Supporting information for:

Synthesis and Electronic Structure Analysis of the Actinide Allenylidenes, $[\{(NR_2)_3\}An(CCCPh_2)]^-$ (An = U, Th; R = $SiMe_3$)

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Experimental

General. All reactions and subsequent manipulations were performed under anaerobic and anhydrous conditions under an atmosphere of dinitrogen. Diethyl ether (Et_2O), pentane and hexanes were dried using a Vacuum Atmospheres DRI-SOLV Solvent Purification system and stored over 3Å sieves for 24 h prior to use. Tetrahydrofuran (THF) was distilled over calcium hydride then distilled over sodium benzophenone, collected, and stored over 3Å sieves for 24 h prior to use. Isooctane was distilled over sodium benzophenone, collected, and stored over 3Å sieves for 24 h prior to use. THF-d_8 and C_6D_6 were stored over 3Å sieves for 24 h prior to use. $[UCl(NR_2)_3]$ (R = $SiMe_3$), $[ThCl(NR_2)_3]$, LDA, and 3,3-diphenylcyclopropene were synthesized according to previously reported literature procedures.[1-4] All other reagents were purchased from commercial vendors and used as received.

^1H, $^{13}C\{^1H\}$, and $^7Li\{^1H\}$ NMR spectra were recorded on a Varian UNITY INOVA 400 MHz or a Varian UNITY INOVA 500 MHz spectrometer. ^1H and $^{13}C\{^1H\}$ NMR spectra were referenced to external $SiMe_4$ using the residual protio solvent peaks as internal standards.[5, 6] $^7Li\{^1H\}$ spectra were referenced to a saturated LiCl solution in D_2O. IR spectra were recorded on a Nicolet 6700 FT-IR spectrometer with a NXR FT Raman Module. Electronic absorption spectra were recorded on a Shimadzu UV3600 UV-NIR Spectrometer. Elemental analyses were performed by the Microanalytical Laboratory at University of California (Berkeley, CA).

Caution! *Depleted uranium (isotope ^{238}U) and natural thorium are weak alpha emitters with a half-life of 4.47×10^9 years and 1.41×10^{10} years, respectively. Manipulations and reactions should be carried out in a fume hood or inert atmosphere glovebox in a laboratory equipped with α- and β-counting equipment.*

X-ray Crystallography: Data for complexes **1** - **4**·C_5H_{12} were collected on a Bruker KAPPA APEX II diffractometer equipped with an APEX II CCD detector using a TRIUMPH monochromater with a Mo Kα X-ray source (α = 0.71073 Å). The crystals were mounted on a cryoloop under Paratone-N oil, and data were collected at 110(2) K using an Oxford nitrogen gas cryostream system. X-ray data for **1**, **2**, **3**·C_5H_{12}, and **4**·C_5H_{12} were collected utilizing frame exposures of 5, 10, 10, and 20 s, respectively. Data collection and cell parameter determination were conducted using the SMART program.[7] Integration of the data frames and final cell parameter refinement were performed using SAINT software.[8] Absorption corrections of the data

were carried out using the multi-scan method SADABS.[9] Subsequent calculations were carried out using SHELXTL.[10] Structure determination was done using direct or Patterson methods and difference Fourier techniques. All hydrogen atom positions were idealized, and rode on the atom of attachment. Structure solution, refinement, graphics, and creation of publication materials were performed using SHELXTL.[10]

The pentane solvate molecule in **3**·C_5H_{12} exhibited positional disorder, as a result the pentane carbon atoms were constrained using the SADI and EADP commands and refined isotopically. The cryptand ligand in **3**·C_5H_{12} also contained slight positional disorder and as a result the temperature factors of the carbon and nitrogen atoms were constrained using the EADP command. The pentane solvate in **4**·C_5H_{12} contained more severe disorder, as a result it was constrained using the SADI and EADP commands, refined isotopically and hydrogen atoms were not assigned. The cryptand moiety in **4**·C_5H_{12} contained unresolved positional disorder, as a result carbon and oxygen atom temperature factors were constrained using the EADP command, and the lithium and nitrogen atoms were refined isotopically. Bond distances on the cryptand moiety were constrained using SADI command and three hydrogen-hydrogen distances were constrained, using the DFIX command, to a distance of 1.99 Å.

Further crystallographic details can be found in Tables S8. Complexes **1**-**4** have been deposited in the Cambridge Structural Database (**1**: CCDC 2098903; **2**: CCDC 2098904; **3**·C_5H_{12}: CCDC 2098905; **4**·C_5H_{12}: CCDC 2098906).

Synthesis of [{$(NR_2)_3$}U(CH=C=CPh_2)] (1). To a cold (-25 °C), colorless Et_2O solution (0.5 mL) of 3,3-diphenylcyclopropene (42.1 mg, 0.0.219 mmol) was added quickly a cold (-25 °C), colorless Et_2O solution (0.5 mL) of LDA (22.3 mg, 0.208 mmol). Immediately, the solution turned light yellow. This solution was then added drop wise to a cold (-25 °C) stirring pink slurry of [$UCl(NR_2)_3$] (157.2 mg, 0.208 mmol) in Et_2O (3 mL). The stirring solution immediately turned red-brown concomitant with the deposition of a light tan precipitate. After stirring for 45 min the resulting brown solution was filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) and the volatiles were removed from the filtrate *in vacuo*. The resulting brown oil was extracted into pentane (4 mL), filtered through a Celite column supported on glass wool (0.5 cm ×

2 cm) and the volatiles were removed from the filtrate *in vacuo*, yielding a brown solid. The resulting brown powder was extracted again into pentane (2 mL), filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) and transferred to a 4 mL scintillation vial. This vial was placed into a 20 mL scintillation vial and the solution was concentrated to 0.5 mL. Isooctane (2 mL) was added to the outer vial and storage of this two-vial system at -25 ºC for 24 h resulted in the deposition of brown blocks. Decanting the supernatant, rinsing the crystals with cold (-25 °C) pentane (2 mL), and drying *in vacuo* afforded **1** (137.3 mg, 72.4 % yield) Anal. Calcd for $UN_3Si_6C_{33}H_{65}$: C, 43.53; H, 7.20; N, 4.62. Found: C, 43.34; H, 7.03; N, 4.68. ^{1}H NMR (C_6D_6/THF-d_8, 298 K, 500 MHz): δ 3.34 (t, *J* = 7.1 Hz, 4H, *m*-CH), 3.03 (t, *J* = 7.2 Hz, 2H, *p*-CH), -1.84 (br. s, 54H, C*H*$_3$), -9.13 (d, *J* = 7.1 Hz, 4H, *o*-CH), -174.80 (s, 1H, α-C*H*). IR (KBr pellet, cm^{-1}): 2954 (m), 2897 (w), 1936 (w, C_α–C_β stretch), 1871 (w, C_β–C_γ stretch), 1400 (w), 1250 (s), 1182 (w), 904 (s), 847 (s), 769 (m), 681 (w), 656 (w), 611 (m).

Synthesis of [{(NR$_2$)$_3$}Th(CH=C=CPh$_2$)] (2). To a cold (-25 °C), colorless Et_2O solution (0.5 mL) of 3,3-diphenylcyclopropene (40.5 mg, 0.211 mmol) was added quickly a cold (-25 °C), colorless Et_2O solution (0.5 mL) of LDA (21.4 mg, 0.200 mmol). Immediately, the solution turned light yellow. This solution was then added drop wise to a cold (-25 °C) stirring colorless slurry of [Th(Cl)(NR$_2$)$_3$] (150.2 mg, 0.200 mmol) in Et_2O (3 mL). The stirring solution immediately turned yellow-orange concomitant with the deposition of a light tan precipitate. After stirring for 45 min the resulting orange suspension was filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) and the volatiles were removed from the filtrate *in vacuo*. The resulting orange oil was extracted into pentane (4 mL), filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) and the volatiles were removed from the filtrate *in vacuo*, yielding a brown solid. The resulting orange oil was extracted again into pentane (2 mL), filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) and transferred to a 4 mL scintillation vial. This vial was placed into a 20 mL scintillation vial and the solution was concentrated to 0.5 mL. Isooctane (2 mL) was added to the outer vial and storage of this two-vial system at -25 ºC for 24 h resulted in the deposition of off-white blocks. Decanting the supernatant, rinsing the crystals with cold (-25 °C) pentane (2 mL), and drying *in vacuo* afforded **2** (112.0 mg, 61.7 % yield) Anal. Calcd for $ThN_3Si_6C_{33}H_{65}$: C, 43.82; H, 7.24; N, 4.65. Found: C, 43.71; H, 7.03; N, 4.57. ^{1}H NMR (C_6D_6/THF-d_8, 298 K, 500 MHz): δ = 7.42 (d, *J* = 7.3 Hz, 4H,

o-CH), 7.17 (t, *J* = 7.7 Hz, 4H, *m*-CH), 6.96 (d, *J* = 7.4 Hz, 2H, *p*-CH), 5.77 (s, 1H, α-C*H*), 0.27 (s, 54H, CH_3). $^{13}C\{^1H\}$ NMR (C_6D_6/THF-d_8, 298 K, 126 MHz) δ 204.67 (C_β), 150.63 (C_{ipso}), 139.38 (C_α), 128.89 (C_{ortho}), 128.39 (C_{meta}), 125.63 (C_{para}), 96.72 (C_γ), 4.57. IR (KBr pellet, cm^{-1}): 2953 (m), 2895 (w), 1934 (w, C_α–C_β stretch), 1869 (m, C_β–C_γ stretch), 1597 (m), 1491 (m), 1450 (m), 1252 (s), 1182 (m), 1113 (w), 1072 (w), 1030 (w), 931 (s), 847 (s), 768 (s), 696 (s), 658 (w), 640 (w), 609 (m).

Synthesis of [Li(2.2.2-Cryptand)][{(NR$_2$)$_3$}U(CCCPh$_2$)] (3). To a cold (-25 °C), dark brown Et_2O solution (3 mL) of **1** (65.8 mg, 0.072 mmol) and 2.2.2-cryptand (27.2 mg, 0.072 mmol) was added dropwise a cold (-25 °C), colorless Et_2O solution (0.5 mL) of LDA (7.74 mg, 0.072 mmol). Upon addition, the solution turned dark purple-red. After 2 min, the solution was concentrated *in vacuo* to 0.5 mL and filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) into a 4 mL scintillation vial. This vial was placed into a 20 mL scintillation vial and isooctane (2 mL) was added to the outer vial. Storage of this two-vial system at -25 ºC for 48 h resulted in the deposition of dark purple solid. Decanting off the supernatant, rinsing with pentane (2 mL), and drying *in vacuo* afforded **3** as an analytically pure dark purple solid (50.0 mg, 53.5 % yield). X-ray quality crystals of **3** were grown by dissolving 40 mg of this material in THF:pentane (0.25:2.5 mL). Storage of this solution at -25 ºC for 24 h resulted in the deposition of dark purple needles. Anal. Calcd for $C_{51}H_{100}LiN_5O_6Si_6U$: C, 47.38; H, 7.80; N, 5.42. Found: C, 47.33; H, 7.59; N, 5.04. ^1H NMR (C_6D_6/THF-d_8, 298 K, 500 MHz,) δ 2.78 (br s, 12H, CH_2), 2.59 (br s, 12H, CH_2), 2.07 (t, *J* = 8.3 Hz, 4H, *m*-CH), 1.86 (br s, 12H, CH_2), -1.60 (br s, 54H, CH_3), -1.74 (t, *J* = 8.3 Hz, 2H, *p*-CH), -12.89 (d, *J* = 8.8 Hz, 4H, *o*-CH). $^7Li\{^1H\}$ NMR (C_6D_6/THF-d_8, 25 °C, 155 MHz): δ –1.59. IR (KBr pellet, cm^{-1}): 2954 (m), 2887 (m), 2862 (w), 2050 (w, C_α–C_β stretch), 1911 (w, C_β–C_γ stretch), 1514 (w), 1477 (m), 1385 (m), 1356 (s), 1263 (m), 1255 (s), 1136 (m), 1101 (s), 1088 (w), 933 (s), 862 (w), 841 (s), 894 (w), 694 (w).

