Supporting Information

1. NMR Spectrum of the prepared compounds

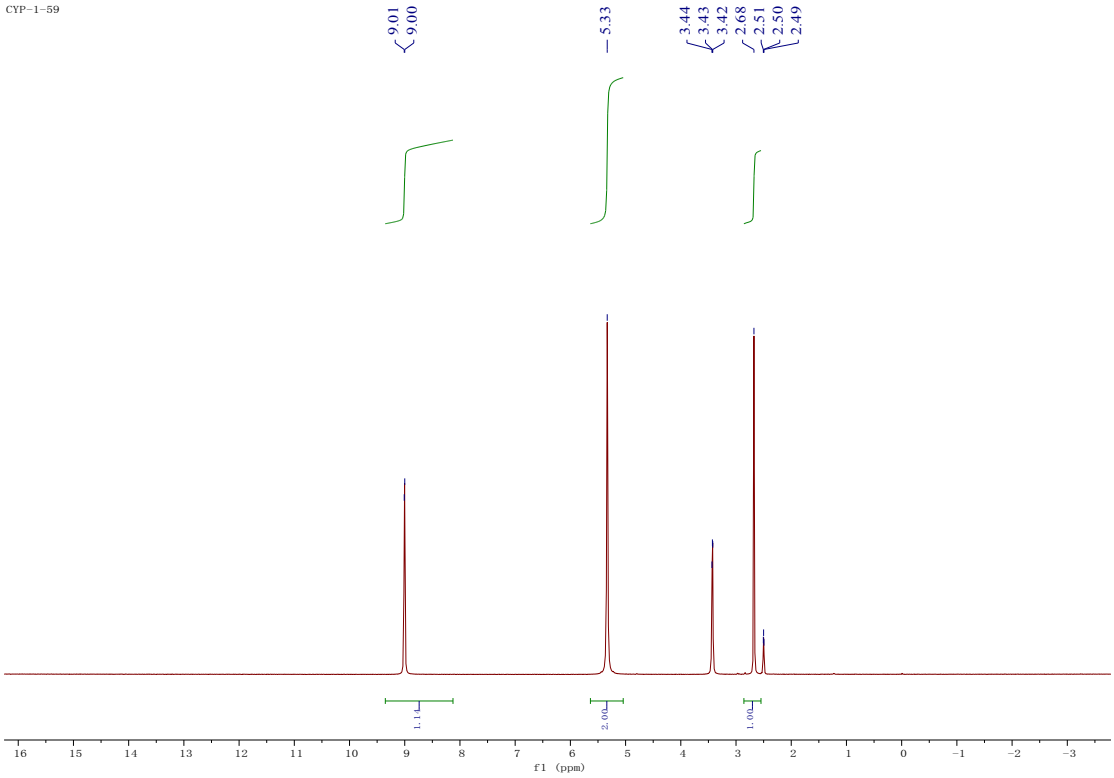


Figure S1 ^{1}H NMR spectrum of dihydroxyimino-malonic acid diamide.

