Supporting Information:

Synthesis, Characterization and Reactivity of Dinuclear Organo-Rare-Earth Metal Alkyl Complexes Supported by 2-Amidate-Functionalized Indolyl Ligands: Substituent Effects on Coordination and Reactivity

Dongjing Hong,[a] Xiancui Zhu,*[a] Shaowu Wang,*[a,b,c] Yun Wei,[a] Shuangliu Zhou,[a] Zeming Huang,[a] Shan Zhu,[a] Ruru Wang,[a] Wenrun Yue[a] and Xiaolong Mu[a]

[a] Laboratory of Functional Molecular Solids, Ministry of Education, Anhui Laboratory of Molecule-Based Materials, College of Chemistry and Materials Science, Anhui Normal University, Wuhu, Anhui 241002, P. R. China

[b] College of Biological and Chemical Engineering, Anhui Polytechnic University, Wuhu, Anhui 241000, P. R. China.

[c] State Key Laboratory of Organometallic Chemistry, Shanghai Institute of Organic Chemistry, Chinese Academy of Sciences, Shanghai 200032, P. R. China

Contents

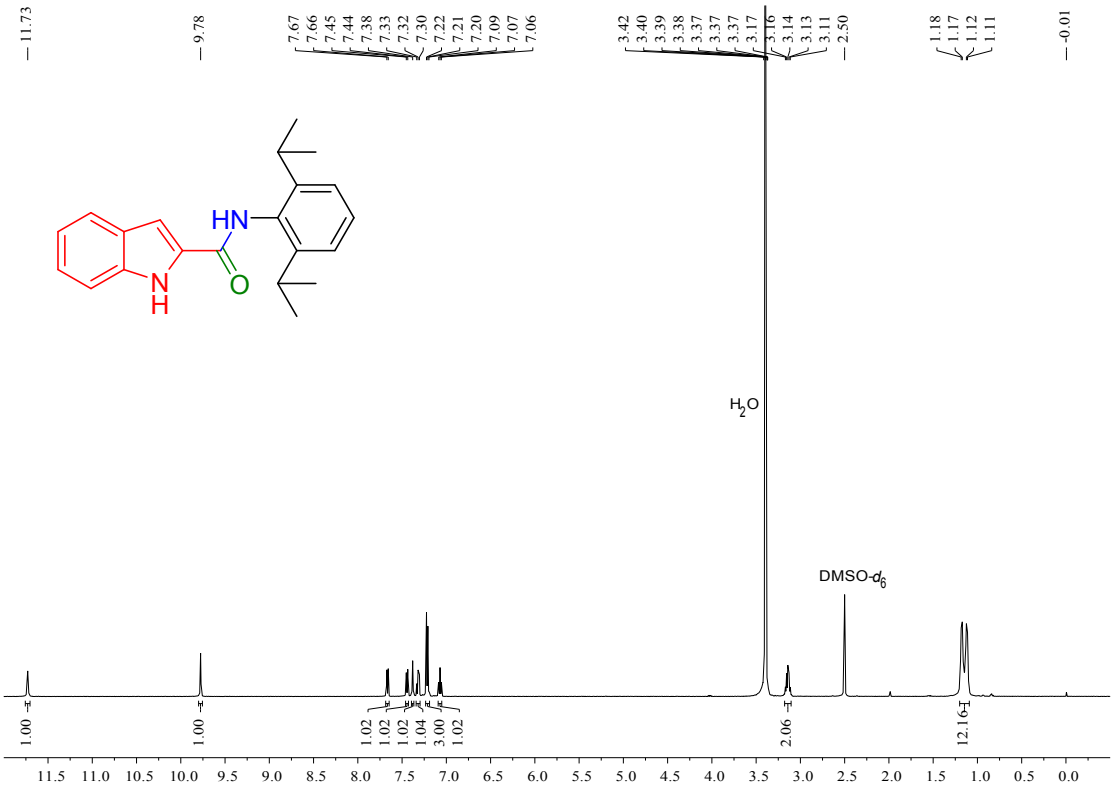

Figure S1. ^{1}H NMR spectrum of pro-ligand $\mathbf{H_2L^1}$ in DMSO-d_6

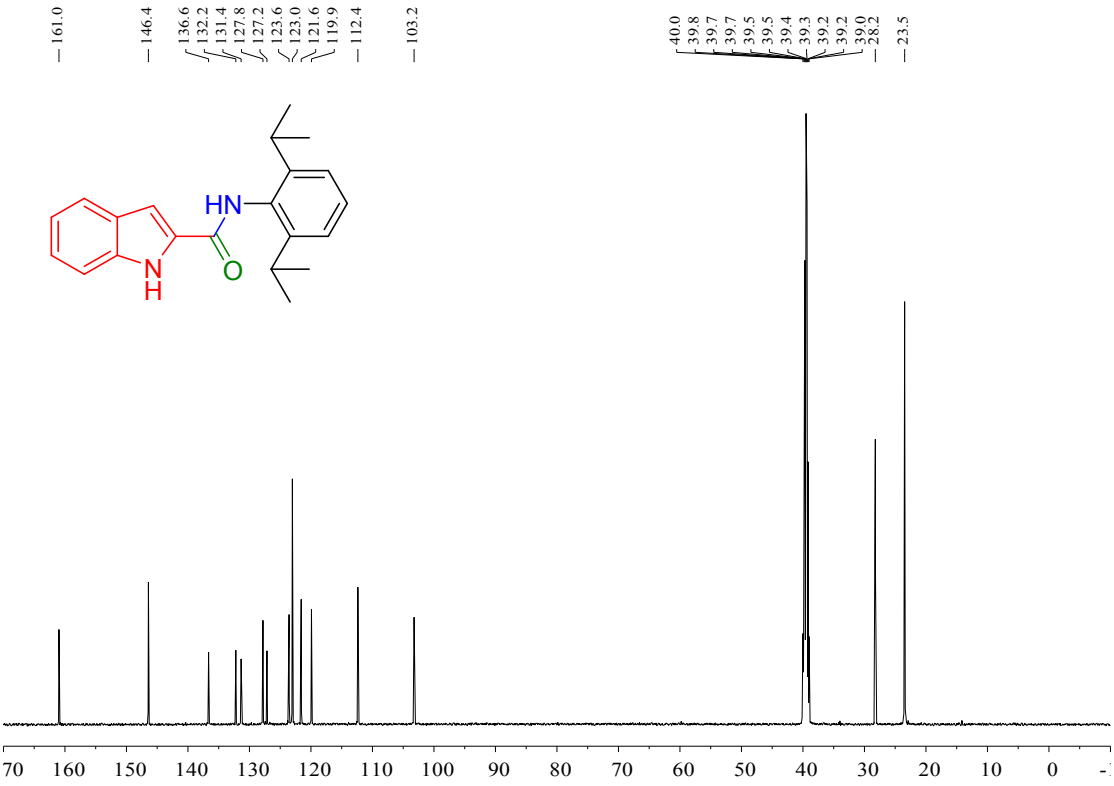

Figure S2. ^{13}C NMR spectrum of pro-ligand $\mathbf{H_2L^1}$ in DMSO-d_6

