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Supplementary Table 1 | Coordinates of the T6/T6' model of α-Fe(II).

atom	constrained?	x	y	z
Fe	n	0.00000	0.00000	-2.47276
O	n	1.15229	-1.52880	-1.91041
O	n	-1.15226	1.52880	-1.91040
O	n	-1.73483	-0.98535	-2.30171
O	n	1.73481	0.98537	-2.30166
O	y	2.74016	-6.56042	2.39464
O	y	2.76340	-6.57312	-2.15917
O	y	1.80947	-4.10494	2.25568
O	y	-1.80947	4.10494	2.25569
O	y	4.39357	-4.53745	2.10031
O	y	5.91337	-0.42657	3.98753
O	y	-6.04696	2.51448	2.39464
O	y	-5.91338	0.42657	3.98754
O	y	6.04696	-2.51448	2.39464
O	y	2.82114	-5.25124	0.11582
O	y	-2.82113	5.25123	0.11583
O	y	3.55990	3.46457	-6.24818
O	n	1.84300	-4.11495	-2.10243
O	y	-3.55991	-3.46457	-6.24819
O	n	-1.84304	4.11492	-2.10243
O	y	4.39358	-4.53747	-1.86990
O	y	2.75126	2.85040	3.96227
O	y	-6.02376	2.50179	-2.15918
O	y	-2.75125	-2.85040	3.96227
O	y	6.02375	-2.50178	-2.15919
O	y	5.96486	-0.46463	-3.81650
O	y	-0.61079	-3.08485	2.45770
O	y	-5.96487	0.46463	-3.81650
O	y	0.61079	3.08485	2.45771
O	y	4.79800	1.67777	-4.78035
O	n	0.50446	-3.28147	0.06511
O	y	-4.79801	-1.67777	-4.78035
O	n	-0.50444	3.28148	0.06511
O	y	1.42117	-1.53923	1.80730
O	y	-1.42117	1.53923	1.80730
O	n	2.62780	2.91323	-3.90330
O	n	-0.63667	-3.40168	-2.33915

O	n	-2.62780	-2.91326	-3.90331
O	n	0.63666	3.40170	-2.33915
O	y	4.91037	4.23063	-4.12474
O	y	-4.91037	-4.23063	-4.12474
O	y	5.01945	4.33630	0.07416
O	y	6.12905	-0.05072	-1.22824
O	y	-2.11968	-0.94291	2.27467
O	y	-6.12904	0.05071	-1.22825
O	y	2.11967	0.94291	2.27467
O	y	4.76873	1.72679	0.14745
O	n	-2.81145	0.23326	0.05746
O	y	-4.76872	-1.72680	0.14745
O	n	2.81153	-0.23331	0.05747
O	y	6.20870	-0.08786	1.39798
O	y	-3.92390	0.96556	2.35680
O	y	-6.20870	0.08785	1.39798
O	y	3.92390	-0.96556	2.35680
O	n	3.04240	3.14903	-1.27205
O	n	-3.04242	-3.14897	-1.27206
O	y	3.00755	3.24816	1.37389
O	n	-3.91090	1.02242	-2.46496
O	y	-3.00755	-3.24816	1.37389
O	n	3.91090	-1.02235	-2.46497
Si	y	2.94130	-5.11549	1.71378
Si	y	-2.94131	5.11549	1.71379
Si	y	2.94544	-5.12357	-1.48342
Si	y	-2.94546	5.12356	-1.48341
Si	y	0.78718	-3.01111	1.66452
Si	y	-0.78718	3.01111	1.66452
Si	y	3.98932	3.06983	-4.74737
Si	n	0.73592	-3.08425	-1.53023
Si	y	-3.98932	-3.06982	-4.74737
Si	n	-0.73592	3.08425	-1.53023
Si	y	-2.58978	0.44990	1.61762
Si	y	2.58978	-0.44990	1.61762
Si	y	3.96384	3.11887	0.08552
Al	n	-2.58789	0.45986	-1.59306
Si	y	-3.96385	-3.11887	0.08551
Al	n	2.58788	-0.45983	-1.59305
Si	y	5.49817	-0.99539	-2.36885
Si	y	-2.12381	-2.53466	2.51472