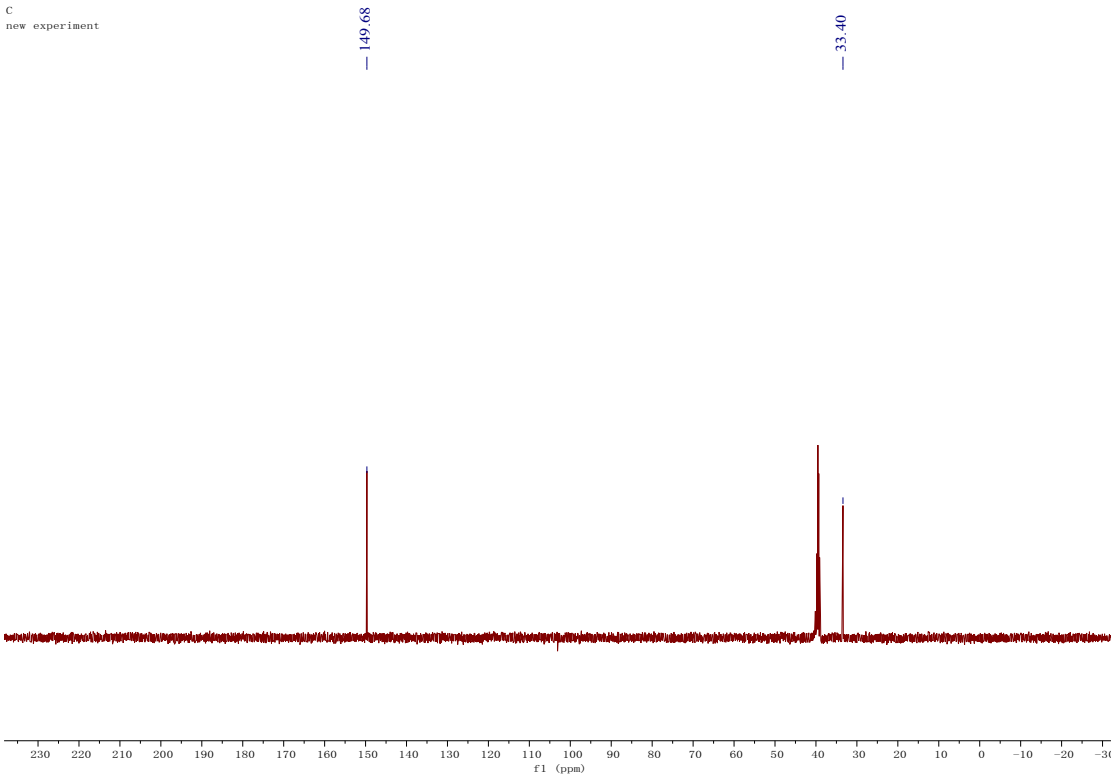


Figure S2 ^{13}C NMR spectrum of dihydroxyimino-malonic acid diamide.

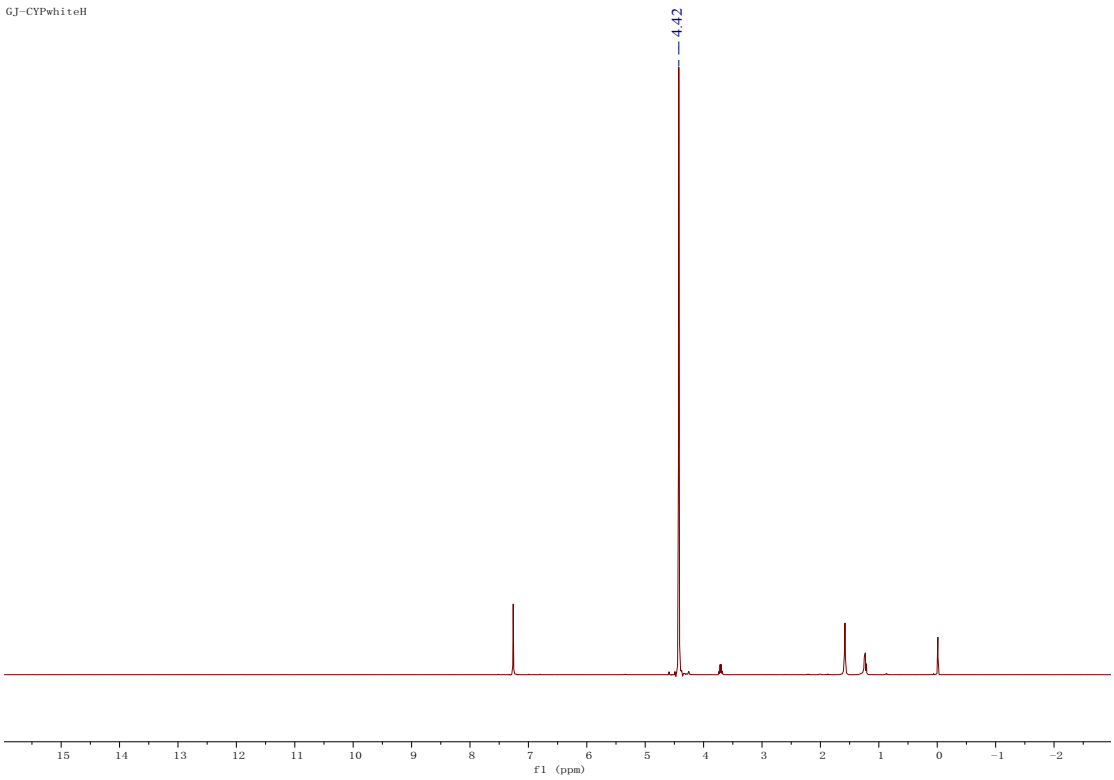


Figure S3 ^{1}H NMR spectrum of compound **1**.