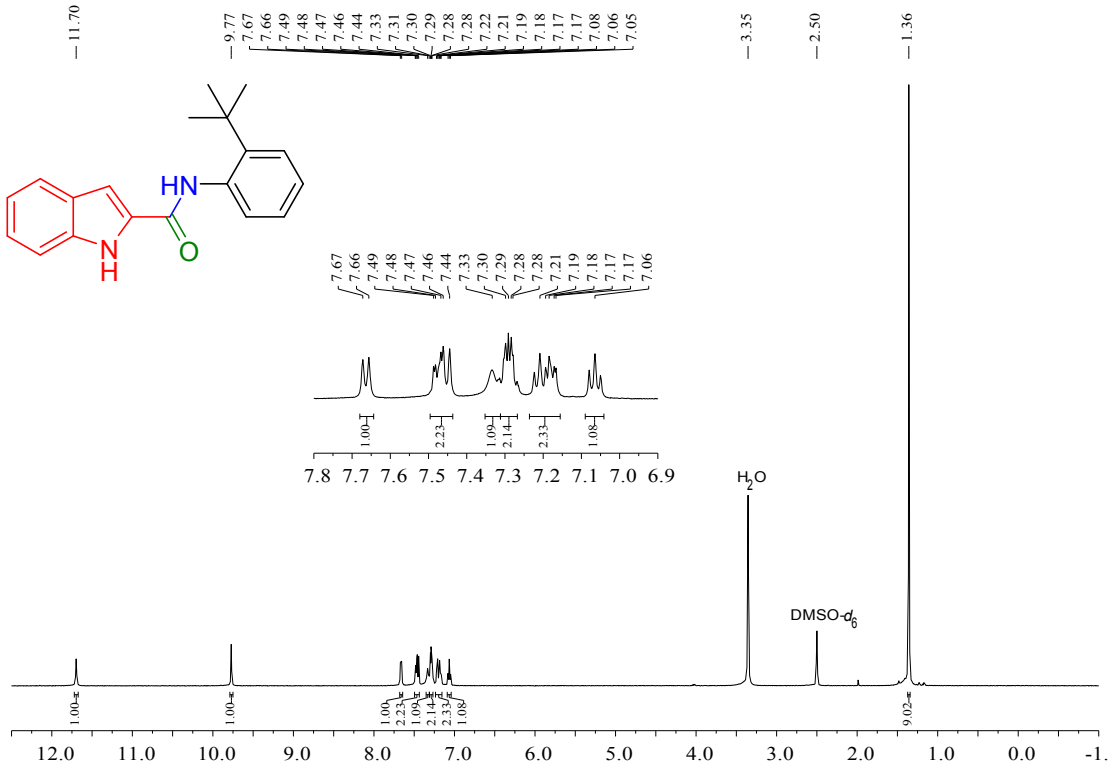

Figure S3. ^{1}H NMR spectrum of pro-ligand $\mathbf{H_2L^2}$ in DMSO-d_6

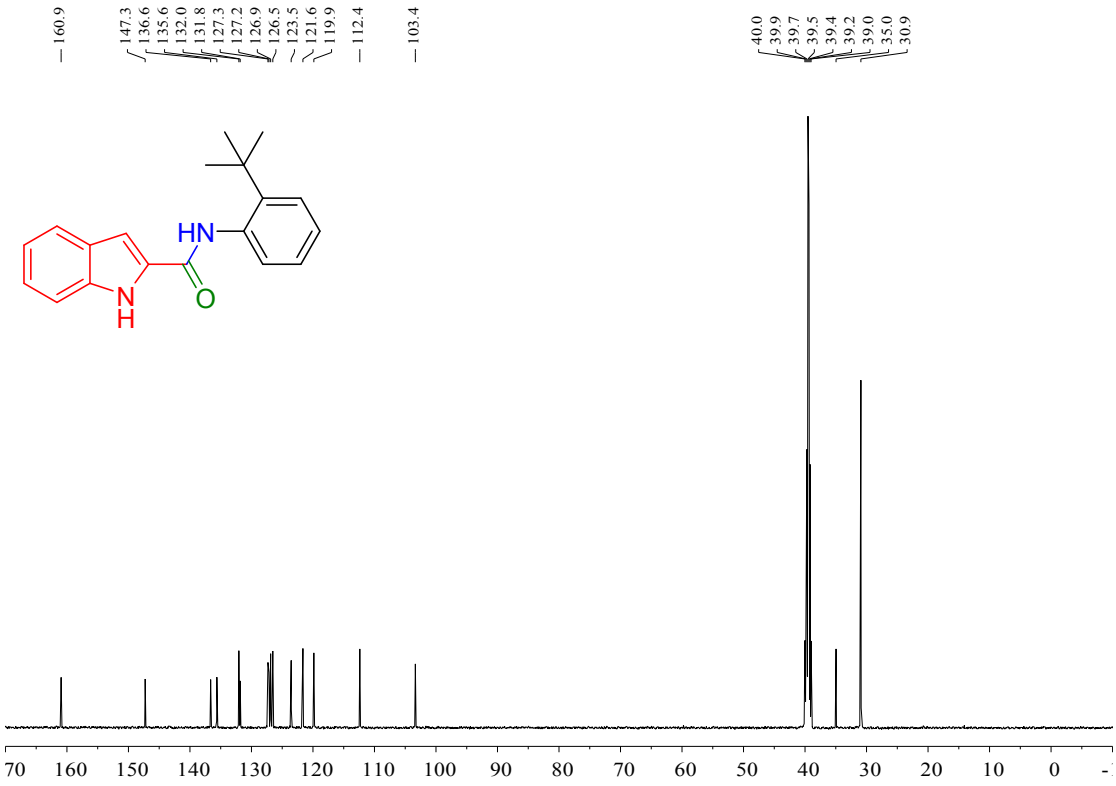

Figure S4. ^{13}C NMR spectrum of pro-ligand $\mathbf{H_2L^2}$ in DMSO-d_6

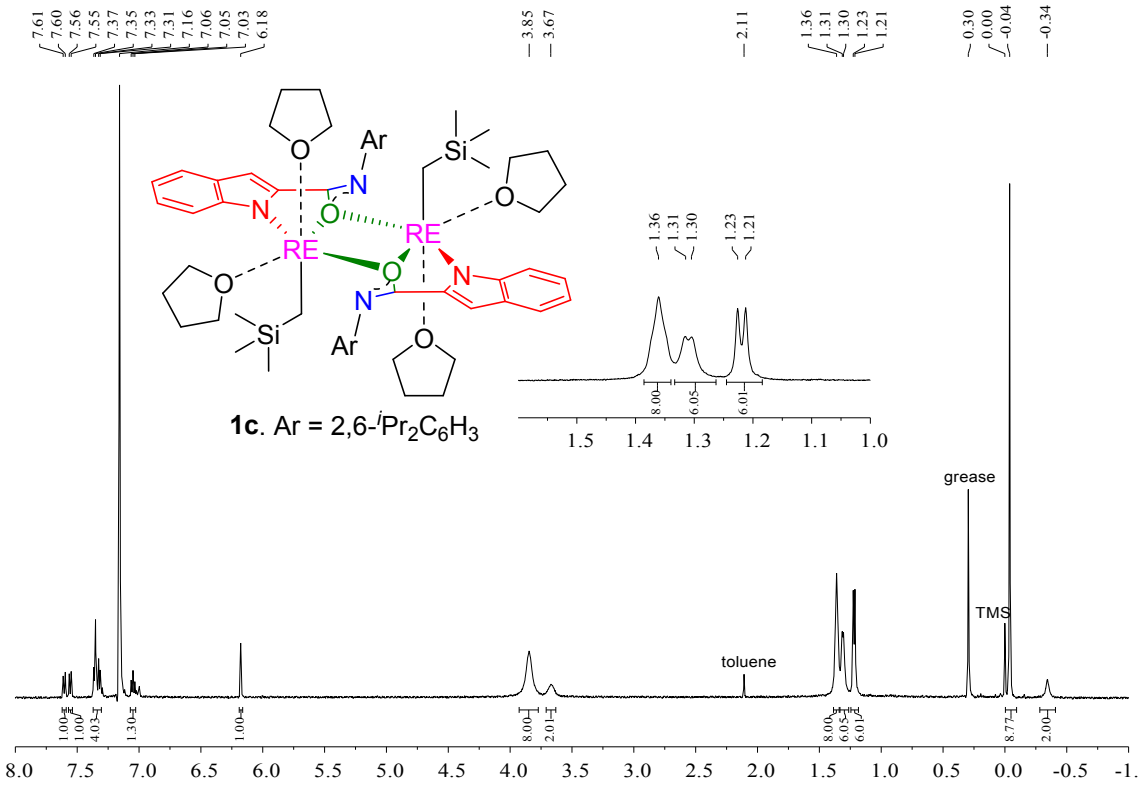

Figure S5. ^{1}H NMR spectrum of complex **1c** in C_6D_6

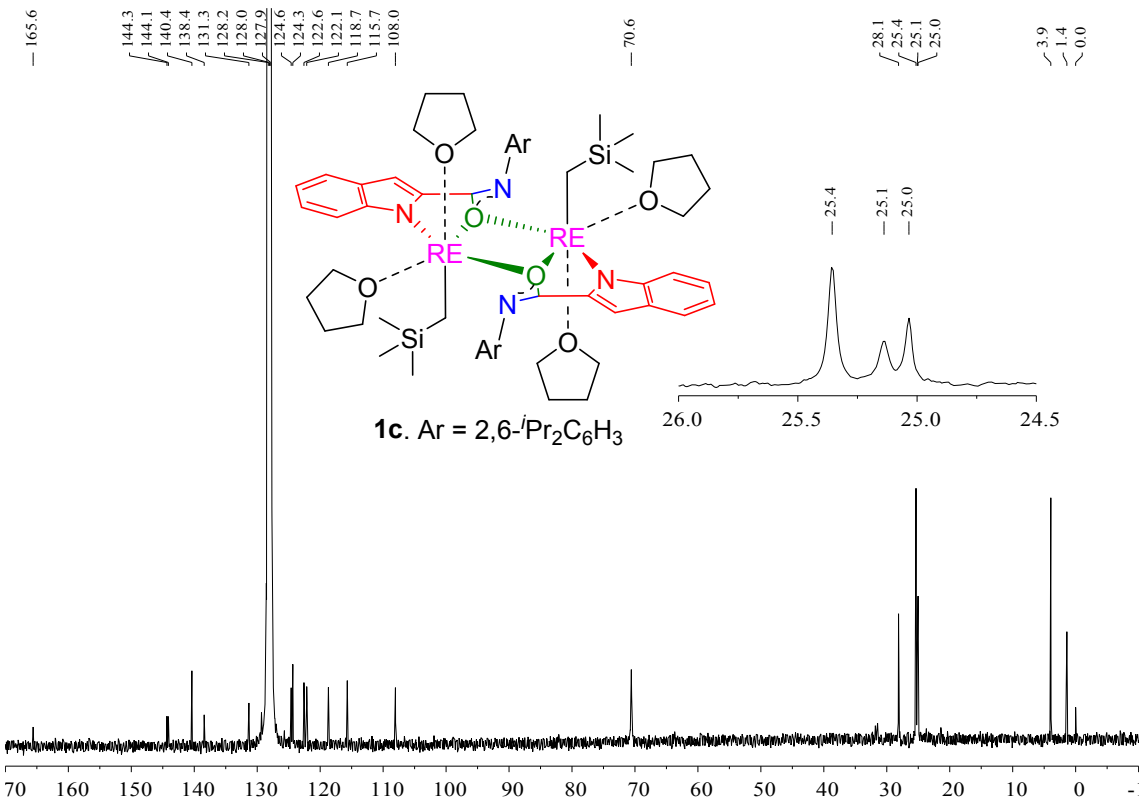

Figure S6. ^{13}C NMR spectrum of complex **1c** in C_6D_6

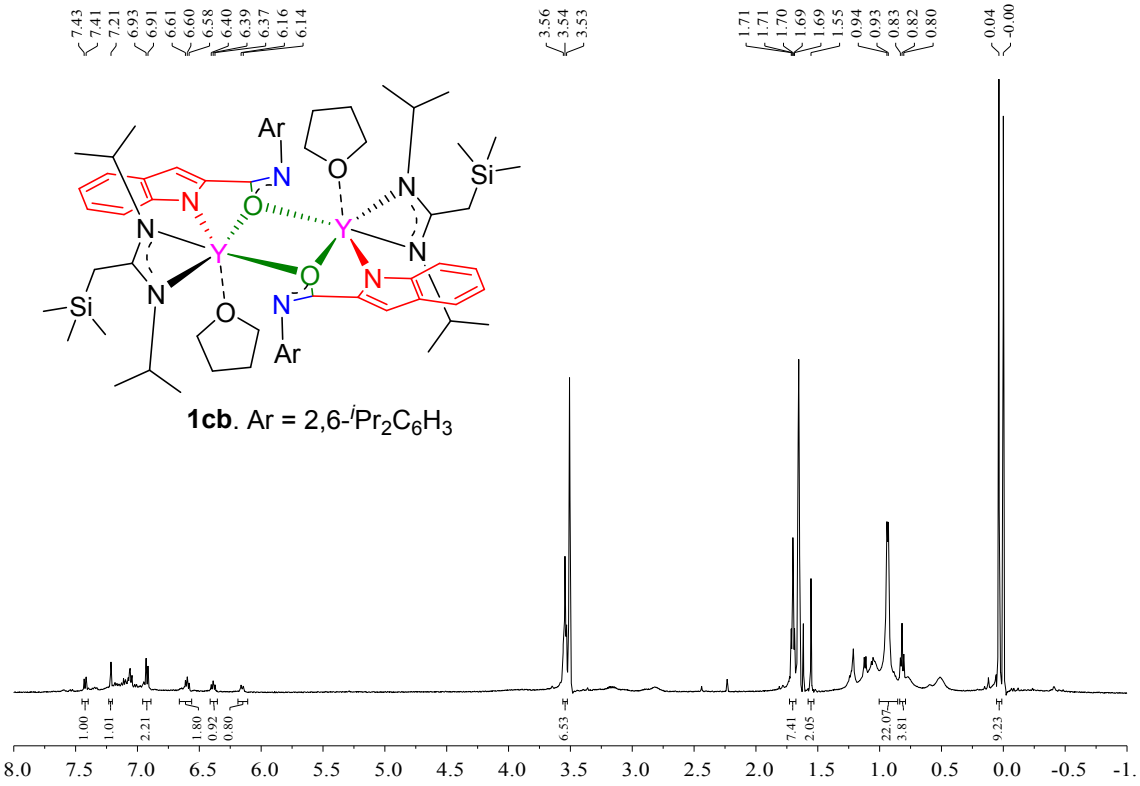

Figure S7. ^{1}H NMR spectrum of complex **1cb** in THF-d_8

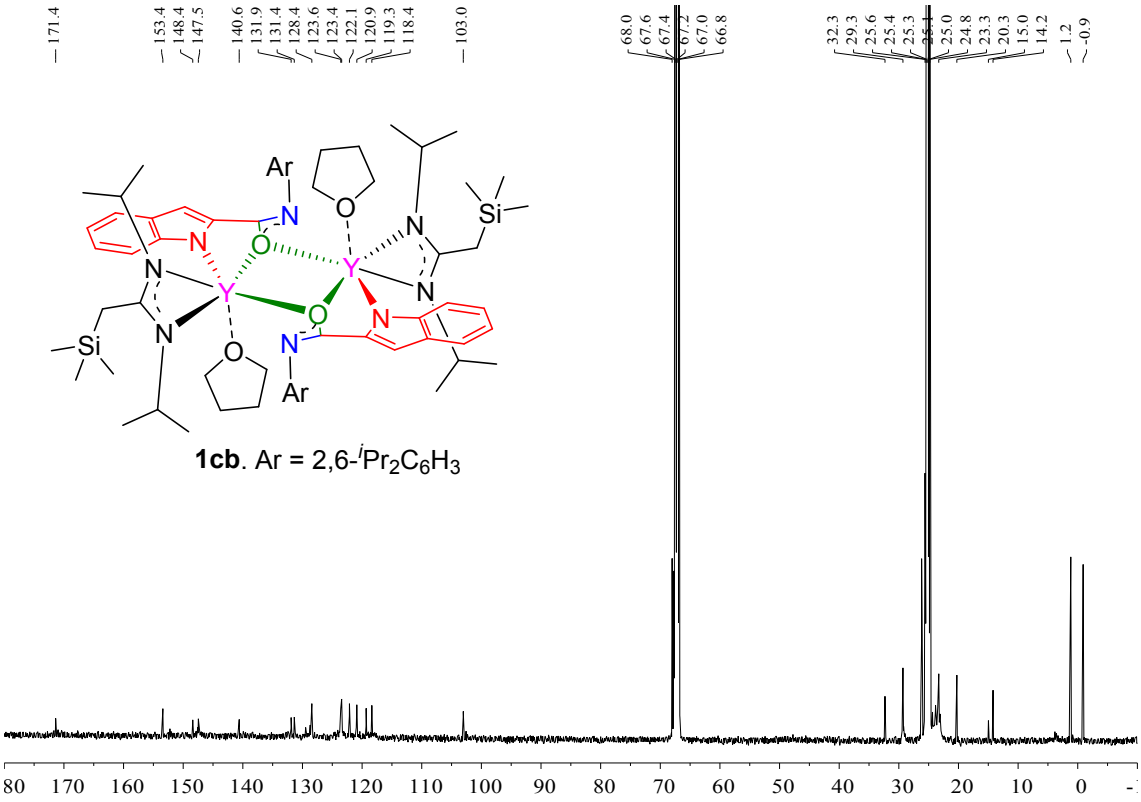

Figure S8. ^{13}C NMR spectrum of complex **1cb** in THF-d_8

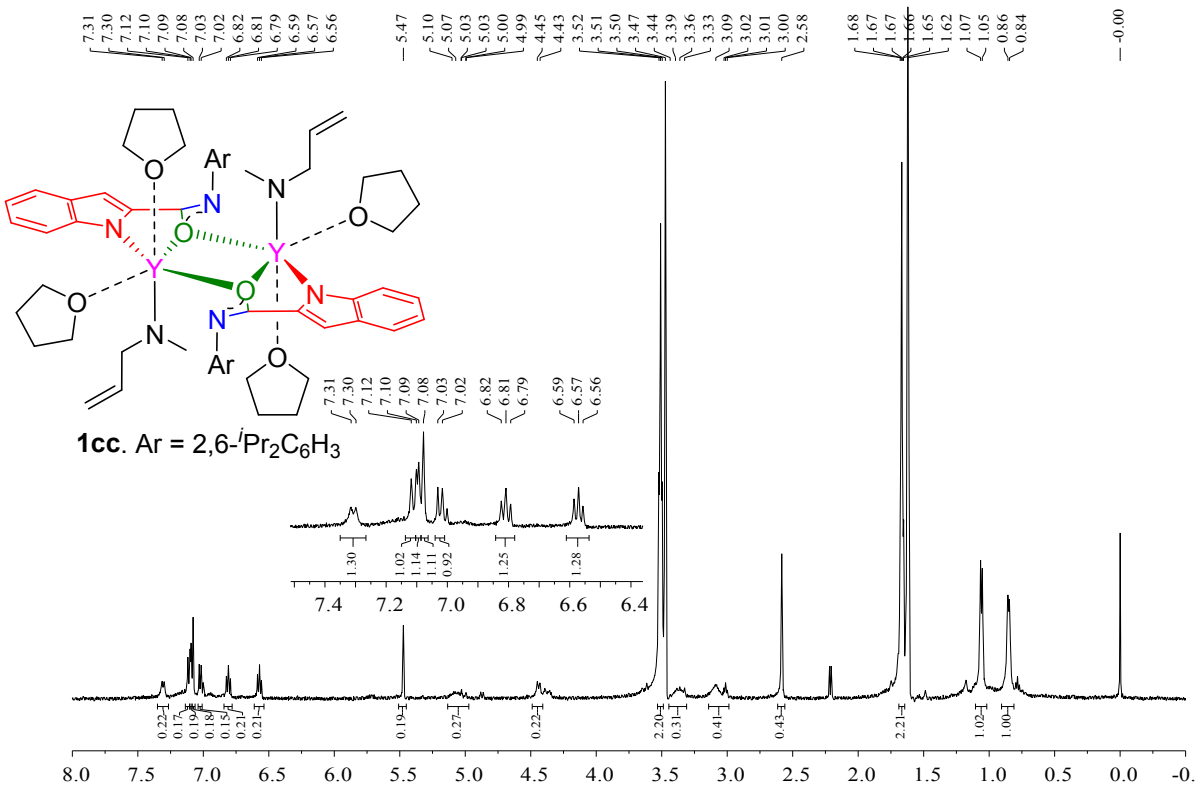

Figure S9. ^{1}H NMR spectrum of complex **1cc** in THF-d_8

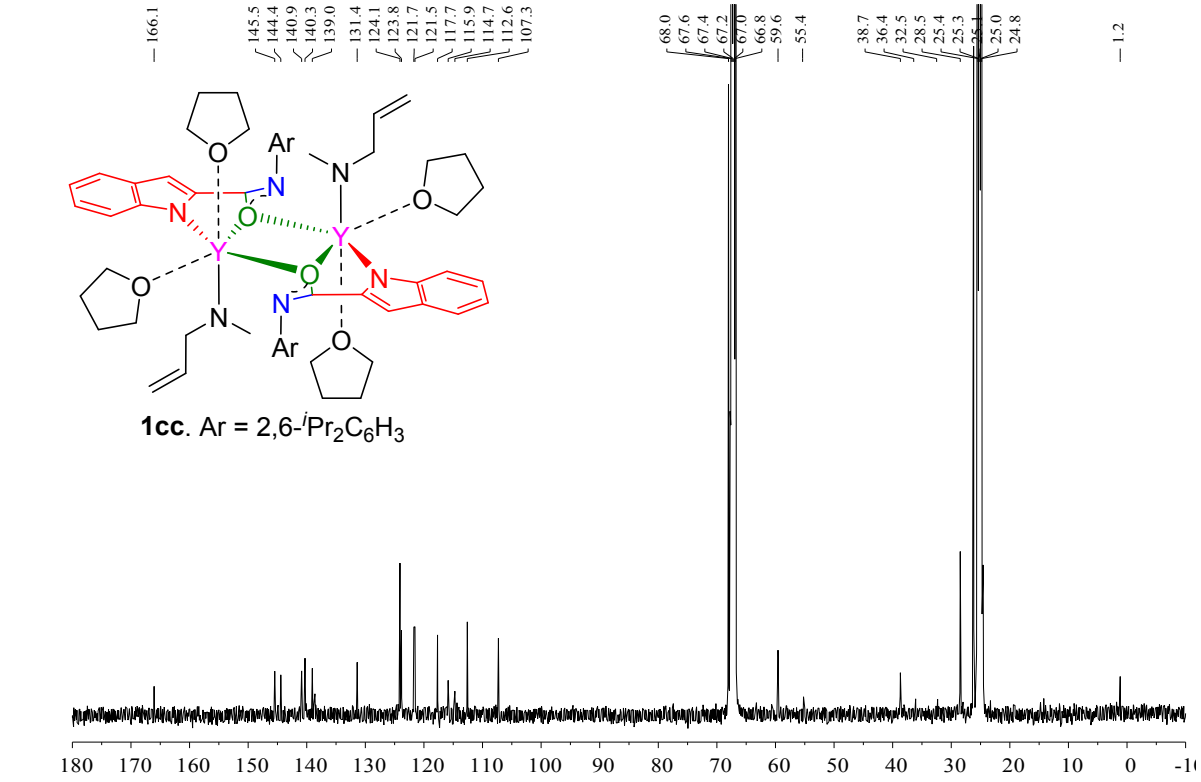

Figure S10. ^{13}C NMR spectrum of complex **1cc** in THF-d_8

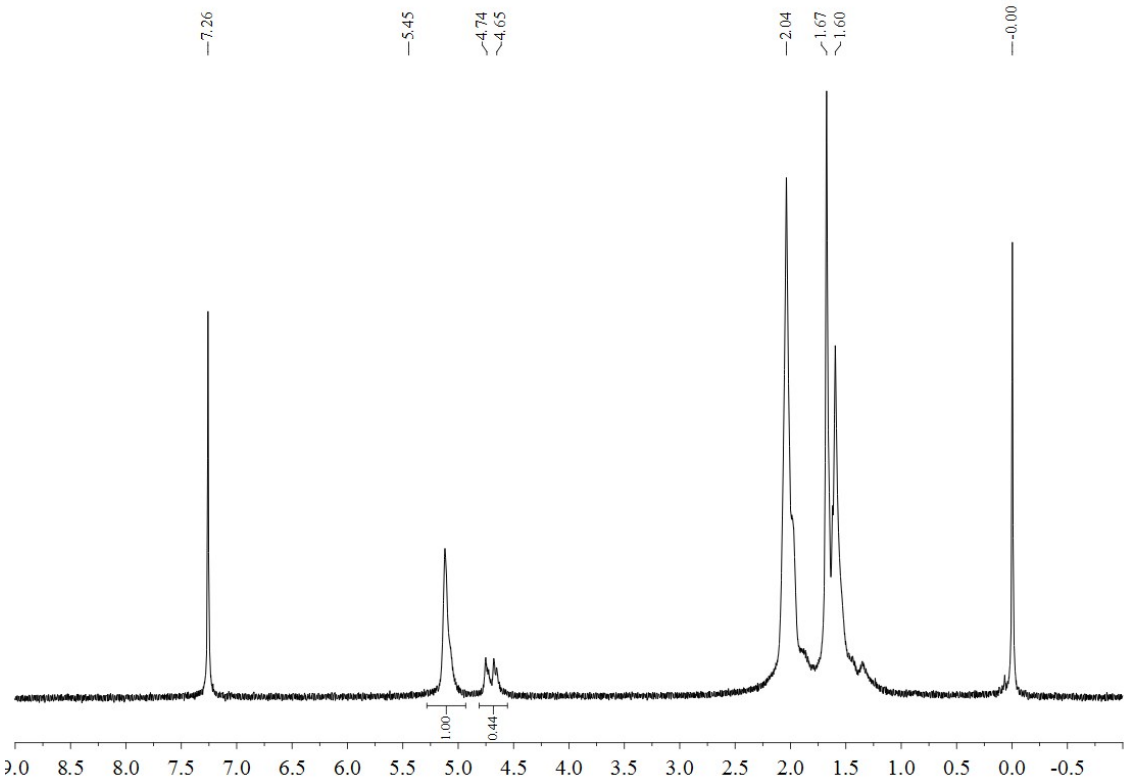

Figure S11. ^{1}H NMR Spectrum of Polyisoprene for Table 3, Entry 4

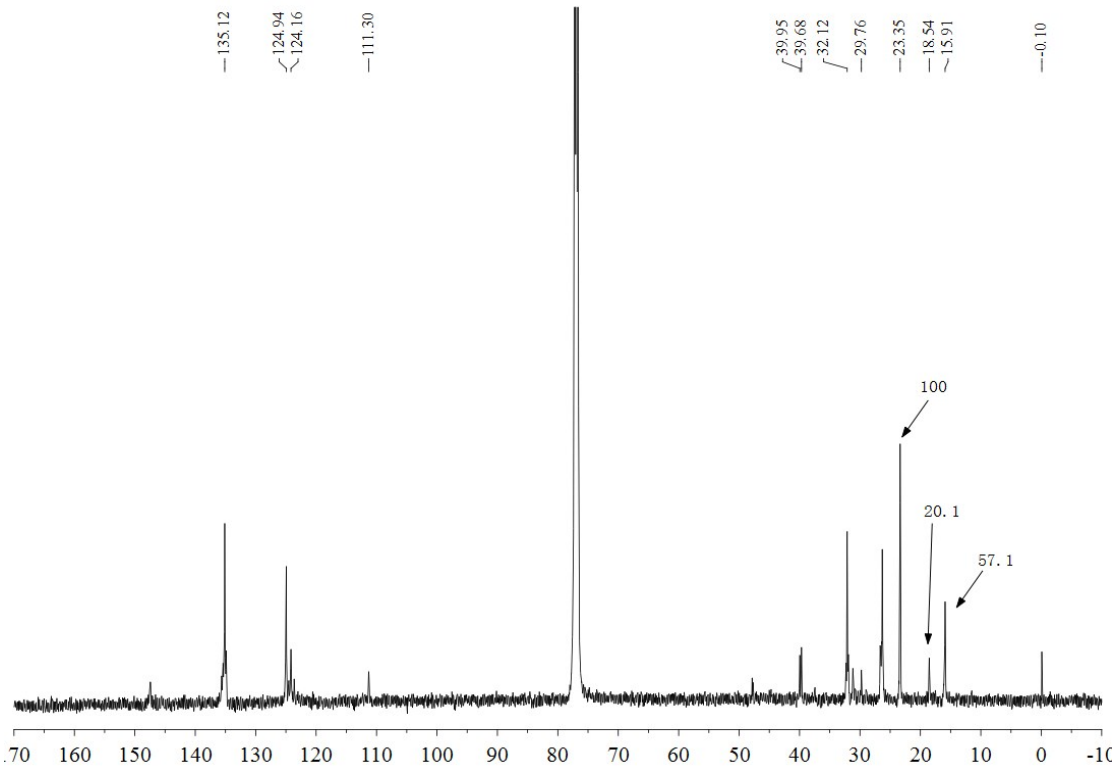

Figure S12. ^{13}C-int NMR Spectrum of Polyisoprene for Table 3, Entry 4

(100 for *cis*-1,4-PIP; 57.1 for *trans*-1,4-PIP; 20.1 for 3,4-PIP)

