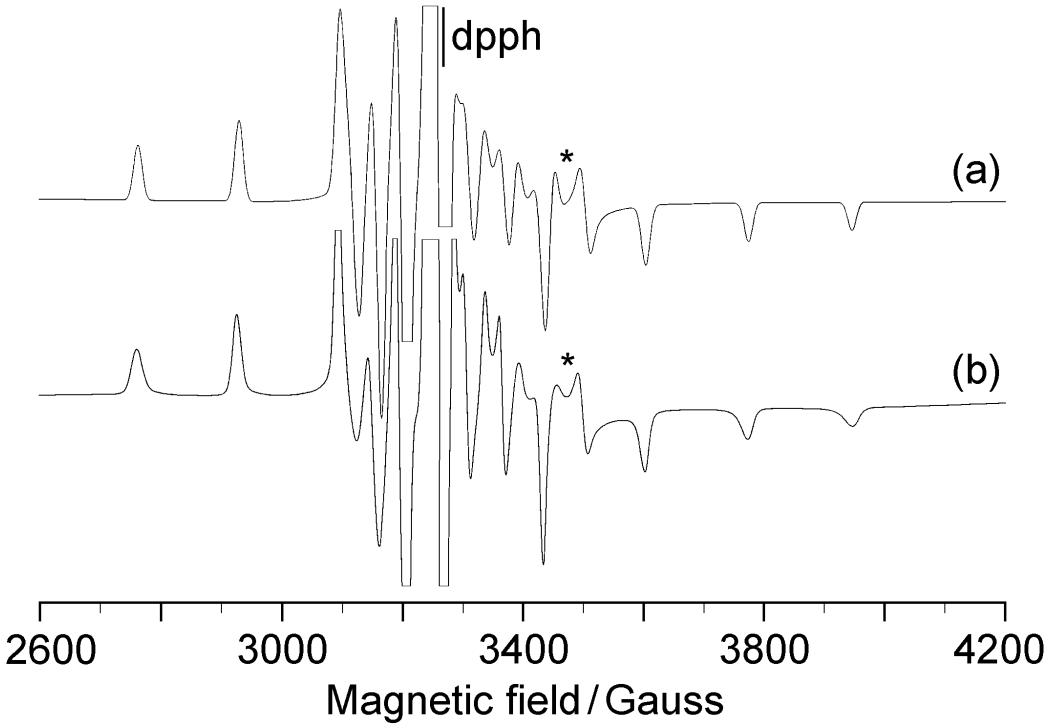


Fig. S-1 X-band anisotropic EPR spectrum of $[VO(citrH_{-1})_2]^{6-}$ recorded at 120 K on aqueous solution of VO(IV) and citrate at pH 8.30 with a ligand to metal molar ratio of 500:1 and a VO(IV) concentration of 4 mM: (a) simulated spectrum and (b) experimental spectrum. The region indicated with an asterisk is that where it is more easily observable the anisotropy of x and y axes.