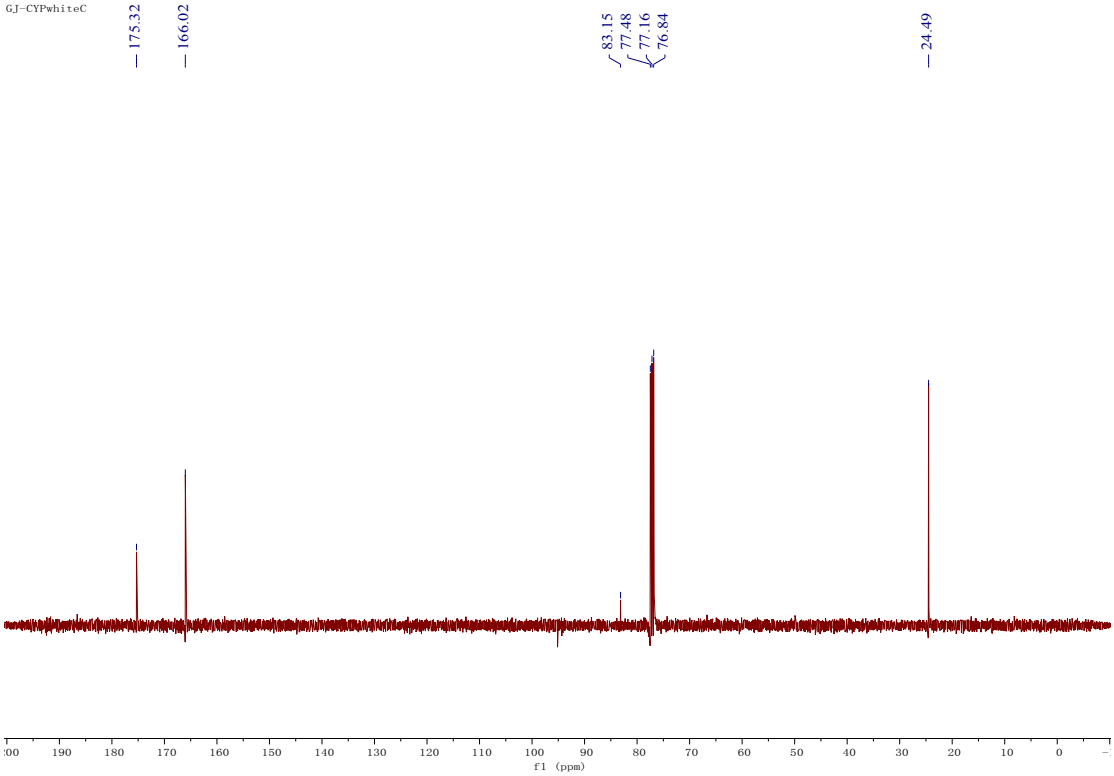


Figure S4 ^{13}C NMR spectrum of compound **1**.