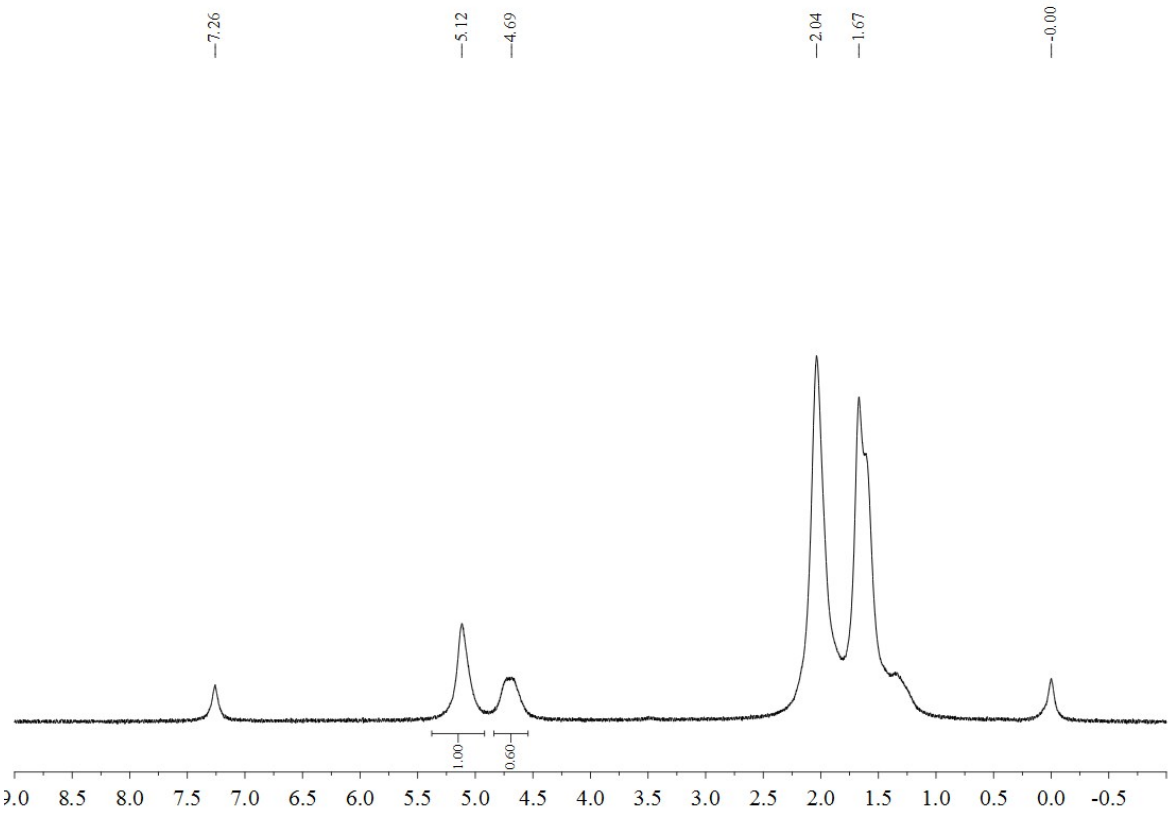

Figure S13. [1]H NMR Spectrum of Polyisoprene for Table 3, Entry 7

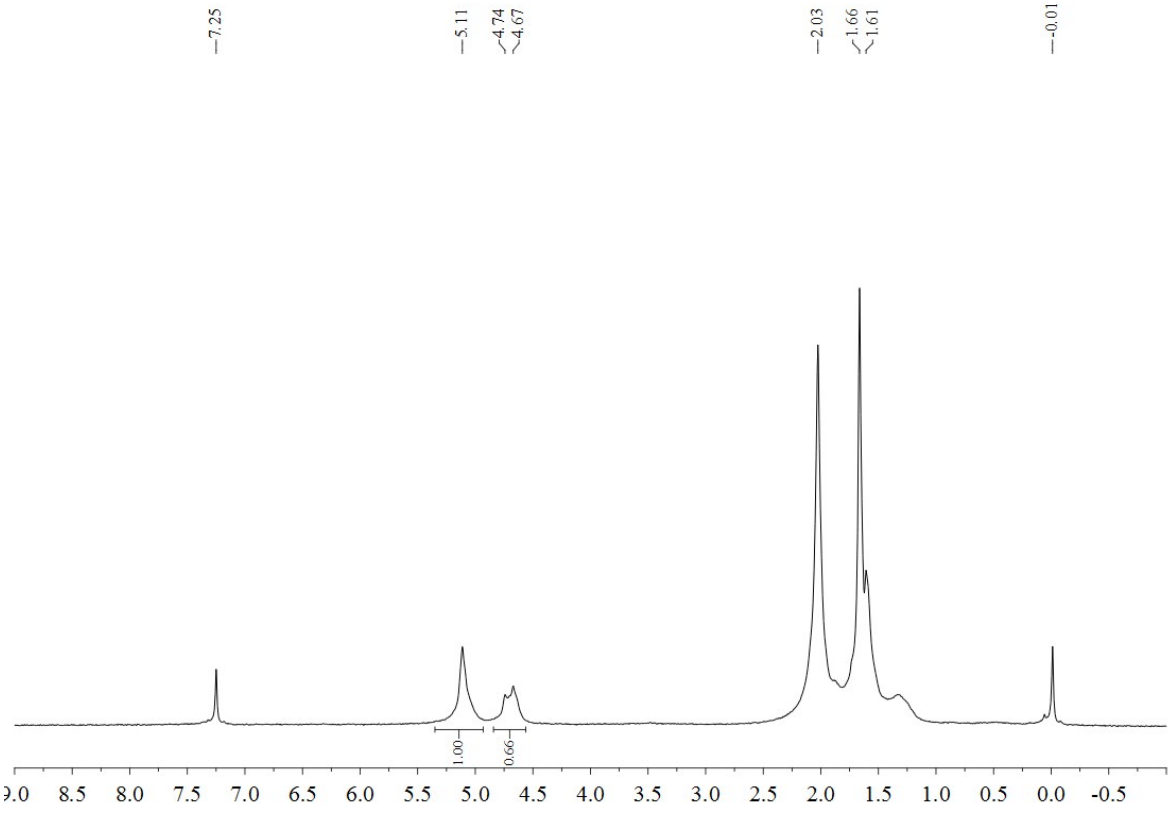

Figure S14. [1]H NMR Spectrum of Polyisoprene for Table 3, Entry 8

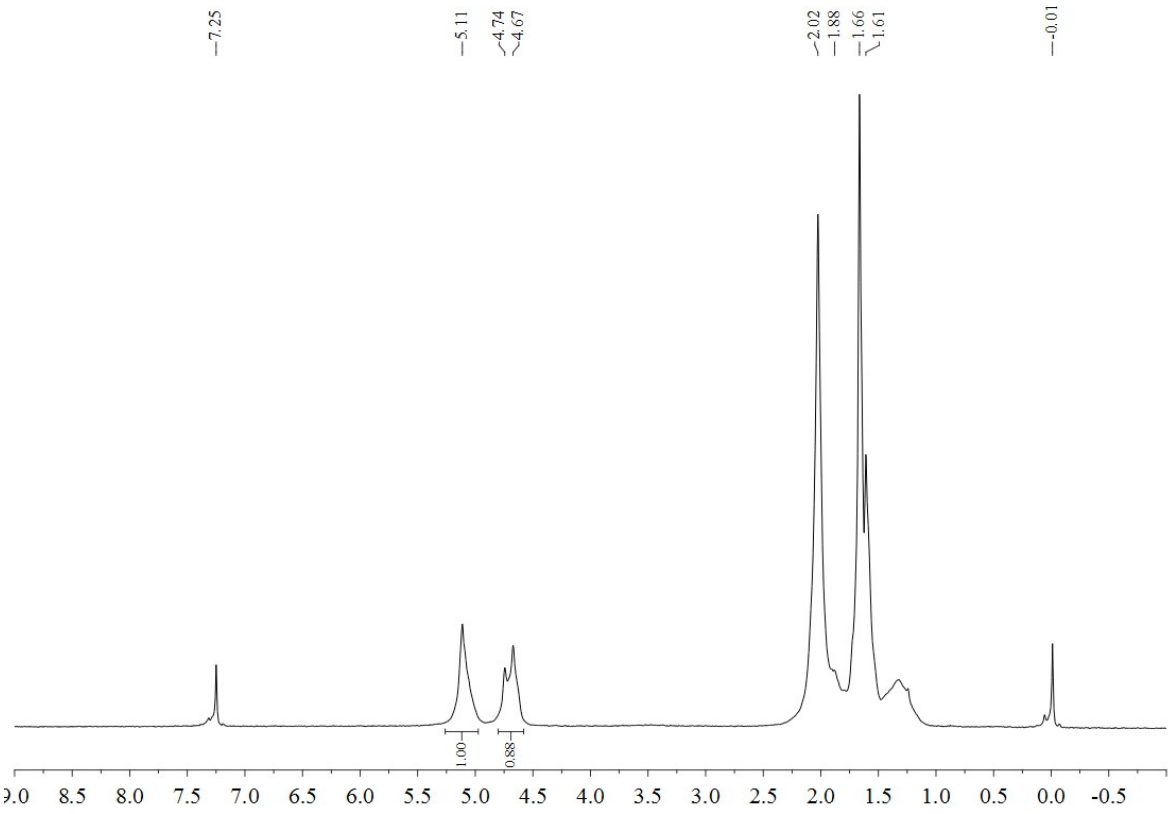

Figure S15. [1]H NMR Spectrum of Polyisoprene for Table 3, Entry 9

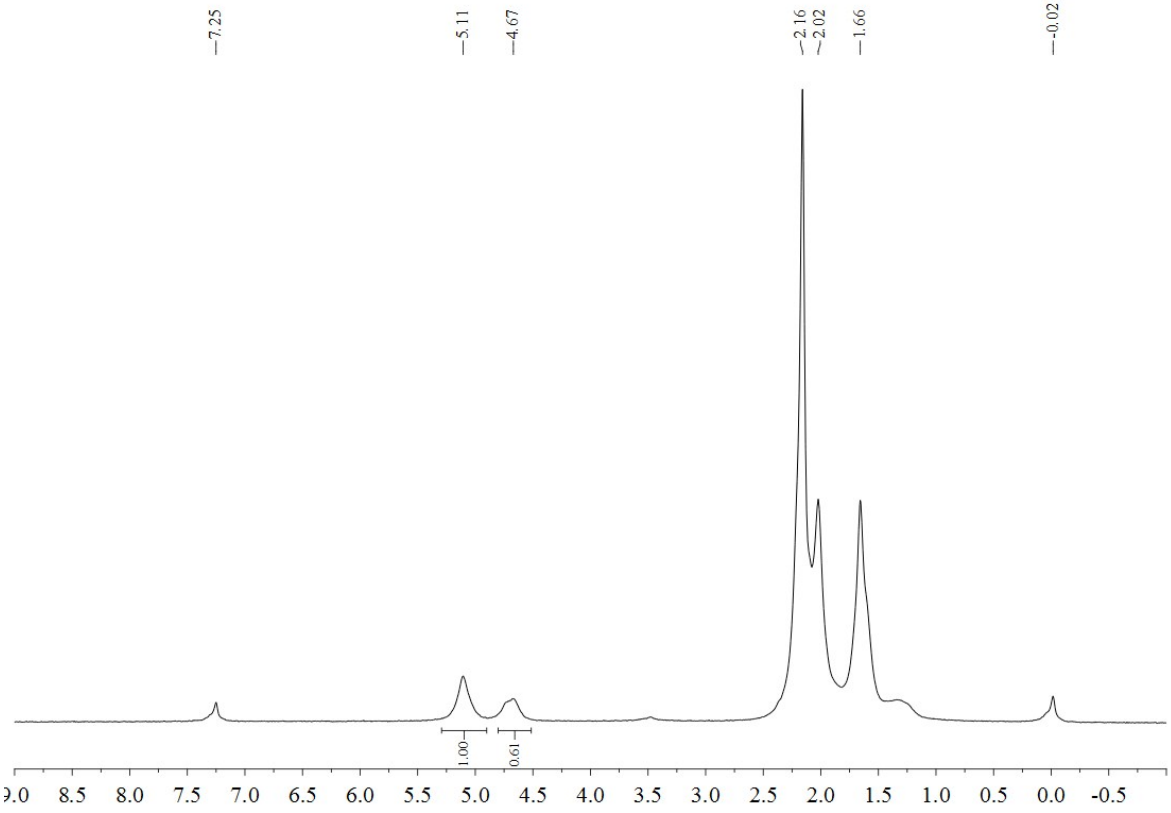

Figure S16. [1]H NMR Spectrum of Polyisoprene for Table 3, Entry 10

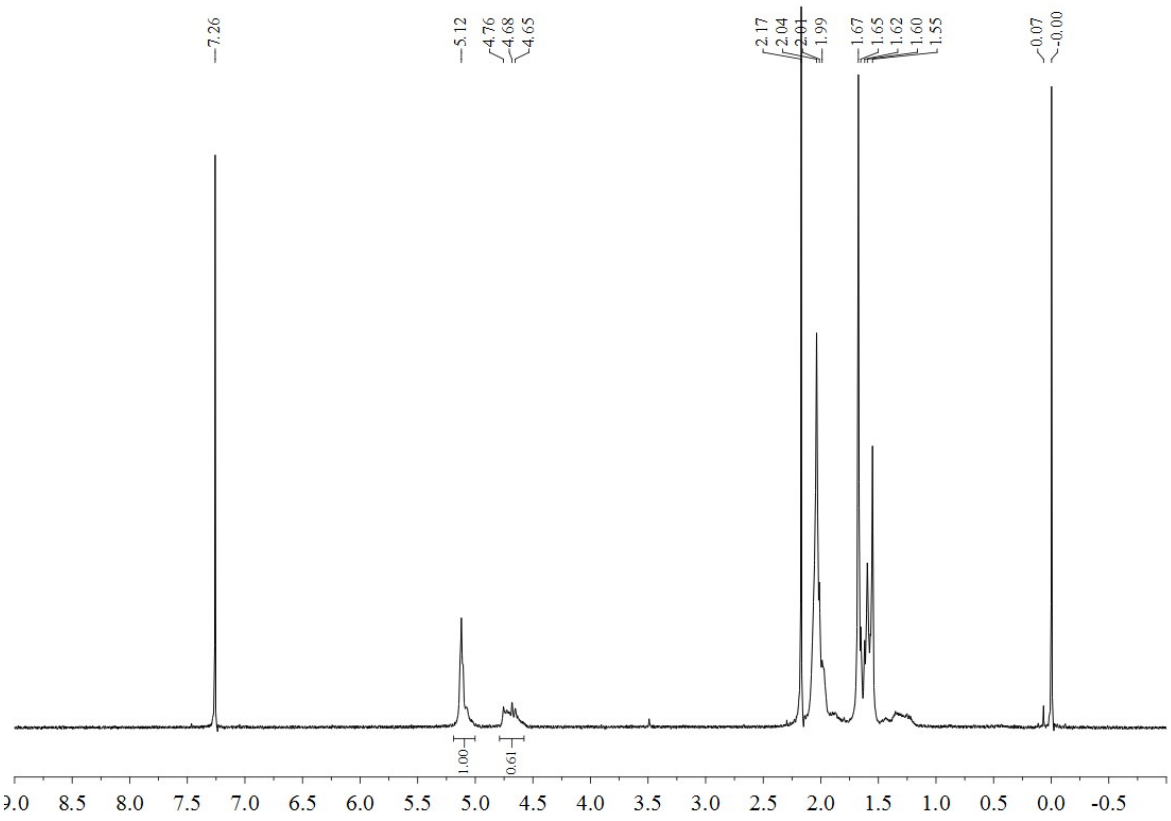

Figure S17. [1]H NMR Spectrum of Polyisoprene for Table 3, Entry 11

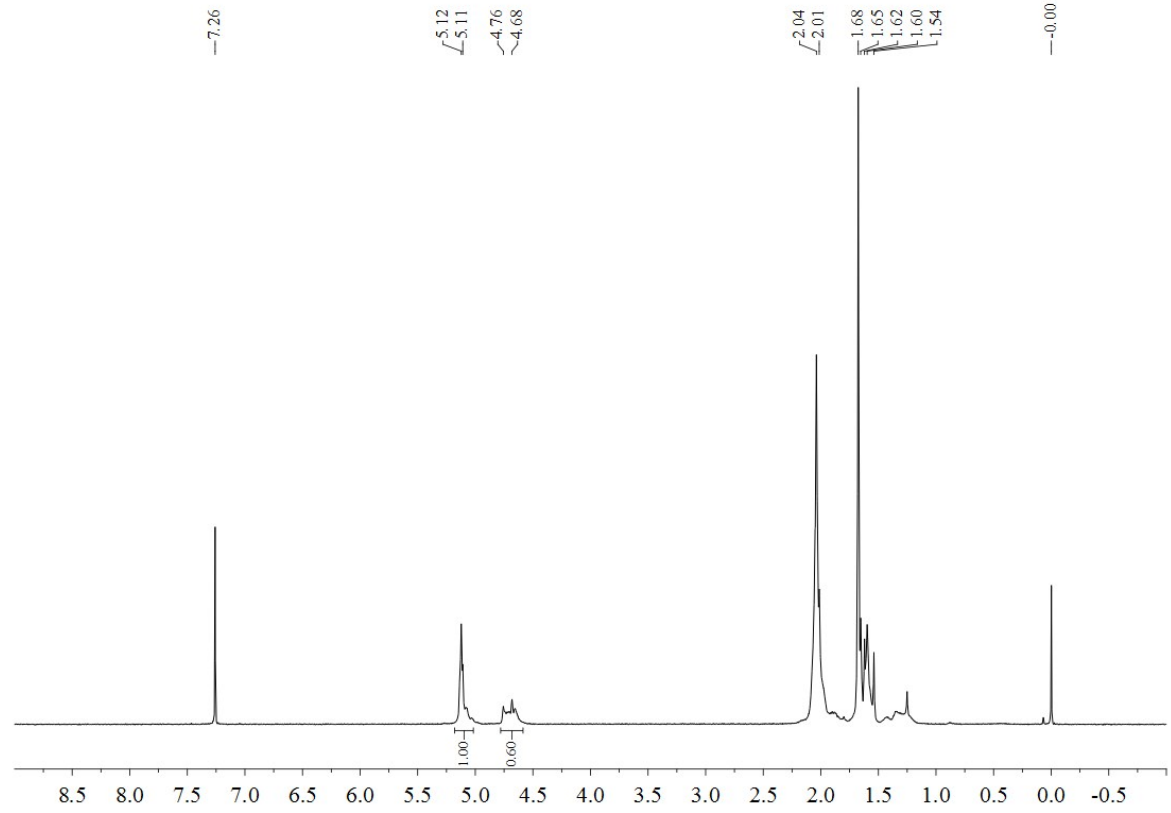

Figure S18. [1]H NMR Spectrum of Polyisoprene for Table 3, Entry 12

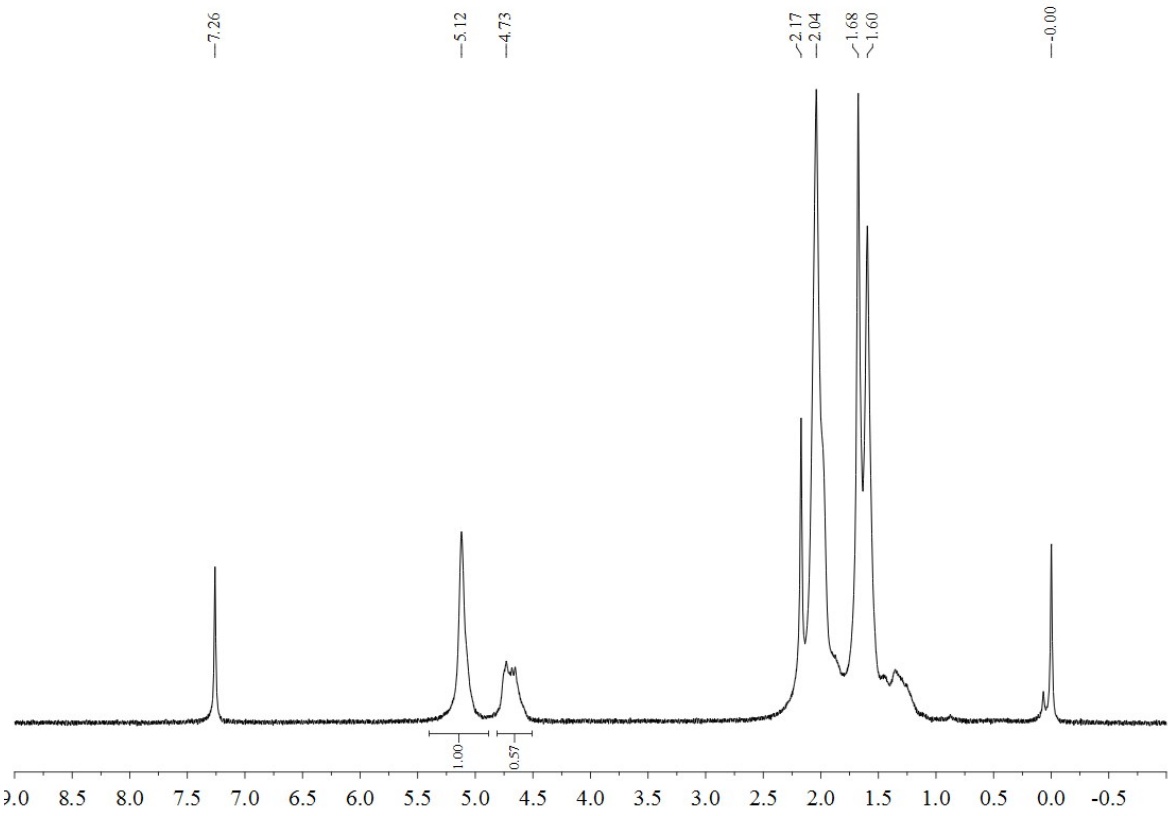

Figure S19. ^{1}H NMR Spectrum of Polyisoprene for Table 3, Entry 13

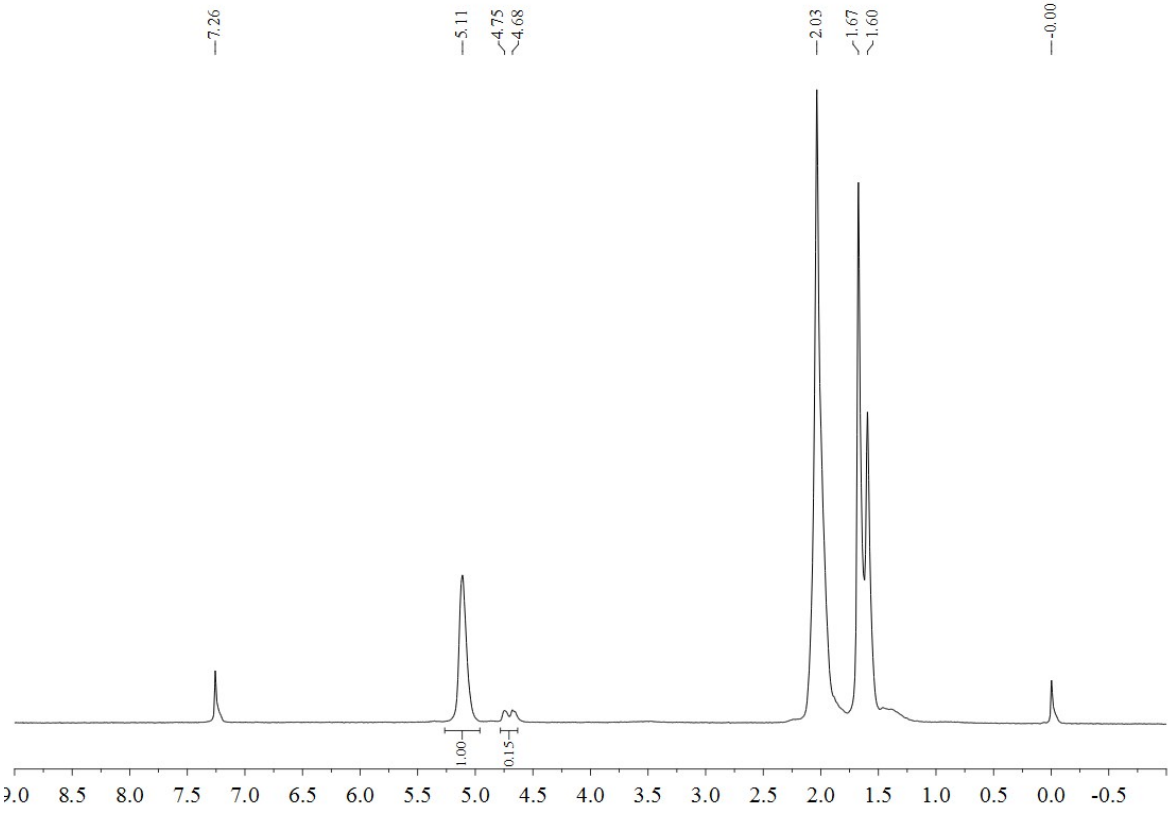

Figure S20. ^{1}H NMR Spectrum of Polyisoprene for Table 4, Entry 2

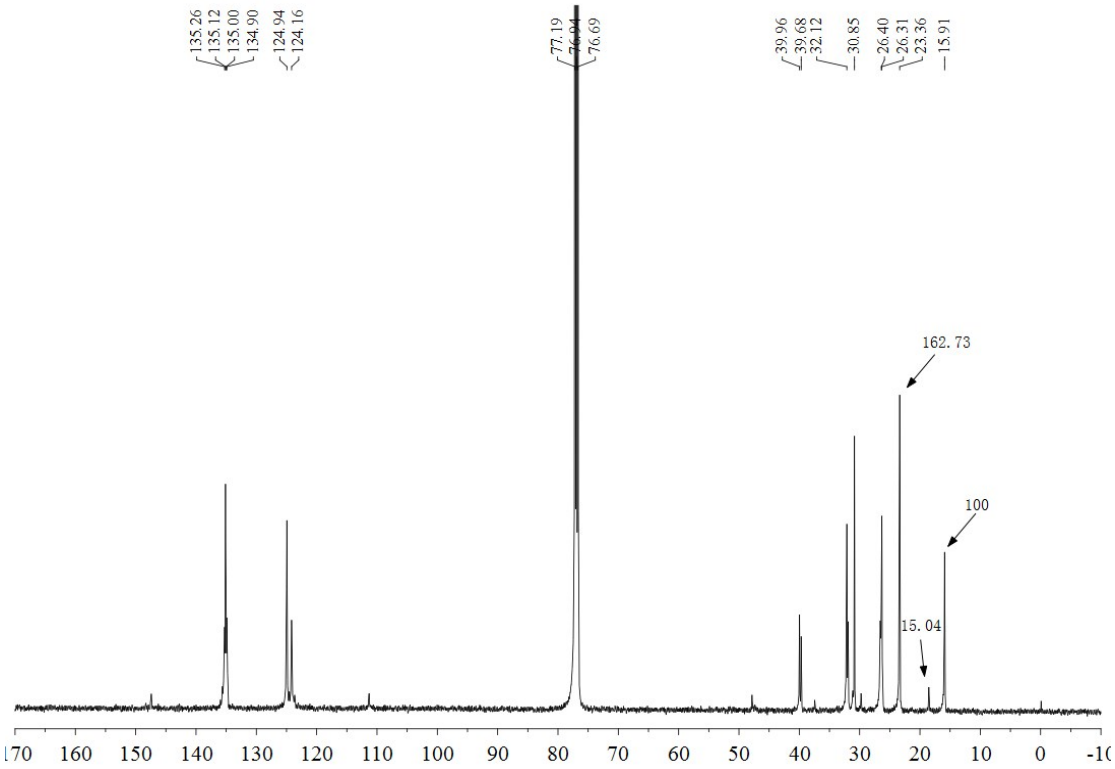

Figure S21. ^{13}C-int NMR Spectrum of Polyisoprene for Table 4, Entry 2 (162.73 for *cis*-1,4-PIP; 100 for *trans*-1,4-PIP; 15.04 for 3,4-PIP)

