

Table S2: The table contains the complete set of frequency data for $[\text{Co}([\text{3}^5\text{adz})]^{2+}$ obtained by a UB3LYP/TZV calculation using Gaussian 98 Rev. A7. Symmetry of the vibrations is indicated by the irreducible representation within the C_2 point group. Tentative assignments of the FT-IR KBr pellet spectra of $[\text{Co}([\text{3}^5\text{adz})]\text{ZnCl}_4$ and $[\text{Co}([\text{3}^5\text{adz})](\text{PF}_6)_2$ are provided along with approximate descriptions of selected vibrations.

No.	Sym	Frequency / cm^{-1}	Red. mass / AMU	Force c. / $\text{mDyne} \times \text{\AA}^{-1}$	IR int. / $\text{km} \times \text{mol}^{-1}$	$[\text{Co}([\text{3}^5\text{adz})]\text{ZnCl}_4$	$[\text{Co}([\text{3}^5\text{adz})](\text{PF}_6)_2$	Approximate description
1	A	93.79	3.11	0.02	0.13			
2	B	107.76	2.70	0.02	0.06			
3	A	108.75	4.00	0.03	0.00			Nsec-Co-Nsec bend
4	B	133.45	2.71	0.03	0.11			
5	A	148.26	3.74	0.05	0.02			
6	A	156.67	3.55	0.05	0.48			Ntert-Co-Ntert bend
7	B	180.29	2.00	0.04	0.45			
8	B	196.32	6.14	0.14	1.84			Nsec-Co-Ntert bend
9	A	208.23	2.79	0.07	0.08			
10	B	232.10	4.74	0.15	0.80			Nsec-Co-Ntert bend
11	A	248.60	2.79	0.10	0.04			
12	B	257.92	2.76	0.11	0.35			
13	A	272.75	2.76	0.12	0.58			
14	B	274.12	3.14	0.14	0.94			
15	A	285.71	2.46	0.12	0.01			
16	B	287.58	3.07	0.15	2.05			
17	B	298.23	2.20	0.12	1.47			
18	B	311.47	2.45	0.14	0.80			
19	A	314.96	2.72	0.16	0.64			
20	A	338.54	2.31	0.16	0.04			
21	A	381.70	1.89	0.16	0.19			
22	B	387.21	2.35	0.21	1.10	373	376	Ntert-Co-Ntert stretch
23	B	402.52	2.39	0.23	1.51	396	401	Nsec-Co-Nsec stretch
24	A	422.54	2.99	0.31	0.08	419	417	
25	B	462.32	3.55	0.45	5.79	468	467	N-Co-N stretch
26	B	472.25	3.14	0.41	1.16			N-Co-N stretch
27	A	472.71	3.65	0.48	0.15			
28	A	479.39	2.48	0.34	6.55		484	
29	A	491.59	2.32	0.33	2.00			
30	B	493.54	2.83	0.41	4.71	498		N-Co-N stretch
31	B	511.52	2.19	0.34	7.52	517	515	
32	B	537.98	2.05	0.35	7.65	548		
33	A	541.46	2.06	0.36	0.39			
34	A	634.82	3.72	0.88	4.25	635	632	
35	B	709.72	1.84	0.55	22.69	720	716	
36	A	735.04	2.45	0.78	2.52	745	740	
37	B	772.99	2.75	0.97	19.54	781		
38	A	789.76	2.85	1.05	4.31			
39	A	801.44	2.37	0.90	2.02			
40	B	806.04	1.84	0.70	2.57			
41	B	850.96	2.45	1.04	15.84	843		
42	A	854.22	3.31	1.42	0.53			
43	B	858.74	2.57	1.12	19.84	857		
44	B	892.20	1.66	0.78	0.22			
45	A	897.61	2.55	1.21	0.20			
46	A	911.08	1.64	0.80	2.87			
47	B	911.90	1.83	0.90	22.62	903		
48	A	927.20	1.67	0.85	0.00			
49	B	928.14	2.36	1.20	3.05			
50	A	939.30	2.34	1.22	0.70			
51	A	945.27	2.61	1.37	0.78			
52	B	971.78	2.30	1.28	49.22	941		
53	B	981.49	2.67	1.51	35.41	956	956	
54	A	981.80	2.20	1.25	9.21	973	972	
55	B	1017.05	2.43	1.48	77.07	1020	1018	
56	B	1029.01	2.37	1.48	26.55	1047	1043	
57	A	1034.06	2.76	1.74	14.82			
58	A	1038.26	3.07	1.95	0.27			
59	A	1057.69	2.45	1.61	10.19	1059	1058	
60	B	1074.18	2.40	1.63	22.62	1071	1066	
61	A	1080.94	1.92	1.32	6.94			
62	B	1088.36	2.75	1.92	3.29			
63	B	1097.72	2.39	1.69	35.59	1089	1089	
64	A	1099.13	1.92	1.37	0.24			
65	A	1103.58	3.11	2.23	0.05			
66	B	1114.24	2.24	1.64	15.14			
67	B	1128.45	2.35	1.76	1.36			
68	A	1154.25	2.16	1.70	1.28			
69	B	1161.51	1.89	1.50	40.41	1144	1140	
70	B	1184.50	1.89	1.56	1.63	1163	1166	