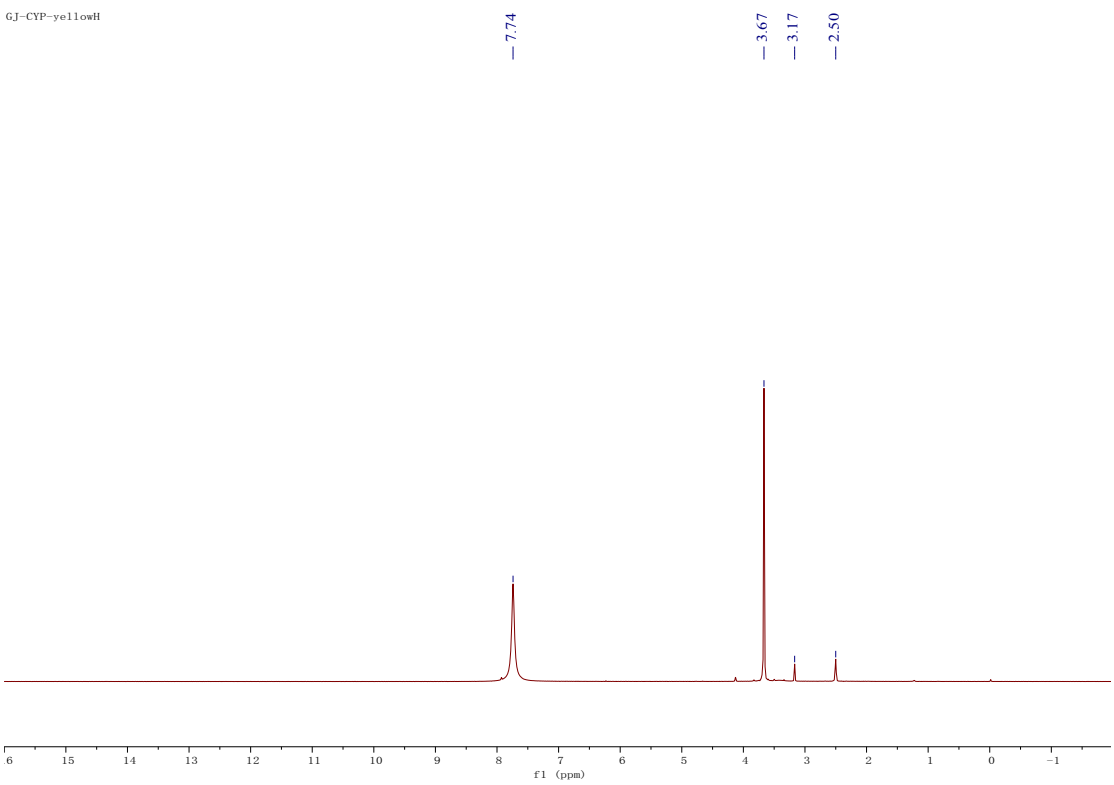


Figure S5 ^{1}H NMR spectrum of compound **2**.

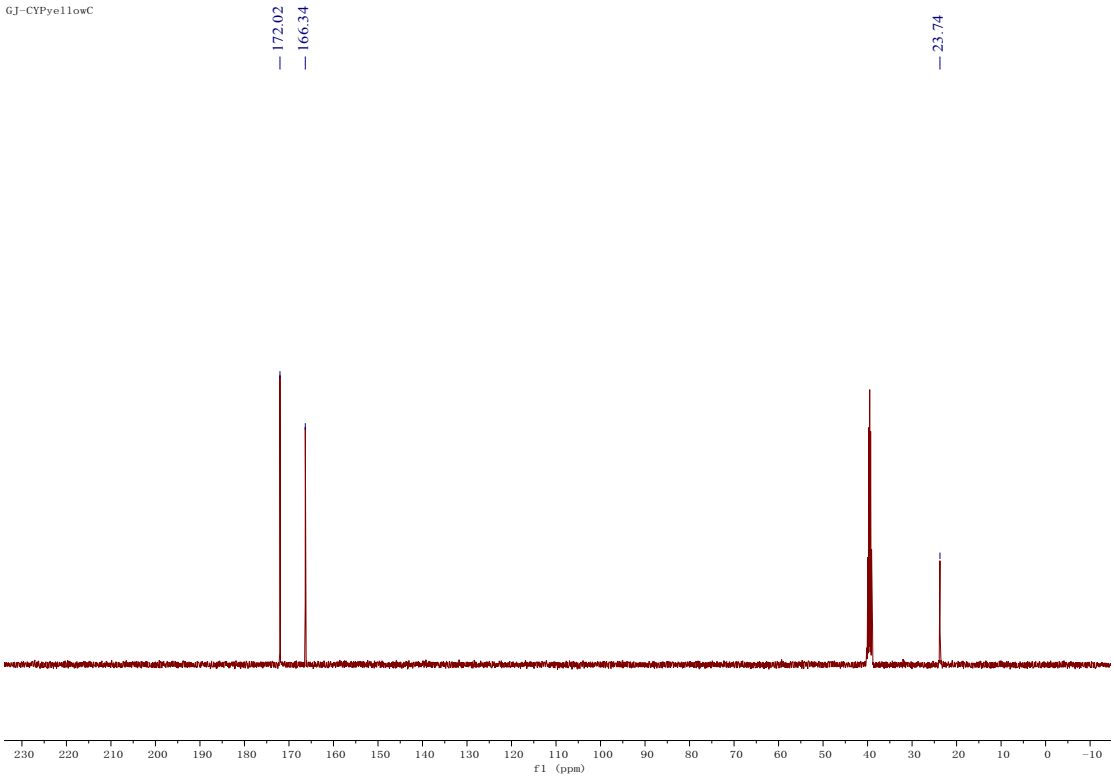


Figure S6 ^{13}C NMR spectrum of compound **2**.