Synthesis of [Li(2.2.2-cryptand)][{(NR$_2$)$_3$}Th(CCCPh$_2$)] (4). To a cold (-25 °C), dark brown Et_2O solution (3 mL) of **2** (82.1 mg, 0.091 mmol) and 2.2.2-cryptand (29.0 mg, 0.091 mmol) was added dropwise a cold (-25 °C), colorless Et_2O solution (0.5 mL) of LDA (9.7 mg, 0.091 mmol). Upon addition, the solution turned dark red-orange. After 2 min, the solution was concentrated *in vacuo* to 0.5 mL and filtered through a Celite column supported on glass wool (0.5 cm × 2 cm) into a 4 mL scintillation vial. This vial was placed into a 20 mL scintillation vial and isooctane (2

mL) was added to the outer vial. Storage of this two-vial system at -25 ºC for 48 h resulted in the deposition of dark orange blocks. Decanting off the supernatant, rinsing with pentane (2 mL), and drying *in vacuo* afforded **4** as an analytically pure dark orange solid (53.4 mg, 45.7 % yield). X-ray quality crystals of **4** were grown by dissolving 36 mg of this material into THF:pentane (0.25:2.5 mL). Storage of this vial at -25 ºC for 24 h resulted in the deposition of orange needles. Anal. Calcd for $C_{51}H_{100}LiN_5O_6Si_6Th$: C, 47.60; H, 7.83; N, 5.44. Found: C, 47.30; H, 7.46; N, 5.14. ^{1}H NMR (C_6D_6/THF-d_8, 298 K, 500 MHz): δ 7.85 (d, J = 8.3 Hz, 4H, *o*-CH), 7.09 (t, J = 7.7 Hz, 4H, *m*-CH), 6.45 (t, J = 7.0 Hz, 2H, *p*-CH), 3.08 (m, 12H, CH_2), 3.03 (t, J = 5.0 Hz, 12H, CH_2), 2.08 (t, J = 5.0 Hz, 12H, CH_2), 0.53 (s, 54H, CH_3). ^{7}Li{^{1}H} NMR (C_6D_6/THF-d_8, 25 °C, 155 MHz): δ –1.81. ^{13}C{^{1}H} NMR (C_6D_6/THF-d_8, 298 K, 126 MHz) δ: 205.40 (C_α), 145.79 (C_{ipso}), 128.49 (C_β), 127.04 (C_{ortho}), 122.53 (C_{meta}), 114.29 (C_{para}), 70.58 (C_γ), 67.87 ($C_{cryptand}$), 67.84 ($C_{cryptand}$), 53.07 ($C_{cryptand}$), 4.70 (CH_3). UV-Vis/NIR (C_6H_6, 0.263 mM, 25 °C, L·mol^{-1}·cm^{-1}): 403 nm (ε = 8310) 537 nm (ε = 15,030). IR (KBr pellet, cm^{-1}): 2954 (m), 2883 (m), 2816 (w), 2044 (m, C_α–C_β stretch), 1921 (s, C_β–C_γ stretch), 1585(w), 1479 (m), 1444 (w), 1356 (m), 1296 (w), 1250 (s), 1144 (w), 1115 (m), 1101 (s), 933 (s), 837 (s), 771 (m), 696 (w), 663 (w), 607 (w).

Computational Details.

Kohn-Sham density functional calculations were employed for **1-4** with the Gaussian 16 package.[11] The crystal structure coordinates were optimized for hydrogen positions using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional.[12] Small-core Stuttgart energy-consistent relativistic pseudopotentials, ECP60MWB for Th and U, were utilized with matching valence basis sets.[13] The 6-31G(d) basis set was used for the Si, C, N, and H atoms.[14] Atom-pairwise corrections for dispersion forces were considered via Grimme's D3 model augmented with the Becke-Johnson (BJ) damping.[15] To quantify the compositions of the chemical bonds of interest, natural localized molecular orbital (NLMO) analyses were carried out with the NBO program, version 6.0.[16] The quantum theory of atoms in molecules (QTAIM) analysis was performed with Multiwfn 3.6.[17]

NMR shielding constants (σ, ppm) for **2** and **4** were calculated with the NMR module of the ADF package (version 2017),[18] using the scalar relativistic and spin-orbit all electron Zeroth-Order Regular Approximation (ZORA) Hamiltonian,[19] in conjunction with all-electron doubly polarized triple-ξ (TZ2P)[20] Slater-type basis set. The conductor-like screening model (COSMO) was used to describe solvent effect (tetrahydrofuran).[21] Functionals used for the NMR calculations were BP86, PBE, PBE0 (25% exact exchange), and PBE0 (40% exact exchange) The ^{13}C chemical shifts (δ, ppm) were obtained by subtracting the C_α, C_β, C_γ nuclear magnetic shielding of interest from the reference compound (Tetramethylsilane, TMS), with the latter calculated at the same level of theory. The localized molecular orbital (LMO) analysis of the NMR shielding and the character of specific chemical bonds quantified on the basis of orbital localizations were described elsewhere.[22, 23] It helps to provide useful information on how spin-orbit coupling affects the chemical shifts. Note that the NLMOs produced from ECP60MWB valence basis set and Slater-type basis set (TZ2P) are qualitatively comparable to each other.

Table S1. % compositions of the An-C (An = Th, U) bonding NLMOs in **1-4**.

Complex	Orbital	Total C_α	2s	2p	Total C_β	2s	2p	Total An	7s	7p	6d	5f
1	σ(U-C)	75	31	69	4	0	100	16	12	1	69	18
	π(U-C)	50	0	100	44	0	100	4	0	0	53	47
2	σ(Th-C)	77	30	70	5	0	100	14	13	1	73	13
	π(Th-C)	50	0	100	44	0	100	4	0	0	53	47
3	σ(U-C)	78	47	53	0	0	0	20	16	1	67	16
	π(U-C)	47	0	100	47	0	100	5	0	0	55	45
	π(U-C)	31	0	100	28	0	100	11	0	0	32	68
4	σ(Th-C)	80	47	53	0	0	0	18	16	1	71	12
	π(Th-C)	48	0	100	45	0	100	5	0	0	60	40
	π(Th-C)	53	0	100	37	0	100	7	0	0	62	38

Table S2. The Wiberg Bond Orders for the selected bonds in **1-4** and $[An(C{\equiv}CH)(NR_2)_3]$ (An = U or Th) complexes.

Complexes	An-C_α	C_α-C_β	C_β-Cγ
1	0.597	2.005	1.638
2	0.565	2.003	1.634
3	0.983	2.401	1.281
4	0.912	2.355	1.305
$[U(C{\equiv}CH)(NR_2)_3]$[24]	0.709		
$[Th(C{\equiv}CH)(NR_2)_3]$[24]	0.674		

Table S3. Calculated carbon shielding (σ) and chemical shift (δ) for TMS, Allene, Acetylene, and the C_α, C_β, and C_γ nuclei of **2** and **4** using various functionals.

Complex	Method	σ_{calc}(ppm)	δ_{calc}(ppm)	Δ_{SO}(ppm)	δ_{expt}(ppm)
TMS	PB86/SO-BP86	186.9 / 187.8			
	PBE/SO-PBE	187.5 / 188.4			
	PBE0/SO-PBE0 (25%)[a]	192.2 / 193.0			
	PBE0/SO-PBE0 (40%)	194.7 / 195.5			
Allene	PBE/SO-PBE	107.9, -41.1, 107.9 / 108.5, -40.4, 108.5	79.6, 228.6, 79.6 / 79.9, 228.8, 79.9	0.3, 0.2, 0.3	
	MPW1PW91	109, -41, 109[b]			73.9, 208.5, 73.9[c]
Acetylene	PBE/SO-PBE	108.3 / 108.9	79.2 / 79.5	0.3	
2[d]	PB86/SO-BP86	69.9, -23.4, 82.2 / 43.7, -26.2, 82.8	117.0, 210.3, 104.7 / 144.1, 214.0, 105.0	27.1, 3.7, 0.3	139.4, 204.7, 96.7
	PBE/SO-PBE	70.4, -22.9, 82.8 / 44.5, -25.7, 83.4	117.1, 210.4, 104.7 / 143.9, 214.1, 105.0	26.8, 3.7, 0.3	
	PBE0/SO-PBE0 (25%)	75.3, -26.5, 88.7 / 46.2, -28.8, 89.1	116.9, 218.7, 103.6 / 146.8, 221.8, 103.9	29.9, 3.1, 0.3	
	PBE0/SO-PBE0 (40%)	78.5, -27.5, 91.8 / 47.2, -29.4, 92.0	116.2, 222.2, 102.9 148.3, 224.9, 103.5	32.1, 2.7, 0.6	
4	PB86/SO-BP86	12.3, 59.3, 104.3 / -23.2, 51.2, 105.2	174.6, 127.6, 82.6 / 211.0, 136.6, 82.0	36.4, 9.0, 0.0	205.4, 128.5, 70.6
	PBE/SO-PBE	12.6, 59.6, 104.8 / -22.7, 51.4, 105.8	174.9, 127.9, 82.7 / 211.1, 137.0, 82.6	36.2, 9.1, -0.1	
	PBE0/SO-PBE0 (25%)	19.4, 62.8, 115.1 / -19.3, 55.1, 115.5	172.8, 129.4, 77.1/ 212.3, 137.9, 77.5	39.5, 8.5, 0.4	
	PBE0/SO-PBE0 (40%)	23.7, 65.0, 120.4/ -16.8, 57.4, 120.6	171.0, 129.7, 74.3/ 212.3, 138.1, 74.9	41.3, 8.4, 0.6	

[a] Fraction of exact exchange in the functional in parentheses.

[b] Values taken from Ref 25.

[c] Values taken from Ref 26.

[d] The shielding and chemical shifts are averaged from two experimental geometries.

Table S4. QTAIM analysis of the complexes **3** and **4**.

Complex	BCP[a]	$\rho(r)$[b]	$\nabla^2\rho(r)$[c]	$H(r)$[d]	$\varepsilon(r)$[e]
3	U-C_α	0.102	0.147	-0.038	0.300
	C_α-C_β	0.399	0.939	-0.693	0.026
	C_β-C_γ	0.295	0.751	-0.293	0.201
4	Th-C_α	0.093	0.116	-0.033	0.193
	C_α-C_β	0.388	0.958	-0.662	0.012
	C_β-C_γ	0.292	0.739	-0.293	0.229

[a] The bond critical points.
[b] The electron density ($\rho(\boldsymbol{r})$, au).
[c] Laplacian of electron density ($\nabla^2\rho(\boldsymbol{r})$, au).
[d] Total electronic energy density ($H(\boldsymbol{r})$, au).
[e] Ellipticity of electron density ($\varepsilon(\boldsymbol{r})$, au).

Table S5. Localized Molecular Orbital(LMO) Analysis of NMR Shielding for allene.

LMO type	**SR/ C_α(L+NL)**	**SOC/ C_α(L+NL)**	**SR/ C_β(L+NL)**	**SOC/ C_β(L+NL)**	**SR/ C_γ(L+NL)**	**SOC/ C_γ(L+NL)**
$\sigma(C_\alpha$-$C_\beta)$	-57.9	-57.9	-77.1	-77.1	-6.3	-6.3
$\pi(C_\alpha$-$C_\beta)$	32.2	32.2	-40.7	-40.8	2.0	2.0
$\sigma(C_\alpha$-$H_1)$	-32.1	-32.1	-2.0	-2.0	-0.6	-0.5
$\sigma(C_\alpha$-$H_2)$	-32.1	-32.1	-2.0	-2.0	-0.6	-0.5
$\sigma(C_\beta$-$C_\gamma)$	-6.3	-6.3	-77.1	-77.1	-57.9	-57.9
$\pi(C_\beta$-$C_\gamma)$	2.0	2.0	-40.7	-40.7	32.2	32.2
C_α(core)	203.4	204.1	-0.6	-0.6	-0.1	-0.1
C_β(core)	-0.1	-0.1	203.6	204.4	-0.1	-0.1
C_γ(core)	-0.1	-0.1	-0.6	-0.6	203.4	204.1
$\sigma(C_\alpha$-$H_3)$	-0.6	-0.5	-2.0	-2.0	-32.1	-32.1
$\sigma(C_\alpha$-$H_4)$	-0.6	-0.5	-2.0	-2.0	-32.1	-32.1
$\sum$other	0.0	-0.2	0.0	0.1	0.0	-0.2
Total calc.	**107.9**	**108.5**	**-41.1**	**-40.4**	**107.9**	**108.5**

Table S6. Localized Molecular Orbital(LMO) Analysis of NMR Shielding for complex **2**.

