Supporting Information

Electrochemical, Spectroscopic, Structural and Magnetic Characterization of the Reduced and Protonated α-Dawson Anions in $[Fe(\eta^5\text{-}C_5Me_5)_2]_5[HS_2Mo_{18}O_{62}].3HCONMe_2.2Et_2O$ and $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

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Tables S1 – S4. X-Ray crystallographic data related to atomic coordinates, anisotropic and displacement parameters, intramolecular distances and intramolecular bond angles.

Table S5. Conductance data for dmf solutions.

Figure S1. X-band EPR spectrum of powdered $[Fe(cp^*)_2]_5[HS_2Mo_{18}O_{62}].3dmf.2Et_2O$ at 6 K. Microwave frequency = 9.23 GHz. The weak sharp lines at about 2000 G and 4000 G arise from impurities in cavity.

Figure S2. Mössbauer spectrum at room temperature of powdered $[Fe(cp^*)_2]_5[HS_2Mo_{18}O_{62}].3dmf.2Et_2O$. The fitted solid line assumes a quadrupole doublet (see Experimental Section).

Table S1. Atomic coordinates and Biso/Beq for $[NBu_4]_5[HS_2Mo_{18}O_{62}] \cdot 2H_2O$

atom	x	y	z	B(eq)
Mo(1)	0.47123(3)	0.59009(4)	0.10780(2)	2.26(1)
Mo(2)	0.53223(2)	0.38801(4)	0.11223(2)	1.96(1)
Mo(3)	0.39194(2)	0.40586(4)	0.10110(2)	2.05(1)
Mo(4)	0.41192(2)	0.65690(4)	0.18804(2)	2.16(1)
Mo(5)	0.56386(2)	0.65964(4)	0.20387(2)	2.01(1)
Mo(6)	0.63129(2)	0.46545(4)	0.20316(2)	1.77(1)
Mo(7)	0.55790(2)	0.25706(4)	0.20510(2)	1.60(1)
Mo(8)	0.41538(2)	0.25749(4)	0.18532(2)	1.84(1)
Mo(9)	0.34152(2)	0.46387(4)	0.18776(2)	1.69(1)
S(1)	0.48679(6)	0.4597(1)	0.19336(5)	1.31(3)
O(1)	0.5148(2)	0.5071(3)	0.0916(1)	2.1(1)
O(2)	0.4578(2)	0.3626(3)	0.0867(1)	1.9(1)
O(3)	0.4082(2)	0.5129(3)	0.0833(1)	2.2(1)
O(4)	0.4226(2)	0.6271(3)	0.1382(1)	2.1(1)
O(5)	0.5291(2)	0.6285(3)	0.1478(1)	2.0(1)
O(6)	0.5912(2)	0.4482(3)	0.1509(1)	1.9(1)
O(7)	0.5380(2)	0.3023(3)	0.1486(1)	2.0(1)
O(8)	0.4086(2)	0.3032(3)	0.1360(1)	1.8(1)
O(9)	0.3568(2)	0.4485(3)	0.1348(1)	1.9(1)
O(10)	0.4890(2)	0.6785(3)	0.2038(1)	1.8(1)
O(11)	0.6241(2)	0.5870(3)	0.1998(1)	1.8(1)
O(12)	0.6072(2)	0.3515(3)	0.2125(1)	1.7(1)
O(13)	0.4850(2)	0.2068(3)	0.1888(1)	1.6(1)
O(14)	0.3714(2)	0.3503(3)	0.1948(1)	1.8(1)
O(15)	0.3485(2)	0.5839(3)	0.1785(1)	1.8(1)
O(16)	0.4147(2)	0.6537(3)	0.2424(1)	2.0(1)
O(17)	0.4365(2)	0.2442(3)	0.2439(1)	1.8(1)
O(18)	0.3508(2)	0.4785(3)	0.2392(1)	1.9(1)
O(19)	0.4641(2)	0.6683(3)	0.0739(1)	2.7(1)
O(20)	0.5666(2)	0.3469(3)	0.0819(1)	2.3(1)
O(21)	0.3399(2)	0.3684(3)	0.0640(1)	2.6(1)
O(22)	0.3831(2)	0.7559(3)	0.1774(1)	2.4(1)
O(23)	0.5862(2)	0.7587(3)	0.1962(1)	2.5(1)
O(24)	0.6955(2)	0.4500(3)	0.1996(1)	2.0(1)
O(25)	0.5958(2)	0.1731(3)	0.1967(1)	2.0(1)
O(26)	0.3727(2)	0.1726(3)	0.1751(1)	2.3(1)
O(27)	0.2727(2)	0.4499(3)	0.1706(1)	2.2(1)
O(28)	0.4759(2)	0.4604(2)	0.1503(1)	1.48(9)
O(29)	0.5394(2)	0.5046(3)	0.2102(1)	1.57(9)
O(30)	0.4899(2)	0.3694(2)	0.2065(1)	1.44(9)
O(31)	0.4413(2)	0.5046(3)	0.2034(1)	1.47(9)
O(32)	0.2687(3)	0.0885(5)	-0.0827(2)	7.5(2)
N(1)	0.0879(2)	0.2264(4)	0.0247(2)	2.7(1)
N(2)	0.7143(3)	0.2156(4)	0.1309(2)	3.6(2)
N(3)	0.5	-0.0382(5)	0.2500	2.9(2)
C(1)	0.0407(3)	0.2841(5)	0.0286(2)	3.7(2)
C(2)	-0.0085(4)	0.2908(6)	-0.0071(3)	5.2(2)
C(3)	-0.0595(5)	0.3321(9)	0.0004(4)	8.2(3)
C(4)	-0.0529(7)	0.424(1)	0.0083(5)	12.4(5)
C(5)	0.0664(3)	0.1336(5)	0.0171(2)	2.9(1)

Table S1.(cont.) Atomic coordinates and Biso/Beq for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	x	y	z	B(eq)
C(6)	0.1098(3)	0.0654(5)	0.0177(2)	2.7(1)
C(7)	0.0818(3)	-0.0234(5)	0.0091(2)	3.0(2)
C(8)	0.1233(3)	-0.0942(5)	0.0090(2)	3.6(2)
C(9)	0.1087(3)	0.2541(5)	-0.0093(2)	3.3(2)
C(10)	0.1249(4)	0.3488(5)	-0.0093(2)	4.2(2)
C(11)	0.1491(5)	0.3676(8)	-0.0425(3)	7.4(3)
C(12)	0.2091(8)	0.339(1)	-0.0300(5)	14.1(6)
C(13)	0.1339(3)	0.2341(5)	0.0610(2)	3.3(2)
C(14)	0.1222(4)	0.2004(6)	0.0975(3)	4.5(2)
C(15)	0.1667(5)	0.2196(8)	0.1340(3)	7.1(3)
C(16a)	0.206(1)	0.165(1)	0.1292(6)	7.0(6)
C(16)	0.1564(9)	0.322(1)	0.1435(6)	6.9(5)
C(17)	0.6571(3)	0.1770(5)	0.1217(2)	3.3(2)
C(18)	0.6544(4)	0.0797(6)	0.1276(3)	5.3(2)
C(19)	0.5950(5)	0.0465(8)	0.1126(3)	7.4(3)
C(20)	0.5994(9)	-0.053(1)	0.1175(6)	17.2(7)
C(21)	0.7484(4)	0.1734(6)	0.1066(3)	5.8(2)
C(22)	0.7203(5)	0.1725(8)	0.0624(4)	8.1(3)
C(23)	0.7615(7)	0.135(1)	0.0408(5)	12.4(5)
C(24)	0.7261(9)	0.114(1)	-0.0050(7)	18.6(8)
C(25)	0.7059(4)	0.3114(6)	0.1220(3)	4.6(2)
C(26)	0.7589(5)	0.3621(8)	0.1265(4)	8.2(3)
C(27)	0.7447(5)	0.4559(8)	0.1185(3)	7.7(3)
C(28)	0.7988(9)	0.502(1)	0.1190(6)	15.1(6)
C(29)	0.7469(3)	0.1991(5)	0.1718(2)	4.0(2)
C(30)	0.7233(3)	0.2363(5)	0.2020(2)	3.6(2)
C(31)	0.7632(4)	0.2206(7)	0.2430(3)	6.3(3)
C(32)	0.7357(7)	0.252(1)	0.2737(5)	11.8(5)
C(33)	0.4899(3)	-0.0968(5)	0.2152(2)	3.2(2)
C(34)	0.4743(5)	-0.0508(7)	0.1763(4)	7.2(3)
C(35)	0.4704(7)	-0.120(1)	0.1444(5)	10.5(4)
C(36)	0.436(1)	-0.091(2)	0.1089(8)	23.0(9)
C(37)	0.5305(7)	0.037(1)	0.2466(4)	4.3(3)
C(37a)	0.561(1)	0.004(1)	0.2554(6)	3.0(4)
C(38)	0.5911(8)	0.010(1)	0.2594(5)	12.0(6)
C(39)	0.6206(7)	-0.031(1)	0.2296(5)	5.5(4)
C(39a)	0.6102(8)	-0.068(1)	0.2572(6)	3.1(4)
C(40)	0.6810(6)	-0.0123(9)	0.2357(4)	4.2(3)
C(40a)	0.671(1)	-0.020(2)	0.2639(8)	5.3(5)
H(1)	0.0276	0.2620	0.0492	4.4
H(2)	0.0550	0.3409	0.0350	4.4
H(3)	0.0023	0.3247	-0.0259	6.3
H(4)	-0.0180	0.2340	-0.0169	6.3
H(5)	-0.0899	0.3238	-0.0218	9.8
H(6)	-0.0669	0.3043	0.0221	9.8
H(7)	-0.0224	0.4329	0.0304	14.9
H(8)	-0.0463	0.4524	-0.0135	14.9
H(9)	-0.0856	0.4461	0.0130	14.9
H(10)	0.0484	0.1190	0.0364	3.5
H(11)	0.0402	0.1322	-0.0077	3.5