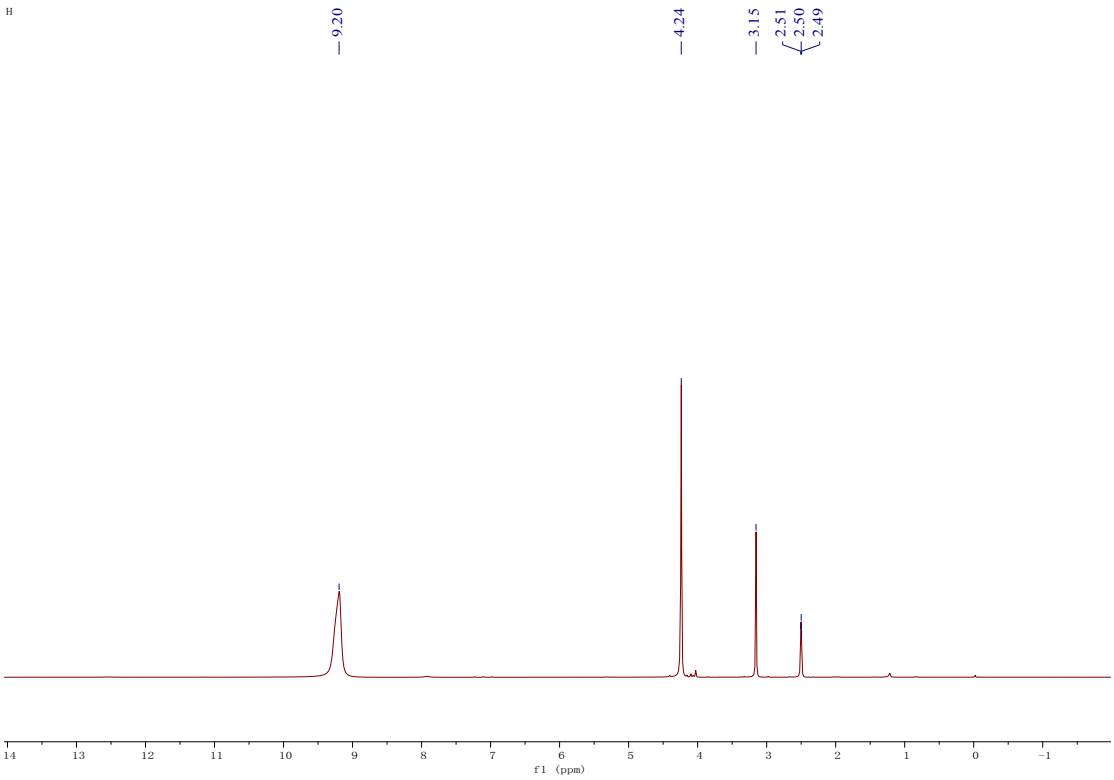


Figure S7 ^{1}H NMR spectrum of compound **3**.