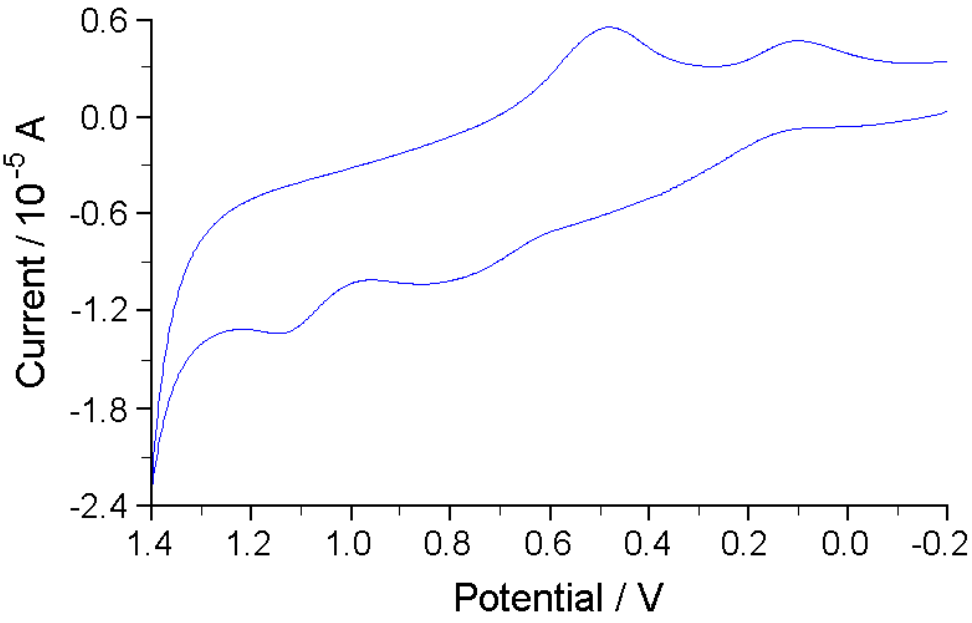


Fig. S-2 Cyclic voltammogram of $[(VO)_2(L\text{-}tartH_{-1})_2]^{4-}$ complex recorded at 25 °C on an aqueous solution of VO(IV) and L-tartrate: pH 9.0, ligand to metal molar ratio 1:1, VO(IV) concentration 5 mM. Au was used as working electrode and the potential *E* was measured vs. Ag/AgCl reference.

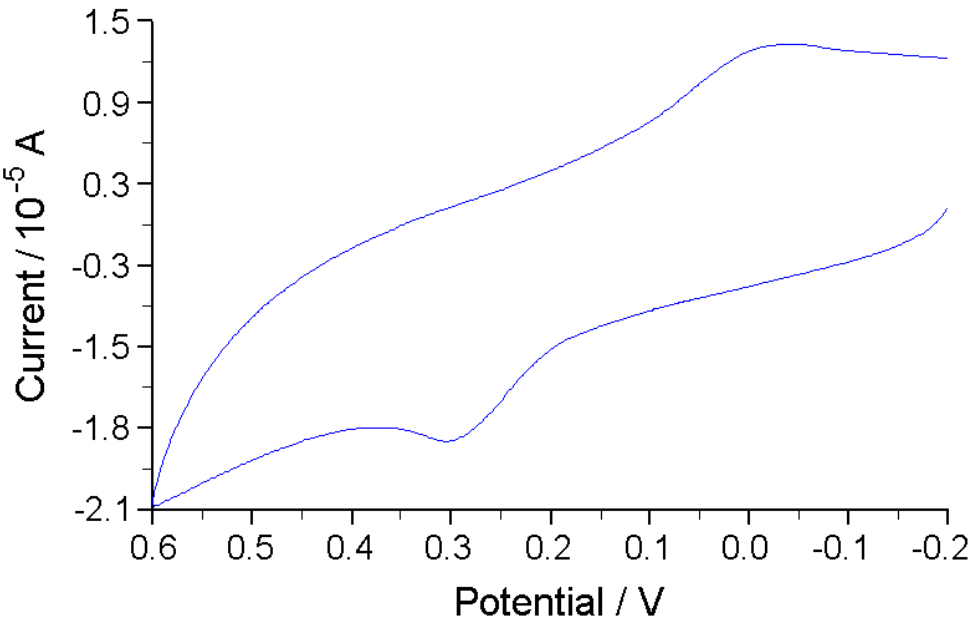


Fig. S-3 Cyclic voltammogram of $[VO(L\text{-}tartH_{-1})_2]^{4-}$ complex recorded at 25 °C on an aqueous solution of VO(IV) and L-tartrate: pH 8.5, ligand to metal molar ratio 100:1, VO(IV) concentration 5 mM. Au was used as working electrode and the potential *E* was measured vs. Ag/AgCl reference