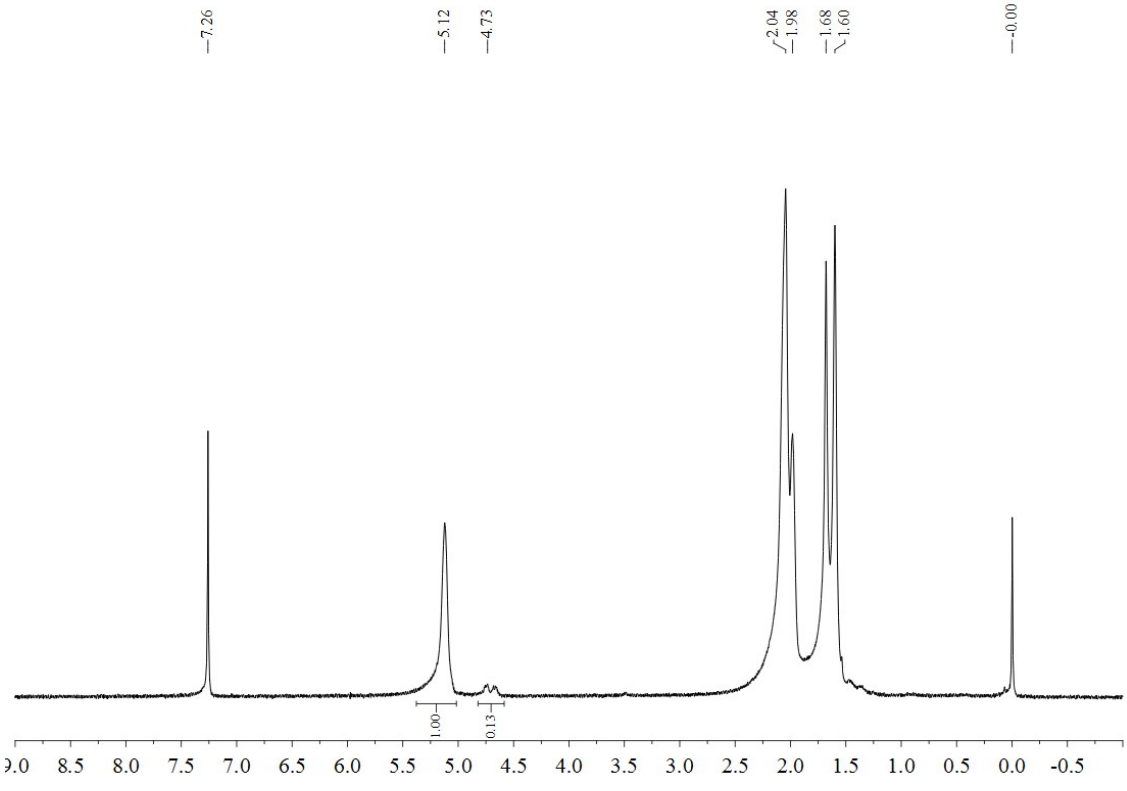

Figure S22. ^{1}H NMR Spectrum of Polyisoprene for Table 4, Entry 3

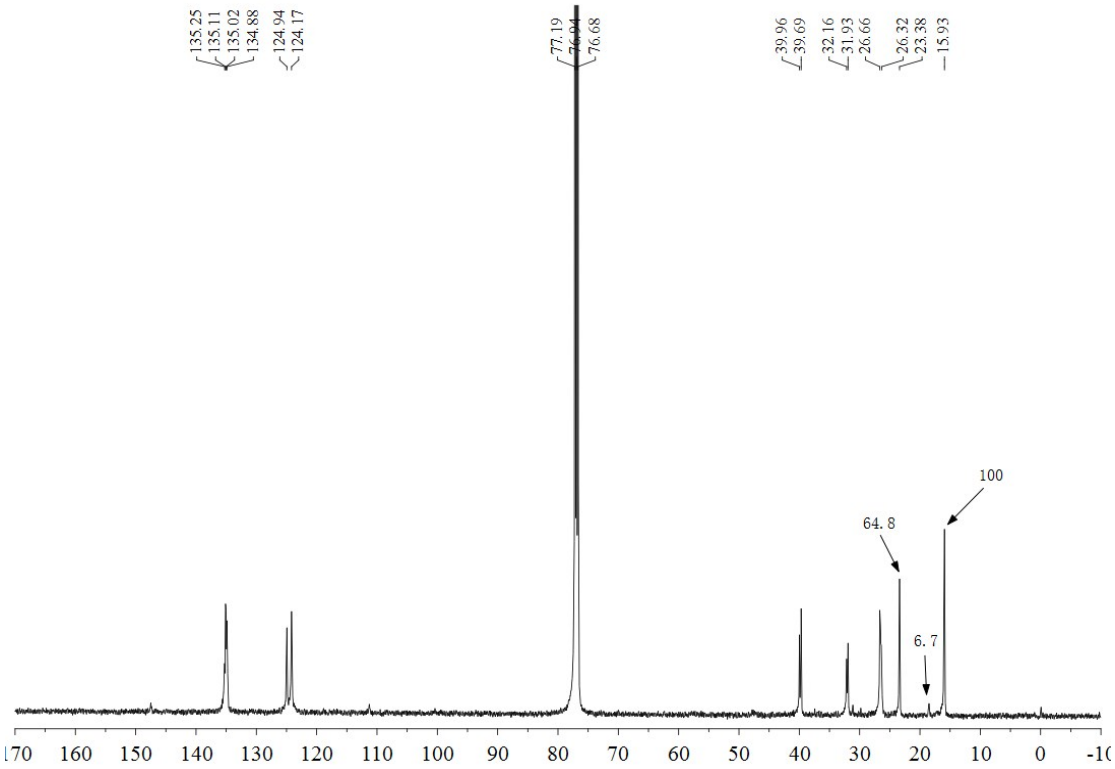

Figure S23. ^{13}C-int NMR Spectrum of Polyisoprene for Table 4, Entry 3 (64.8 for *cis*-1,4-PIP; 100 for *trans*-1,4-PIP; 6.7 for 3,4-PIP)