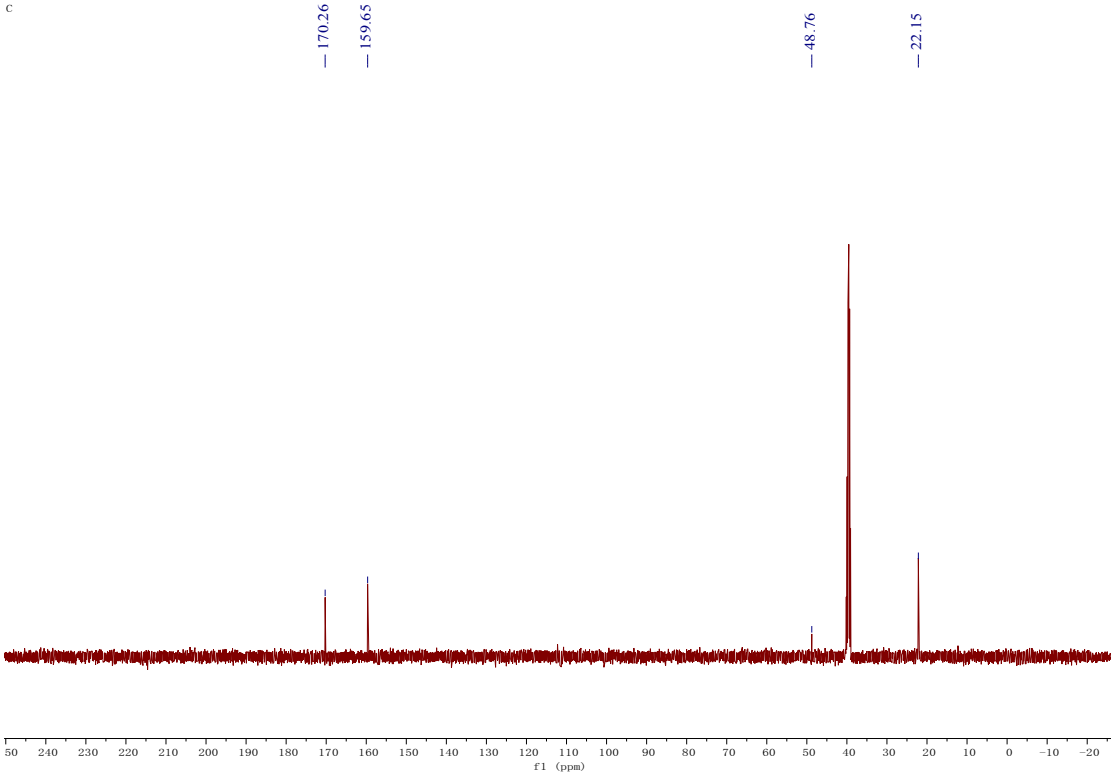


Figure S8 ^{13}C NMR spectrum of compound **3**.

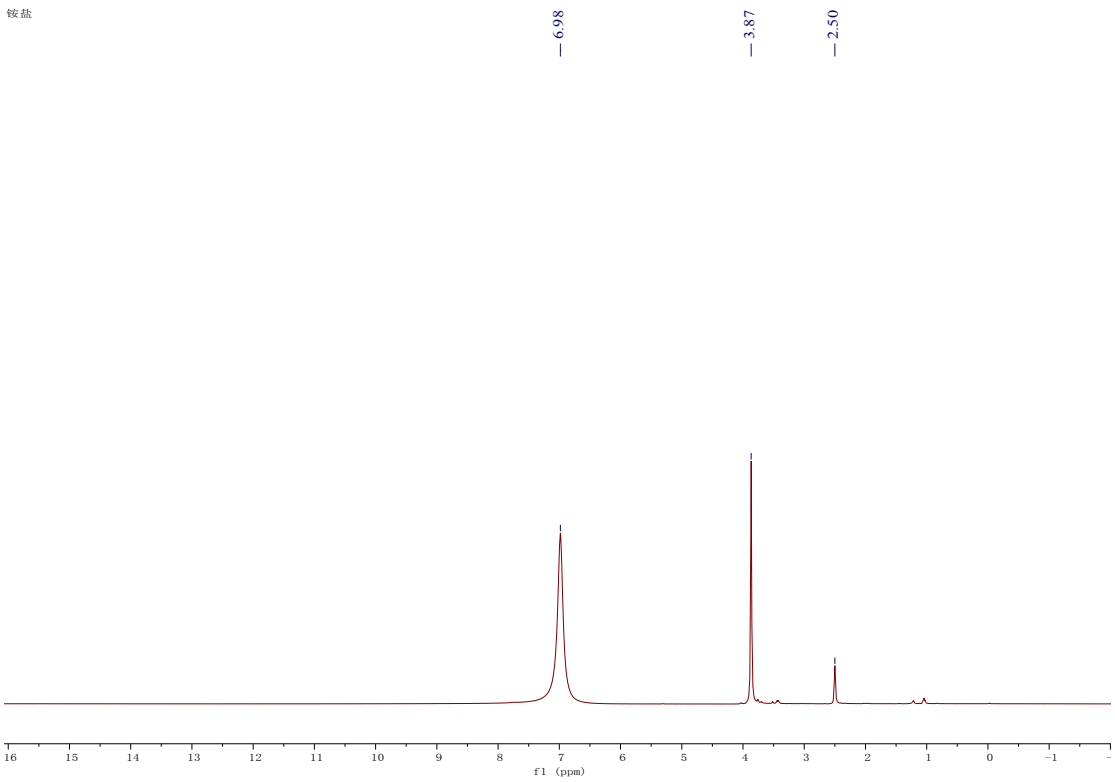


Figure S9 ^{1}H NMR spectrum of compound **4**.