LMO type	**SR/ C_α(L+NL)**	**SOC/ C_α(L+NL)**	**Δ^{SO}/ C_α**	**SR/ C_β(L+NL)**	**SOC/ C_β(L+NL)**	**Δ^{SO}/ C_β**	**SR/ C_γ(L+NL)**	**SOC/ C_γ(L+NL)**	**Δ^{SO}/ C_γ**
$\sigma(C_\alpha\text{-}C_\beta)$	-49.2	-48.3	0.9	-85.8	-86.3	-0.5	-6.0	-6.0	0.0
$\pi(C_\alpha\text{-}C_\beta)$/ π(Th-C)	26.3	26.4	0.1	-15.7	-15.8	-0.1	1.6	1.6	0.0
$\sigma(C_\alpha\text{-H})$	-48.1	-46.9	1.2	-4.2	-4.1	0.1	0.4	0.4	0.0
$\sigma(C_\beta\text{-}C_\gamma)$	-6.6	-6.6	0.0	-89.6	-89.7	-0.1	-37.3	-37.3	0.0
$\pi(C_\beta\text{-}C_\gamma)$	0.2	0.2	0.0	-29.9	-29.8	0.1	5.6	5.5	-0.1
$\sigma(C_\gamma\text{-}C_{ipso1})$	-0.3	-0.3	0.0	1.0	1.1	0.1	-38.1	-38.1	0.0
$\sigma(C_\gamma\text{-}C_{ipso2})$	-0.3	-0.3	0.0	0.5	0.5	0.0	-35.4	-35.4	0.0
C_α(core)	203.6	189.5	-14.1	-0.5	-0.5	0.0	-0.1	-0.1	0.0
C_β(core)	-0.3	-0.3	0.0	203.7	203.1	-0.6	0.0	0.0	0.0
C_γ(core)	-0.2	-0.2	0.0	-1.0	-1.0	0.0	203.3	204.0	0.7
σ(Th-C_α)	-50.1	-61.0	-10.9	2.7	1.5	-1.2	2.0	2.0	0.0
Th(core)	-1.9	-3.9	-2.0	-0.4	-0.3	0.1	-0.1	-0.1	0.0
$\sum$other	-3.1	-3.5	-0.4	-5.4	-6.1	-0.7	-13.1	-13.1	0.0
Total calc.	**70.0**	**44.8**	**-25.2**	**-24.6**	**-27.4**	**-2.8**	**82.8**	**83.4**	**0.6**

Table S7. Localized Molecular Orbital(LMO) Analysis of NMR Shielding for complex **4**

LMO type	**SR/ C_α(L+NL)**	**SOC/ C_α(L+NL)**	**Δ^{SO}/ C_α**	**SR/ C_β(L+NL)**	**SOC/ C_β(L+NL)**	**Δ^{SO}/ C_β**	**SR/ C_γ(L+NL)**	**SOC/ C_γ(L+NL)**	**Δ^{SO}/ C_γ**
$\sigma(C_\alpha\text{-}C_\beta)$	-69.2	-68.0	1.2	-68.5	-69.1	-0.6	-2.6	-2.7	-0.1
$\pi(C_\alpha\text{-}C_\beta)$	-2.2	-1.8	0.4	13.4	13.0	-0.4	1.7	1.7	0.0
$\pi(C_\alpha\text{-}C_\beta)$	-12.5	-12.3	0.2	37.6	37.5	-0.1	-0.4	-0.5	-0.1
$\sigma(C_\beta\text{-}C_\gamma)$	-7.7	-8.0	-0.3	-72.1	-72.6	-0.5	-26.5	-26.4	0.1
$\sigma(C_\gamma\text{-}C_{ipso1})$	-1.0	-1.0	0.0	0.5	0.5	0.0	-37.7	-37.8	-0.1
$\sigma(C_\gamma\text{-}C_{ipso2})$	-0.9	-0.9	0.0	0.1	0.0	-0.1	-37.6	-37.8	-0.2
C_α(core)	203.7	183.4	-20.3	-0.5	-0.4	0.1	-0.1	-0.1	0.0
C_β(core)	-0.7	-0.6	0.0	203.6	201.1	-2.5	0.2	0.2	0.0
C_γ(core)	-0.2	-0.2	0.0	-0.7	-0.7	0.0	203.3	204.1	0.8
σ(Th-C_α)	-81.4	-95.2	-13.8	-12.9	-15.9	-3.0	2.0	2.0	0.0
C_γ LP	-7.6	-7.6	0.0	-27.9	-28.1	-0.2	17.9	18.2	0.3
Th(core)	-4.5	-6.9	-2.4	-1.2	-1.6	-0.4	-0.3	-0.2	0.1
∑other	-3.2	-3.6	-0.4	-11.8	-12.3	-0.5	-15.1	-14.9	0.2
Total calc.	**12.6**	**-22.7**	**-35.3**	**59.6**	**51.4**	**-8.2**	**104.8**	**105.8**	**1.0**

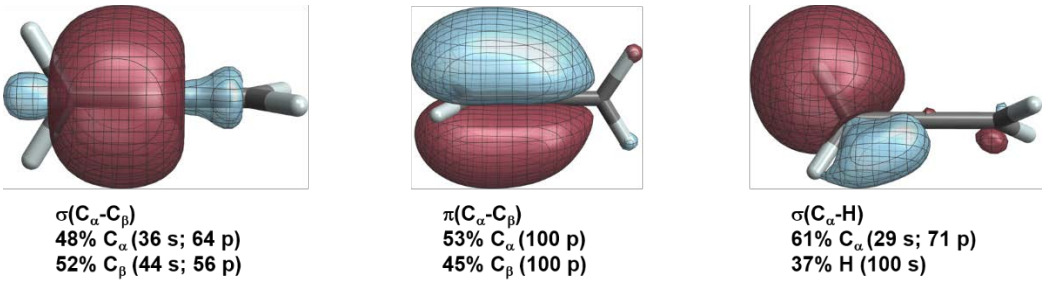

Figure S1. Isosurfaces (±0.03 a.u.) of representative bonding NLMOs in allene.

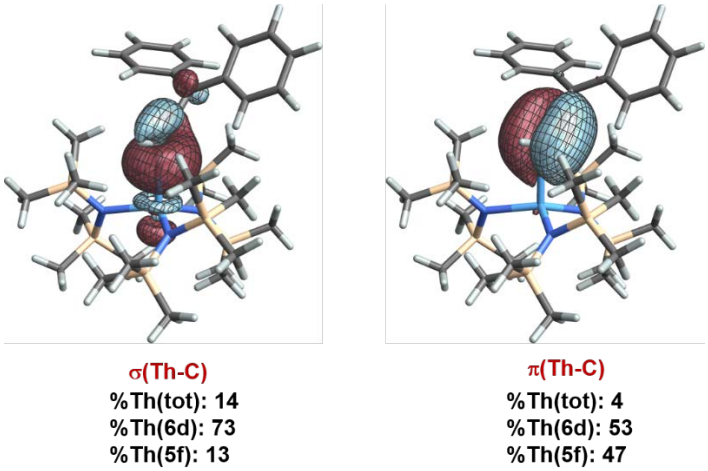

Figure S2. Isosurfaces (±0.03 a.u.) of two bonding NLMOs in complex **2**, along with weight-% metal and ligand character and 6d vs. 5f contributions at the metal.

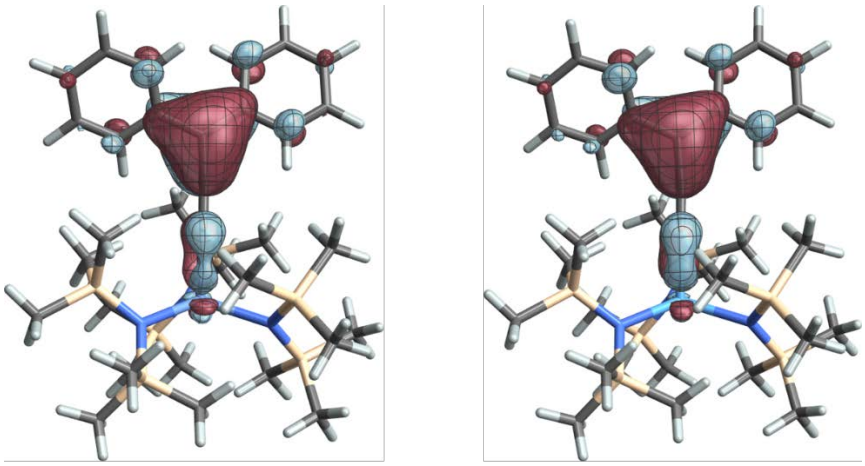

Figure S3. Isosurfaces (±0.03 a.u.) of representative π-LP(C_γ) bonding NLMO in **3** (left) and **4** (right) along with weight-% metal and ligand character and 6d vs. 5f contributions at the metal.

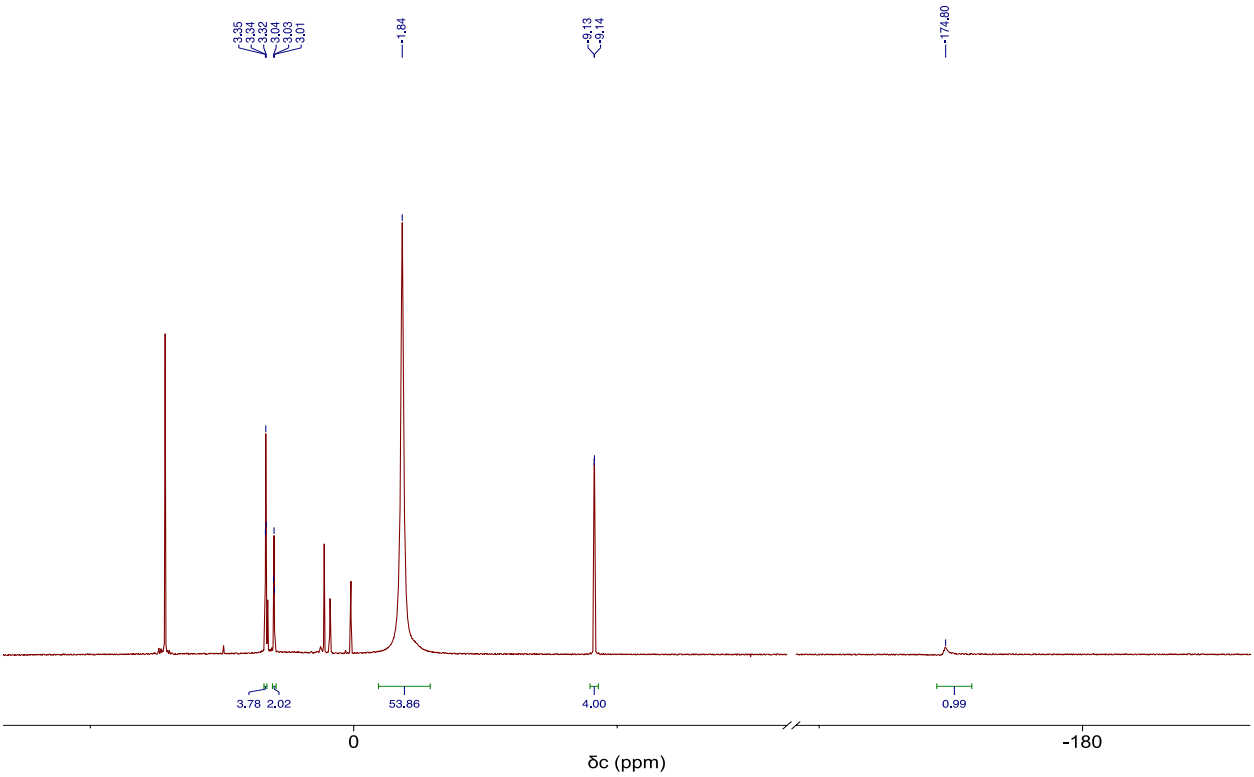

Figure S4. ^{1}H NMR spectrum of [{$(NR_2)_3$}U(CH=C=CPh_2)] (**1**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature.