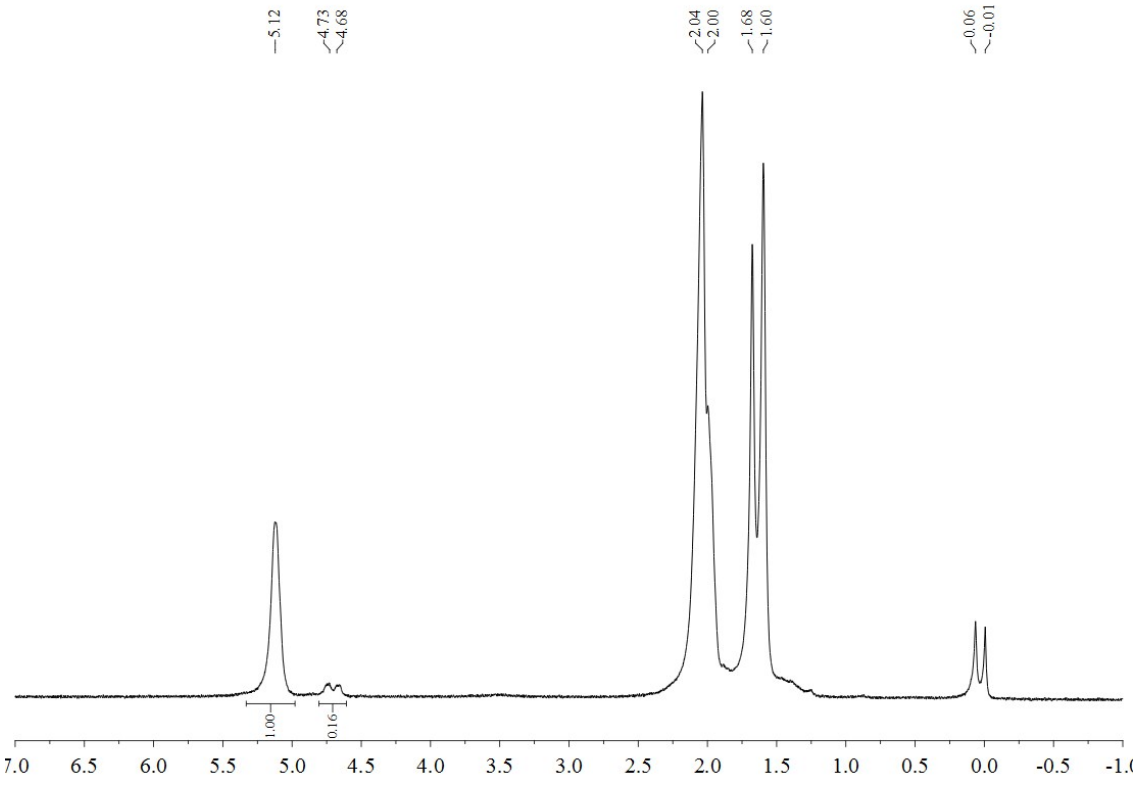

Figure S24. ^{1}H NMR Spectrum of Polyisoprene for Table 4, Entry 4

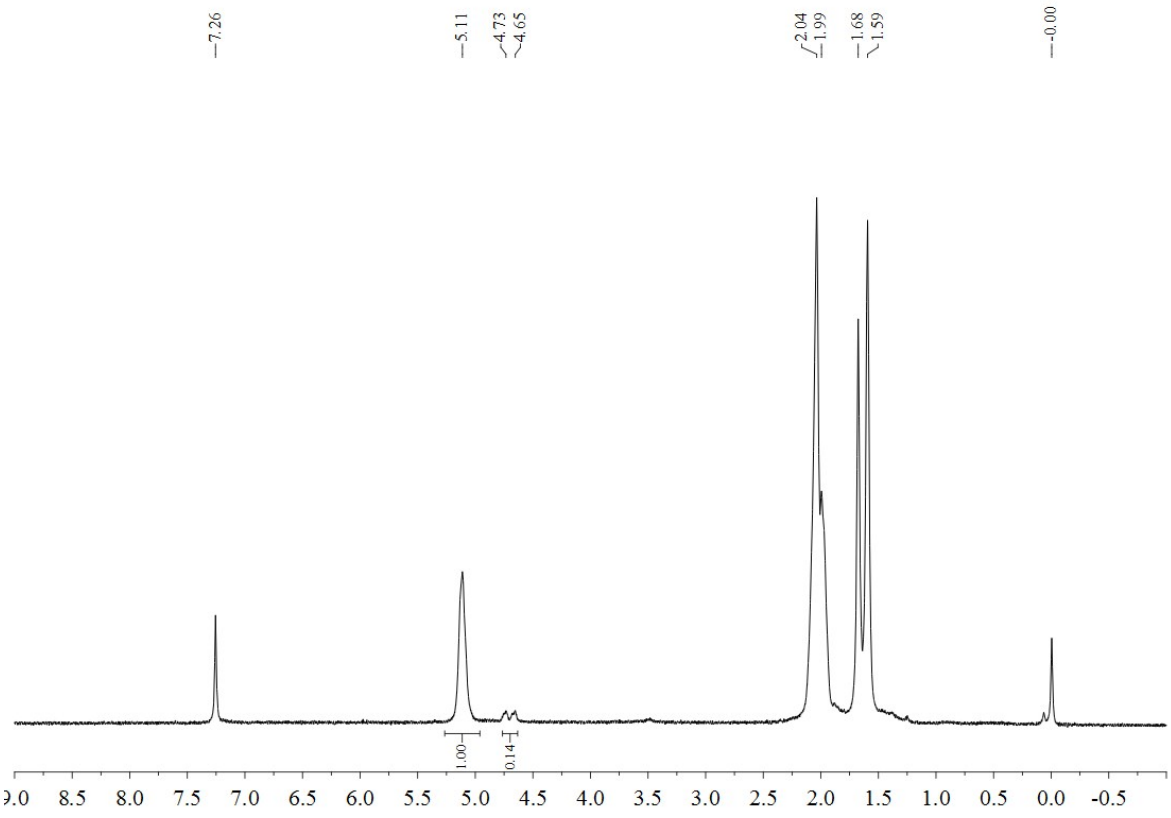

Figure S25. [1]H NMR Spectrum of Polyisoprene for Table 4, Entry 5

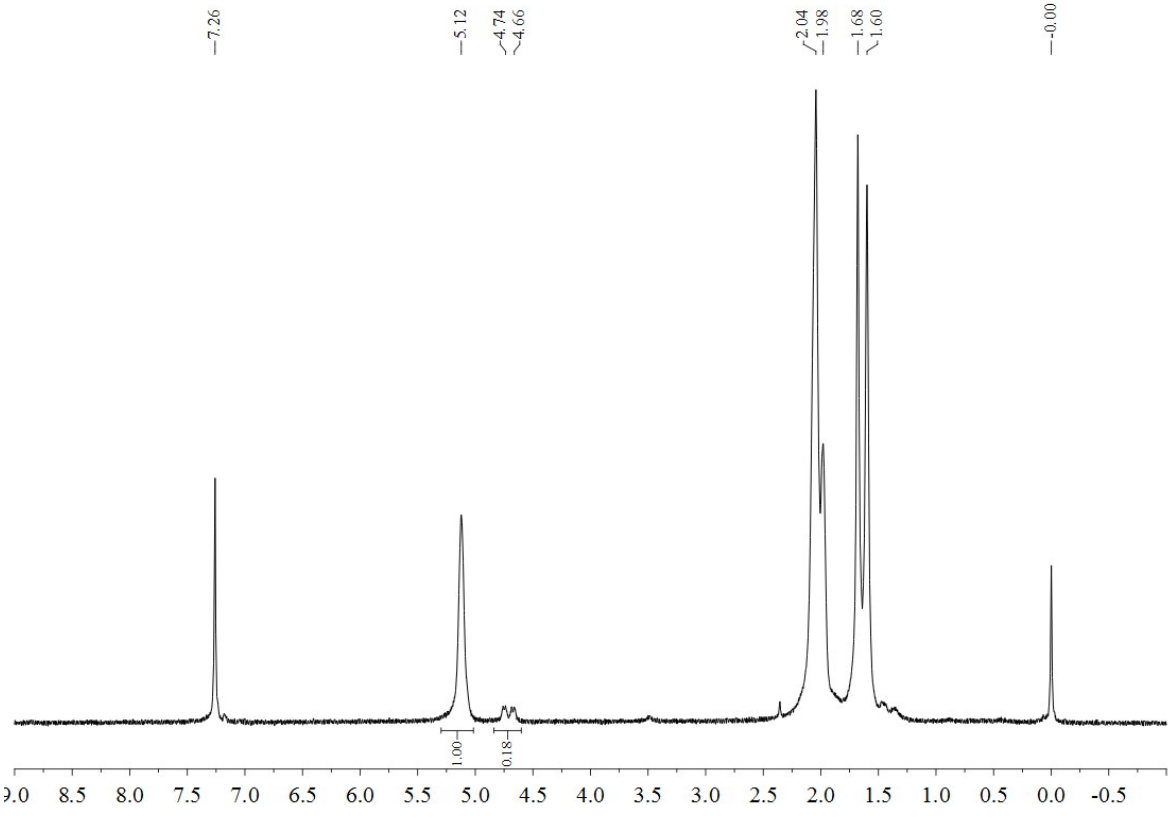

Figure S26. [1]H NMR Spectrum of Polyisoprene for Table 4, Entry 6

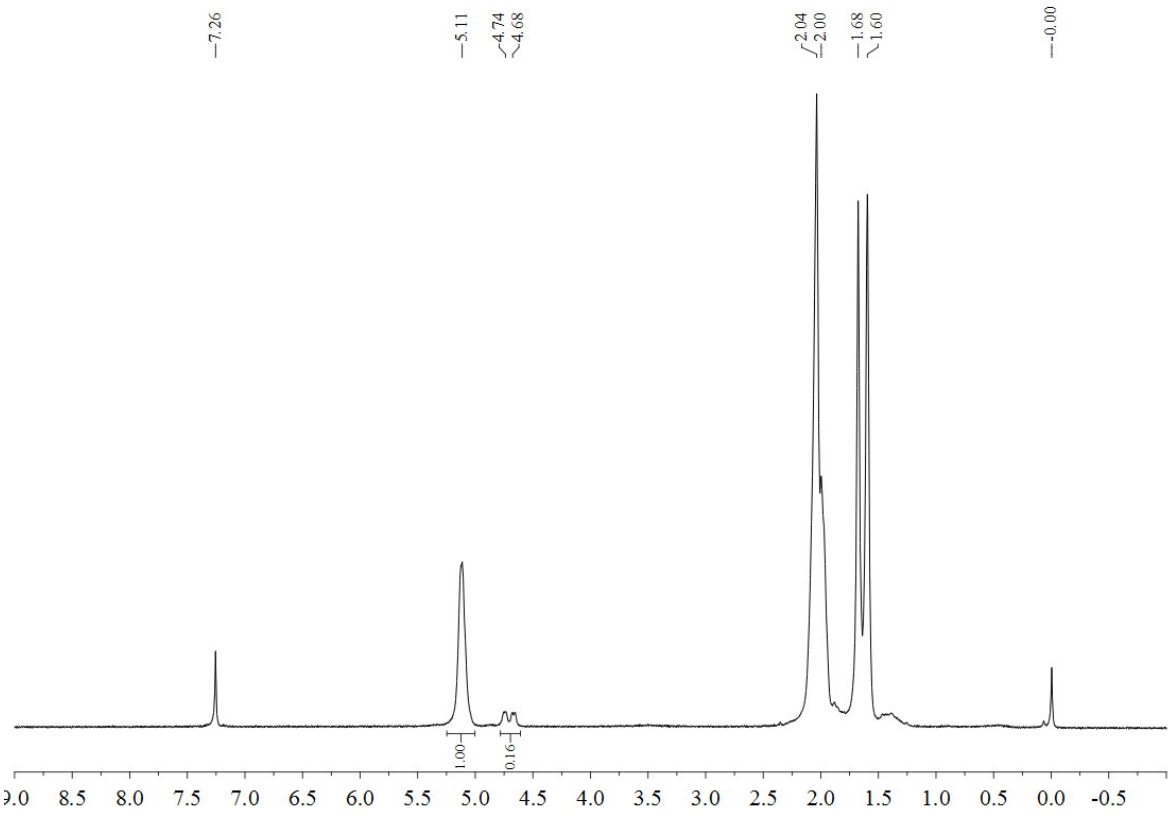

Figure S27. [1]H NMR Spectrum of Polyisoprene for Table 4, Entry 7

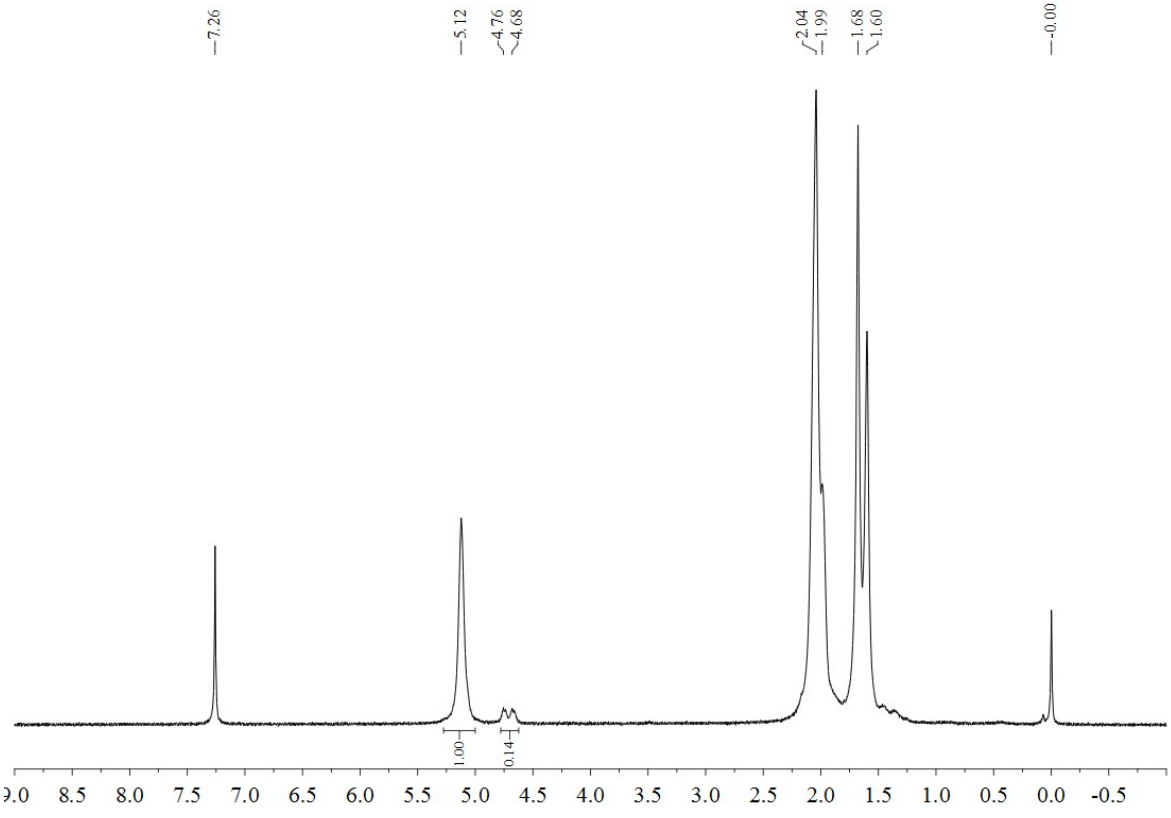

Figure S28. [1]H NMR Spectrum of Polyisoprene for Table 4, Entry 8

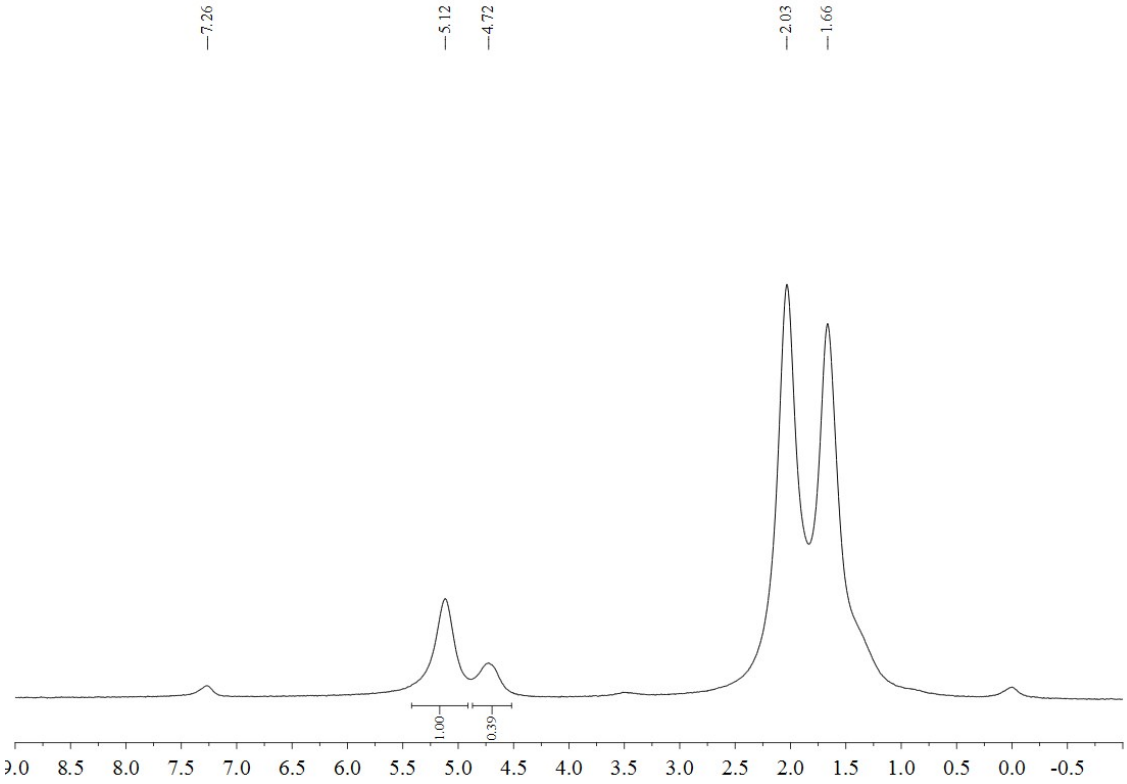

Figure S29. ^{1}H NMR Spectrum of Polyisoprene for Table 4, Entry 9

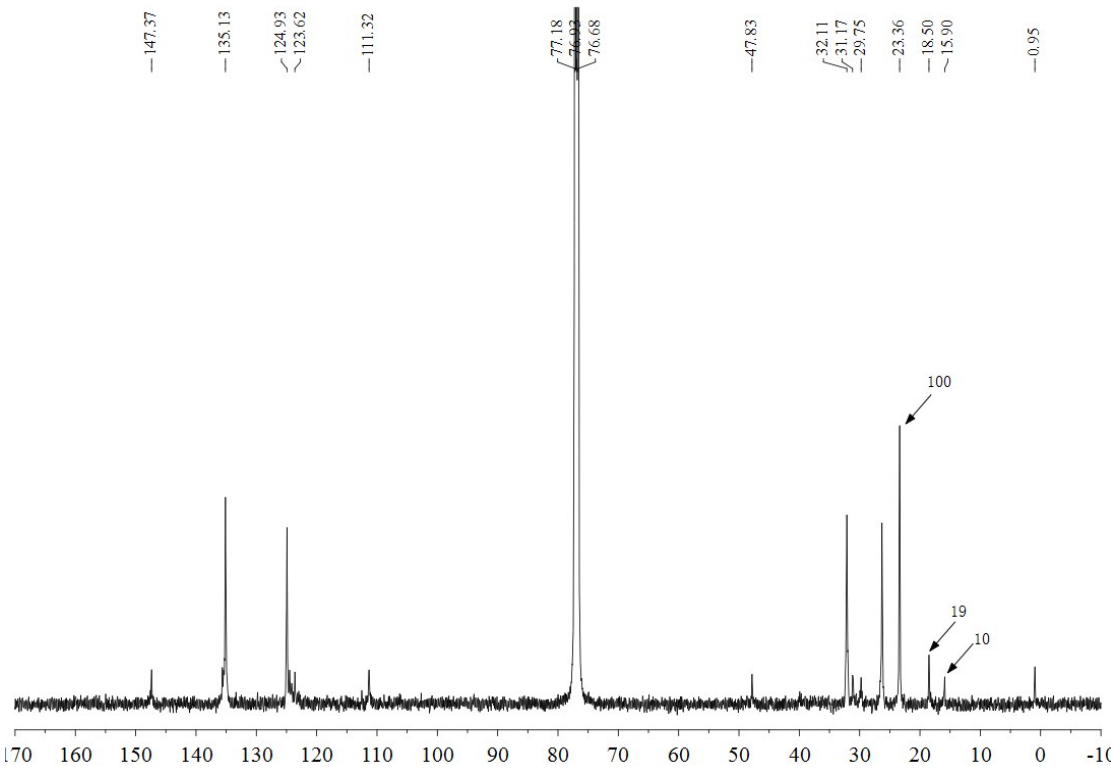

Figure S30. ^{13}C-int NMR Spectrum of Polyisoprene for Table 4, Entry 9

(100 for *cis*-1,4-PIP; 10 for *trans*-1,4-PIP; 19 for 3,4-PIP)

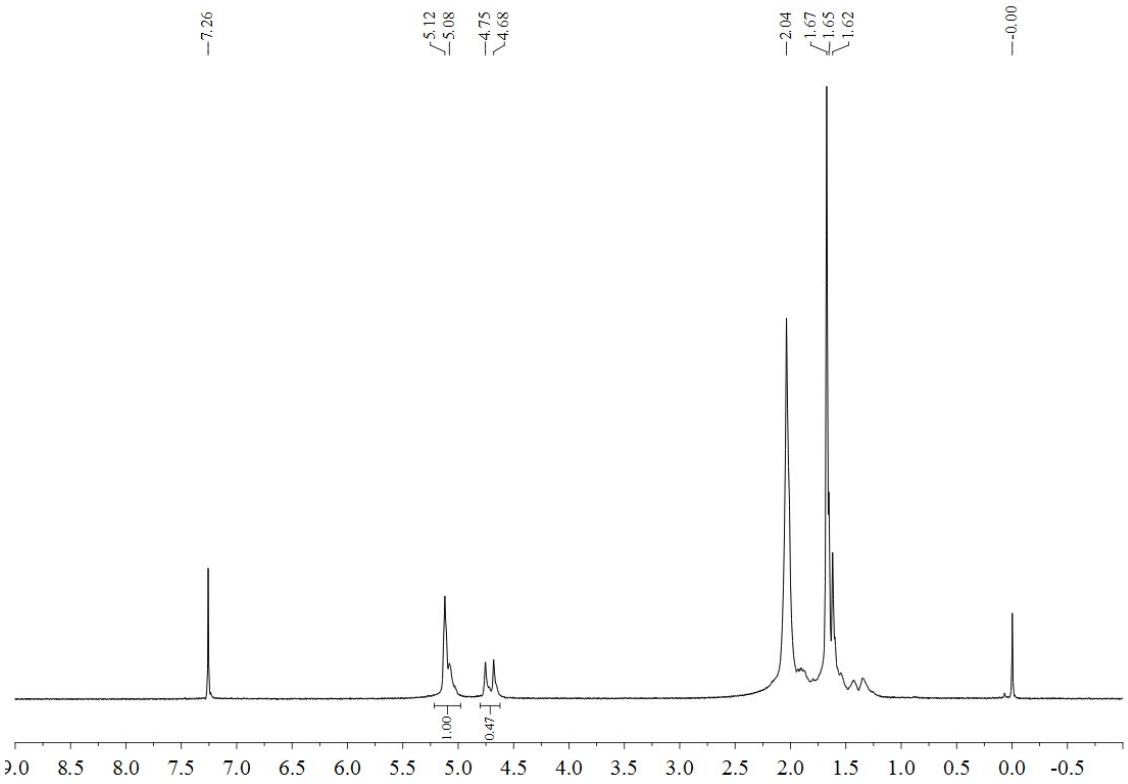

Figure S31. ^{1}H NMR Spectrum of Polyisoprene for Table 4, Entry 17

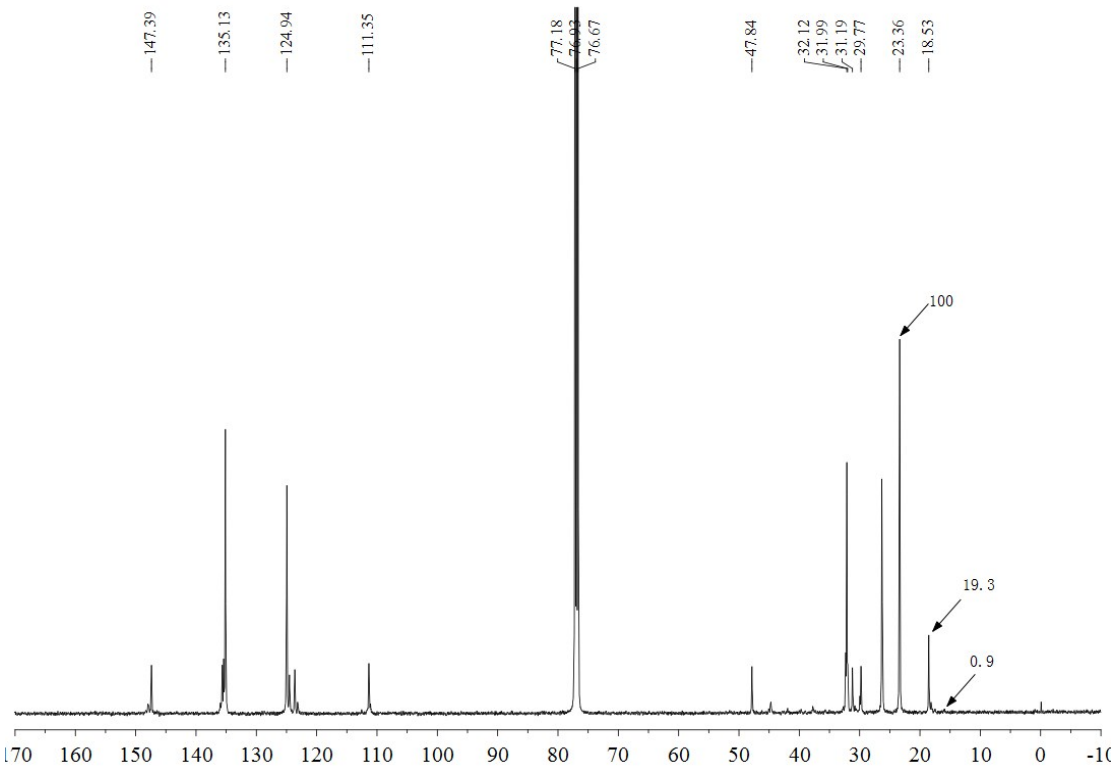

Figure S32. ^{13}C-int NMR Spectrum of Polyisoprene for Table 4, Entry 17

(100 for *cis*-1,4-PIP; 0.9 for *trans*-1,4-PIP; 19.3 for 3,4-PIP)

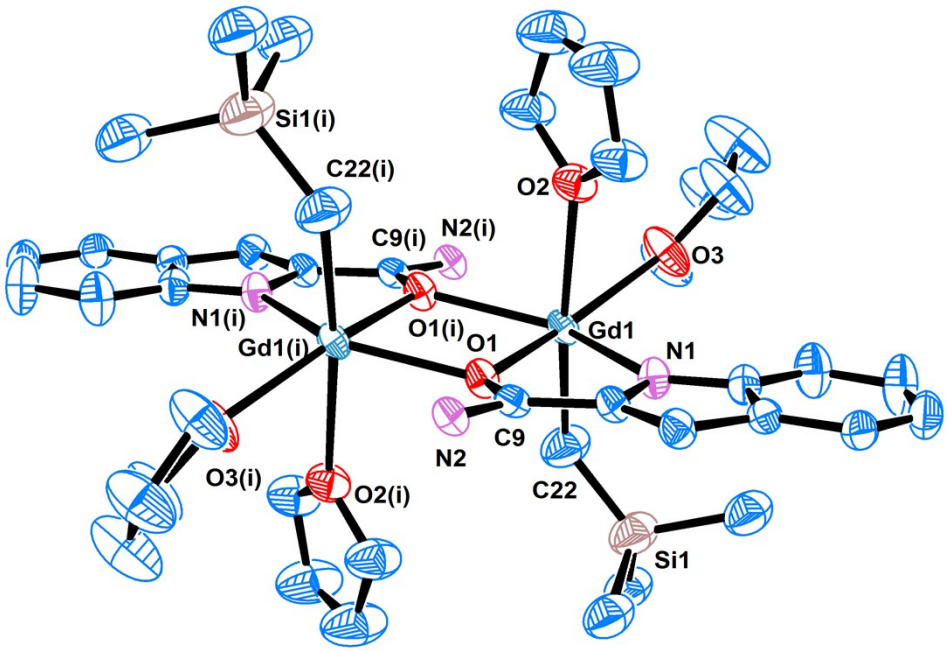

Figure S33. Single-crystal structure of the complex **1a** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Gd1-C22 2.416(7), Gd1-O1 2.298(3), Gd1-N1 2.410(4), O1-Gd1-N1 66.59(12).

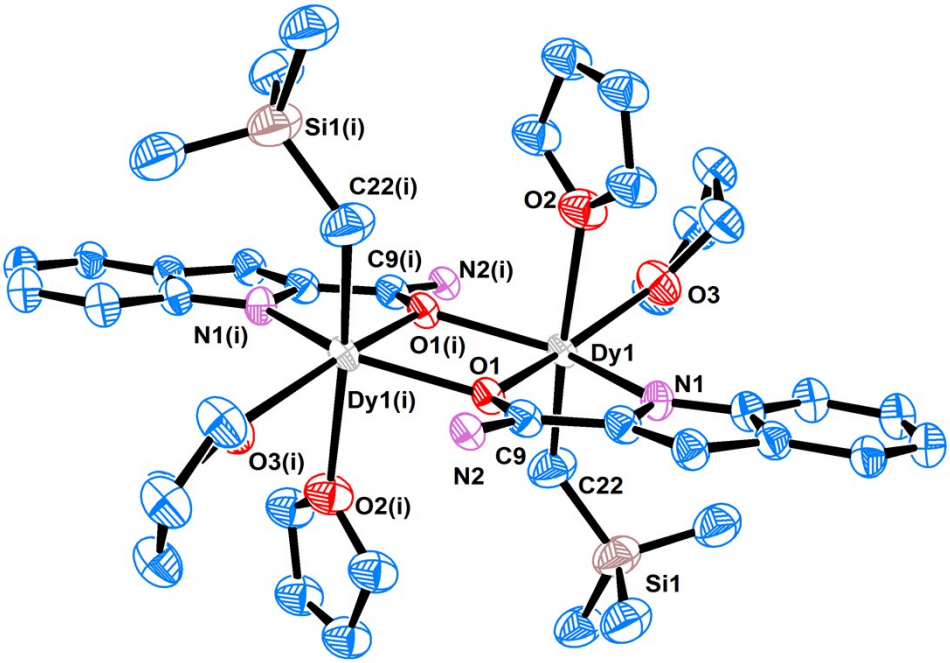

Figure S34. Single-crystal structure of the complex **1b** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Dy1-C22 2.388(8), Dy1-O1 2.284(3), Dy1-N1 2.371(4), O1-Dy1-N1 67.23(13).

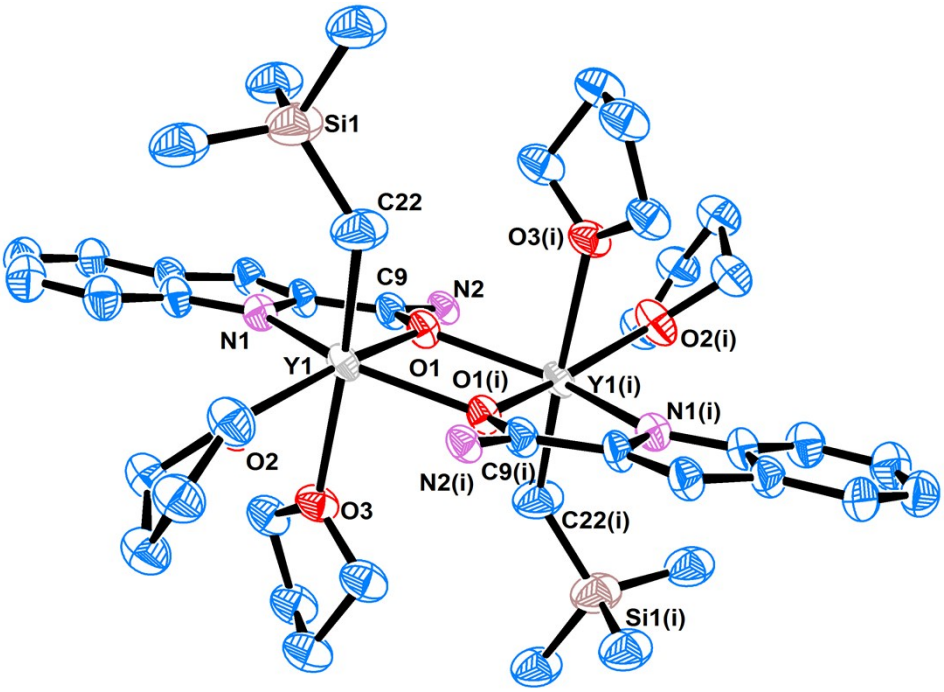

Figure S35. Single-crystal structure of the complex **1c** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Y1-C22 2.376(6), Y1-O1 2.269(4), Y1-N1 2.367(6), O1-Y1-N1 67.97(17).