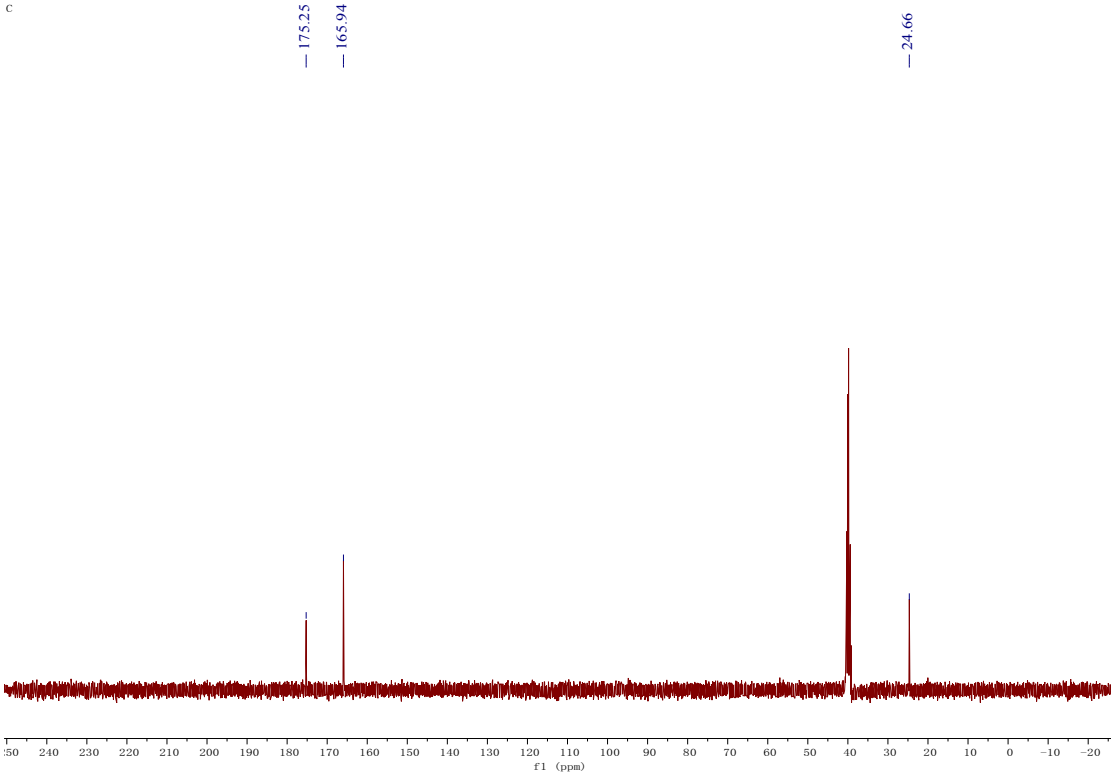


Figure S10 ^{13}C NMR spectrum of compound **4**.