Table S1.(cont.) Atomic coordinates and Biso/Beq for
$[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	x	y	z	B(eq)
H(12)	0.1283	0.0786	-0.0014	3.3
H(13)	0.1358	0.0644	0.0425	3.3
H(14)	0.0635	-0.0361	0.0284	3.6
H(15)	0.0554	-0.0215	-0.0156	3.6
H(16)	0.1047	-0.1484	0.0037	4.3
H(17)	0.1415	-0.0823	-0.0104	4.3
H(18)	0.1498	-0.0965	0.0336	4.3
H(19)	0.1403	0.2199	-0.0091	4.0
H(20)	0.0803	0.2430	-0.0324	4.0
H(21)	0.0931	0.3839	-0.0117	5.0
H(22)	0.1517	0.3619	0.0145	5.0
H(23)	0.1295	0.3363	-0.0649	8.9
H(24)	0.1469	0.4279	-0.0481	8.9
H(25)	0.2109	0.2787	-0.0239	16.9
H(26)	0.2255	0.3487	-0.0505	16.9
H(27)	0.2285	0.3710	-0.0079	16.9
H(28)	0.1647	0.2029	0.0573	4.0
H(29)	0.1431	0.2938	0.0648	4.0
H(30)	0.0887	0.2261	0.0996	5.4
H(31)	0.1177	0.1393	0.0953	5.4
H(32)	0.6378	0.2045	0.1378	4.0
H(33)	0.6391	0.1892	0.0953	4.0
H(34)	0.6769	0.0513	0.1139	6.4
H(35)	0.6678	0.0670	0.1545	6.4
H(36)	0.5724	0.0701	0.1274	8.9
H(37)	0.5800	0.0614	0.0861	8.9
H(38)	0.5634	-0.0778	0.1097	20.6
H(39)	0.6211	-0.0758	0.1019	20.6
H(40)	0.6162	-0.0667	0.1439	20.6
H(41)	0.7823	0.2040	0.1110	6.9
H(42)	0.7557	0.1150	0.1150	6.9
H(43)	0.6881	0.1369	0.0571	9.7
H(44)	0.7102	0.2298	0.0537	9.7
H(45)	0.7896	0.1764	0.0411	14.8
H(46)	0.7778	0.0832	0.0531	14.8
H(47)	0.7093	0.1657	-0.0171	22.4
H(48)	-0.7506	0.0920	-0.0188	22.4
H(49)	0.6983	0.0720	-0.0053	22.4
H(50)	0.6839	0.3174	0.0960	5.5
H(51)	0.6868	0.3356	0.1390	5.5
H(52)	0.7820	0.3557	0.1522	9.9
H(53)	0.7776	0.3411	0.1087	9.9
H(54)	0.7187	0.4623	0.0938	9.2
H(55)	0.7297	0.4792	0.1380	9.2
H(56)	0.8249	0.4931	0.1435	18.1
H(57)	0.8129	0.4789	0.0991	18.1
H(58)	0.7920	0.5622	0.1148	18.1
H(59)	0.7498	0.1381	0.1755	4.7
H(60)	0.7828	0.2231	0.1752	4.7
H(61)	0.7179	0.2970	0.1978	4.3

Table S1.(cont.) Atomic coordinates and Biso/Beq for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	x	y	z	B(eq)
H(62)	0.6887	0.2094	0.2004	4.3
H(63)	0.7710	0.1604	0.2466	7.6
H(64)	0.7966	0.2516	0.2455	7.6
H(65)	0.7602	0.2439	0.2988	14.2
H(66)	0.7271	0.3122	0.2698	14.2
H(67)	0.7026	0.2204	0.2715	14.2
H(68)	0.5228	-0.1290	0.2171	3.8
H(69)	0.4607	-0.1353	0.2159	3.8
H(70)	0.4397	-0.0224	0.1725	8.7
H(71)	0.5019	-0.0092	0.1753	8.7
H(72)	0.5063	-0.1319	0.1419	12.6
H(73)	0.4552	-0.1721	0.1517	12.6
H(74)	0.4531	-0.0428	0.1003	27.4
H(75)	0.4319	-0.1368	0.0902	27.4
H(76)	0.4013	-0.0754	0.1117	27.4

Table S2. Anisotropic Displacement Parameters for $[NBu_4]_5[HS_2Mo_{18}O_{62}] \cdot 2H_2O$