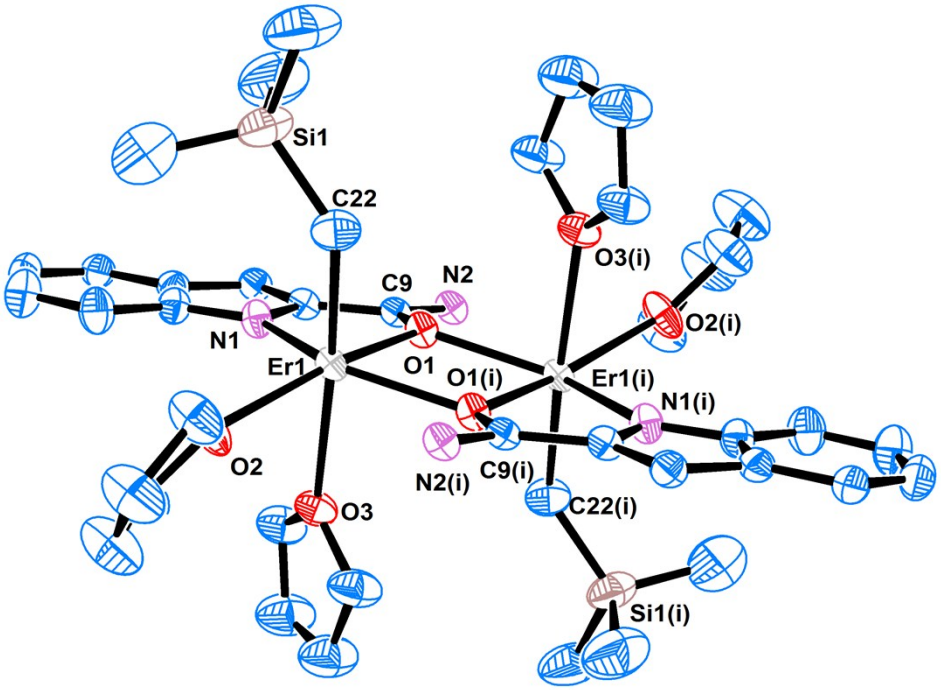

Figure S36. Single-crystal structure of the complex **1d** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Er1-C22 2.367(7), Er1-O1 2.259(3), Er1-N1 2.331(5), O1-Er1-N1 68.61(15).

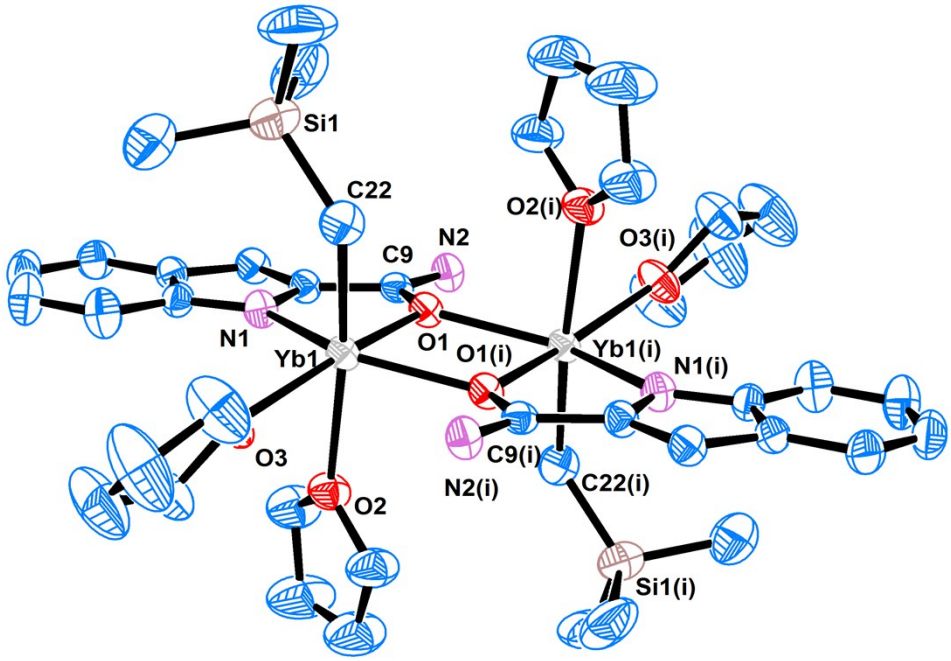

Figure S37. Single-crystal structure of the complex **1e** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-C22 2.326(5), Yb1-O1 2.237(2), Yb1-N1 2.315(3), O1-Yb1-N1 70.02(10).

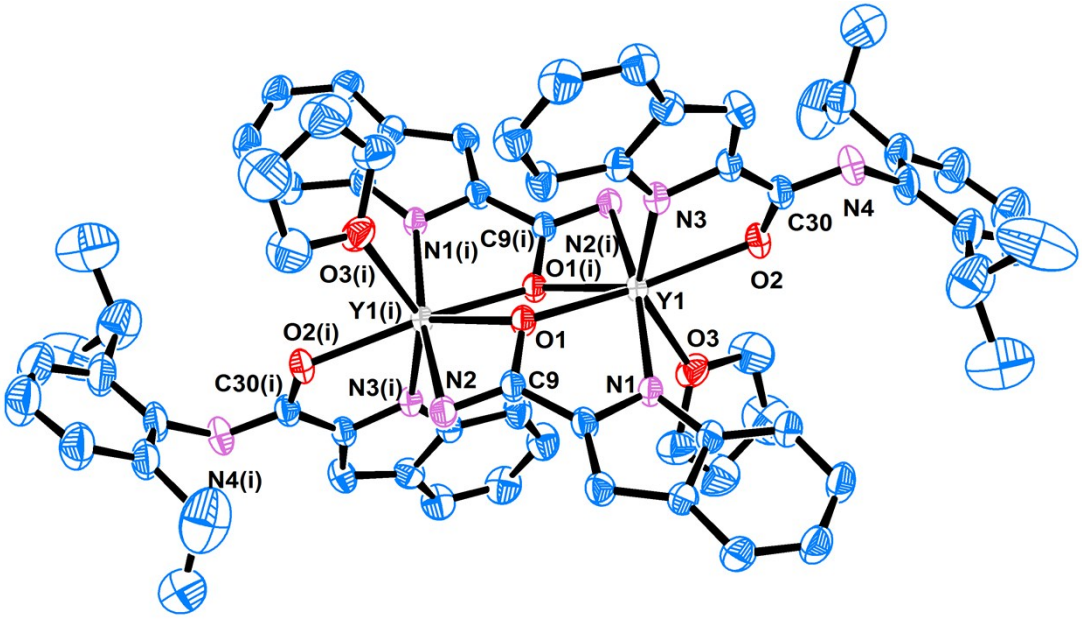

Figure S38. Single-crystal structure of the complex **1ca** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Y1-N1 2.345(3), Y1-O1 2.307(2), Y1-N3 2.340(3), Y1-O2 2.332(3), N1-Y1-O1 67.94(9), N3-Y1-O2 70.05(11).

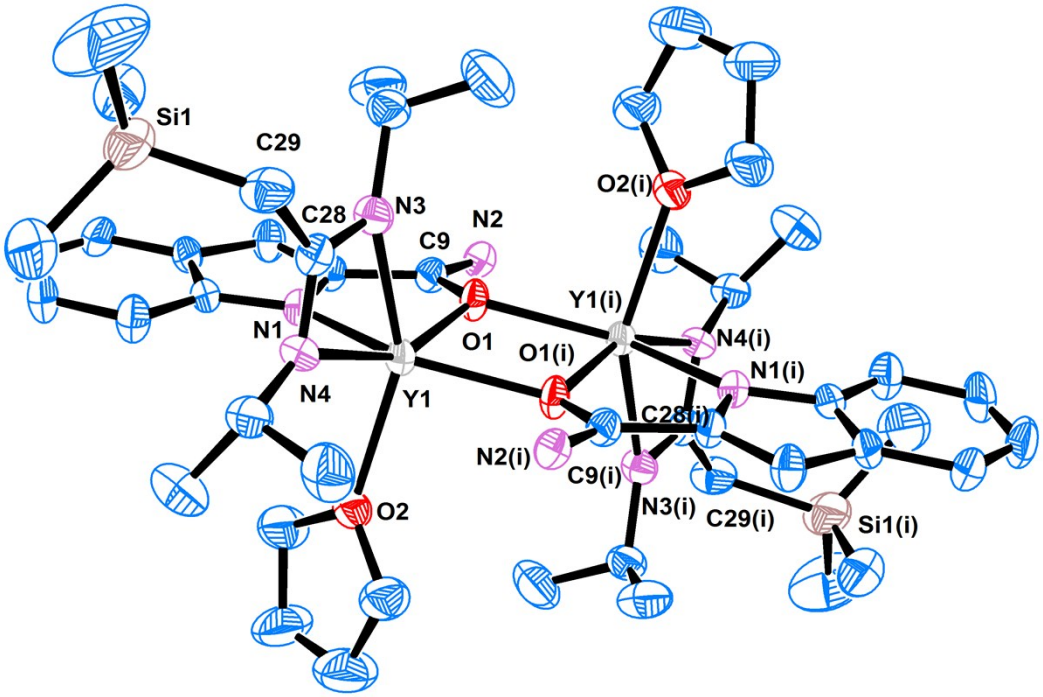

Figure S39. Single-crystal structure of the complex **1cb** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Y1-N1 2.340(4), Y1-O1 2.298(3), Y1-N3 2.293(4), Y1-N4 2.345(4), O1-Y1-N1 67.52(13), N3-Y1-N4 57.67(15).

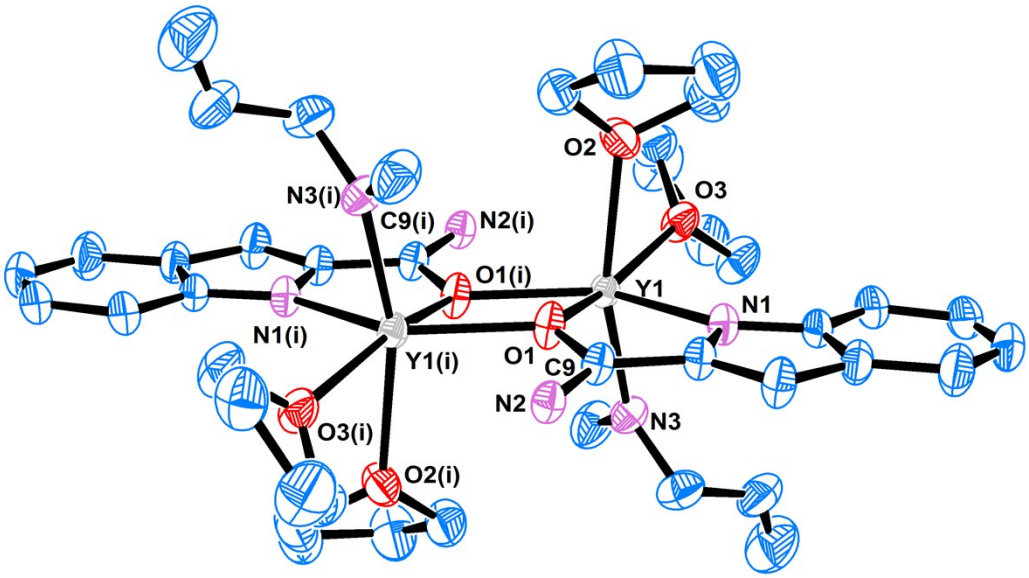

Figure S40. Single-crystal structure of the complex **1cc** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Y1-N1 2.396(2), Y1-O1 2.2874(17), Y1-N3 2.171(2), N1-Y1-O1 67.07(7).

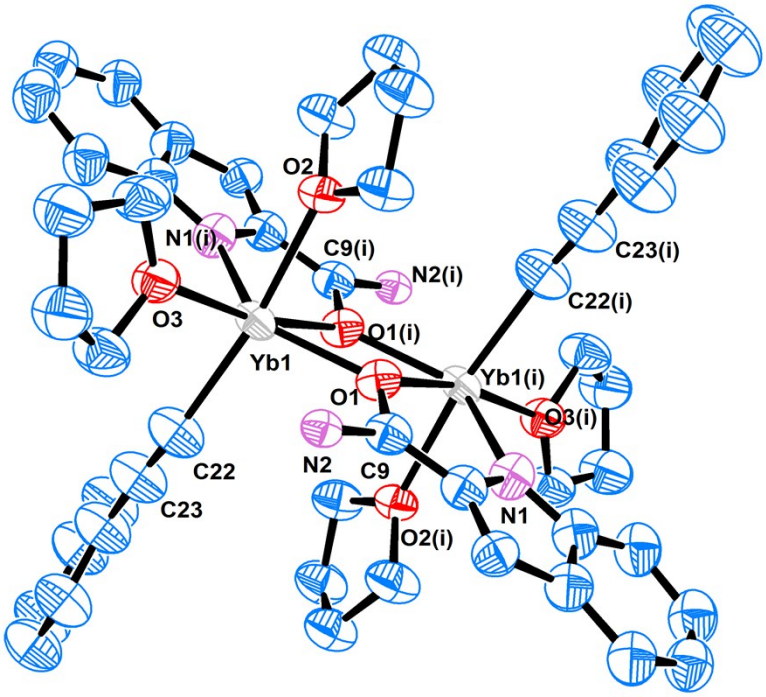

Figure S41. Single-crystal structure of the complex **1ea** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1(i) 2.326(9), Yb1-O1(i) 2.251(6), Yb1-C22 2.314(12), C22-C23 1.192(14), O1(i)-Yb1-N1(i) 68.9(3).

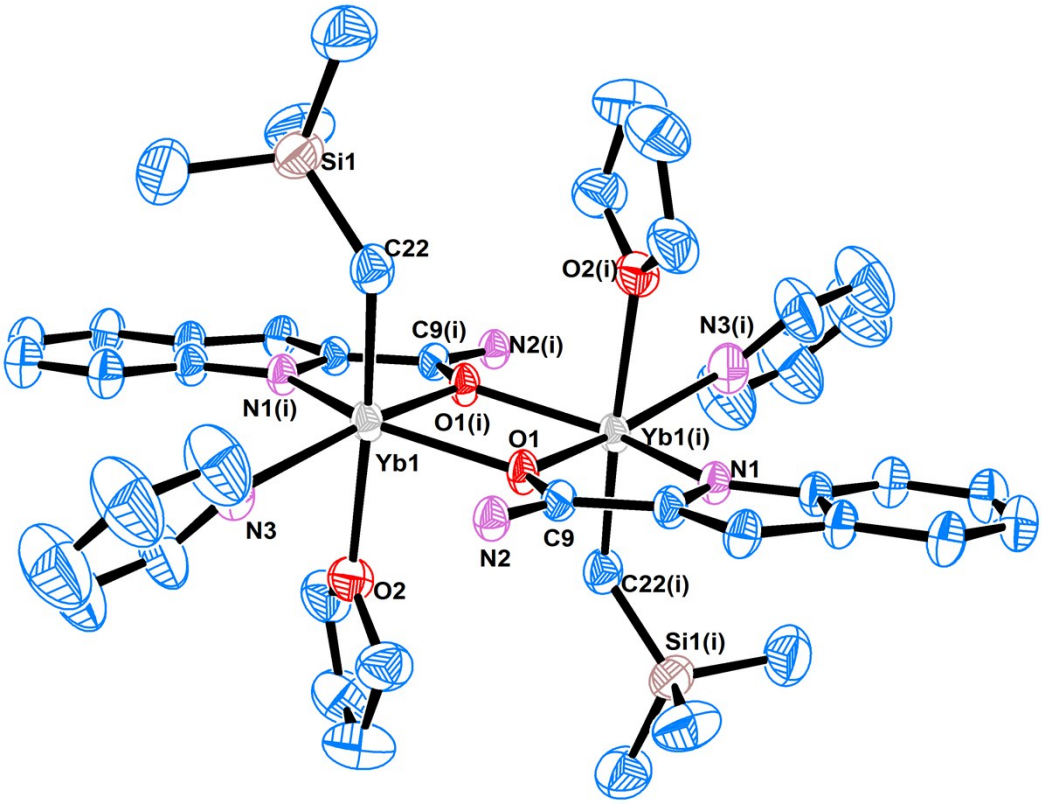

Figure S42. Single-crystal structure of the complex **1eb** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1(i) 2.310(3), Yb1-O1(i) 2.236(3), Yb1-N3 2.394(3), N1(i)-Yb1-O1(i) 69.75(11).

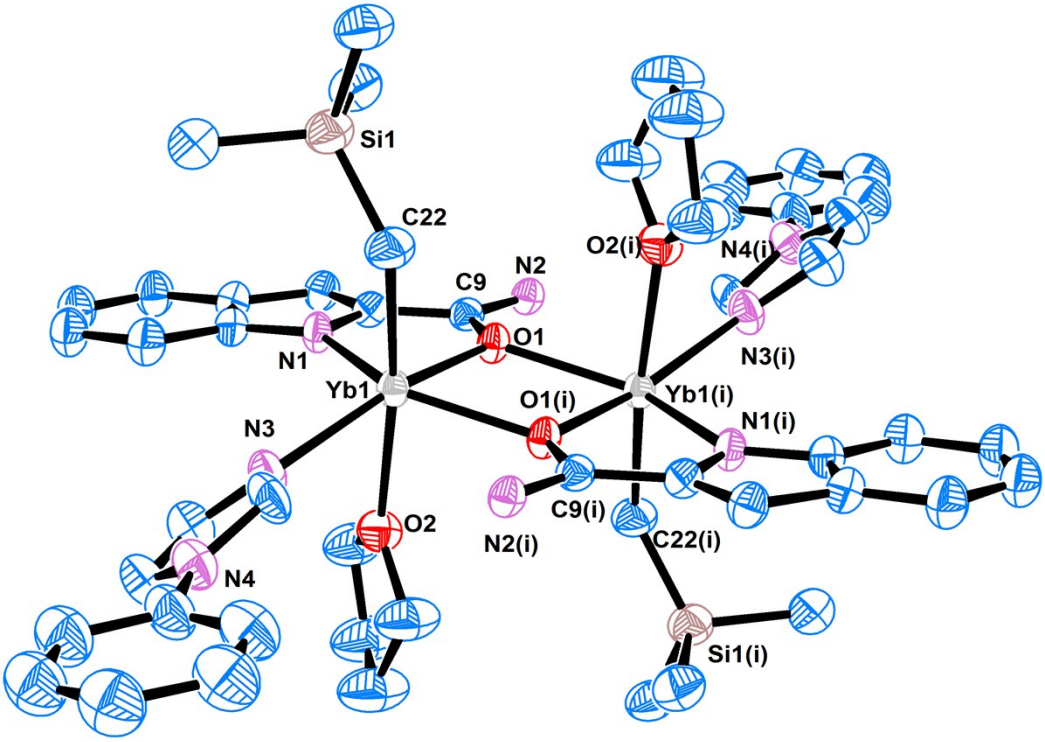

Figure S43. Single-crystal structure of the complex **1ec** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1 2.310(5), Yb1-O1 2.241(4), Yb1-N3 2.419(6), O1-Yb1-N1 69.66(16).