Si	y	-5.49816	0.99540	-2.36884
Si	y	2.12381	2.53466	2.51472
Si	n	2.05479	2.61177	-2.42434
Si	n	-2.05479	-2.61175	-2.42436
Si	y	5.52280	-0.99820	2.53357
Si	y	-5.52280	0.99820	2.53357
O	y	-5.01945	-4.33631	0.07415
O	y	-4.39357	4.53745	2.10032
O	y	-2.74016	6.56042	2.39464
O	y	-4.39359	4.53747	-1.86989
O	y	-2.76339	6.57311	-2.15917
O	y	5.02422	1.88452	4.87177
O	y	-5.02422	-1.88453	4.87178
O	y	3.55988	3.46457	6.38382
O	y	-3.55988	-3.46458	6.38383
O	y	4.81113	4.44229	4.29070
Si	y	4.03970	3.15726	4.87683
Si	y	-4.03969	-3.15728	4.87682
O	y	-4.81112	-4.44229	4.29070
H	y	3.42138	-7.16912	2.06856
H	y	3.44970	-7.17302	-1.82760
H	y	5.07909	-5.14238	1.77625
H	y	6.87727	-0.44610	4.09439
H	y	-7.01088	2.53441	2.50122
H	y	-6.87727	0.44609	4.09440
H	y	7.01088	-2.53442	2.50122
H	y	4.35460	3.57923	-6.79240
H	y	-4.35460	-3.57922	-6.79240
H	y	5.08294	-5.13307	-1.53692
H	y	-6.99161	2.51823	-2.22139
H	y	6.99160	-2.51823	-2.22139
H	y	6.93297	-0.48655	-3.87236
H	y	-6.93298	0.48654	-3.87235
H	y	5.59305	1.77844	-5.32679
H	y	-5.59306	-1.77843	-5.32679
H	y	5.70109	4.34598	-4.67457
H	y	-5.70109	-4.34598	-4.67457
H	y	4.54070	5.17909	0.03688
H	y	7.09681	-0.07080	-1.29038
H	y	-7.09681	0.07080	-1.29037
H	y	5.72204	1.90500	0.16512

H	y	-5.72203	-1.90502	0.16553
H	y	7.17250	-0.10828	1.50562
H	y	-7.17249	0.10827	1.50563
H	y	-4.54070	-5.17910	0.03687
H	y	-5.07909	5.14237	1.77626
H	y	-3.42139	7.16912	2.06858
H	y	-5.08294	5.13307	-1.53691
H	y	-3.44971	7.17303	-1.82758
H	y	5.79926	2.07307	5.42375
H	y	-5.79925	-2.07308	5.42375
H	y	4.33398	3.65005	6.93814
H	y	-4.33398	-3.65006	6.93814
H	y	5.58862	4.63068	4.83927
H	y	-5.58861	-4.63069	4.83927

Supplementary Table 2 | Coordinates of the T6/T6' model of α-O.

atom	constrained?	x	y	z
Fe	n	-0.00001	-0.00095	-2.69180
O	n	1.09485	-1.48564	-1.85966
O	n	-1.09482	1.48507	-1.86123
O	n	-1.68098	-0.98421	-2.24666
O	n	1.68084	0.98359	-2.24700
O	y	2.73947	-6.56012	2.43709
O	y	2.76281	-6.57370	-2.11672
O	y	1.80900	-4.10458	2.29765
O	y	-1.80917	4.10563	2.29598
O	y	4.39307	-4.53736	2.14241
O	y	5.91321	-0.42626	4.02887
O	y	-6.04682	2.51559	2.43514
O	y	-5.91346	0.42797	4.02844
O	y	6.04664	-2.51449	2.43638
O	y	2.82061	-5.25138	0.15802
O	y	-2.82068	5.25160	0.15587
O	y	3.56032	3.46312	-6.20766
O	n	1.86143	-4.08169	-2.04996
O	y	-3.56013	-3.46536	-6.20648
O	n	-1.86147	4.08098	-2.05157
O	y	4.39316	-4.53815	-1.82781
O	y	2.75140	2.85100	4.00290
O	y	-6.02351	2.50201	-2.11868
O	y	-2.75164	-2.84929	4.00388
O	y	6.02353	-2.50267	-2.11745
O	y	5.96487	-0.46584	-3.77516
O	y	-0.61118	-3.08424	2.49939
O	y	-5.96477	0.46453	-3.77560
O	y	0.61099	3.08537	2.49823
O	y	4.79822	1.67649	-4.73945
O	n	0.50579	-3.27217	0.10430
O	y	-4.79809	-1.67816	-4.73901
O	n	-0.50577	3.27198	0.10290
O	y	1.42099	-1.53895	1.84882
O	y	-1.42114	1.53982	1.84816
O	n	2.63192	2.90393	-3.85713
O	n	-0.63176	-3.41847	-2.30387