atom	U11	U22	U33	U12	U13	U23
Mo(1)	0.0333(4)	0.0246(3)	0.0254(3)	0.0046(3)	0.0036(3)	0.0076(3)
Mo(2)	0.0252(3)	0.0291(3)	0.0186(3)	0.0048(3)	0.0033(3)	-0.0004(3)
Mo(3)	0.0225(3)	0.0331(4)	0.0183(3)	0.0011(3)	-0.0011(3)	-0.0016(3)
Mo(4)	0.0216(3)	0.0169(3)	0.0398(4)	0.0050(3)	0.0023(3)	0.0039(3)
Mo(5)	0.0220(3)	0.0149(3)	0.0375(4)	-0.0005(3)	0.0050(3)	-0.0003(3)
Mo(6)	0.0187(3)	0.0179(3)	0.0293(3)	0.0010(3)	0.0046(3)	-0.0005(3)
Mo(7)	0.0209(3)	0.0149(3)	0.0235(3)	0.0033(2)	0.0035(3)	0.0000(2)
Mo(8)	0.0205(3)	0.0162(3)	0.0301(4)	-0.0019(3)	0.0018(3)	-0.0041(3)
Mo(9)	0.0176(3)	0.0200(3)	0.0236(3)	0.0026(3)	0.0007(3)	0.0009(3)
S(1)	0.0152(8)	0.0138(8)	0.0178(8)	0.0011(7)	-0.0007(6)	0.0002(7)
O(1)	0.029(3)	0.026(3)	0.024(3)	0.002(2)	0.007(2)	0.004(2)
O(2)	0.024(3)	0.027(3)	0.018(2)	0.007(2)	-0.002(2)	-0.001(2)
O(3)	0.026(3)	0.034(3)	0.020(3)	0.005(2)	0.000(2)	0.004(2)
O(4)	0.026(3)	0.024(3)	0.029(3)	0.005(2)	0.005(2)	0.006(2)
O(5)	0.027(3)	0.024(3)	0.027(3)	0.003(2)	0.007(2)	0.004(2)
O(6)	0.023(3)	0.023(2)	0.025(3)	0.001(2)	0.005(2)	0.001(2)
O(7)	0.029(3)	0.021(2)	0.024(3)	0.005(2)	0.006(2)	0.003(2)
O(8)	0.027(3)	0.020(2)	0.021(2)	-0.001(2)	0.003(2)	-0.002(2)
O(9)	0.021(3)	0.027(3)	0.021(3)	0.001(2)	-0.001(2)	0.001(2)
O(10)	0.025(3)	0.016(2)	0.026(3)	0.002(2)	0.004(2)	-0.002(2)
O(11)	0.021(2)	0.021(2)	0.025(3)	-0.001(2)	0.005(2)	0.000(2)
O(12)	0.020(2)	0.024(2)	0.020(2)	0.001(2)	0.003(2)	0.002(2)
O(13)	0.024(3)	0.016(2)	0.016(2)	0.000(2)	0.001(2)	-0.003(2)
O(14)	0.022(2)	0.026(3)	0.017(2)	-0.002(2)	0.000(2)	-0.003(2)
O(15)	0.020(2)	0.019(2)	0.026(3)	0.004(2)	0.002(2)	0.003(2)
O(16)	0.022(3)	0.020(2)	0.031(3)	0.001(2)	0.005(2)	0.003(2)
O(17)	0.023(3)	0.019(2)	0.023(2)	-0.001(2)	0.003(2)	-0.001(2)
O(18)	0.025(3)	0.020(2)	0.025(3)	0.000(2)	0.002(2)	0.001(2)
O(19)	0.038(3)	0.033(3)	0.028(3)	0.008(2)	0.002(2)	0.014(2)
O(20)	0.028(3)	0.038(3)	0.021(3)	0.006(2)	0.005(2)	0.001(2)
O(21)	0.029(3)	0.044(3)	0.021(3)	-0.004(2)	-0.001(2)	-0.002(2)
O(22)	0.029(3)	0.022(3)	0.039(3)	0.006(2)	0.007(2)	0.003(2)
O(23)	0.030(3)	0.019(3)	0.044(3)	-0.003(2)	0.008(2)	-0.003(2)
O(24)	0.018(2)	0.027(3)	0.032(3)	0.002(2)	0.005(2)	0.002(2)
O(25)	0.026(3)	0.022(3)	0.029(3)	0.006(2)	0.005(2)	-0.002(2)
O(26)	0.031(3)	0.021(3)	0.032(3)	-0.007(2)	0.005(2)	-0.006(2)
O(27)	0.022(3)	0.035(3)	0.025(3)	0.004(2)	0.003(2)	0.001(2)
O(28)	0.019(2)	0.018(2)	0.018(2)	0.000(2)	0.002(2)	-0.001(2)
O(29)	0.021(2)	0.016(2)	0.021(2)	-0.001(2)	0.002(2)	0.000(2)
O(30)	0.018(2)	0.014(2)	0.019(2)	-0.001(2)	-0.001(2)	0.000(2)
O(31)	0.018(2)	0.016(2)	0.019(2)	0.002(2)	0.002(2)	-0.002(2)
O(32)	0.047(4)	0.158(7)	0.074(5)	-0.024(5)	0.009(4)	-0.036(5)
N(1)	0.040(4)	0.032(4)	0.030(4)	-0.006(3)	0.007(3)	-0.003(3)
N(2)	0.039(4)	0.062(5)	0.044(4)	0.023(4)	0.023(3)	0.019(4)
N(3)	0.074(7)	0.012(4)	0.032(5)	0	0.028(5)	0

Table S3. Interatomic distances involving the nonhydrogen atoms for $[NBu_4]_5[HS_2Mo_{18}O_{62}] \cdot 2H_2O$

atom	atom	distance	ADC(*)	atom	atom	distance	ADC(*)
Mo(1)	Mo(3)	3.4388(9)	1	Mo(4)	Mo(9)	3.4594(8)	1
Mo(1)	Mo(2)	3.4553(8)	1	Mo(4)	Mo(5)	3.7024(8)	1
Mo(1)	Mo(4)	3.7306(9)	1	Mo(4)	Mo(5)	3.7558(9)	65502
Mo(1)	Mo(5)	3.7458(9)	1	Mo(5)	Mo(6)	3.4425(8)	1
Mo(2)	Mo(3)	3.4479(8)	1	Mo(6)	Mo(7)	3.7134(8)	1
Mo(2)	Mo(6)	3.7143(8)	1	Mo(6)	Mo(9)	3.7823(8)	65502
Mo(2)	Mo(7)	3.7950(8)	1	Mo(7)	Mo(8)	3.4539(8)	1
Mo(3)	Mo(8)	3.7042(8)	1	Mo(7)	Mo(8)	3.8009(8)	65502
Mo(3)	Mo(9)	3.7715(8)	1	Mo(8)	Mo(9)	3.6955(8)	1
Mo(1)	S(1)	3.598(2)	1	Mo(6)	S(1)	3.551(2)	1
Mo(2)	S(1)	3.580(2)	1	Mo(7)	S(1)	3.565(2)	1
Mo(3)	S(1)	3.603(2)	1	Mo(8)	S(1)	3.568(2)	1
Mo(4)	S(1)	3.551(2)	1	Mo(9)	S(1)	3.601(2)	1
Mo(5)	S(1)	3.603(2)	1	S(1)	S(1)	3.933(3)	65502
Mo(1)	O(1)	1.876(4)	1	Mo(5)	O(11)	1.919(4)	1
Mo(1)	O(3)	1.983(5)	1	Mo(5)	O(16)	1.855(4)	65502
Mo(1)	O(4)	1.933(4)	1	Mo(5)	O(23)	1.675(4)	1
Mo(1)	O(5)	1.837(4)	1	Mo(5)	O(29)	2.494(4)	1
Mo(1)	O(19)	1.686(4)	1	Mo(6)	O(6)	1.886(4)	1
Mo(1)	O(28)	2.496(4)	1	Mo(6)	O(11)	1.883(4)	1
Mo(2)	O(1)	1.982(4)	1	Mo(6)	O(12)	1.918(4)	1
Mo(2)	O(2)	1.885(4)	1	Mo(6)	O(18)	2.002(4)	65502
Mo(2)	O(6)	1.963(4)	1	Mo(6)	O(24)	1.670(4)	1
Mo(2)	O(7)	1.834(4)	1	Mo(6)	O(29)	2.467(4)	1
Mo(2)	O(20)	1.688(4)	1	Mo(7)	O(7)	2.071(4)	1
Mo(2)	O(28)	2.483(4)	1	Mo(7)	O(12)	1.882(4)	1
Mo(3)	O(2)	1.980(4)	1	Mo(7)	O(13)	1.926(4)	1
Mo(3)	O(3)	1.853(4)	1	Mo(7)	O(17)	1.807(4)	65502
Mo(3)	O(8)	1.989(4)	1	Mo(7)	O(25)	1.682(4)	1
Mo(3)	O(9)	1.805(4)	1	Mo(7)	O(30)	2.444(4)	1
Mo(3)	O(21)	1.689(4)	1	Mo(8)	O(8)	1.868(4)	1
Mo(3)	O(28)	2.495(4)	1	Mo(8)	O(13)	1.888(4)	1
Mo(4)	O(4)	1.935(4)	1	Mo(8)	O(14)	1.895(4)	1
Mo(4)	O(10)	1.893(4)	1	Mo(8)	O(17)	2.031(4)	1
Mo(4)	O(15)	1.904(4)	1	Mo(8)	O(26)	1.667(4)	1
Mo(4)	O(16)	1.932(4)	1	Mo(8)	O(30)	2.505(4)	1
Mo(4)	O(22)	1.688(4)	1	Mo(9)	O(9)	2.053(4)	1
Mo(4)	O(31)	2.477(4)	1	Mo(9)	O(14)	1.893(4)	1
Mo(5)	O(5)	2.018(4)	1	Mo(9)	O(15)	1.895(4)	1
Mo(5)	O(10)	1.902(4)	1	Mo(9)	O(18)	1.809(4)	1
Mo(9)	O(27)	1.682(4)	1	O(9)	O(14)	2.574(6)	1
Mo(9)	O(31)	2.495(4)	1	O(9)	O(15)	2.651(6)	1
S(1)	O(28)	1.494(4)	1	O(10)	O(16)	2.640(6)	1
S(1)	O(29)	1.466(4)	1	O(10)	O(16)	2.675(6)	65502
S(1)	O(30)	1.464(4)	1	O(11)	O(18)	2.688(6)	65502
S(1)	O(31)	1.463(4)	1	O(11)	O(29)	2.592(6)	1
O(1)	O(2)	2.627(6)	1	O(12)	O(17)	2.706(6)	65502
O(1)	O(3)	2.614(6)	1	O(12)	O(18)	2.635(6)	65502
O(1)	O(5)	2.700(6)	1	O(13)	O(17)	2.653(6)	1
O(1)	O(6)	2.605(6)	1	O(13)	O(30)	2.580(5)	1
O(1)	O(28)	2.649(6)	1	O(14)	O(17)	2.621(6)	1
O(2)	O(3)	2.617(6)	1	O(14)	O(18)	2.675(6)	1
O(2)	O(8)	2.587(6)	1	O(15)	O(16)	2.663(6)	1
O(2)	O(28)	2.666(6)	1	O(15)	O(18)	2.704(6)	1
O(3)	O(4)	2.590(6)	1	O(15)	O(31)	2.567(6)	1
O(3)	O(28)	2.659(6)	1	O(28)	O(29)	2.397(6)	1