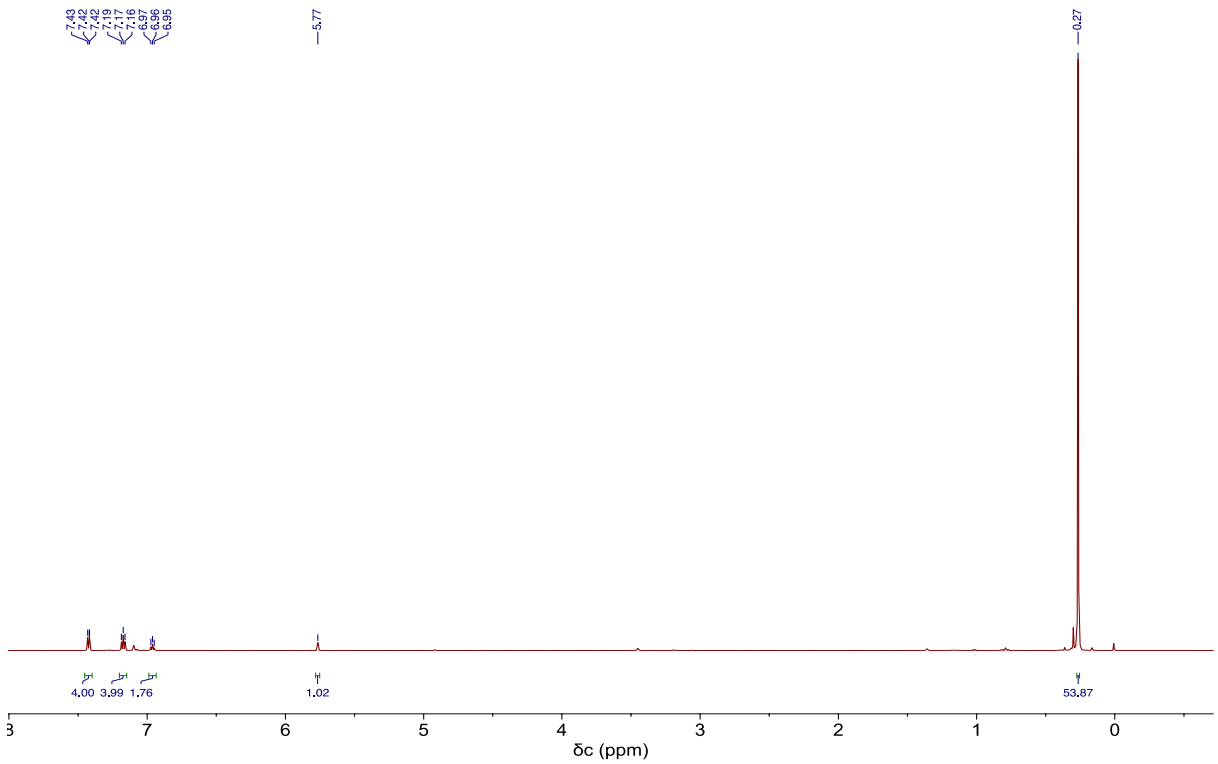

Figure S5. ^{1}H NMR spectrum of [{$(NR_2)_3$}Th(CH=C=CPh_2)] (**2**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature.

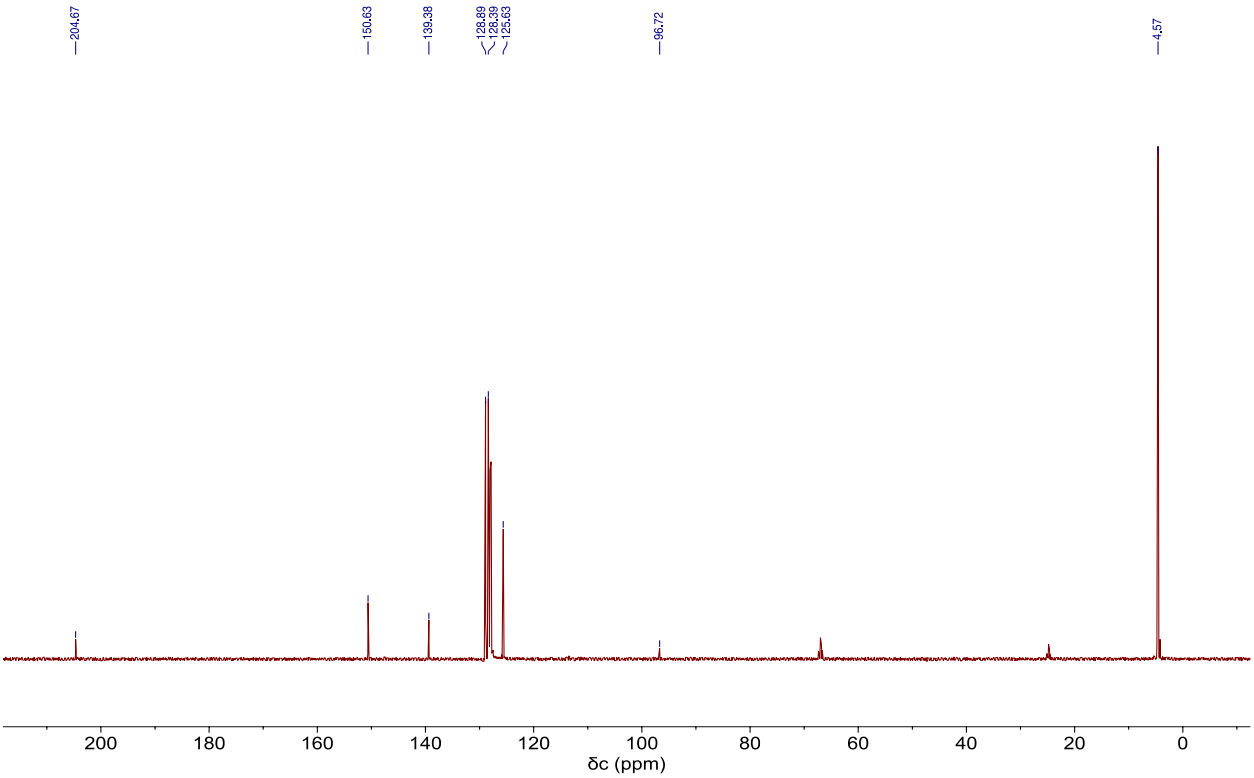

Figure S6. $^{13}C\{^{1}H\}$ NMR spectrum of [{$(NR_2)_3$}Th(CH=C=CPh_2)] (**2**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature.

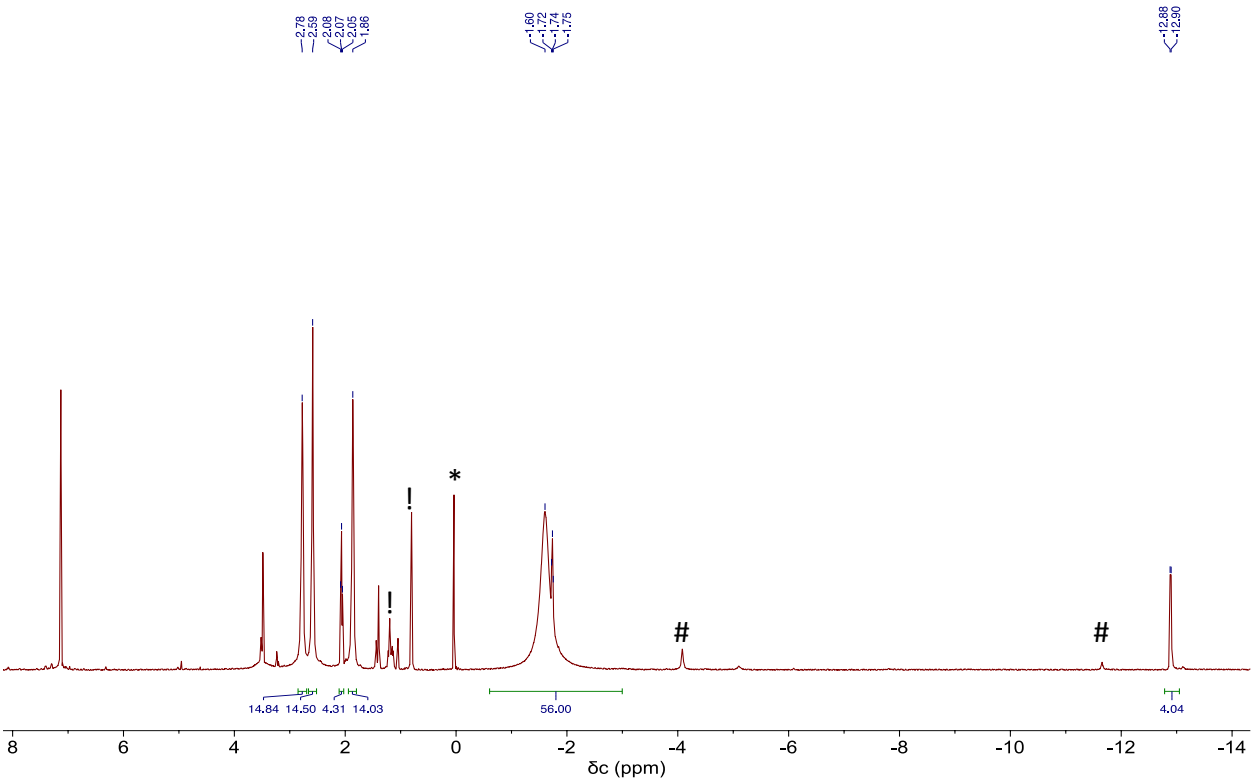

Figure S7. ^{1}H NMR spectrum of [Li(2.2.2-Cryptand)][{$(NR_2)_3$}U($CCCPh_2$)] (**3**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature. (*) indicates free $HN(SiMe_3)_2$, (#) indicates an unidentified impurity. (!) indicates pentane.

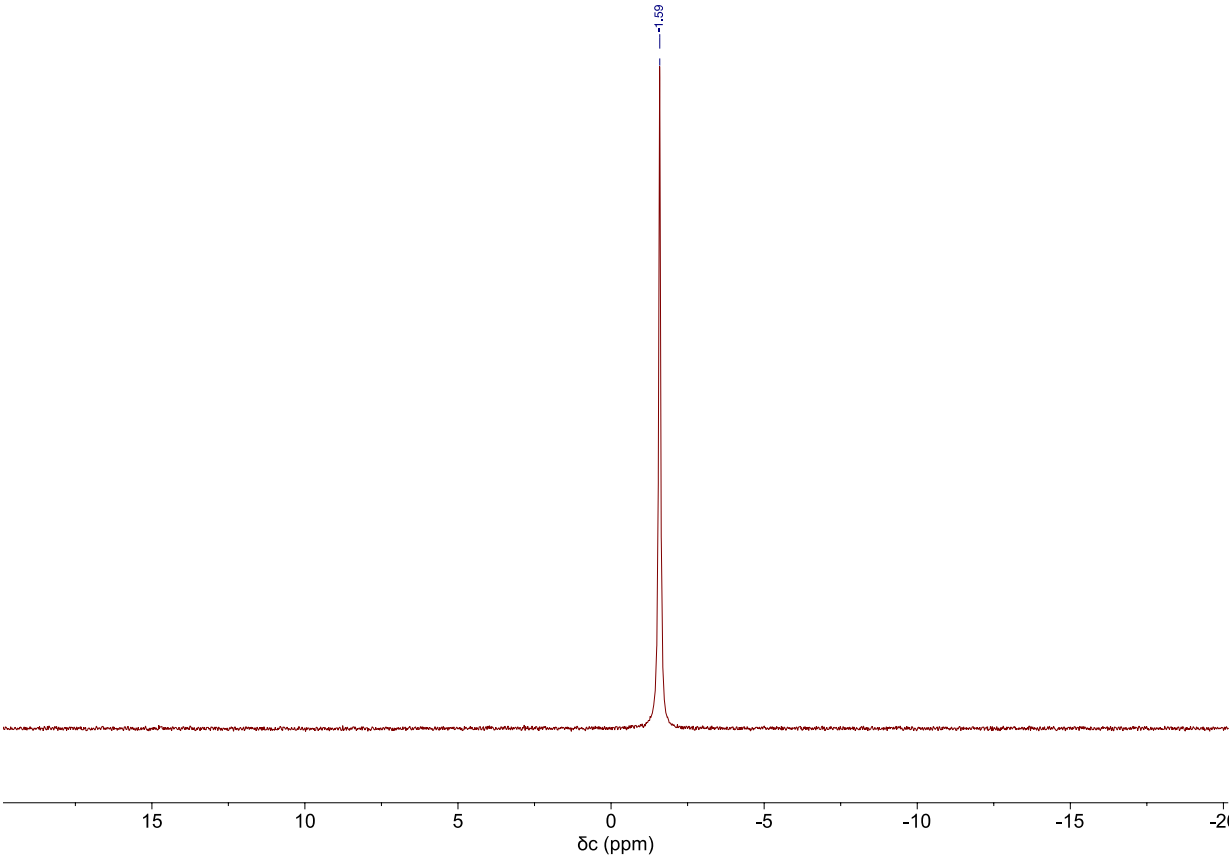

Figure S8. ^{7}Li**{**^{1}H} NMR spectrum of [Li(2.2.2-Cryptand)][{$(NR_2)_3$}U(CCCPh$_2$)] (**3**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature.