O	n	-2.63177	-2.90539	-3.85616
O	n	0.63174	3.41781	-2.30538
O	y	4.91081	4.22946	-4.08433
O	y	-4.91071	-4.23089	-4.08291
O	y	5.01981	4.33594	0.11456
O	y	6.12903	-0.05144	-1.18698
O	y	-2.11991	-0.94217	2.31599
O	y	-6.12904	0.05113	-1.18727
O	y	2.11972	0.94324	2.31570
O	y	4.76884	1.72646	0.18834
O	n	-2.80449	0.23295	0.09724
O	y	-4.76891	-1.72624	0.18880
O	n	2.80462	-0.23259	0.09752
O	y	6.20863	-0.08807	1.43925
O	y	-3.92389	0.96645	2.39767
O	y	-6.20876	0.08879	1.43895
O	y	3.92372	-0.96537	2.39821
O	n	3.03041	3.12137	-1.22648
O	n	-3.03052	-3.12179	-1.22549
O	y	3.00778	3.24823	1.41444
O	n	-3.90477	1.00622	-2.41152
O	y	-3.00790	-3.24752	1.41556
O	n	3.90482	-1.00697	-2.41078
Si	y	2.94076	-5.11534	1.75596
Si	y	-2.94091	5.11618	1.75385
Si	y	2.94496	-5.12407	-1.44126
Si	y	-2.94498	5.12366	-1.44336
Si	y	0.78682	-3.01075	1.70621
Si	y	-0.78696	3.01156	1.70501
Si	y	3.98967	3.06865	-4.70676
Si	n	0.72940	-3.06971	-1.49063
Si	y	-3.98952	-3.07032	-4.70575
Si	n	-0.72938	3.06921	-1.49197
Si	y	-2.58975	0.45053	1.65850
Si	y	2.58962	-0.44973	1.65880
Si	y	3.96412	3.11862	0.12613
Al	n	-2.56556	0.45420	-1.55103
Si	y	-3.96421	-3.11842	0.12716
Al	n	2.56551	-0.45444	-1.55073
Si	y	5.49811	-0.99627	-2.32742
Si	y	-2.12410	-2.53389	2.55626

Si	y	-5.49807	0.99554	-2.32804
Si	y	2.12393	2.53503	2.55538
Si	n	2.03992	2.61164	-2.38882
Si	n	-2.03994	-2.61244	-2.38796
Si	y	5.52262	-0.99814	2.57500
Si	y	-5.52280	0.99930	2.57436
O	y	-5.01988	-4.33574	0.11601
O	y	-4.39323	4.53835	2.14046
O	y	-2.73964	6.56122	2.43443
O	y	-4.39316	4.53759	-1.82975
O	y	-2.76278	6.57303	-2.11939
O	y	5.02425	1.88509	4.91264
O	y	-5.02454	-1.88303	4.91315
O	y	3.56002	3.46557	6.42435
O	y	-3.56037	-3.46293	6.42554
O	y	4.81141	4.44276	4.33107
Si	y	4.03985	3.15792	4.91743
Si	y	-4.04013	-3.15587	4.91847
O	y	-4.81166	-4.44093	4.33258
H	y	3.42064	-7.16895	2.11115
H	y	3.44905	-7.17361	-1.78501
H	y	5.07854	-5.14242	1.81848
H	y	6.87711	-0.44585	4.13575
H	y	-7.01074	2.53563	2.54169
H	y	-6.87735	0.44761	4.13528
H	y	7.01056	-2.53449	2.54298
H	y	4.35504	3.57759	-6.75188
H	y	-4.35482	-3.58004	-6.75068
H	y	5.08247	-5.13374	-1.49469
H	y	-6.99136	2.51854	-2.18091
H	y	6.99138	-2.51922	-2.17963
H	y	6.93298	-0.48785	-3.83099
H	y	-6.93288	0.48652	-3.83148
H	y	5.59330	1.77697	-5.28589
H	y	-5.59314	-1.77886	-5.28545
H	y	5.70156	4.34463	-4.63416
H	y	-5.70143	-4.34627	-4.63273
H	y	4.54114	5.17877	0.07709
H	y	7.09679	-0.07162	-1.24910
H	y	-7.09681	0.07129	-1.24943
H	y	5.72218	1.90459	0.20600