Table S3 (cont.). Interatomic distances involving the nonhydrogen atoms for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

O(4)	O(5)	2.602(6)	1	O(28)	O(30)	2.401(6)	1
O(4)	O(10)	2.607(6)	1	O(28)	O(31)	2.402(6)	1
O(5)	O(10)	2.598(6)	1	O(29)	O(30)	2.411(6)	1
O(5)	O(11)	2.671(6)	1	O(29)	O(31)	2.407(6)	1
O(6)	O(7)	2.606(6)	1	O(30)	O(31)	2.401(6)	1
O(6)	O(12)	2.603(6)	1	O(8)	O(9)	2.584(6)	1
O(7)	O(12)	2.583(6)	1	O(8)	O(14)	2.634(6)	1
O(7)	O(13)	2.660(6)	1	N(1)	C(1)	1.52(1)	1
N(1)	C(5)	1.527(9)	1	N(1)	C(9)	1.512(9)	1
N(1)	C(13)	1.488(9)	1	C(19)	C(20)	1.54(2)	1
N(2)	C(17)	1.51(1)	1	C(21)	C(22)	1.55(1)	1
N(2)	C(21)	1.53(1)	1	C(22)	C(23)	1.56(2)	1
N(2)	C(25)	1.51(1)	1	C(23)	C(24)	1.67(2)	1
N(2)	C(29)	1.49(1)	1	C(25)	C(26)	1.51(1)	1
N(3)	C(33)	1.504(9)	1	C(26)	C(27)	1.50(2)	1
N(3)	C(33)	1.504(9)	65502	C(27)	C(28)	1.53(2)	1
N(3)	C(37)	1.41(1)	1	C(29)	C(30)	1.49(1)	1
N(3)	C(37)	1.41(1)	65502	C(30)	C(31)	1.56(1)	1
N(3)	C(37a)	1.63(2)	1	C(31)	C(32)	1.53(2)	1
N(3)	C(37a)	1.63(2)	65502	C(33)	C(34)	1.52(1)	1
C(1)	C(2)	1.52(1)	1	C(34)	C(35)	1.55(2)	1
C(2)	C(3)	1.52(1)	1	C(35)	C(36)	1.40(3)	1
C(3)	C(4)	1.44(2)	1	C(37)	C(37)	1.62(3)	65502
C(5)	C(6)	1.51(1)	1	C(6)	C(7)	1.53(1)	1
C(37)	C(38)	1.52(2)	1	C(7)	C(8)	1.51(1)	1
C(9)	C(10)	1.52(1)	1	C(37a)	C(39a)	1.66(3)	1
C(10)	C(11)	1.51(1)	1	C(38)	C(39)	1.59(2)	1
C(11)	C(12)	1.52(2)	1	C(38)	C(39a)	1.31(2)	1
C(13)	C(14)	1.51(1)	1	C(39)	C(39a)	1.23(2)	1
C(14)	C(15)	1.50(1)	1	C(39)	C(40)	1.50(2)	1
C(15)	C(16a)	1.36(2)	1	C(39)	C(40a)	1.52(3)	1
C(15)	C(16)	1.65(2)	1	C(39a)	C(40a)	1.66(3)	1
C(17)	C(18)	1.52(1)	1	C(18)	C(19)	1.53(1)	1

Distances are in angstroms. Estimated standard deviations in the least significant figure are given in parentheses.

Table S4. Intramolecular Bond Angles for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	atom	atom	angle
O(1)	Mo(1)	O(3)	85.2(2)
O(1)	Mo(1)	O(4)	152.6(2)
O(1)	Mo(1)	O(5)	93.3(2)
O(1)	Mo(1)	O(19)	102.7(2)
O(1)	Mo(1)	O(28)	73.0(2)
O(3)	Mo(1)	O(4)	82.8(2)
O(3)	Mo(1)	O(5)	154.6(2)
O(3)	Mo(1)	O(19)	101.3(2)
O(3)	Mo(1)	O(28)	71.8(2)
O(4)	Mo(1)	O(5)	87.2(2)
O(4)	Mo(1)	O(19)	103.7(2)
O(4)	Mo(1)	O(28)	79.9(2)
O(5)	Mo(1)	O(19)	103.7(2)
O(5)	Mo(1)	O(28)	83.5(2)
O(19)	Mo(1)	O(28)	172.0(2)
O(1)	Mo(2)	O(2)	85.6(2)
O(1)	Mo(2)	O(6)	82.7(2)
O(1)	Mo(2)	O(7)	153.9(2)
O(1)	Mo(2)	O(20)	102.0(2)
O(1)	Mo(2)	O(28)	71.8(2)
O(2)	Mo(2)	O(6)	153.8(2)
O(2)	Mo(2)	O(7)	94.0(2)
O(2)	Mo(2)	O(20)	102.9(2)
O(2)	Mo(2)	O(28)	73.8(2)
O(6)	Mo(2)	O(7)	86.6(2)
O(6)	Mo(2)	O(20)	102.4(2)
O(10)	Mo(4)	O(22)	104.4(2)
O(10)	Mo(4)	O(31)	83.2(2)
O(15)	Mo(4)	O(16)	87.9(2)
O(15)	Mo(4)	O(22)	102.0(2)
O(15)	Mo(4)	O(31)	70.3(2)
O(16)	Mo(4)	O(22)	98.4(2)
O(16)	Mo(4)	O(31)	79.9(2)
O(22)	Mo(4)	O(31)	172.2(2)
O(5)	Mo(5)	O(10)	83.0(2)
O(5)	Mo(5)	O(11)	85.4(2)
O(5)	Mo(5)	O(16)	161.1(2)
O(5)	Mo(5)	O(23)	97.3(2)
O(5)	Mo(5)	O(29)	79.2(2)
O(10)	Mo(5)	O(11)	152.7(2)
O(10)	Mo(5)	O(16)	90.8(2)
O(10)	Mo(5)	O(23)	103.6(2)
O(10)	Mo(5)	O(29)	83.0(2)
O(11)	Mo(5)	O(16)	92.3(2)
O(11)	Mo(5)	O(23)	102.3(2)
O(11)	Mo(5)	O(29)	70.6(2)
O(16)	Mo(5)	O(23)	101.5(2)
O(16)	Mo(5)	O(29)	82.3(2)
O(23)	Mo(5)	O(29)	172.2(2)
O(6)	Mo(6)	O(11)	93.5(2)
O(6)	Mo(6)	O(12)	86.4(2)
O(6)	Mo(6)	O(18)	161.2(2)
O(25)	Mo(7)	O(30)	169.2(2)
O(8)	Mo(8)	O(13)	93.5(2)
O(8)	Mo(8)	O(14)	88.8(2)
O(8)	Mo(8)	O(17)	161.4(2)
O(8)	Mo(8)	O(26)	101.6(2)
O(6)	Mo(2)	O(28)	80.3(2)
O(7)	Mo(2)	O(20)	103.5(2)
O(7)	Mo(2)	O(28)	83.0(2)
O(20)	Mo(2)	O(28)	173.0(2)
O(2)	Mo(3)	O(3)	86.0(2)
O(2)	Mo(3)	O(8)	81.4(2)
O(2)	Mo(3)	O(9)	153.5(2)
O(2)	Mo(3)	O(21)	101.6(2)
O(2)	Mo(3)	O(28)	72.1(2)
O(3)	Mo(3)	O(8)	153.6(2)
O(3)	Mo(3)	O(9)	95.8(2)
O(3)	Mo(3)	O(21)	103.2(2)
O(3)	Mo(3)	O(28)	73.7(2)
O(8)	Mo(3)	O(9)	85.7(2)
O(8)	Mo(3)	O(21)	102.0(2)
O(8)	Mo(3)	O(28)	80.3(1)
O(9)	Mo(3)	O(21)	103.7(2)
O(9)	Mo(3)	O(28)	83.0(2)
O(21)	Mo(3)	O(28)	173.0(2)
O(4)	Mo(4)	O(10)	85.8(2)
O(4)	Mo(4)	O(15)	90.8(2)
O(4)	Mo(4)	O(16)	161.9(2)
O(4)	Mo(4)	O(22)	99.5(2)
O(4)	Mo(4)	O(31)	82.7(2)
O(10)	Mo(4)	O(15)	153.6(2)
O(10)	Mo(4)	O(16)	87.3(2)
O(6)	Mo(6)	O(24)	99.7(2)
O(6)	Mo(6)	O(29)	82.9(2)
O(11)	Mo(6)	O(12)	153.3(2)
O(11)	Mo(6)	O(18)	87.5(2)
O(11)	Mo(6)	O(24)	102.4(2)
O(11)	Mo(6)	O(29)	71.7(2)
O(12)	Mo(6)	O(18)	84.5(2)
O(12)	Mo(6)	O(24)	104.0(2)
O(12)	Mo(6)	O(29)	81.7(2)
O(18)	Mo(6)	O(24)	98.4(2)
O(18)	Mo(6)	O(29)	79.6(2)
O(24)	Mo(6)	O(29)	173.8(2)
O(7)	Mo(7)	O(12)	81.5(2)
O(7)	Mo(7)	O(13)	83.4(2)
O(7)	Mo(7)	O(17)	163.5(2)
O(7)	Mo(7)	O(25)	94.6(2)
O(7)	Mo(7)	O(30)	78.4(1)
O(12)	Mo(7)	O(13)	152.7(2)
O(12)	Mo(7)	O(17)	94.4(2)
O(12)	Mo(7)	O(25)	103.7(2)
O(12)	Mo(7)	O(30)	83.5(2)
O(13)	Mo(7)	O(17)	93.8(2)
O(13)	Mo(7)	O(25)	100.0(2)
O(13)	Mo(7)	O(30)	71.3(1)
O(17)	Mo(7)	O(25)	101.9(2)
O(17)	Mo(7)	O(30)	85.3(2)
O(15)	Mo(9)	O(27)	101.3(2)
O(15)	Mo(9)	O(31)	70.0(2)
O(18)	Mo(9)	O(27)	102.8(2)
O(18)	Mo(9)	O(31)	84.2(2)
O(27)	Mo(9)	O(31)	169.5(2)