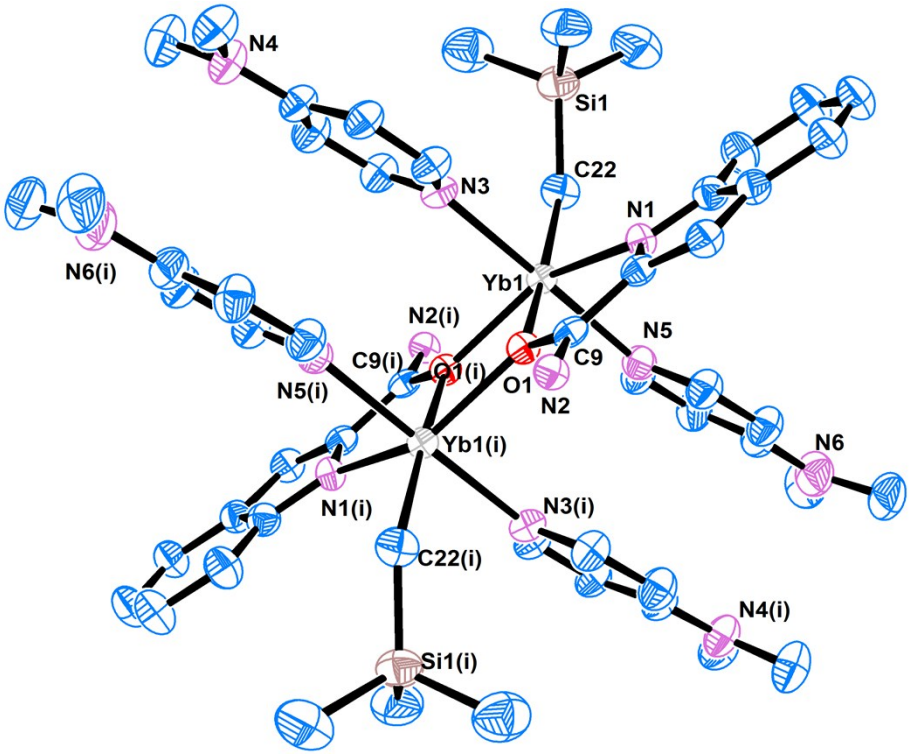

Figure S44. Single-crystal structure of the complex **1ed** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1 2.307(4), Yb1-O1 2.339(4), Yb1-N3 2.405(5), Yb1-N5 2.392(5), Yb1-C22 2.362(7), O1-Yb1-N1 68.66(14).

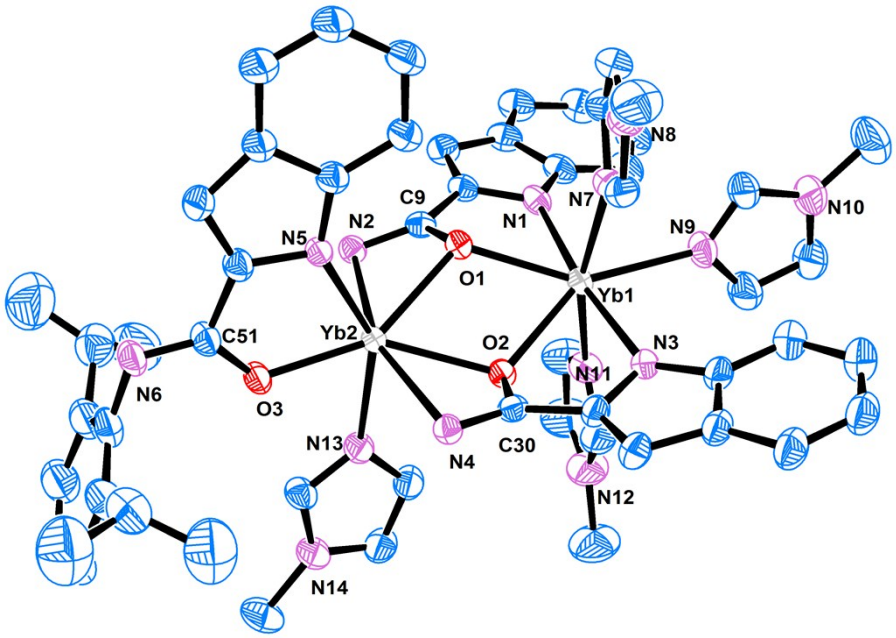

Figure S45. Single-crystal structure of the complex **1ee** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N(2) and N(4) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1 2.413(4), Yb1-O1 2.281(3), Yb1-N3 2.416(4), Yb1-O2 2.266(3), Yb1-N7 2.390(4), Yb1-N9 2.424(4), Yb1-N11 2.392(4), Yb2-N2 2.452(4), Yb2-O1 2.345(3), Yb2-N4 2.475(4), Yb2-O2 2.339(3), Yb2-N5 2.312(4), Yb2-O3 2.156(3), Yb2-N13 2.408(4), N1-Yb1-O1 68.13(12), N3-Yb1-O2 67.88(12), N5-Yb2-O3 72.35(12).

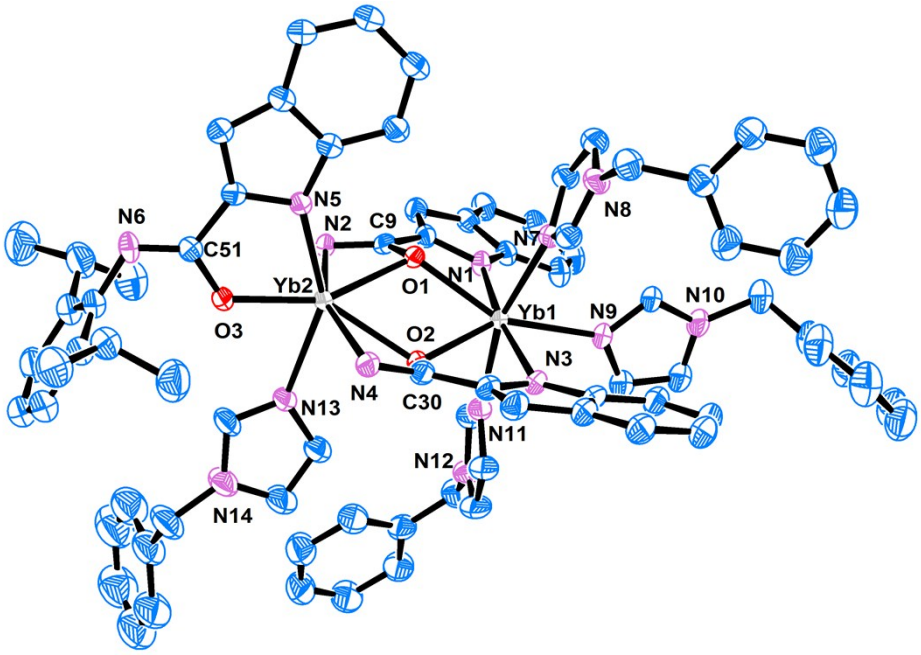

Figure S46. Single-crystal structure of the complex **1ef** with thermal ellipsoids at the 30% level, all hydrogen atoms and Dipp on N(2) and N(4) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1 2.396(4), Yb1-O1 2.283(3), Yb1-N3 2.424(4), Yb1-O2 2.263(3), Yb1-N7 2.389(4), Yb1-N9 2.429(4), Yb1-N11 2.392(4), Yb2-N2 2.435(4), Yb2-O1 2.360(3), Yb2-N4 2.466(4), Yb2-O2 2.368(3), Yb2-N5 2.307(4), Yb2-O3 2.139(3), Yb2-N13 2.415(4), N1-Yb1-O1 68.68(12), N3-Yb1-O2 67.74(13), N5-Yb2-O3 72.30(13).

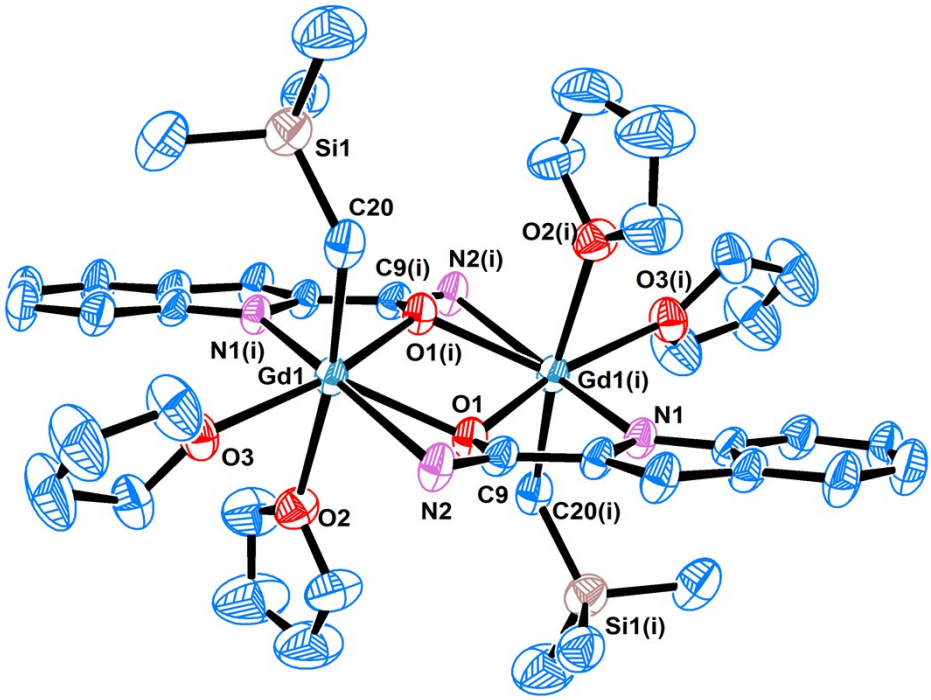

Figure S47. Single-crystal structure of the complex **2a** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Gd1-C20 2.424(4), Gd1-O1(i) 2.324(2), Gd1-N1(i) 2.398(3), O1(i)-Gd1-N1(i) 66.39(9).

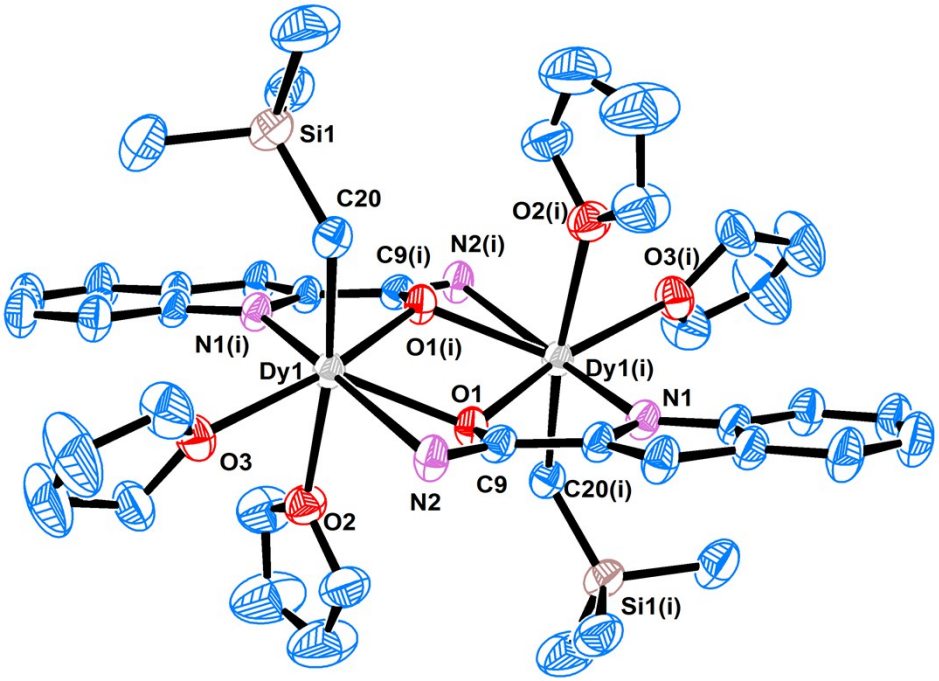

Figure S48. Single-crystal structure of the complex **2b** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Dy1-C20 2.413(3), Dy1-O1(i) 2.2973(18), Dy1-N1(i) 2.364(2), O1(i)-Dy1-N1(i) 67.07(7).

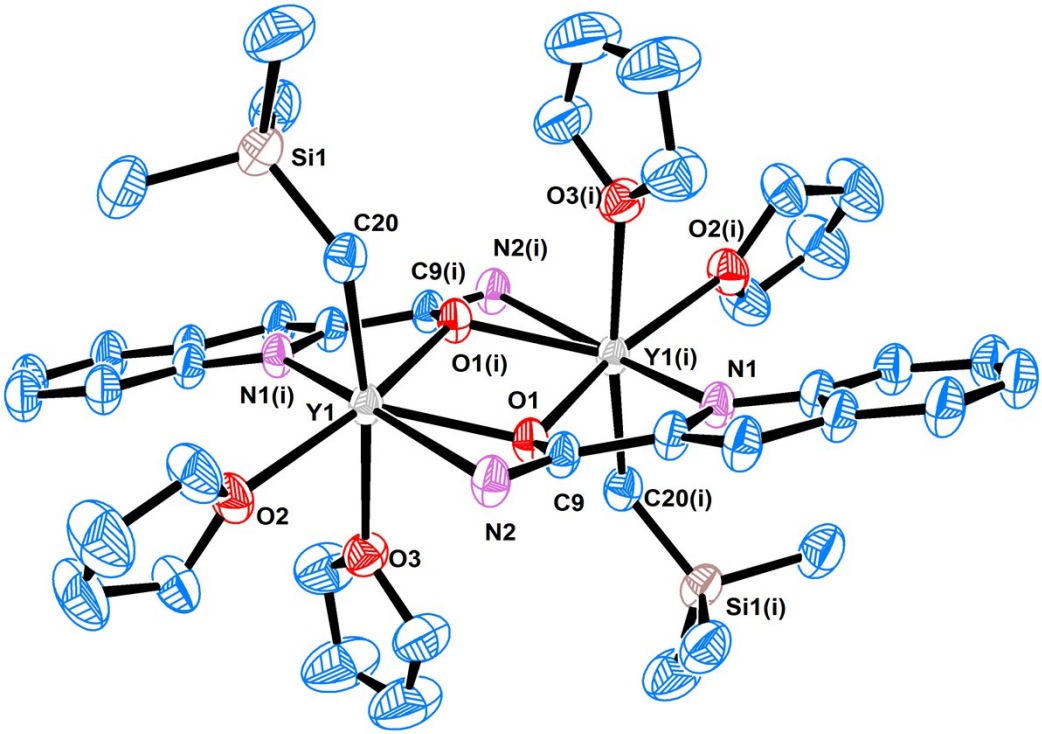

Figure S49. Single-crystal structure of the complex **2c** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Y1-C20 2.412(3), Y1-O1(i) 2.2785(18), Y1-N1(i) 2.367(2), O1(i)-Y1-N1(i) 67.43(7).

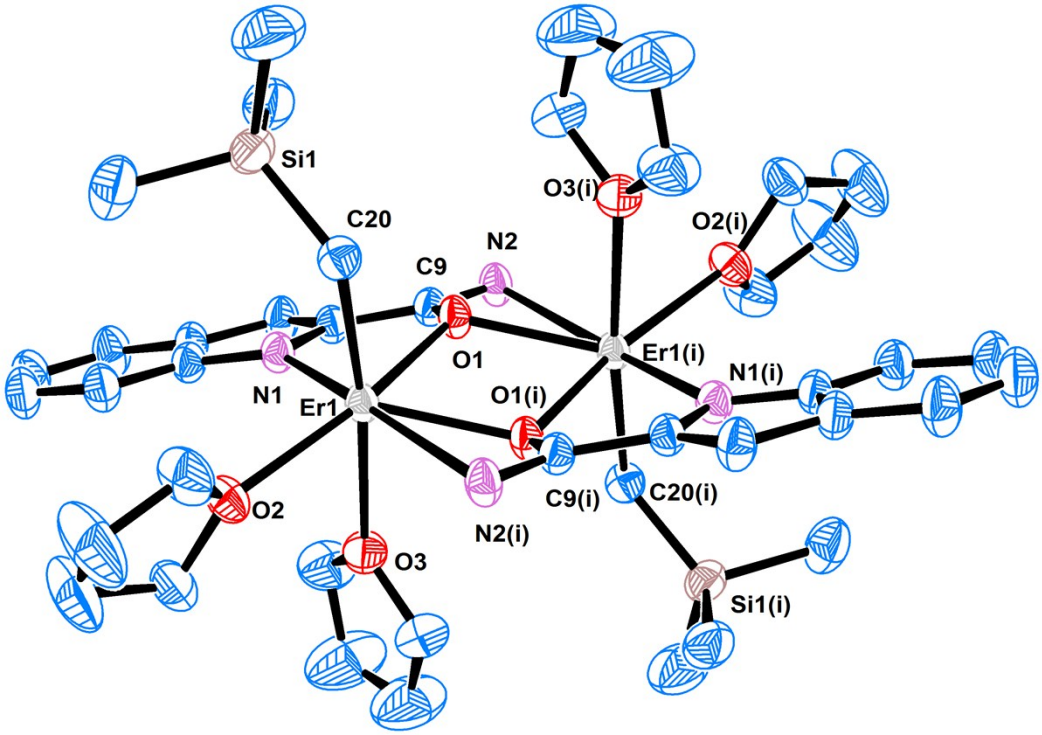

Figure S50. Single-crystal structure of the complex **2d** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Er1-C20 2.395(4), Er1-O1 2.2708(19), Er1-N1 2.341(3), O1-Er1-N1 67.75(8).

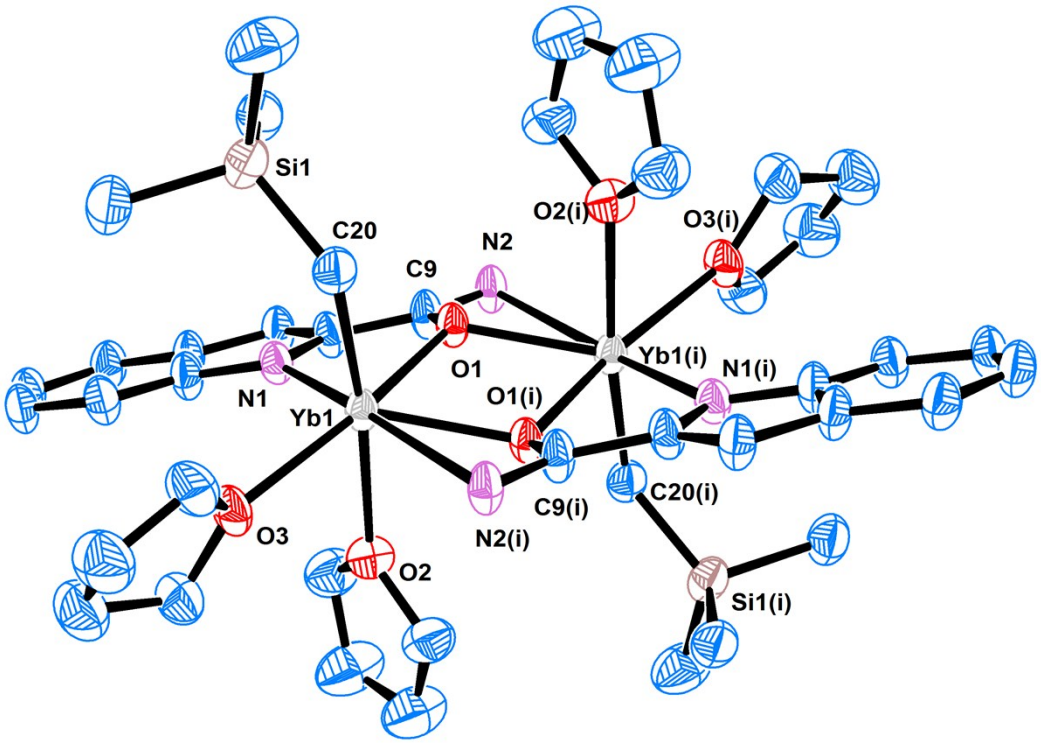

Figure S51. Single-crystal structure of the complex **2e** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-C20 2.346(9), Yb1-O1 2.254(4), Yb1-N1 2.304(6), O1-Yb1-N1 68.3(2).

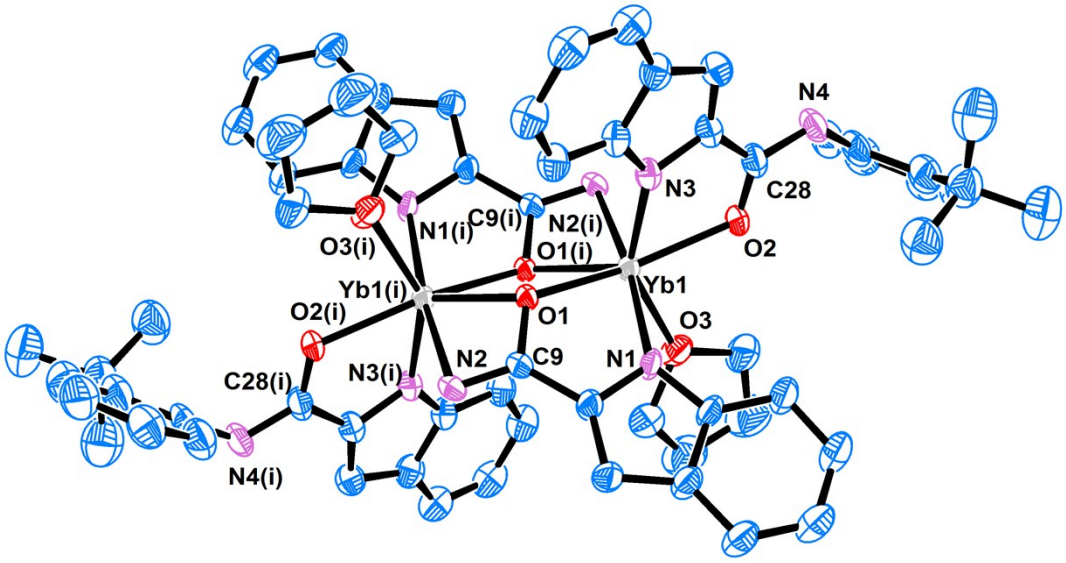

Figure S52. Single-crystal structure of the complex **2ea** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1 2.319(6), Yb1-O1 2.268(4), Yb1-N3 2.307(6), Yb1-O2 2.247(4), Yb1-N2(i) 2.534(6), Yb1-O1(i) 2.328(4), N1-Yb1-O1 68.38(16), N3-Yb1-O2 71.60(19), N2(i)-Yb1-O1(i) 53.27(16).