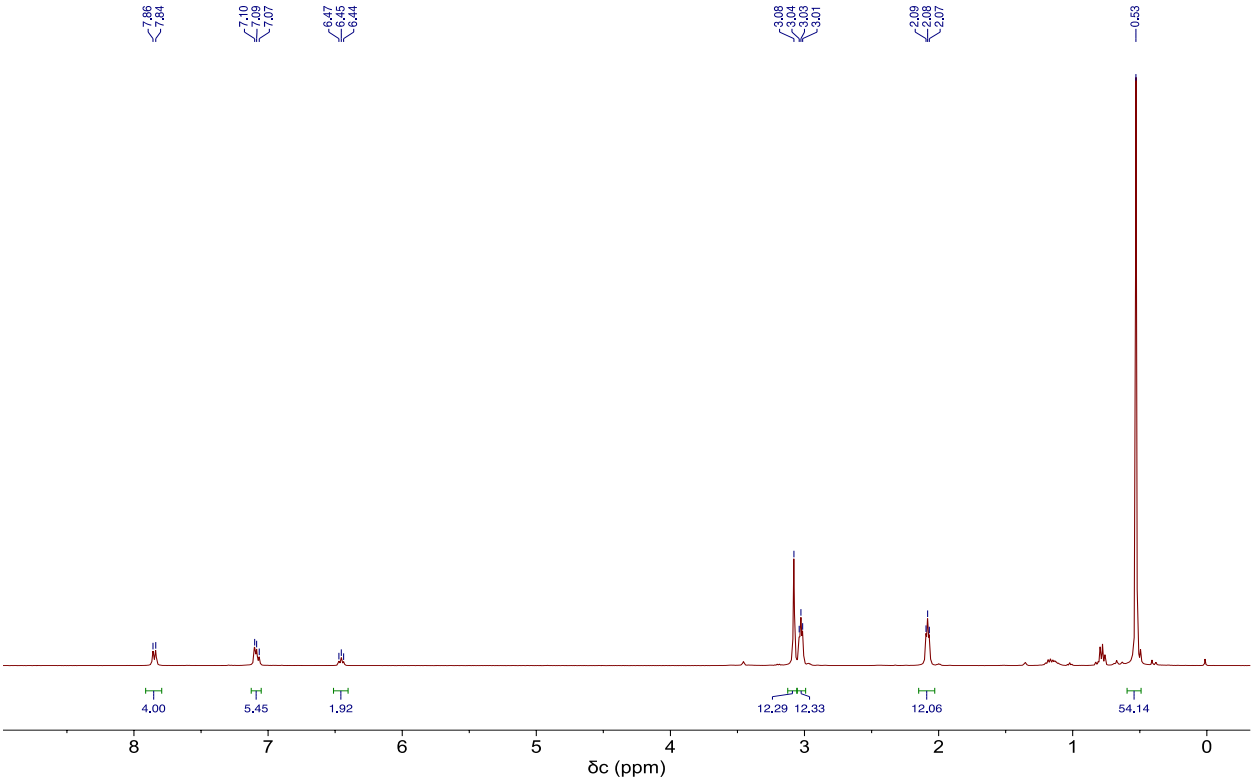

Figure S9. ^{1}H NMR spectrum of [Li(2.2.2-Cryptand)][{$(NR_2)_3$}Th($CCCPh_2$)] (**4**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature.

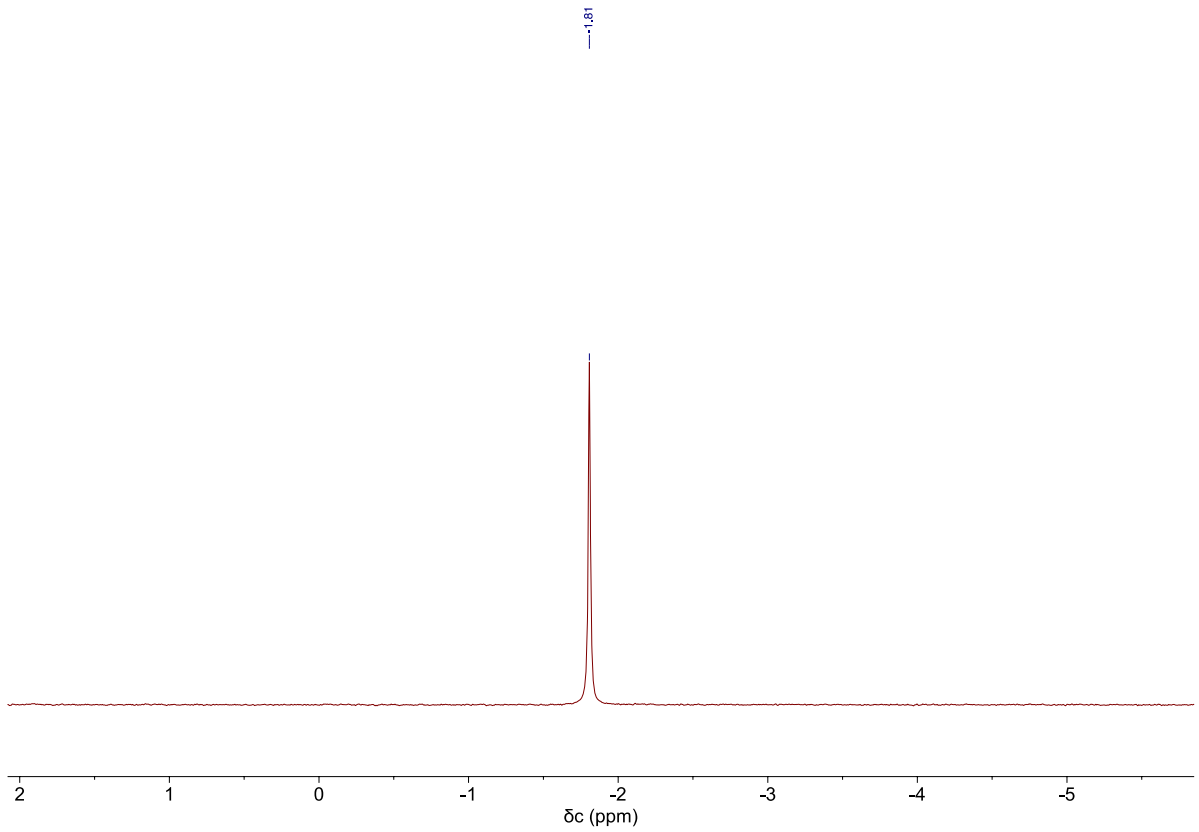

Figure S10. ^{7}Li**{**^{1}H} NMR spectrum of [Li(2.2.2-Cryptand)][{$(NR_2)_3$}Th($CCCPh_2$)] (**4**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature.

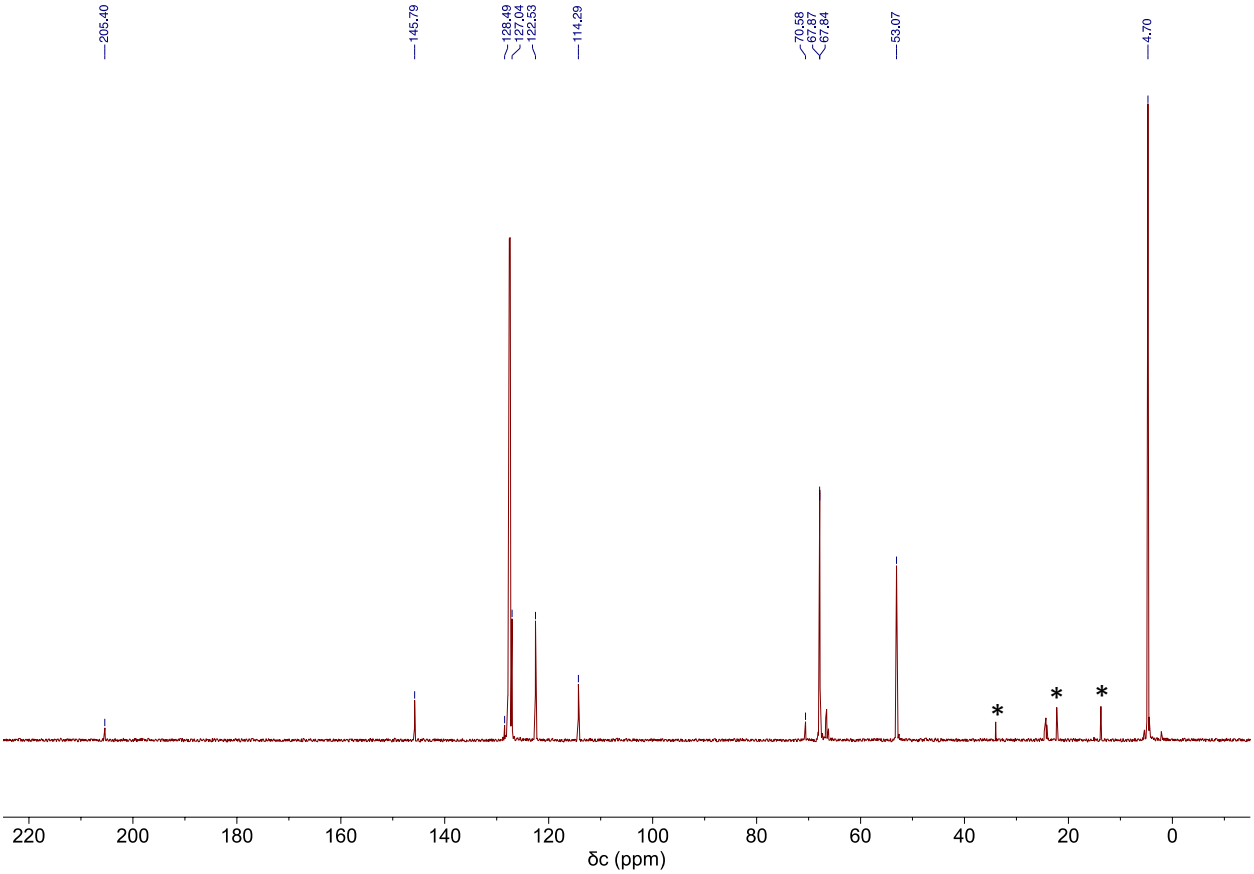

Figure S11. $^{13}C\{^{1}H\}$ NMR spectrum of [Li(2.2.2-Cryptand)][{$(NR_2)_3$}Th($CCCPh_2$)] (**4**) in a 10:1 mixture of C_6D_6 and THF-d_8 at room temperature. (*) indicates pentane.

Table S8. X-ray Crystallographic Data for Complexes **1**, **2**, **3**·C_5H_{12}, and **4**·C_5H_{12}

	1	**2**	**3**·C_5H_{12}	**4**·C_5H_{12}
empirical formula	$C_{33}H_{65}N_3Si_6U$	$C_{33}H_{65}N_3Si_6Th$	$C_{56}H_{112}LiN_5O_6Si_6U$	$C_{56}H_{112}LiN_5O_6Si_6Th$
Crystal habit, color	Block, Brown	Block, Colorless	Needle, Dark-purple	Needle, Orange
crystal size (mm)	0.30 × 0.20 × 0.10	0.30 × 0.20 × 0.10	0.40 × 0.10 × 0.05	0.40 × 0.10 × 0.05
crystal system	Triclinic	Triclinic	Monoclinic	Monoclinic
space group	P–1	P–1	$P2_1$	$P2_1$
vol ($Å^3$)	2162.2(9)	4339.3(18)	3449.1(13)	3520.1(16)
a (Å)	11.486(3)	11.816(3)	12.471(3)	12.595(3)
b (Å)	11.561(3)	19.383(5)	10.829(2)	10.892(3)
c (Å)	18.978(5)	19.538(5)	25.657(6)	25.756(7)
α (deg)	77.340(4)	97.533(3)	90.00	90.00
β (deg)	80.647(4)	101.555(3)	95.484(3)	94.977(4)
γ (deg)	61.820(4)	91.603(3)	90.00	90.00
Z	2	4	2	2
fw (g/mol)	910.45	904.46	1365.01	1359.02
density (calcd) (Mg/m^3)	1.398	1.384	1.314	1.282
abs coeff (mm^{-1})	3.944	3.626	2.503	2.265
F_{000}	920	1832	1420	1416
Total no. reflections	23271	43276	36346	28848
Unique reflections	9281	18382	14410	14458
R_{int}	0.0399	0.0376	0.0343	0.0444
final R indices [$I > 2\sigma(I)$]	R_1 = 0.0237 wR_2 = 0.0556	R_1 = 0.0381 wR_2 = 0.0810	R_1 = 0.0404, wR_2 = 0.1001	R_1 = 0.0623, wR_2 = 0.1615
largest diff peak and hole ($e^-Å^{-3}$)	1.235 and -0.448	6.071and -1.692	1.180 and -0.840	1.859 and -1.978
GOF	1.149	1.005	1.012	1.049