Table S4 Intramolecular Bond Angles for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	atom	atom	angle
O(8)	Mo(8)	O(30)	83.9(2)
O(13)	Mo(8)	O(14)	151.2(2)
O(13)	Mo(8)	O(17)	85.2(2)
O(13)	Mo(8)	O(26)	102.6(2)
O(13)	Mo(8)	O(30)	70.4(1)
O(14)	Mo(8)	O(17)	83.7(2)
O(14)	Mo(8)	O(26)	105.1(2)
O(14)	Mo(8)	O(30)	81.4(2)
O(17)	Mo(8)	O(26)	96.8(2)
O(17)	Mo(8)	O(30)	78.2(1)
O(26)	Mo(8)	O(30)	171.5(2)
O(9)	Mo(9)	O(14)	81.3(2)
O(9)	Mo(9)	O(15)	84.3(2)
O(9)	Mo(9)	O(18)	162.5(2)
O(9)	Mo(9)	O(27)	94.6(2)
O(9)	Mo(9)	O(31)	78.8(1)
O(14)	Mo(9)	O(15)	150.8(2)
O(14)	Mo(9)	O(18)	92.5(2)
O(14)	Mo(9)	O(27)	105.1(2)
O(14)	Mo(9)	O(31)	82.3(2)
O(15)	Mo(9)	O(18)	93.7(2)
O(3)	O(1)	O(5)	89.1(2)
O(3)	O(1)	O(6)	127.4(2)
O(3)	O(1)	O(28)	60.7(2)
O(5)	O(1)	O(6)	73.4(2)
O(5)	O(1)	O(28)	66.3(2)
O(6)	O(1)	O(28)	66.8(2)
Mo(2)	O(2)	Mo(3)	126.3(2)
Mo(2)	O(2)	O(1)	48.8(1)
Mo(2)	O(2)	O(3)	103.6(2)
Mo(2)	O(2)	O(8)	109.7(2)
Mo(2)	O(2)	O(28)	63.4(1)
Mo(3)	O(2)	O(1)	100.2(2)
Mo(3)	O(2)	O(3)	45.0(1)
Mo(3)	O(2)	O(8)	49.5(1)
Mo(3)	O(2)	O(28)	63.0(1)
O(1)	O(2)	O(3)	59.8(2)
O(1)	O(2)	O(8)	127.4(2)
O(1)	O(2)	O(28)	60.1(2)
O(3)	O(2)	O(8)	91.9(2)
O(3)	O(2)	O(28)	60.4(2)
O(8)	O(2)	O(28)	67.5(2)
Mo(1)	O(3)	Mo(3)	127.3(2)
Mo(1)	O(3)	O(1)	45.6(1)
Mo(1)	O(3)	O(2)	101.4(2)
Mo(1)	O(3)	O(4)	47.8(1)
Mo(1)	O(3)	O(28)	63.1(1)
Mo(5)	O(5)	O(4)	106.4(2)
Mo(5)	O(5)	O(10)	46.6(1)
Mo(5)	O(5)	O(11)	45.7(1)
O(1)	O(5)	O(4)	88.5(2)
O(1)	O(5)	O(10)	141.4(2)
O(1)	O(5)	O(11)	105.4(2)
O(4)	O(5)	O(10)	60.2(2)
O(4)	O(5)	O(11)	141.2(2)
O(10)	O(5)	O(11)	89.6(2)
Mo(2)	O(6)	Mo(6)	149.6(2)
Mo(2)	O(6)	O(1)	49.0(1)
O(28)	S(1)	O(29)	108.1(2)
O(28)	S(1)	O(30)	108.5(2)
O(28)	S(1)	O(31)	108.6(2)
O(29)	S(1)	O(30)	110.7(2)
O(29)	S(1)	O(31)	110.5(2)
O(30)	S(1)	O(31)	110.2(2)
Mo(1)	O(1)	Mo(2)	127.2(2)
Mo(1)	O(1)	O(2)	104.2(2)
Mo(1)	O(1)	O(3)	49.1(1)
Mo(1)	O(1)	O(5)	42.8(1)
Mo(1)	O(1)	O(6)	110.3(2)
Mo(1)	O(1)	O(28)	64.3(1)
Mo(2)	O(1)	O(2)	45.7(1)
Mo(2)	O(1)	O(3)	100.9(2)
Mo(2)	O(1)	O(5)	113.2(2)
Mo(2)	O(1)	O(6)	48.3(1)
Mo(2)	O(1)	O(28)	62.9(1)
O(2)	O(1)	O(3)	59.9(2)
O(2)	O(1)	O(5)	126.6(2)
O(2)	O(1)	O(6)	91.5(2)
O(2)	O(1)	O(28)	60.7(2)
Mo(3)	O(3)	O(1)	104.2(2)
Mo(3)	O(3)	O(2)	49.0(1)
Mo(3)	O(3)	O(4)	110.0(2)
Mo(3)	O(3)	O(28)	64.3(1)
O(1)	O(3)	O(2)	60.3(2)
O(1)	O(3)	O(4)	90.6(2)
O(1)	O(3)	O(28)	60.3(2)
O(2)	O(3)	O(4)	127.0(2)
O(2)	O(3)	O(28)	60.7(2)
O(4)	O(3)	O(28)	66.4(2)
Mo(1)	O(4)	Mo(4)	149.4(2)
Mo(1)	O(4)	O(3)	49.4(1)
Mo(1)	O(4)	O(5)	44.9(1)
Mo(1)	O(4)	O(10)	104.6(2)
Mo(4)	O(4)	O(3)	146.8(2)
Mo(4)	O(4)	O(5)	105.9(2)
Mo(4)	O(4)	O(10)	46.4(1)
O(3)	O(4)	O(5)	91.8(2)
O(3)	O(4)	O(10)	144.5(2)
O(5)	O(4)	O(10)	59.8(2)
Mo(1)	O(5)	Mo(5)	152.6(2)
Mo(1)	O(5)	O(1)	43.9(1)
Mo(1)	O(5)	O(4)	47.9(1)
Mo(1)	O(5)	O(10)	107.9(2)
Mo(1)	O(5)	O(11)	146.2(2)
Mo(5)	O(5)	O(1)	147.2(2)
O(6)	O(7)	O(12)	60.2(2)
O(6)	O(7)	O(13)	142.0(2)
O(12)	O(7)	O(13)	89.8(2)
Mo(3)	O(8)	Mo(8)	147.6(2)
Mo(3)	O(8)	O(2)	49.2(1)
Mo(3)	O(8)	O(9)	44.1(1)
Mo(3)	O(8)	O(14)	103.0(2)
Mo(8)	O(8)	O(2)	147.6(2)
Mo(8)	O(8)	O(9)	104.9(2)
Mo(8)	O(8)	O(14)	46.0(1)
O(2)	O(8)	O(9)	90.9(2)