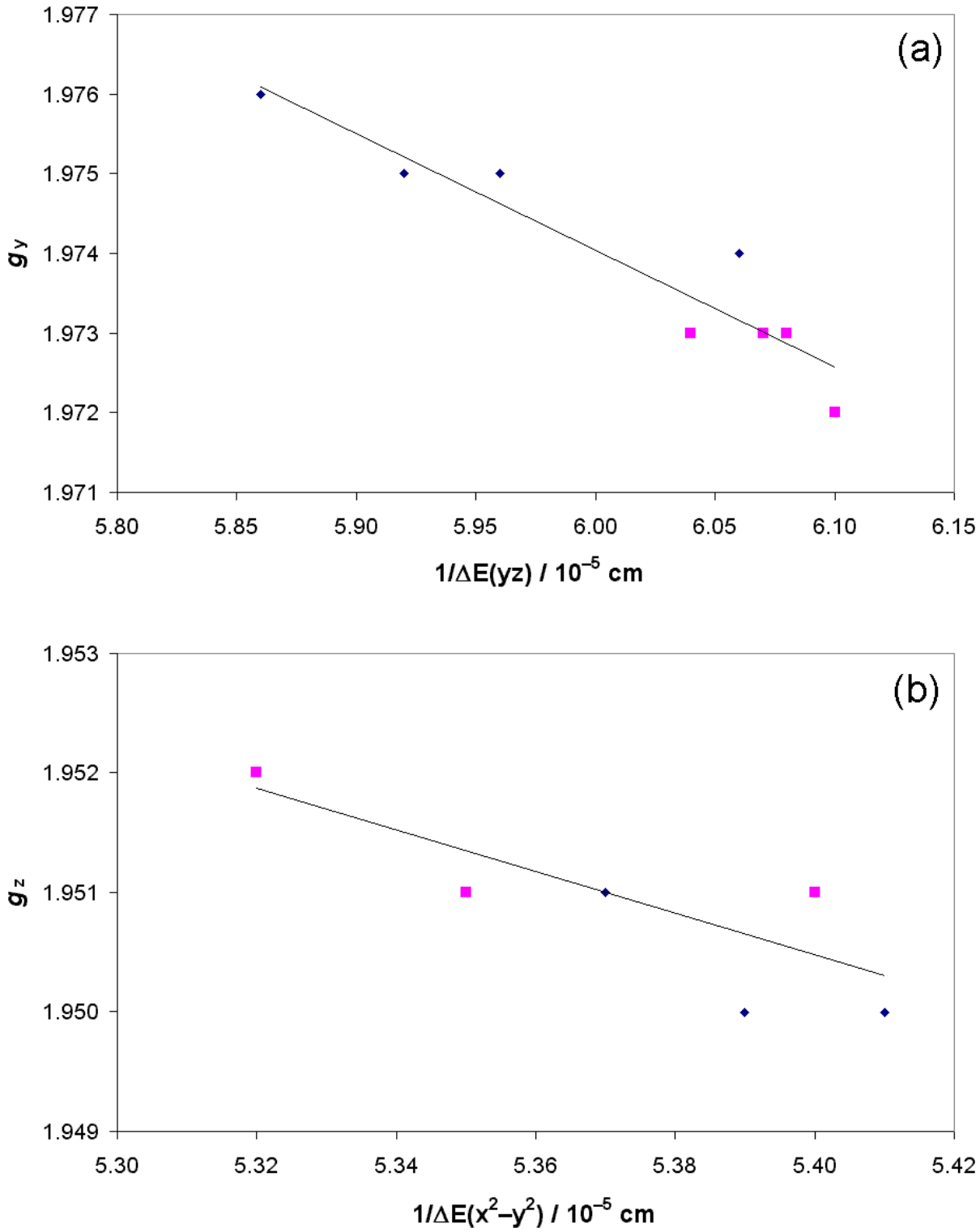
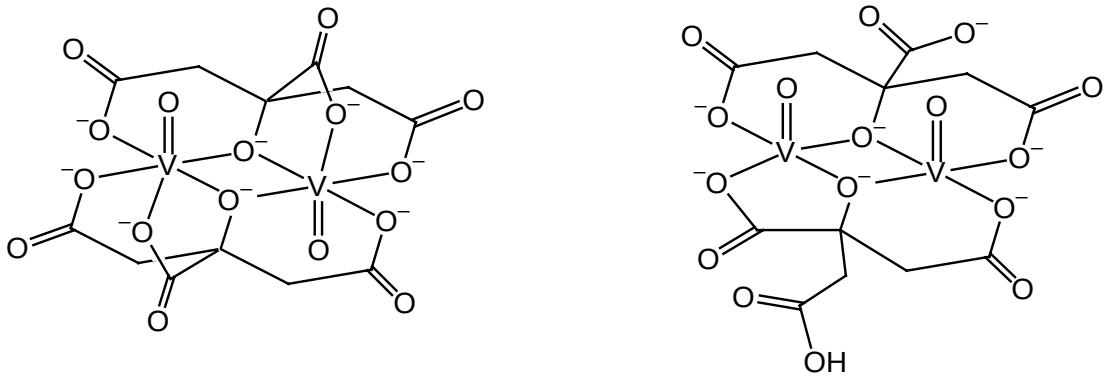
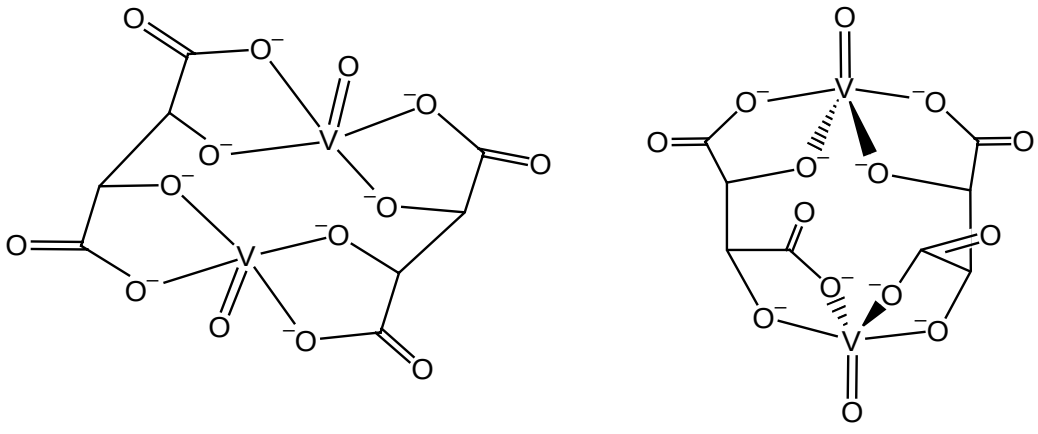


Fig. S-4 Plot of the g_y (a) and g_z value (b) as a function of reciprocal value of energy difference between the d_{yz} and d_{xy} atomic orbital ΔE(yz) (a) and $d_{x^2-y^2}$ and d_{xy} atomic orbital, ΔE(x^2–y^2) (b). With the blue rhombi are represented the values for glycolate, 2-hydroxyisobutirate, 2-ethyl-2-hydroxybutirate and benzilate taken from ref. 25 of the manuscript, and with the pink squares the values for citrate and tartrates (see Tables 3 and 4 of the manuscript).



(a) (b)

Scheme S-1 Dinuclear VO(IV) complexes formed by citrate in an equimolar solution: (a) $[(VO)_2(LH_{-1})_2]^{4-}$ and (b) $[(VO)_2L(LH_{-1})]^{3-}$.



(a) (b)

Scheme S-2. Dinuclear $[(VO)_2(LH_{-1})_2]^{4-}$ complexes formed by tartrates in an equimolar solution: (a) structure formed by D- or L-tartrate and (b) structure formed by DL-tartrate.

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