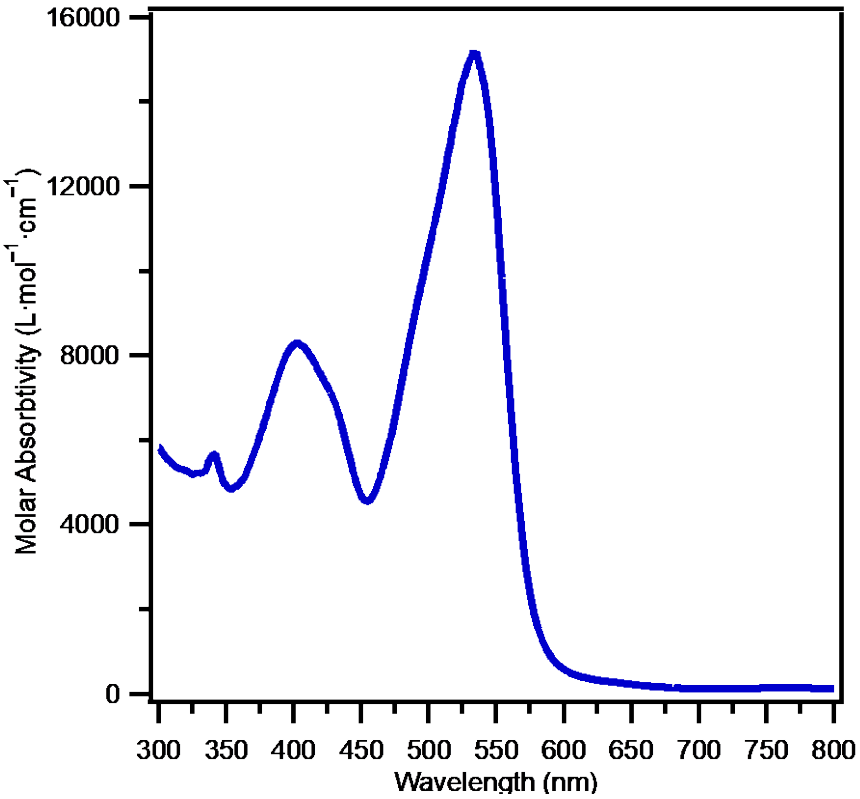

Figure S12. UV-Vis spectra of **4** (0.263 mM) in C_6H_6.

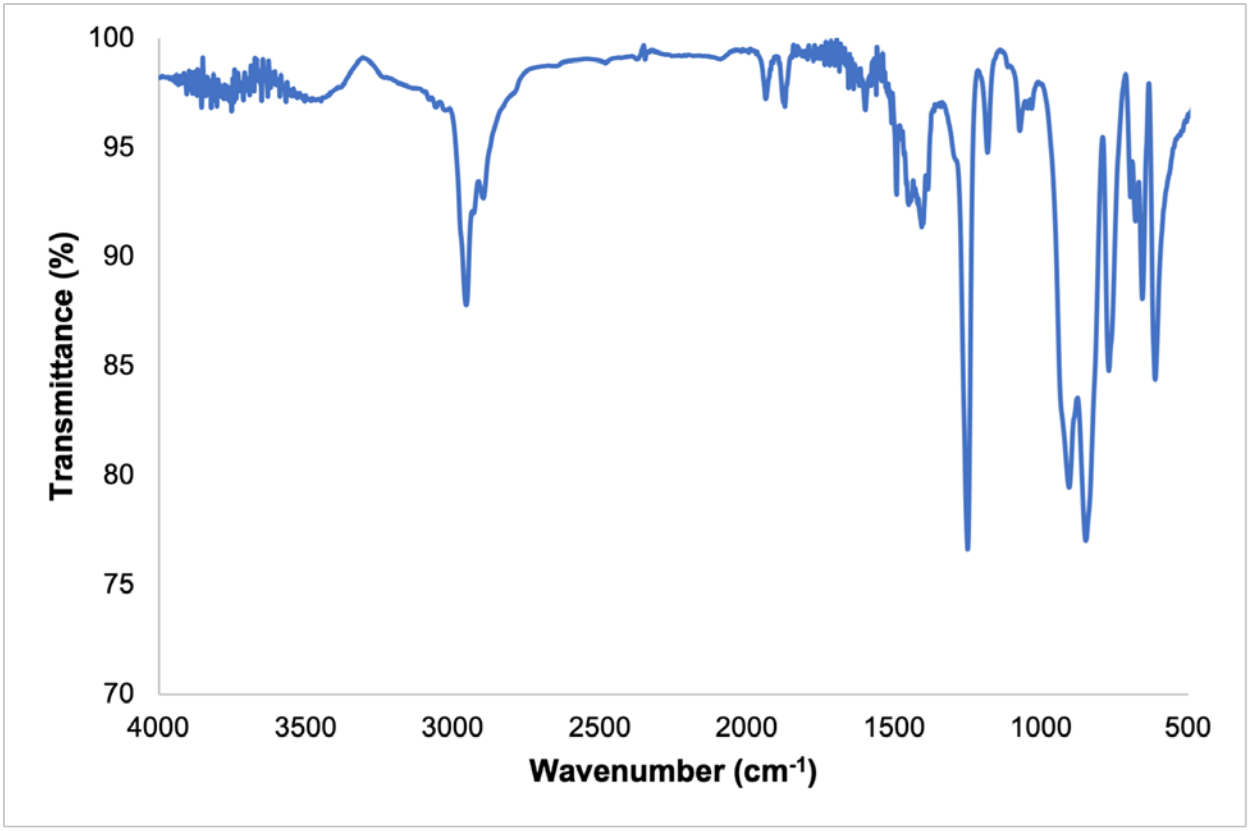

Figure S13. IR spectrum of **1** (KBr Pellet).

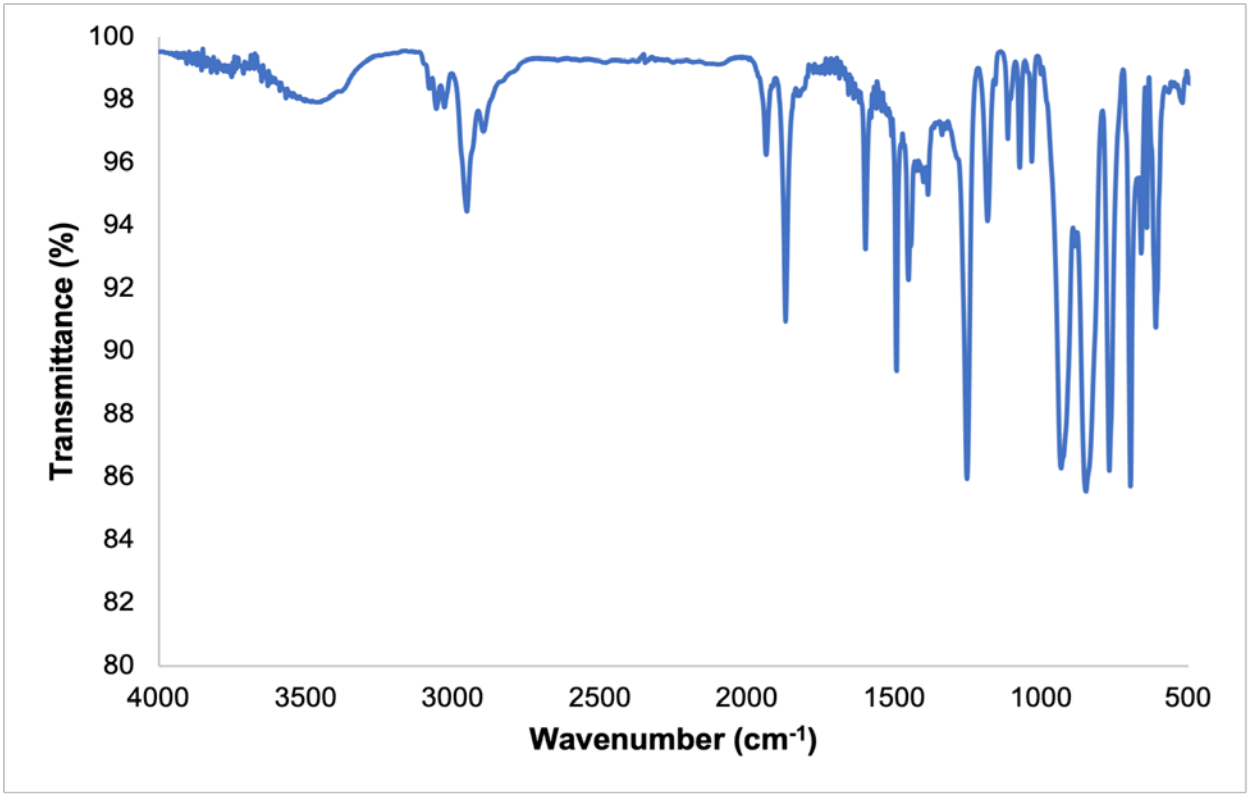

Figure S14. IR spectrum of **2** (KBr Pellet).

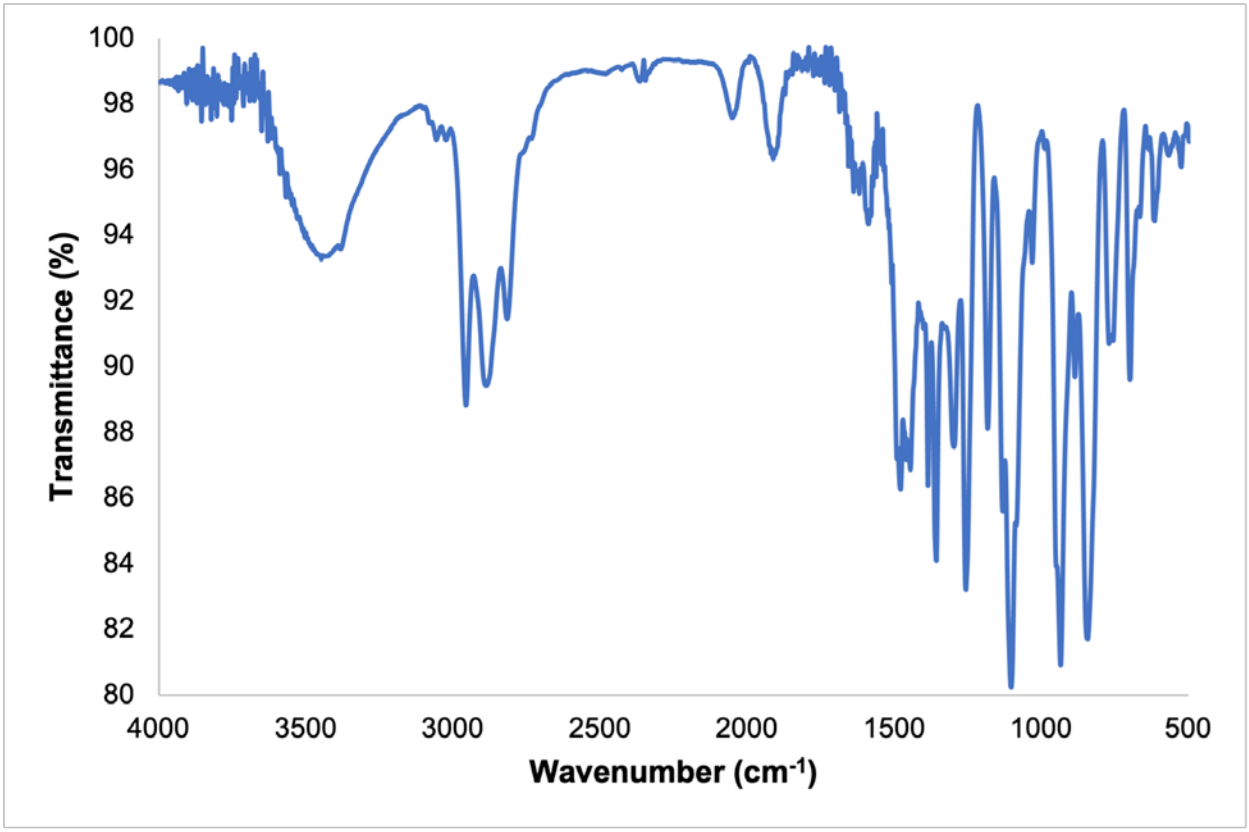

Figure S15. IR spectrum of **3** (KBr Pellet).