Table S4. Intramolecular Bond Angles for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	atom	atom	angle
Mo(2)	O(6)	O(7)	44.6(1)
Mo(2)	O(6)	O(12)	103.9(2)
Mo(6)	O(6)	O(1)	146.6(2)
Mo(6)	O(6)	O(7)	106.5(2)
Mo(6)	O(6)	O(12)	47.3(1)
O(1)	O(6)	O(7)	91.0(2)
O(1)	O(6)	O(12)	143.1(2)
O(7)	O(6)	O(12)	59.5(2)
Mo(2)	O(7)	Mo(7)	152.8(2)
Mo(2)	O(7)	O(6)	48.8(1)
Mo(2)	O(7)	O(12)	108.8(2)
Mo(2)	O(7)	O(13)	146.2(2)
Mo(7)	O(7)	O(6)	105.9(2)
Mo(7)	O(7)	O(12)	46.1(1)
Mo(7)	O(7)	O(13)	46.0(1)
Mo(4)	O(10)	O(16)	47.0(1)
Mo(4)	O(10)	O(16)	147.0(2)
Mo(5)	O(10)	O(4)	110.0(2)
Mo(5)	O(10)	O(5)	50.4(1)
Mo(5)	O(10)	O(16)	144.9(2)
Mo(5)	O(10)	O(16)	43.9(1)
O(4)	O(10)	O(5)	60.0(2)
O(4)	O(10)	O(16)	93.4(2)
O(4)	O(10)	O(16)	146.7(2)
O(5)	O(10)	O(16)	147.4(2)
O(5)	O(10)	O(16)	92.9(2)
O(16)	O(10)	O(16)	103.3(2)
Mo(5)	O(11)	Mo(6)	129.8(2)
Mo(5)	O(11)	O(5)	48.9(1)
Mo(5)	O(11)	O(18)	108.3(2)
Mo(5)	O(11)	O(29)	65.1(1)
Mo(6)	O(11)	O(5)	109.6(2)
Mo(6)	O(11)	O(18)	48.1(1)
Mo(6)	O(11)	O(29)	64.6(1)
O(5)	O(11)	O(18)	133.0(2)
O(5)	O(11)	O(29)	66.8(2)
O(18)	O(11)	O(29)	66.2(2)
Mo(6)	O(12)	Mo(7)	155.6(2)
Mo(6)	O(12)	O(6)	46.3(1)
Mo(6)	O(12)	O(7)	106.3(2)
Mo(6)	O(12)	O(17)	149.2(2)
Mo(9)	O(14)	O(8)	111.3(2)
Mo(9)	O(14)	O(9)	52.0(1)
Mo(9)	O(14)	O(17)	144.9(2)
Mo(9)	O(14)	O(18)	42.5(1)
O(8)	O(14)	O(9)	59.5(2)
O(8)	O(14)	O(17)	94.2(2)
O(8)	O(14)	O(18)	148.1(2)
O(9)	O(14)	O(17)	148.0(2)
O(9)	O(14)	O(18)	93.3(2)
O(17)	O(14)	O(18)	104.4(2)
Mo(4)	O(15)	Mo(9)	131.2(2)
Mo(4)	O(15)	O(9)	112.0(2)
Mo(4)	O(15)	O(16)	46.5(1)
Mo(4)	O(15)	O(18)	112.2(2)
Mo(4)	O(15)	O(31)	65.3(1)
Mo(9)	O(15)	O(9)	50.4(1)
Mo(9)	O(15)	O(16)	108.0(2)
O(2)	O(8)	O(14)	142.3(2)
O(9)	O(8)	O(14)	59.1(2)
Mo(3)	O(9)	Mo(9)	155.7(2)
Mo(3)	O(9)	O(8)	50.1(1)
Mo(3)	O(9)	O(14)	111.2(2)
Mo(3)	O(9)	O(15)	144.5(2)
Mo(9)	O(9)	O(8)	107.8(2)
Mo(9)	O(9)	O(14)	46.6(1)
Mo(9)	O(9)	O(15)	45.3(1)
O(8)	O(9)	O(14)	61.4(2)
O(8)	O(9)	O(15)	142.4(2)
O(14)	O(9)	O(15)	89.1(2)
Mo(4)	O(10)	Mo(5)	154.7(2)
Mo(4)	O(10)	O(4)	47.8(1)
Mo(4)	O(10)	O(5)	107.4(2)
Mo(6)	O(12)	O(18)	49.1(1)
Mo(7)	O(12)	O(6)	112.3(2)
Mo(7)	O(12)	O(7)	52.5(1)
Mo(7)	O(12)	O(17)	41.7(1)
Mo(7)	O(12)	O(18)	142.8(2)
O(6)	O(12)	O(7)	60.3(2)
O(6)	O(12)	O(17)	147.9(2)
O(6)	O(12)	O(18)	94.2(2)
O(7)	O(12)	O(17)	93.0(2)
O(7)	O(12)	O(18)	148.4(2)
O(17)	O(12)	O(18)	103.1(2)
Mo(7)	O(13)	Mo(8)	129.8(2)
Mo(7)	O(13)	O(7)	50.6(1)
Mo(7)	O(13)	O(17)	106.2(2)
Mo(7)	O(13)	O(30)	63.7(1)
Mo(8)	O(13)	O(7)	109.1(2)
Mo(8)	O(13)	O(17)	49.7(1)
Mo(8)	O(13)	O(30)	66.1(1)
O(7)	O(13)	O(17)	133.2(2)
O(7)	O(13)	O(30)	66.3(2)
O(17)	O(13)	O(30)	66.8(2)
Mo(8)	O(14)	Mo(9)	154.5(2)
Mo(8)	O(14)	O(8)	45.2(1)
Mo(8)	O(14)	O(9)	104.5(2)
Mo(8)	O(14)	O(17)	50.4(1)
Mo(8)	O(14)	O(18)	151.9(2)
Mo(4)	O(16)	O(10)	45.8(1)
Mo(4)	O(16)	O(10)	120.0(2)
Mo(4)	O(16)	O(15)	45.6(1)
Mo(5)	O(16)	O(10)	119.5(2)
Mo(5)	O(16)	O(10)	45.3(1)
Mo(5)	O(16)	O(15)	148.1(2)
O(10)	O(16)	O(10)	74.2(2)
O(10)	O(16)	O(15)	88.4(2)
O(10)	O(16)	O(15)	155.0(2)
Mo(7)	O(17)	Mo(8)	164.1(2)
Mo(7)	O(17)	O(12)	43.9(1)
Mo(7)	O(17)	O(13)	148.6(2)
Mo(7)	O(17)	O(14)	118.2(2)
Mo(8)	O(17)	O(12)	120.2(2)
Mo(8)	O(17)	O(13)	45.1(1)
Mo(8)	O(17)	O(14)	45.9(1)
O(12)	O(17)	O(13)	154.8(2)