H	y	-5.72225	-1.90437	0.20690
H	y	7.17242	-0.10856	1.54692
H	y	-7.17255	0.10932	1.54657
H	y	-4.54121	-5.17858	0.07888
H	y	-5.07869	5.14327	1.81627
H	y	-3.42080	7.16993	2.10823
H	y	-5.08247	5.13332	-1.49689
H	y	-3.44904	7.17308	-1.78793
H	y	5.79929	2.07367	5.46460
H	y	-5.79960	-2.07140	5.46514
H	y	4.33413	3.65109	6.97865
H	y	-4.33451	-3.64823	6.97987
H	y	5.58890	4.63119	4.87961
H	y	-5.58918	-4.62915	4.88116
O	n	0.00459	-0.00862	-4.28135

Supplementary Table 3 | Coordinates of α-^{5}Fe(IV)=O/CH_4 HAA transition state.

atom	constrained?	x	y	z
Fe	n	0.03480	0.00047	-2.69430
O	n	-1.07131	1.48819	-1.84463
O	n	1.11537	-1.47751	-1.78882
O	n	1.73402	1.00072	-2.12279
O	n	-1.66769	-0.98079	-2.22784
O	y	-2.80607	6.55478	2.46285
O	y	-2.73115	6.58256	-2.09034
O	y	-1.87518	4.09873	2.33582
O	y	1.73426	-4.11516	2.38648
O	y	-4.45488	4.53465	2.12624
O	y	-6.01933	0.41927	3.96658
O	y	5.96946	-2.52994	2.62199
O	y	5.80377	-0.44715	4.21859
O	y	-6.11636	2.51259	2.37814
O	y	-2.83929	5.25322	0.17847
O	y	2.79061	-5.25551	0.26510
O	y	-3.44990	-3.44066	-6.22894
O	n	-1.83202	4.08964	-2.01026
O	y	3.67554	3.48044	-6.05247
O	n	1.88180	-4.07480	-1.95800
O	y	-4.36932	4.54780	-1.84304
O	y	-2.86085	-2.86115	3.99855
O	y	6.04441	-2.50217	-1.93120
O	y	2.64637	2.83344	4.13609
O	y	-5.99503	2.51491	-2.17415
O	y	-5.90258	0.48319	-3.83659
O	y	0.53909	3.07528	2.58652
O	y	6.02339	-0.45948	-3.58262
O	y	-0.68871	-3.09305	2.53967
O	y	-4.71747	-1.65733	-4.78219
O	n	-0.52995	3.25874	0.16428
O	y	4.87983	1.68740	-4.56424
O	n	0.48308	-3.27005	0.16759
O	y	-1.48005	1.53411	1.88744
O	y	1.35846	-1.54757	1.93844
O	n	-2.57193	-2.90033	-3.85699
O	n	0.66841	3.42681	-2.21368