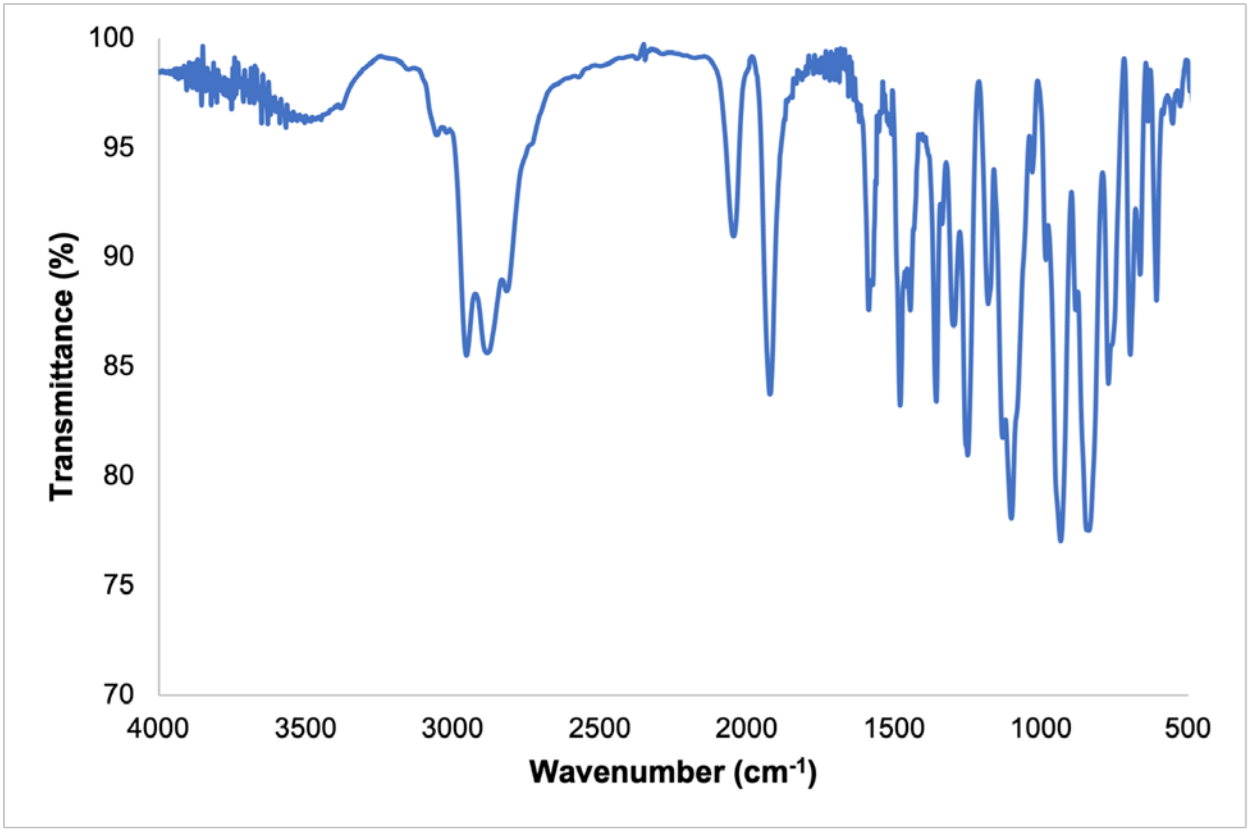

Figure S16. IR spectrum of **4** (KBr Pellet).

Cartesian Coordinates (unit: angstrom)

Compound **1**

U	0.00000000	0.00000000	2.45728744
C	0.00000000	0.00000000	0.00000000
C	-0.94729212	0.00067491	-0.88888076
C	-1.95677151	0.06762573	-1.75096813
H	1.02426414	0.16068345	-0.39023598
Si	-2.82509481	0.15671689	4.58269330
Si	-2.71430029	-1.95642368	2.40080172
Si	2.98538026	-1.44081347	1.62519121
Si	-0.40475195	3.24212554	1.72209668
Si	1.75985096	-2.50481724	4.17161513
Si	1.71589791	2.79481333	3.82169218
N	0.49052334	2.15684460	2.75974148
N	1.71581054	-1.38471360	2.83812883
N	-2.00027299	-0.62229209	3.26078731
C	-2.56752014	1.40934869	-1.96568001
C	-2.08026338	2.48355952	1.37372565
H	-2.11579304	1.42540474	1.06042029
H	-2.55519099	3.04070184	0.54604652
H	-2.72636459	2.58934850	2.26168686
C	-2.43508037	-1.11470577	-2.48923794
C	3.33918261	0.29048778	1.00376177
H	2.49027627	0.99681958	1.01130930
H	3.72160710	0.26198790	-0.03309476
H	4.12416456	0.75180852	1.63019883
C	-2.03530220	-2.40012237	-2.12702212
H	-1.39242741	-2.53202451	-1.25101770
C	-1.76935733	1.53744290	5.25309688
H	-0.80235067	1.16891141	5.64524638
H	-2.29282407	2.01047625	6.10461440
H	-1.54114254	2.32752142	4.52074412
C	-1.77315451	2.51657278	-2.19145762
H	-0.69081763	2.37639652	-2.27806421
C	0.55281603	3.60523784	0.16627748
H	1.51264874	4.09794777	0.39892100
H	-0.01584752	4.27225849	-0.50477735
H	0.77565188	2.68056529	-0.39138182
C	-1.31640753	-2.90522036	1.57815470
H	-0.62454100	-2.32048210	0.94319080
H	-1.77121675	-3.64845830	0.89715101
H	-0.71382762	-3.46225850	2.31389789
C	-3.25098740	-0.99523853	-3.62721754
H	-3.56920293	0.00021310	-3.95285072
C	-3.95291715	1.59459313	-1.87666510
H	-4.59608557	0.72726170	-1.69086882
C	0.20234388	-2.33110030	5.18419256
H	0.07521232	-1.31452805	5.60295492

H	0.27156810	-3.01519930	6.05073808
H	-0.72390382	-2.57447587	4.64241451
C	2.48803602	-2.57864208	0.22830948
H	2.28701104	-3.59354518	0.61472475
H	3.28752350	-2.66272364	-0.53036329
H	1.57336669	-2.23495734	-0.28462171
C	-2.31432090	3.78884653	-2.28267941
H	-1.66265876	4.65262403	-2.45378579
C	-3.92813474	-1.40024259	1.10769340
H	-4.80565966	-0.90373646	1.55708189
H	-4.29169448	-2.25498148	0.50903209
H	-3.46540072	-0.68674451	0.40360400
C	3.16084666	-2.22253832	5.37524480
H	4.15086118	-2.47144366	4.96072712
H	3.00673678	-2.86572685	6.26151517
H	3.20081992	-1.17832754	5.72861181
C	-4.46254843	0.83628179	4.03636968
H	-4.38108223	1.45813650	3.12897007
H	-4.91993457	1.45485338	4.83014422
H	-5.17632924	0.02358891	3.80971282
C	4.62740980	-2.06730481	2.26868528
H	5.02531928	-1.44082211	3.08545566
H	5.35916065	-2.02775638	1.44009729
H	4.59779448	-3.11093637	2.62365078
C	-3.68519343	3.94804101	-2.18890048
H	-4.12527156	4.94877154	-2.27094113
C	-3.15083806	-0.94915158	6.06675983
H	-3.89021711	-1.74192798	5.86976634
H	-3.54690577	-0.32712108	6.89080442
H	-2.23090202	-1.43591180	6.43306660
C	-2.42539641	-3.50566026	-2.86758913
H	-2.09043584	-4.50202781	-2.55593366
C	-3.58103389	-3.20027713	3.50482898
H	-2.95100769	-3.54010272	4.34478319
H	-3.84726338	-4.09038876	2.90541594
H	-4.51973956	-2.80219502	3.92769347
C	-4.49784225	2.85648371	-1.98797656
H	-5.58304770	2.98672076	-1.90718200
C	1.85828587	-4.26883245	3.61189385
H	1.03567190	-4.54400907	2.92916459
H	1.80413573	-4.94783468	4.48274410
H	2.80234736	-4.49550317	3.08596531
C	-3.63088117	-2.10375856	-4.36170143
H	-4.25823506	-1.96723999	-5.25001864
C	2.58021061	1.39450121	4.70686267
H	1.87934792	0.81109029	5.33371500
H	3.34318691	1.81144089	5.38972790
H	3.08261011	0.68249029	4.03295292
C	1.06252693	3.88958065	5.19563166

H	0.66011241	4.85196700	4.84221492
H	1.89528168	4.11708616	5.88706582
H	0.27288666	3.39382708	5.78495068
C	-0.79374027	4.88568811	2.51495717
H	-1.32247392	4.77963891	3.47796298
H	-1.45755546	5.45246547	1.83573161
H	0.10138324	5.50815646	2.68603009
C	2.98801907	3.78531176	2.89543571
H	3.43663362	3.21426373	2.06397580
H	3.80702950	4.08763901	3.57354232
H	2.56299679	4.71145130	2.46968655
C	-3.21793336	-3.36055309	-3.98870166
H	-3.51732002	-4.23969060	-4.57022724

Compound **2**

Th	0.00000000	0.00000000	2.52835735
C	-0.00000000	0.00000000	0.00000000
C	0.33030625	-0.90060817	-0.86551977
C	0.65710632	-1.89596474	-1.67909978
H	-0.29854831	0.81636964	-0.38607021
Si	-2.86057349	-0.45218505	4.52234631
Si	0.70867323	-3.30941995	2.56062547
Si	2.42048980	-1.78122193	4.53812057
Si	-3.30971938	0.75964206	1.76194118
Si	0.54263407	3.39981979	3.68123742
Si	2.49226541	2.20289657	1.75913413
N	-2.25159540	0.14807475	3.00596917
N	1.15640247	-1.81785636	3.33882169
N	1.05341604	2.03186860	2.73395043
C	-0.95432542	-3.02752350	1.71767757
H	-1.62496795	-2.78474761	2.38869919
H	-1.23248623	-3.84964639	1.25991897
H	-0.87106751	-2.30196577	1.06199327
C	-0.06848652	-4.28769173	-2.02626431
H	0.83388168	-4.56577657	-1.92150451
C	-2.51699642	2.28396900	0.98208319
H	-2.73742707	3.07627035	1.51500044
H	-2.85676432	2.39947782	0.06860113
H	-1.54322261	2.16879292	0.95499284
C	-3.60889123	-0.54588670	0.46013008
H	-2.75786701	-0.78862363	0.03992047
H	-4.22121558	-0.19355631	-0.21920377
H	-4.00622030	-1.33749246	0.87776442
C	-0.38665030	-2.92456686	-1.96151019
C	1.92610462	-3.83189236	1.25074403
H	2.08195801	-3.08606806	0.63498981
H	1.56519126	-4.59529887	0.75472508
H	2.77174184	-4.08756966	1.67490135
C	-1.72554120	-2.56067474	-2.09139695

H	-1.96574828	-1.64298339	-2.04264368
C	3.26908009	0.50497175	1.49498703
H	3.80635609	0.26773207	2.27963515
H	3.84170648	0.52790277	0.70011269
H	2.56207577	-0.16074312	1.36925778
C	-3.85605290	0.75770371	5.54266347
H	-3.38959363	1.61905963	5.57704608
H	-3.96269327	0.40683336	6.45118482
H	-4.73892625	0.88133689	5.13361372
C	-1.06132874	-5.23607877	-2.23923435
H	-0.82583806	-6.15532824	-2.29665410
C	-4.97068850	1.32563426	2.39646913
H	-5.42033487	0.57945704	2.84495989
H	-5.51974317	1.63399216	1.64496860
H	-4.84398079	2.06247004	3.03059225
C	-2.71902583	-3.51712807	-2.29053000
H	-3.62761007	-3.25026189	-2.37249194
C	0.48073313	-4.73447486	3.74393242
H	1.34732790	-4.97684695	4.13428306
H	0.11092438	-5.50251265	3.26357500
H	-0.13468825	-4.46873339	4.45886255
C	2.01054481	-2.69918666	6.12811578
H	1.22673755	-2.28821115	6.54937703
H	2.77346789	-2.65058290	6.74147907
H	1.81686831	-3.63736283	5.92430363
C	-1.15686632	3.14007374	4.39635850
H	-1.78169717	2.90507060	3.67966866
H	-1.45754182	3.96624460	4.83232916
H	-1.12674041	2.41647658	5.05570426
C	2.27002039	-2.73542561	-3.42133797
H	1.57440745	-3.25517428	-3.80716904
C	-3.92379910	-1.97361141	4.31420218
H	-4.62950008	-1.79258930	3.65907750
H	-4.32962596	-2.20872345	5.17435308
H	-3.37012823	-2.71977849	4.00155822
C	2.00395922	-1.96685595	-2.30499840
C	-2.38195736	-4.86634829	-2.36742018
H	-3.05476476	-5.52262174	-2.50673361
C	-1.35684294	-0.86733448	5.57155492
H	-0.91187935	-1.66027916	5.20596692
H	-1.64033943	-1.04557186	6.49125300
H	-0.73402124	-0.11089686	5.56204275
C	2.67020145	-0.01503601	5.11643615
H	2.87506924	0.55425114	4.34736164
H	3.41440829	0.01738373	5.75349184
H	1.85322894	0.30507555	5.55167729
C	4.02339323	-2.49487489	3.88792911
H	3.94030400	-3.46721973	3.81494803
H	4.75268152	-2.27436787	4.50361758