Table S4. Intramolecular Bond Angles for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	atom	atom	angle
Mo(9)	O(15)	O(18)	41.9(1)
Mo(9)	O(15)	O(31)	66.0(1)
O(9)	O(15)	O(16)	134.0(2)
O(9)	O(15)	O(18)	90.9(2)
O(9)	O(15)	O(31)	67.7(2)
O(16)	O(15)	O(18)	71.3(2)
O(16)	O(15)	O(31)	66.3(2)
O(18)	O(15)	O(31)	67.5(2)
Mo(4)	O(16)	Mo(5)	165.2(2)
Mo(9)	O(18)	O(14)	45.0(1)
Mo(9)	O(18)	O(15)	44.4(1)
O(11)	O(18)	O(12)	88.0(2)
O(11)	O(18)	O(14)	154.6(2)
O(11)	O(18)	O(15)	103.4(2)
O(12)	O(18)	O(14)	74.6(2)
O(12)	O(18)	O(15)	153.1(2)
O(14)	O(18)	O(15)	85.9(2)
Mo(1)	O(28)	Mo(2)	87.9(1)
Mo(1)	O(28)	Mo(3)	87.1(1)
Mo(1)	O(28)	S(1)	127.0(2)
Mo(1)	O(28)	O(1)	42.6(1)
Mo(1)	O(28)	O(2)	87.8(2)
Mo(1)	O(28)	O(3)	45.1(1)
Mo(1)	O(28)	O(29)	103.2(2)
Mo(1)	O(28)	O(30)	162.1(2)
Mo(1)	O(28)	O(31)	106.9(2)
Mo(2)	O(28)	Mo(3)	87.7(1)
Mo(2)	O(28)	S(1)	126.6(2)
Mo(2)	O(28)	O(1)	45.3(1)
Mo(2)	O(28)	O(2)	42.8(1)
Mo(2)	O(28)	O(3)	87.8(2)
Mo(2)	O(28)	O(29)	106.4(2)
Mo(2)	O(28)	O(30)	103.1(2)
Mo(2)	O(28)	O(31)	161.6(2)
Mo(3)	O(28)	S(1)	127.4(2)
O(30)	O(28)	O(31)	60.0(2)
Mo(5)	O(29)	Mo(6)	87.9(1)
Mo(5)	O(29)	S(1)	129.2(2)
Mo(5)	O(29)	O(11)	44.3(1)
Mo(5)	O(29)	O(28)	108.1(2)
Mo(5)	O(29)	O(30)	163.6(2)
Mo(5)	O(29)	O(31)	105.2(2)
Mo(6)	O(29)	S(1)	127.2(2)
Mo(6)	O(29)	O(11)	43.6(1)
Mo(6)	O(29)	O(28)	104.4(2)
Mo(6)	O(29)	O(30)	105.5(2)
Mo(6)	O(29)	O(31)	161.9(2)
S(1)	O(29)	O(11)	148.6(3)
S(1)	O(29)	O(28)	36.3(1)
S(1)	O(29)	O(30)	34.6(1)
S(1)	O(29)	O(31)	34.7(1)
O(11)	O(29)	O(28)	112.3(2)
O(11)	O(29)	O(30)	147.9(2)
O(11)	O(29)	O(31)	147.5(2)
O(28)	O(29)	O(30)	59.9(2)
O(28)	O(29)	O(31)	60.0(2)
O(30)	O(29)	O(31)	59.8(2)
Mo(7)	O(30)	Mo(8)	88.5(1)
O(12)	O(17)	O(14)	74.3(2)
O(13)	O(17)	O(14)	88.0(2)
Mo(6)	O(18)	Mo(9)	166.0(2)
Mo(6)	O(18)	O(11)	44.4(1)
Mo(6)	O(18)	O(12)	46.4(1)
Mo(6)	O(18)	O(14)	121.0(2)
Mo(6)	O(18)	O(15)	147.3(2)
Mo(9)	O(18)	O(11)	147.6(2)
Mo(9)	O(18)	O(12)	119.6(2)
Mo(3)	O(28)	O(1)	87.4(2)
Mo(3)	O(28)	O(2)	45.0(1)
Mo(3)	O(28)	O(3)	42.0(1)
Mo(3)	O(28)	O(29)	162.7(2)
Mo(3)	O(28)	O(30)	107.3(2)
Mo(3)	O(28)	O(31)	103.7(2)
S(1)	O(28)	O(1)	145.1(3)
S(1)	O(28)	O(2)	145.2(2)
S(1)	O(28)	O(3)	145.6(3)
S(1)	O(28)	O(29)	35.5(1)
S(1)	O(28)	O(30)	35.3(1)
S(1)	O(28)	O(31)	35.3(1)
O(1)	O(28)	O(2)	59.2(2)
O(1)	O(28)	O(3)	59.0(2)
O(1)	O(28)	O(29)	109.6(2)
O(1)	O(28)	O(30)	145.6(2)
O(1)	O(28)	O(31)	147.7(2)
O(2)	O(28)	O(3)	58.9(2)
O(2)	O(28)	O(29)	147.5(2)
O(2)	O(28)	O(30)	109.9(2)
O(2)	O(28)	O(31)	145.6(2)
O(3)	O(28)	O(29)	145.7(2)
O(3)	O(28)	O(30)	147.7(2)
O(3)	O(28)	O(31)	110.4(2)
O(29)	O(28)	O(30)	60.3(2)
O(29)	O(28)	O(31)	60.2(2)
Mo(7)	O(30)	O(29)	105.1(2)
Mo(7)	O(30)	O(31)	164.4(2)
Mo(8)	O(30)	S(1)	126.0(2)
Mo(8)	O(30)	O(13)	43.6(1)
Mo(8)	O(30)	O(28)	102.9(2)
Mo(8)	O(30)	O(29)	160.6(2)
Mo(8)	O(30)	O(31)	104.8(2)
S(1)	O(30)	O(13)	148.1(3)
S(1)	O(30)	O(28)	36.1(1)
S(1)	O(30)	O(29)	34.7(1)
S(1)	O(30)	O(31)	34.9(1)
O(13)	O(30)	O(28)	112.0(2)
O(13)	O(30)	O(29)	147.3(2)
O(13)	O(30)	O(31)	147.4(2)
O(28)	O(30)	O(29)	59.7(2)
O(28)	O(30)	O(31)	60.0(2)
O(29)	O(30)	O(31)	60.0(2)
Mo(4)	O(31)	Mo(9)	88.2(1)
Mo(4)	O(31)	S(1)	126.7(2)
Mo(4)	O(31)	O(15)	44.3(1)
Mo(4)	O(31)	O(28)	104.0(2)
Mo(4)	O(31)	O(29)	104.9(2)
Mo(4)	O(31)	O(30)	161.6(2)

Table S4. Intramolecular Bond Angles for $[NBu_4]_5[HS_2Mo_{18}O_{62}].2H_2O$

atom	atom	atom	angle
Mo(7)	O(30)	S(1)	129.9(2)
Mo(7)	O(30)	O(13)	45.0(1)
Mo(7)	O(30)	O(28)	109.7(2)
Mo(9)	O(31)	O(29)	163.7(2)
Mo(9)	O(31)	O(30)	105.0(2)
S(1)	O(31)	O(29)	34.8(1)
C(33)	N(3)	C(37a)	101.8(8
C(33)	N(3)	C(37)	113.1(7)
C(33)	N(3)	C(37a)	106.2(8)
C(1)	N(1)	C(9)	111.1(6)
C(1)	N(1)	C(13)	108.0(6)
C(5)	N(1)	C(9)	107.7(5)
C(5)	N(1)	C(13)	112.6(5)
C(9)	N(1)	C(13)	108.9(6)
C(17)	N(2)	C(21)	110.9(7)
C(17)	N(2)	C(25)	105.6(6)
C(17)	N(2)	C(29)	112.5(6)
C(21)	N(2)	C(25)	111.3(6)
C(21)	N(2)	C(29)	104.2(6)
C(25)	N(2)	C(29)	112.5(7)
C(33)	N(3)	C(33)	106.2(8)
C(33)	N(3)	C(37)	113.1(7)
C(33)	N(3)	C(37)	126.3(7)
C(33)	N(3)	C(37a)	106.2(8)
C(17)	C(18)	C(19)	111.0(9)
C(37)	C(38)	C(39)	121(1)
C(37)	C(38)	C(39a)	126(2)
C(37a)	C(38)	C(39)	118(3)
C(37a)	C(38)	C(39a)	105(3)
C(25)	C(26)	C(27)	109(1)
C(38)	C(39)	C(40)	118(1)
C(29)	C(30)	C(31)	110.3(7)
C(30)	C(31)	C(32)	109(1)
C(33)	C(34)	C(35)	108(1)
C(37a)	C(39a)	C(40a)	111(2)
C(37)	C(37)	C(37a)	135(2)
N(3)	C(37a)	C(38)	163(3)
N(3)	C(37a)	C(39a)	114(1)
C(37)	C(37a)	C(39a)	161(3)