O	n	2.69731	2.90660	-3.72730
O	n	-0.60603	-3.40904	-2.26232
O	y	-4.84662	-4.21222	-4.13766
O	y	4.98073	4.23796	-3.89788
O	y	-5.04628	-4.33164	0.05755
O	y	-6.12294	0.06091	-1.25386
O	y	2.04936	0.93223	2.42900
O	y	6.13218	-0.05430	-0.99006
O	y	-2.19108	-0.94880	2.33136
O	y	-4.79444	-1.72267	0.14491
O	n	2.79727	-0.25270	0.22408
O	y	4.74440	1.72018	0.36191
O	n	-2.82390	0.24143	0.10141
O	y	-6.25914	0.08946	1.37015
O	y	3.84933	-0.97849	2.54360
O	y	6.15519	-0.10021	1.63714
O	y	-3.99461	0.96141	2.38091
O	n	-3.03020	-3.12629	-1.23709
O	n	3.04951	3.15104	-1.09022
O	y	-3.06171	-3.25005	1.40393
O	n	3.93365	-0.99496	-2.27343
O	y	2.95880	3.23945	1.55516
O	n	-3.87244	1.01981	-2.42115
Si	y	-2.99402	5.11234	1.77301
Si	y	2.87647	-5.12518	1.86572
Si	y	-2.92924	5.13102	-1.42350
Si	y	2.94950	-5.12273	-1.33066
Si	y	-0.84153	3.00569	1.76315
Si	y	0.72610	-3.01820	1.77703
Si	y	-3.91114	-3.05042	-4.73643
Si	n	-0.71426	3.06464	-1.43917
Si	y	4.07208	3.08028	-4.54407
Si	n	0.73883	-3.05759	-1.42248
Si	y	2.53194	-0.45891	1.77744
Si	y	-2.64536	0.44669	1.66884
Si	y	-3.98992	-3.11545	0.09570
Al	n	2.57382	-0.44775	-1.42977
Si	y	3.94255	3.11337	0.28729
Al	n	-2.54540	0.45854	-1.54290
Si	y	-5.46665	1.00863	-2.37746
Si	y	2.04991	2.52319	2.67429

Si	y	5.52506	-0.99450	-2.14713
Si	y	-2.20200	-2.54132	2.56590
Si	n	-2.01488	-2.60452	-2.37388
Si	n	2.07856	2.61848	-2.26062
Si	y	-5.59691	0.99527	2.52327
Si	y	5.44402	-1.01354	2.75464
O	y	4.99940	4.32964	0.30273
O	y	4.32066	-4.55006	2.28538
O	y	2.65917	-6.57213	2.53728
O	y	4.40624	-4.53695	-1.68389
O	y	2.78053	-6.56980	-2.01500
O	y	-5.15187	-1.89572	4.86208
O	y	4.89820	1.86201	5.09115
O	y	-3.72212	-3.48241	6.40006
O	y	3.40327	3.43870	6.57655
O	y	-4.92900	-4.45180	4.27722
Si	y	-4.16903	-3.16958	4.88411
Si	y	3.91513	3.13584	5.07922
O	y	4.70036	4.42192	4.51413
H	y	-3.47947	7.16532	2.12420
H	y	-3.42381	7.18214	-1.77163
H	y	-5.13262	5.14143	1.78950
H	y	-6.98529	0.43953	4.05270
H	y	6.93084	-2.55130	2.74925
H	y	6.76511	-0.46811	4.34612
H	y	-7.08233	2.53325	2.46398
H	y	-4.23281	-3.55262	-6.79054
H	y	4.48190	3.59599	-6.57904
H	y	-5.06508	5.14306	-1.52300
H	y	7.01336	-2.51949	-1.97259
H	y	-6.96129	2.53265	-2.25714
H	y	-6.86924	0.50638	-3.91321
H	y	6.99247	-0.48229	-3.61766
H	y	-5.50066	-1.75530	-5.34597
H	y	5.68658	1.78898	-5.09309
H	y	-5.62544	-4.32485	-4.70478
H	y	5.78324	4.35423	-4.43015
H	y	-4.56773	-5.17484	0.02778
H	y	-7.08911	0.08228	-1.33677
H	y	7.10105	-0.07527	-1.03139
H	y	-5.74811	-1.89987	0.14144

H	y	5.69729	1.89727	0.40113
H	y	-7.22501	0.11060	1.45707
H	y	7.11642	-0.12207	1.76546
H	y	4.52245	5.17308	0.25792
H	y	5.01237	-5.15468	1.97416
H	y	3.34662	-7.18052	2.22394
H	y	5.08764	-5.13442	-1.33810
H	y	3.45890	-7.17158	-1.67070
H	y	-5.93882	-2.08522	5.39660
H	y	5.66135	2.04786	5.66032
H	y	-4.50819	-3.66885	6.93695
H	y	4.16544	3.62148	7.14802
H	y	-5.71833	-4.64113	4.80827
H	y	5.46604	4.60763	5.07995
O	n	0.11261	-0.00505	-4.40301
C	n	2.02376	0.09533	-6.09743
H	n	2.01048	1.09939	-6.51606
H	n	1.09882	0.05682	-5.35102
H	n	2.90542	-0.09543	-5.48946
H	n	1.82309	-0.68508	-6.82916