No.	Sym	Frequency / cm ⁻¹	Red. mass / AMU	Force c./ mDyne × Å ⁻¹	IR int. / km × mol ⁻¹	[Co([3 ³]adz)] ZnCl	[Co([3 ³]adz)] (PF ₆) ₂	Approximate description
71	A	1200.28	1.81	1.53	12.22	1184	1177	
72	A	1232.71	1.08	0.97	1.38			
73	B	1254.14	1.23	1.14	11.03	1222	1221	
74	B	1275.49	1.15	1.10	9.77	1244	1240	
75	A	1281.16	1.13	1.09	1.17			
76	B	1297.40	1.20	1.19	2.32			
77	A	1299.09	1.18	1.17	0.01			
78	B	1309.41	1.15	1.16	8.67			
79	A	1312.86	1.19	1.20	0.04			
80	B	1318.62	1.14	1.17	8.80			
81	A	1328.12	1.13	1.17	9.21	1257	1256	
82	A	1335.88	1.15	1.21	5.04	1277	1273	
83	B	1348.46	1.15	1.23	12.23	1288	1288	CH ₂ rocking
84	A	1349.34	1.15	1.24	8.24	1312	1311	
85	A	1356.68	1.12	1.22	8.84	1322	1320	
86	B	1358.82	1.14	1.24	14.91	1357	1354	
87	B	1404.17	1.30	1.51	0.46			
88	A	1405.08	1.39	1.61	0.17			
89	A	1410.63	1.27	1.48	0.04			
90	B	1410.79	1.36	1.59	3.90			
91	B	1411.85	1.25	1.47	1.00	1373	1372	C-C-H bending + N-H
92	B	1418.42	1.23	1.46	8.94	1382	1383	bending
93	B	1419.73	1.24	1.48	4.56	1393	1393	
94	A	1420.42	1.24	1.47	2.62			
95	A	1423.46	1.25	1.49	0.01			
96	B	1423.88	1.24	1.48	1.15			
97	A	1425.77	1.24	1.48	0.25			
98	B	1428.60	1.29	1.55	1.22			
99	A	1435.36	1.26	1.53	1.13			
100	B	1441.65	1.29	1.58	0.16			
101	A	1444.46	1.28	1.57	0.01			
102	B	1479.72	1.24	1.60	16.97	1425	1431	H-C-H angle scissoring + N-H bending
103	A	1480.92	1.23	1.59	3.77			
104	A	1516.21	1.08	1.47	10.92			
105	B	1517.13	1.08	1.47	9.26	1441	1448	H-C-H angle scissoring
106	B	1518.63	1.08	1.47	24.80			
107	A	1520.64	1.09	1.48	0.99			
108	B	1528.59	1.08	1.49	27.52			
109	A	1528.74	1.08	1.49	11.79	1457		H-C-H angle scissoring
110	B	1531.09	1.08	1.49	12.98			
111	A	1531.79	1.08	1.49	8.86			
112	A	1535.92	1.08	1.50	7.12			
113	A	1536.97	1.08	1.50	26.91			
114	B	1537.08	1.08	1.51	8.55	1465	1468	H-C-H angle scissoring
115	B	1537.48	1.08	1.50	11.79			
116	A	1545.93	1.07	1.51	0.43			
117	B	1551.15	1.08	1.53	60.01			
118	A	1553.26	1.08	1.54	36.91	1480	1481	H-C-H angle scissoring
119	A	3008.32	1.07	5.70	1.93			
120	B	3009.66	1.07	5.70	7.97			
121	A	3010.44	1.07	5.69	0.41			
122	B	3010.60	1.07	5.69	26.93			
123	B	3016.56	1.07	5.72	5.72			
124	A	3016.68	1.07	5.72	17.15	2879	2886	Sym C-H stretch
125	B	3018.76	1.06	5.69	3.74			
126	A	3018.77	1.06	5.69	20.92			
127	B	3023.27	1.07	5.74	21.14			
128	A	3023.36	1.07	5.74	0.16			
129	A	3030.68	1.06	5.76	1.63			
130	B	3031.00	1.06	5.76	8.02			
131	B	3035.93	1.07	5.78	4.79			
132	A	3036.18	1.06	5.78	0.15			
133	A	3037.85	1.06	5.78	7.11			
134	B	3054.66	1.10	6.04	1.25			
135	A	3057.32	1.10	6.06	1.12			
136	B	3058.02	1.10	6.06	6.15			
137	A	3059.47	1.10	6.04	0.31			
138	B	3059.53	1.10	6.05	3.05			
139	A	3066.84	1.10	6.12	0.19			
140	B	3066.92	1.10	6.11	8.59			
141	A	3079.38	1.10	6.13	19.92			
142	B	3080.02	1.10	6.14	4.99			
143	A	3084.34	1.10	6.15	2.33			
144	A	3087.17	1.10	6.16	0.21		2944	
145	B	3087.46	1.10	6.17	4.32	2926	3068	Assym C-H stretch
146	B	3089.17	1.10	6.17	30.35			
147	A	3095.89	1.10	6.19	29.65			
148	B	3097.64	1.10	6.19	28.47			
149	A	3403.26	1.07	7.33	21.56	3137		Sym N-H stretch
150	B	3403.48	1.07	7.33	1.96	3236	3276	Assym N-H stretch