H	4.21466489	-2.11489240	3.00478431
C	2.12357493	2.95434866	0.09734273
H	1.50066811	2.37596663	-0.39176485
H	2.95528444	3.04404859	-0.41230883
H	1.71918479	3.83856561	0.22201949
C	3.85387771	3.23790945	2.55305061
H	3.52409913	4.14720299	2.70937921
H	4.63000084	3.27177507	1.95524370
H	4.11653018	2.83475730	3.40641100
C	4.55307971	-1.99311263	-3.47093952
H	5.41594674	-2.00647024	-3.87025753
C	1.63424551	3.71774425	5.15687276
H	1.61015465	2.94181216	5.75488813
H	1.31249844	4.50857549	5.63668416
H	2.55427080	3.86860156	4.85610389
C	0.51130861	4.95880086	2.64646575
H	1.39203626	5.10005241	2.24195120
H	0.28658545	5.72427749	3.21550043
H	-0.16320617	4.86817682	1.94175434
C	3.54072349	-2.76360940	-3.98979919
H	3.70814618	-3.31760178	-4.74182438
C	3.05526995	-1.19211357	-1.79890515
H	2.89746578	-0.64394023	-1.03925612
C	4.31246232	-1.20035965	-2.36828177
H	5.00705301	-0.66217783	-2.00368071

Compound **3**

U	0.00000000	-0.00000000	2.30439631
C	0.00000000	0.00000000	0.00000000
C	-0.14324712	-0.00330987	-1.21315422
C	-0.31355157	0.07081719	-2.60142708
N	1.13438998	1.91204657	2.81585501
N	-2.23811023	-0.02828677	2.57235230
N	1.07098940	-1.93540012	2.78572272
Si	2.24652247	2.00469045	4.14723090
Si	-3.17024394	-0.87511506	1.38358011
Si	-2.98361537	0.88693483	3.84624440
Si	2.47428142	-2.29162844	1.81353584
Si	0.53307491	-3.05827448	3.98899166
Si	0.89012387	3.26275611	1.77725536
C	-0.39557568	1.41596947	-3.18213318
C	-0.01752273	1.73170013	-4.49966215
C	-4.14937998	2.20164830	3.20499468
C	-0.81371582	2.47917738	-2.39510652
C	2.39842186	0.34702011	4.97036445
C	-0.07303125	3.05407262	-4.97032265
C	-0.85954361	3.75856532	-2.88984745
C	-0.88908280	-1.25669935	-4.67590301
C	-1.02144849	-2.40974010	4.78823714

C	-0.33990500	-1.14785778	-3.37397410
C	0.16741715	-3.55172031	-3.55853544
C	-0.38213334	-3.54645178	-4.83541062
C	1.72489958	3.16292012	5.51033657
C	-0.89197396	3.31203271	1.23862575
C	-3.93098344	-0.13927019	5.11676656
C	-2.25434031	-2.36174274	0.79880036
C	2.00318812	-3.42767126	0.37904505
C	1.22922885	4.91984279	2.59627494
C	-4.80850701	-1.51882956	2.07060465
C	-0.87922778	-2.43934589	-5.37438416
C	0.13483212	-2.36174824	-2.82780178
C	-1.63914831	1.75260343	4.83068874
C	-3.61119466	0.21354544	-0.07195947
C	1.72221662	-3.32772205	5.38284498
C	-0.46478155	4.04483726	-4.17394929
C	3.88338055	-3.10194484	2.75623719
C	3.18468092	-0.70928394	1.14103349
C	0.15956461	-4.74348810	3.26009938
C	3.93515815	2.53187229	3.58755972
C	1.94930740	3.15611137	0.25562551
H	0.36648446	0.94717647	-5.15772414
H	-3.64489262	2.85716451	2.47454497
H	-4.53486463	2.83645590	4.02440848
H	-5.02147109	1.75590866	2.69337074
H	-1.12398506	2.28200715	-1.36594100
H	1.44175654	0.02299888	5.42306117
H	3.13331651	0.40563641	5.79617323
H	2.71693518	-0.46148443	4.29371911
H	0.24857705	3.24946072	-6.00223380
H	-1.20346619	4.56773473	-2.23099190
H	-1.34929480	-0.37243957	-5.12851165
H	-0.84421345	-1.45564475	5.32131598
H	-1.38766387	-3.12965019	5.54401278
H	-1.83767498	-2.22896065	4.06982256
H	0.57704738	-4.46349205	-3.11093111
H	-0.37552997	-4.48196963	-5.41384860
H	1.73718963	4.22325701	5.20994079
H	2.40978280	3.05783174	6.37352291
H	0.70668211	2.93159845	5.86807207
H	-1.26517726	2.33866165	0.86660540
H	-1.02318206	4.03106907	0.40914858
H	-1.55546884	3.62685813	2.06383814
H	-4.83667614	-0.60994689	4.70081479
H	-4.24717620	0.51185407	5.95366468
H	-3.29984902	-0.94064845	5.53689952
H	-1.32558375	-2.09780156	0.26796406
H	-2.87743757	-2.93374661	0.08435614
H	-1.99933661	-3.04468280	1.62929212

H	1.87555260	-4.47532788	0.70485265
H	2.74831849	-3.41176431	-0.43659168
H	1.03943860	-3.09064221	-0.03569077
H	0.56466557	5.10143608	3.45951977
H	1.04032271	5.72708419	1.86332139
H	2.27033950	5.03693482	2.94286566
H	-4.66609131	-2.20397078	2.92545903
H	-5.33181865	-2.08423384	1.27696448
H	-5.48872174	-0.71148442	2.39635565
H	-1.32075141	-2.45972399	-6.37995619
H	0.56037111	-2.33347622	-1.82051638
H	-0.94936963	1.03956304	5.32259107
H	-2.10138883	2.34746010	5.64157297
H	-1.02194446	2.43435906	4.22400426
H	-4.08008266	1.15865252	0.25645771
H	-4.31856047	-0.29237000	-0.75502618
H	-2.70738248	0.46302886	-0.65163902
H	2.63565471	-3.86684129	5.08339964
H	1.23660600	-3.92413167	6.17884516
H	2.04007486	-2.37230643	5.83601426
H	-0.50469941	5.08003183	-4.53816930
H	4.18015474	-2.51874797	3.64580655
H	4.76573023	-3.16796509	2.09227097
H	3.64842361	-4.12848082	3.08827723
H	2.46039763	-0.12976192	0.53734549
H	4.02793637	-0.94936072	0.46618182
H	3.57554654	-0.05271709	1.93702891
H	-0.52958122	-4.66683865	2.40109236
H	-0.30040560	-5.41748378	4.00680326
H	1.07934169	-5.23586314	2.89557852
H	4.33925984	1.84479801	2.82296622
H	4.65601919	2.55668079	4.42661808
H	3.92431779	3.53911351	3.13578513
H	3.02554647	3.11457889	0.50408722
H	1.78745108	4.00912363	-0.42893118
H	1.69621637	2.23753070	-0.30254106

Compound **4**

Th	0.00000000	0.00000000	2.36739322
C	0.00000000	0.00000000	0.00000000
C	0.17314732	0.00933229	-1.22374792
C	0.32124163	-0.10335879	-2.61335178
N	-1.20462656	-1.90038245	2.88982102
N	2.30229067	-0.11163034	2.71585493
N	-1.01058007	2.03853123	2.85577971
Si	-2.35656244	-1.97145256	4.20258928
Si	3.29631206	0.70610636	1.56332019
Si	2.97100571	-1.04592685	3.99423481
Si	-2.40255158	2.43047026	1.87517206

Si	-0.41260722	3.11732665	4.08834625
Si	-1.03727504	-3.27975563	1.83372106
C	0.41313942	-1.44867451	-3.19950436
C	0.01507098	-1.75226946	-4.53052002
H	-0.37404070	-0.99266983	-5.20894971
C	4.08574293	-2.43056883	3.37748094
H	3.55932319	-3.05938536	2.63871991
H	4.42352898	-3.08289436	4.20397775
H	4.98733122	-2.03042410	2.87998230
C	0.83373897	-2.49710869	-2.41689925
H	1.15961271	-2.27331788	-1.39565609
C	-2.39610880	-0.30327613	4.99733447
H	-1.42552084	-0.04325030	5.46427133
H	-3.13871553	-0.28839549	5.81804881
H	-2.65000821	0.52249830	4.31255096
C	0.09316192	-3.13434981	-4.94131680
H	-0.22490362	-3.33938999	-5.97585983
C	0.85842057	-3.81202351	-2.85478134
H	1.19145122	-4.62133511	-2.19603551
C	0.83609426	1.20760587	-4.72038903
H	1.30224862	0.32033001	-5.16053792
C	1.08962100	2.35165098	4.88053711
H	0.83125884	1.42484431	5.42960042
H	1.51169733	3.04589197	5.63149637
H	1.89912745	2.09423973	4.17718045
C	0.31778182	1.11677091	-3.39952054
C	-0.15963985	3.49000917	-3.55546832
H	-0.55545936	4.40463766	-3.09977447
C	0.25961586	3.45938500	-4.83277515
H	0.20817914	4.40288531	-5.40397537
C	-1.92748242	-3.18531489	5.61152203
H	-2.02723247	-4.24071331	5.31017584
H	-2.61093517	-3.01307762	6.46414391
H	-0.89521845	-3.03259665	5.97159750
C	0.73268002	-3.35080007	1.27409598
H	1.07904115	-2.38932659	0.84668008
H	0.85505815	-4.09576757	0.46670177
H	1.42268927	-3.62206551	2.09269760
C	3.92166344	-0.08063961	5.30654711
H	4.86228361	0.35085992	4.92685021
H	4.17689925	-0.75081626	6.14893022
H	3.31349972	0.74739705	5.71011372
C	2.49594937	2.20718215	0.98351008
H	3.14749741	2.73132308	0.25882321
H	2.28670142	2.92045941	1.80229095
H	1.55097780	2.00727494	0.44493799
C	-1.82658968	3.53154085	0.38340707
H	-1.62864078	4.57463241	0.68715295
H	-2.55874485	3.54188542	-0.44330899

H	-0.88499700	3.10427866	0.00211641
C	-1.47430191	-4.95579130	2.65925683
H	-0.82930582	-5.17267943	3.52862133
H	-1.31916755	-5.76092187	1.91732343
H	-2.52505092	-5.01165661	2.98990460
C	4.99721210	1.26532157	2.29105732
H	4.88278914	1.96193695	3.14082190
H	5.56432901	1.79059974	1.50029815
H	5.61472130	0.41416005	2.62856521
C	0.81328699	2.39705705	-5.43731374
H	1.20542828	2.44388346	-6.45914950
C	-0.14349385	2.29384749	-2.82963333
H	-0.52310770	2.26570872	-1.80489803
C	1.54952842	-1.83916572	4.92984389
H	0.89772439	-1.09095777	5.42406900
H	1.94933314	-2.47422503	5.74327564
H	0.89993791	-2.47592250	4.30670346
C	3.69909317	-0.46973117	0.14530063
H	4.07681393	-1.43117098	0.53513344
H	4.45896825	-0.06001698	-0.54541984
H	2.78603740	-0.67005168	-0.44119524
C	-1.56422621	3.42762637	5.51255762
H	-2.47368202	3.98207352	5.22647757
H	-1.04564157	4.02126362	6.28888761
H	-1.89172010	2.48263611	5.98203095
C	0.42581050	-4.06341706	-4.17845182
H	0.44516446	-5.10731907	-4.52842292
C	-3.80629424	3.29513140	2.80940001
H	-4.13095439	2.70702895	3.68640689
H	-4.67670873	3.40506945	2.13663820
H	-3.53162847	4.30489212	3.16193790
C	-3.09808494	0.88317233	1.19687052
H	-2.36743435	0.32559827	0.57512519
H	-3.94247581	1.12214677	0.52271115
H	-3.49011380	0.19950263	1.97101794
C	0.01177133	4.80307697	3.35278286
H	0.69978926	4.70207146	2.49518814
H	0.48002640	5.47891418	4.09214166
H	-0.90213052	5.29969133	2.97987659
C	-4.05077374	-2.40516306	3.62092838
H	-4.41037018	-1.69639978	2.85363777
H	-4.78126779	-2.39514029	4.45173525
H	-4.08805862	-3.41050498	3.16687362
C	-2.10252997	-3.09690562	0.29809690
H	-3.17657250	-3.00973658	0.54256392
H	-1.97251230	-3.94540684	-0.39825406
H	-1.79732896	-2.18111782	-0.23926795

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