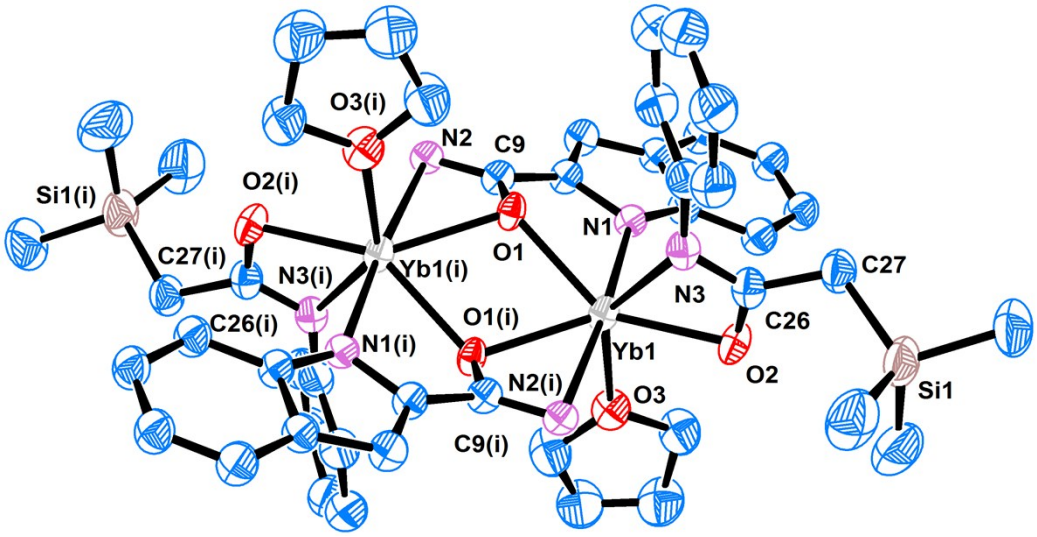

Figure S53 Single-crystal structure of the complex **2eb** with thermal ellipsoids at the 30% level, all hydrogen atoms and *o*-^{t}BuC$_6$H$_5$ on N2 and N2(i) are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-N1 2.333(6), Yb1-O1 2.293(5), Yb1-N2(i) 2.462(6), Yb1-O1(i) 2.321(4), Yb1-N3 2.316(6), Yb1-O2 2.238(5), N1-Yb1-O1 69.71(19), N2(i)-Yb1-O1(i) 55.38(18), N3-Yb1-O2 57.4(2).

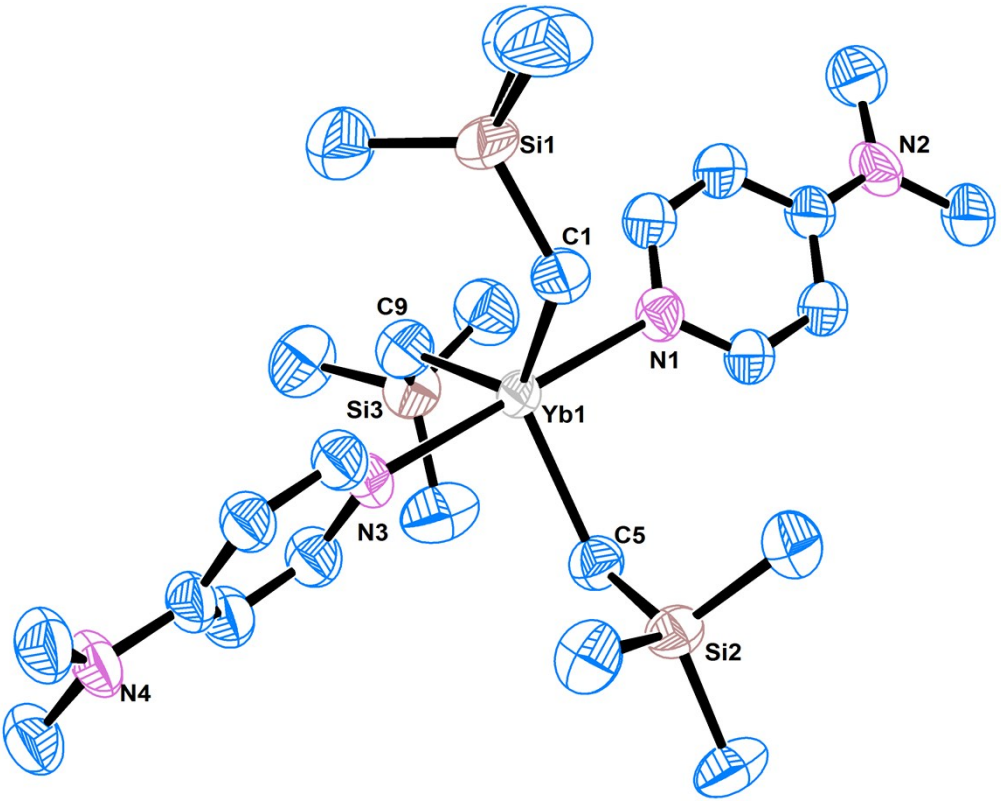

Figure S54 Single-crystal structure of the complex **3a** with thermal ellipsoids at the 30% level, all hydrogen atoms are omitted for clarity. Selected bond lengths (Å) and angles (°): Yb1-C1 2.366(13), Yb1-C5 2.353(12), Yb1-C9 2.306(15), Yb1-N1 2.429(10), Yb1-N3 2.444(5), C1-Yb1-C5 120.2(5), C1-Yb1-C9 125.6(5), C5-Yb1-C9 117.2(5), N1-Yb1-C1 88.2(4), N1-Yb1-C5 92.3(4), N1-Yb1-C9 91.7(4), N3-Yb1-C1 90.9(4), N3-Yb1-C5 87.5(4), N3-Yb1-C9 89.5(4).

Table S1. Crystal data and structure refinement for **1a-1c**

	1a	**1b**	**1c**
Formula	$C_{66}H_{98}N_4O_6Si_2Gd_2$	$C_{66}H_{98}N_4O_6Si_2Dy_2$	$C_{66}H_{98}N_4O_6Si_2Y_2$
FW	1414.16	1424.66	1227.48
T(K)	293(2)	293(2)	293(2)
λ(Å)	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/n$	$P2_1/n$	$P2_1/c$
a(Å)	11.9048(10)	11.8533(6)	11.8210(19)
b(Å)	17.2036(15)	17.2333(9)	17.250(3)
c(Å)	17.5111(15)	17.5236(9)	19.936(3)
α(deg)	90	90	90
β(deg)	97.0820(10)	97.1180(10)	118.885(7)
γ(deg)	90	90	90
v(Å^3)	3559.0(5)	3552.0(3)	3559.4(10)
Z	2	2	2
D_{calcd}(mg/m^3)	1.320	1.332	1.192
μ(mm^{-1})	1.928	2.169	1.703
F(000)	1452	1460	1352
θ range(deg)	1.666-27.585	1.664-27.547	1.660-24.998
Reflections collected/unique	39748/8153	39805/8098	25189/6260
R(int)	0.1049	0.0340	0.1247
Goodness-of-fit on F^2	1.036	1.067	1.016
R_1,wR_2 [$I>2\sigma(I)$]	0.0435, 0.0919	0.0483, 0.1264	0.0686, 0.1436
R_1,wR_2(all data)	0.0719, 0.1173	0.0640, 0.1345	0.1745, 0.1938
Largest diff. peak and hole(e. Å^{-3})	1.498 and -0.827	1.770 and -1.932	0.696 and -0.479

Table S2. Crystal data and structure refinement for **1d-1e**

	1d	**1e**
Formula	$C_{66}H_{98}N_4O_6Si_2Er_2$	$C_{66}H_{98}N_4O_6Si_2Yb_2$
FW	1434.18	1445.74
T(K)	293(2)	293(2)
λ(Å)	0.71073	0.71073
Crystal system	Monoclinic	Triclinic
Space group	$P2_1/n$	$P\bar{1}$
a(Å)	11.8031(10)	11.7367(7)
b(Å)	17.2359(15)	18.4753(11)
c(Å)	17.5507(15)	18.9841(12)
α(deg)	90	87.5120(10)
β(deg)	97.1660(10)	77.8380(10)
γ(deg)	90	75.5950(10)
v(Å^3)	3542.6(5)	3897.4(4)
Z	2	2
D_{calcd}(mg/m^3)	1.345	1.232
μ(mm^{-1})	2.434	2.459
F(000)	1468	1476
θ range(deg)	1.662-27.551	2.195-27.526
Reflections collected/unique	39319/8081	44382/17355
R(int)	0.1276	0.0404
Goodness-of-fit on F^2	1.064	0.822
R_1,wR_2 [$I>2\sigma(I)$]	0.0504, 0.1241	0.0355, 0.0728
R_1,wR_2(all data)	0.0740, 0.1363	0.0692, 0.0808
Largest diff. peak and hole(e. Å^{-3})	1. 984 and -1.138	0.958 and -0.527

Table S3. Crystal data and structure refinement for **1ca-1cc**

	1ca	**1cb**	**1cc**
Formula	$C_{92}H_{106}N_8O_6Y_2$	$C_{72}H_{110}N_8O_4Si_2Y_2$	$C_{66}H_{92}N_6O_6Y_2$
FW	1597.66	1385.67	1243.27
T(K)	293(2)	293(2)	293(2)
λ(Å)	0.71073	0.71073	0.71073
Crystal system	Triclinic	Monoclinic	Monoclinic
Space group	$P\bar{1}$	$P2_1/c$	$P2_1/n$
a(Å)	11.8363(18)	15.081(2)	11.4194(11)
b(Å)	14.411(2)	17.316(3)	15.3671(15)
c(Å)	17.416(4)	17.270(3)	18.6412(18)
α(deg)	108.535(2)	90	90
β(deg)	95.671(2)	103.702(2)	90.1120(10)
γ(deg)	110.241(2)	90	90
v(Å^3)	2568.8(8)	4381.6(12)	3271.2(5)
Z	1	2	2
D_{calcd}(mg/m^3)	1.033	1.050	1.262
μ(mm^{-1})	1.171	1.388	1.818
F(000)	840	1472	1312
θ range(deg)	1.269-27.584	2.364-24.998	1.717-25.450
Reflections collected/unique	28955/11479	41329/7689	32723/6048
R(int)	0.1112	0.1651	0.0447
Goodness-of-fit on F^2	0.980	0.832	1.008
R_1,wR_2 [$I>2\sigma(I)$]	0.0720, 0.1888	0.0615, 0.1152	0.0360, 0.0857
R_1,wR_2(all data)	0.1063, 0.2067	0.1529, 0.1361	0.0578, 0.0953
Largest diff. peak and hole(e. Å^{-3})	1.425 and -0.793	0.444 and -0.259	0.391 and -0.260

Table S4. Crystal data and structure refinement for **1ec-1ea**

	1ea	**1eb**	**1ec**
Formula	$C_{74}H_{86}N_4O_6Yb_2$	$C_{68}H_{92}N_6O_4Si_2Yb_2$	$C_{76}H_{98}N_8O_4Si_2Yb_2$
FW	1473.54	1459.73	1589.88
T(K)	293(2)	293(2)	293(2)
λ(Å)	0.71073	0.71073	0.71073
Crystal system	Triclinic	Monoclinic	Triclinic
Space group	$P\bar{1}$	$C2/c$	$P\bar{1}$
a(Å)	12.8086(16)	27.113(2)	13.0440(10)
b(Å)	16.837(2)	14.1425(11)	13.8601(10)
c(Å)	20.863(3)	24.319(3)	14.8195(11)
α(deg)	79.527(2)	90	97.3780(10)
β(deg)	76.232(2)	116.5790(10)	113.2400(10)
γ(deg)	89.163(2)	90	114.7870(10)
v(Å^3)	4295.1(9)	8839.7(14)	2091.8(3)
Z	2	4	1
D_{calcd}(mg/m^3)	1.139	1.163	1.262
μ(mm^{-1})	2.206	2.298	2.297
F(000)	1492	2968	810
θ range(deg)	1.230-25.409	1.667-25.400	1.598-25.324
Reflections collected/unique	43638/15663	41364/7672	21203/7595
R(int)	0.0788	0.0408	0.0772
Goodness-of-fit on F^2	0.872	1.012	0.937
R_1,wR_2 [$I>2\sigma(I)$]	0.0629, 0.1457	0.0342, 0.0848	0.0531, 0.0976
R_1,wR_2(all data)	0.1222, 0.1646	0.0511, 0.0929	0.0892, 0.1076
Largest diff. peak and hole(e. Å^{-3})	1.014 and -0.810	0.937 and -0.758	0.817 and -1.003

Table S5. Crystal data and structure refinement for **3a** and **1ed**

	3a	**1ed**
Formula	$C_{26}H_{53}N_4Si_3Yb$	$C_{78}H_{106}N_{12}O_2Si_2Yb_2$
FW	679.03	1646.00
T(K)	293(2)	293(2)
λ(Å)	0.71073	0.71073
Crystal system	Triclinic	Triclinic
Space group	*P*$\bar{1}$	*P*$\bar{1}$
a(Å)	10.477(3)	12.1857(9)
b(Å)	12.806(4)	14.6994(10)
c(Å)	13.939(4)	15.6856(11)
α(deg)	93.672(3)	103.6230(10)
β(deg)	105.014(3)	112.0460(10)
γ(deg)	92.303(3)	97.2230(10)
v(Å^3)	1799.5(9)	2458.4(3)
Z	2	1
D_{calcd}(mg/m^3)	1.253	1.112
μ(mm^{-1})	2.716	1.956
F(000)	698	842
θ range(deg)	1.517-25.497	1.472-25.384
Reflections collected/unique	18060/6652	25275/8980
R(int)	0.1655	0.1287
Goodness-of-fit on F^2	1.009	0.959
R_1,wR_2 [$I>2\sigma(I)$]	0.0805, 0.1831	0.0517, 0.1131
R_1,wR_2(all data)	0.1294, 0.2082	0.0718, 0.1204
Largest diff. peak and hole(e. Å^{-3})	1.633 and -1.286	0.995 and -1.006

Table S6. Crystal data and structure refinement for **1ee-1ef**

	1ee	**1ef**
Formula	$C_{79}H_{90}N_{14}O_3Yb_2$	$C_{103}H_{106}N_{14}O_3Yb_2$
FW	1629.72	1934.09
T(K)	293(2)	293(2)
λ(Å)	0.71073	0.71073
Crystal system	Monoclinic	Triclinic
Space group	$P2_1/c$	$P\bar{1}$
a(Å)	14.5555(10)	14.7004(8)
b(Å)	30.698(2)	14.9519(9)
c(Å)	20.3236(14)	23.7312(14)
α(deg)	90	101.4710(10)
β(deg)	92.8790(10)	96.2690(10)
γ(deg)	90	93.2780(10)
v(Å^3)	9069.5(11)	5064.6(5)
Z	4	2
D_{calcd}(mg/m^3)	1.194	1.268
μ(mm^{-1})	2.097	1.888
F(000)	3304	1972
θ range(deg)	1.203-25.399	1.394-25.372
Reflections collected/unique	92129/16660	52092/18488
R(int)	0.0868	0.0916
Goodness-of-fit on F^2	1.026	0.984
R_1,wR_2 [$I>2\sigma(I)$]	0.0417, 0.0965	0.0461, 0.1099
R_1,wR_2(all data)	0.0515, 0.1019	0.0678, 0.1187
Largest diff. peak and hole(e. Å^{-3})	1.385 and -0.904	1.562 and -0.925

Table S7. Crystal data and structure refinement for **2a-2c**

	2a	**2b**	**2c**
Formula	$C_{62}H_{90}N_4O_6Si_2Gd_2$	$C_{62}H_{90}N_4O_6Si_2Dy_2$	$C_{62}H_{90}N_4O_6Si_2Y_2$
FW	1358.05	1368.55	1221.37
T(K)	293(2)	293(2)	293(2)
λ(Å)	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	*C*2/*c*	*C*2/*c*	*C*2/*c*
a(Å)	30.2539(18)	30.336(3)	30.302(4)
b(Å)	14.9846(9)	14.9931(15)	14.9856(15)
c(Å)	22.6708(13)	22.644(2)	22.640(3)
α(deg)	90	90	90
β(deg)	117.5380(10)	118.1020(10)	118.104(2)
γ(deg)	90	90	90
v(Å^3)	9113.2(9)	9085.0(16)	9068.6(17)
Z	4	4	4
D_{calcd}(mg/m^3)	0.990	1.001	0.895
μ(mm^{-1})	1.504	1.693	1.335
F(000)	2776	2792	2576
θ range(deg)	2.026-27.498	1.522-27.480	1.654-27.536
Reflections collected/unique	38364/10322	49894/10299	48881/10204
R(int)	0.0701	0.0386	0.0879
Goodness-of-fit on F^2	0.794	1.078	0.954
R_1,wR_2 [$I>2\sigma(I)$]	0.0402, 0.0711	0.0328, 0.0773	0.0503, 0.1151
R_1,wR_2(all data)	0.0926, 0.0788	0.0506, 0.0828	0.1167, 0.1389
Largest diff. peak and hole(e. Å^{-3})	0.620 and -0.437	2.132 and -0.790	0.513 and -0.376

Table S8. Crystal data and structure refinement for **2d-2e**

	2d	**2e**
Formula	$C_{62}H_{90}N_4O_6Si_2Er_2$	$C_{62}H_{90}N_4O_6Si_2Yb_2$
FW	1378.07	1389.63
T(K)	293(2)	293(2)
λ(Å)	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic
Space group	*C*2/*c*	*C*2/*c*
a(Å)	30.328(5)	30.273(4)
b(Å)	14.9950(19)	15.0332(17)
c(Å)	22.623(4)	22.541(3)
α(deg)	90	90
β(deg)	118.215(3)	118.311(3)
γ(deg)	90	90
v(Å^3)	9066(2)	9031(2)
Z	4	4
D_{calcd}(mg/m^3)	1.010	1.022
μ(mm^{-1})	1.900	2.120
F(000)	2808	2824
θ range(deg)	1.524-27.553	1.555-27.563
Reflections collected/unique	36649/10241	49016/10228
R(int)	0.0417	0.1596
Goodness-of-fit on F^2	1.056	0.953
R_1,wR_2 [$I>2\sigma(I)$]	0.0356, 0.0808	0.0656, 0.1273
R_1,wR_2(all data)	0.0566, 0.0876	0.1642, 0.1610
Largest diff. peak and hole(e. Å^{-3})	2.235 and -0.944	2.770 and -1.374

Table S9. Crystal data and structure refinement for **2ea-2eb**

	2ea	**2eb**
Formula	$C_{84}H_{90}N_8O_6Yb_2$	$C_{68}H_{84}N_6O_6Si_2Yb_2$
FW	1653.71	1483.67
T(K)	293(2)	293(2)
λ(Å)	0.71073	0.71073
Crystal system	Monoclinic	Triclinic
Space group	*C*2/*c*	*P*$\bar{1}$
a(Å)	32.297(7)	11.095(4)
b(Å)	14.388(3)	14.589(6)
c(Å)	25.203(9)	16.114(6)
α(deg)	90	112.592(4)
β(deg)	128.886(2)	91.520(5)
γ(deg)	90	110.684(5)
v(Å^3)	9116(4)	2212.8(15)
Z	4	1
D_{calcd}(mg/m^3)	1.205	1.113
μ(mm^{-1})	2.087	2.168
F(000)	3352	750
θ range(deg)	1.620-25.355	1.394-25.711
Reflections collected/unique	43301/8320	21550/8258
R(int)	0.0635	0.0589
Goodness-of-fit on F^2	1.027	1.084
R_1,wR_2 [$I>2\sigma(I)$]	0.0555, 0.1410	0.0704, 0.1882
R_1,wR_2(all data)	0.0757, 0.1587	0.0826, 0.2032
Largest diff. peak and hole(e. Å^{-3})	3.127 and -1.209	4.153 and -3.401