atom	atom	atom	angle
Mo(9)	O(31)	S(1)	129.0(2)
Mo(9)	O(31)	O(15)	44.0(1)
Mo(9)	O(31)	O(28)	108.1(2)
S(1)	O(31)	O(15)	146.6(3)
S(1)	O(31)	O(28)	36.1(1)
S(1)	O(31)	O(30)	34.9(1)
C(33)	N(3)	C(37)	126.3(7)
C(33)	N(3)	C(37a)	101.8(8)
C(1)	N(1)	C(5)	108.4(6)
N(1)	C(1)	C(2)	115.8(7)
C(1)	C(2)	C(3)	114.2(9)
C(2)	C(3)	C(4)	113(1)
N(1)	C(5)	C(6)	115.4(6)
C(5)	C(6)	C(7)	109.2(6)
C(6)	C(7)	C(8)	111.5(6)
N(1)	C(9)	C(10)	115.1(6)
C(9)	C(10)	C(11)	110.6(8)
C(10)	C(11)	C(12)	107(1)
N(1)	C(13)	C(14)	116.2(7)
C(13)	C(14)	C(15)	114.4(8)
C(14)	C(15)	C(16a)	99(1)
C(14)	C(15)	C(16)	104(1)
C(16a)	C(15)	C(16)	142(2)
N(2)	C(17)	C(18)	115.9(7)
C(18)	C(19)	C(20)	105(1)
N(2)	C(21)	C(22)	114.9(9)
C(21)	C(22)	C(23)	110(1)
C(22)	C(23)	C(24)	108(1)
N(2)	C(25)	C(26)	114.4(8)
C(26)	C(27)	C(28)	106(1)
N(2)	C(29)	C(30)	115.4(7)
C(39a)	C(39)	C(40)	114(2)
N(3)	C(33)	C(34)	115.1(7)
C(34)	C(35)	C(36)	110(2)
N(3)	C(37)	C(38)	106(1)
C(37)	C(37)	C(38)	150(2)
C(39a)	C(40a)	C(40)	110(2)
C(37)	C(37a)	C(38)	137(4)

Angles are in degrees. Estimated standard deviations in the least significant figure are given in parentheses.

Table S5. Conductivity parameters for various electrolytes in dmf solutions.

1:2 Electrolyte $[Ni(o\text{-}phen)_3(PF_6)_2$			1:3 Electrolyte $[NBu_4]_3[Mo_{12}O_{40}]$			1:4 Electrolyte $[NHex_4]_4[S_2Mo_{18}O_{62}]$			1:5 Electrolyte $[NBu_4]_5[HS_2Mo_{18}O_{62}]$			$[Fe(\eta^5\text{-}C_5Me_5)_2]_5$ $[HS_2Mo_{18}O_{62}](dmf)_3$ $(Et_2O)_2$		
$C^{1/2}$	Λ_e	$\Lambda_o-\Lambda_e$	$C^{1/2}$	Λ_e	$\Lambda_o-\Lambda_e$	$C^{1/2}$	Λ_e	$\Lambda_o-\Lambda_e$	$C^{1/2}$	Λ_e	$\Lambda_o-\Lambda_e$	$C^{1/2}$	Λ_e	$\Lambda_o-\Lambda_e$
0.0100	165.00	2.42	0.0100	220.00	55.48	0.0050	332.00	36.57	0.0032	520.00	86.77	0.0032	530.00	61.33
0.0200	160.25	7.17	0.0122	210.00	65.48	0.0071	304.00	64.57	0.0039	480.00	126.77	0.0039	466.67	124.67
0.0300	155.22	12.19	0.0141	198.50	76.98	0.0100	261.00	107.57	0.0050	436.00	170.77	0.0050	412.00	179.33
0.0400	161.88	5.54	0.0150	192.44	83.04	0.0122	252.67	115.90	0.0071	378.00	228.77	0.0071	370.00	221.33
0.0500	157.20	10.22	0.0166	184.73	90.75	0.0132	256.00	112.57	0.0100	294.86	311.92	0.0100	306.00	285.33
0.0600	152.22	15.19	-	-	-	0.0141	248.00	120.57	-	-	-	-	-	-
0.0800	143.28	24.13	-	-	-	-	-	-	-	-	-	-	-	-

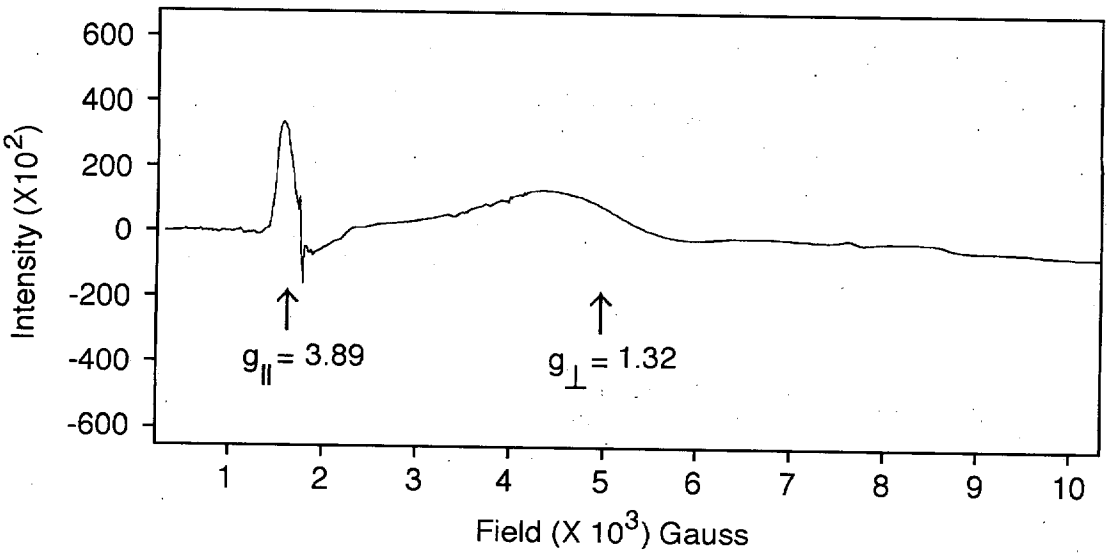

Figure S1

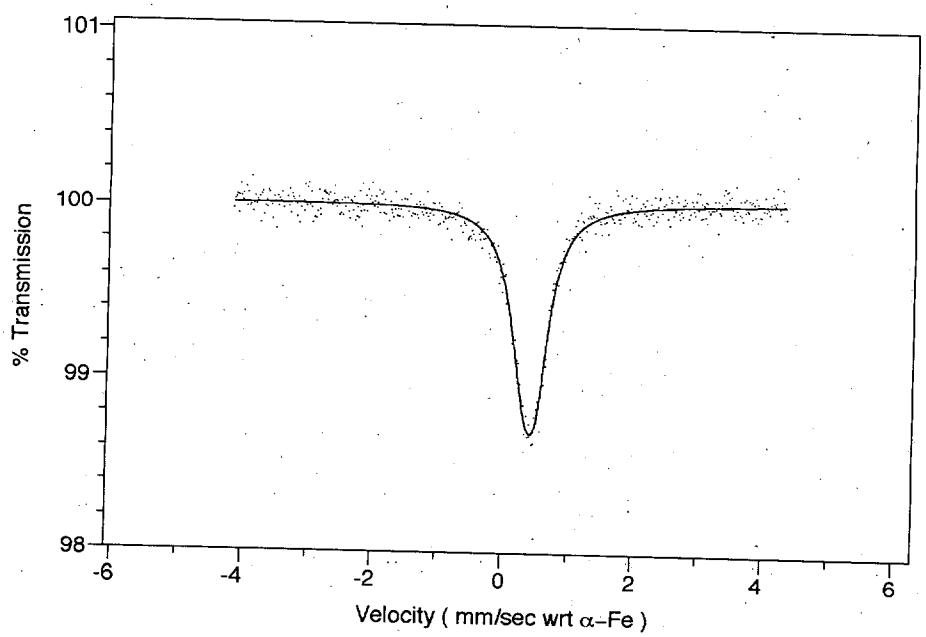

Figure S2