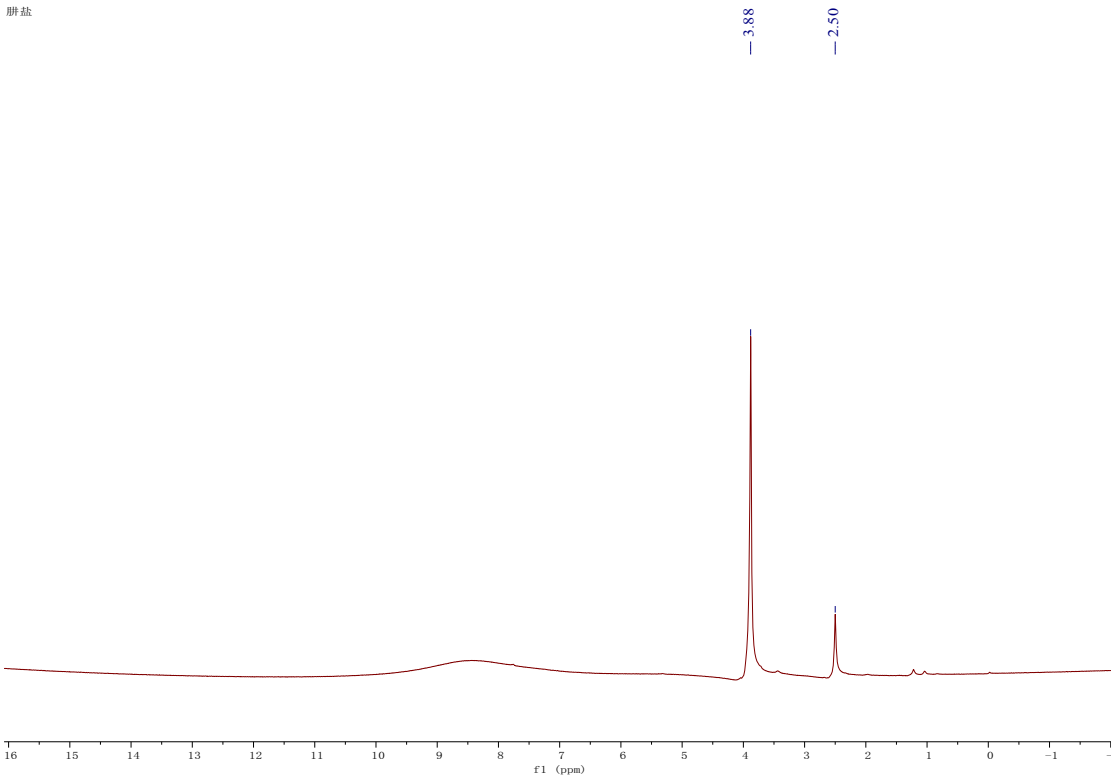


Figure S11 ^{1}H NMR spectrum of compound **5**.

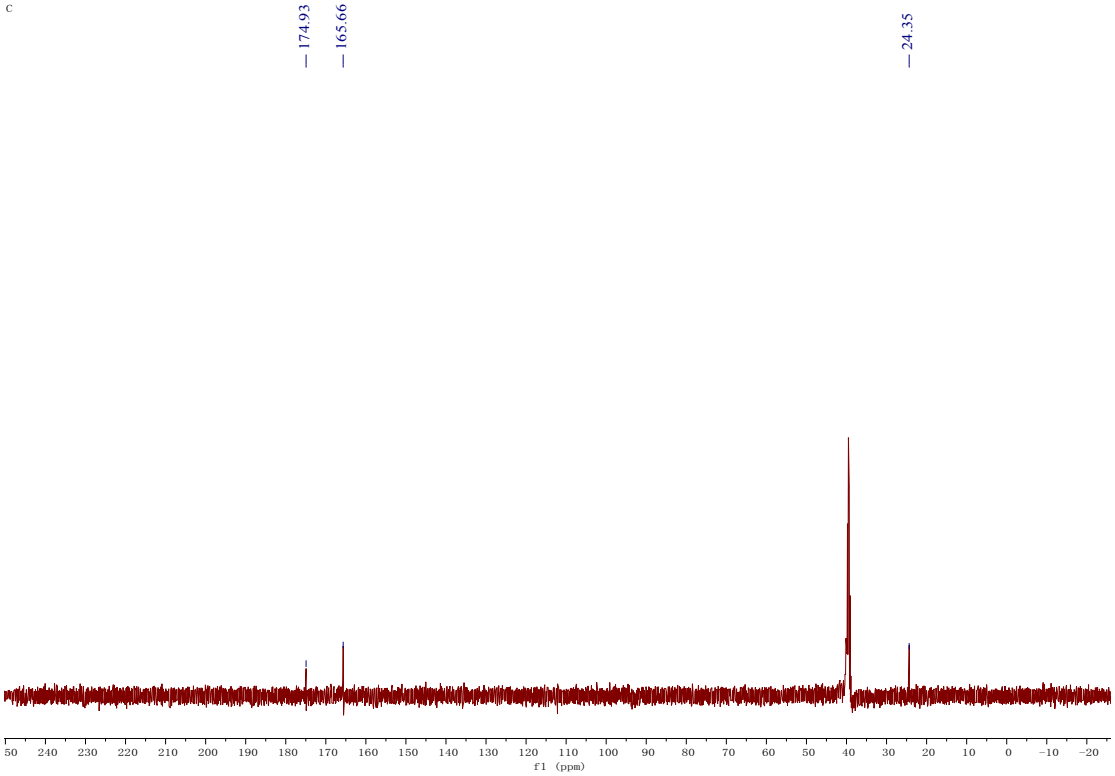


Figure S12 ^{13}C NMR spectrum of compound **5**.

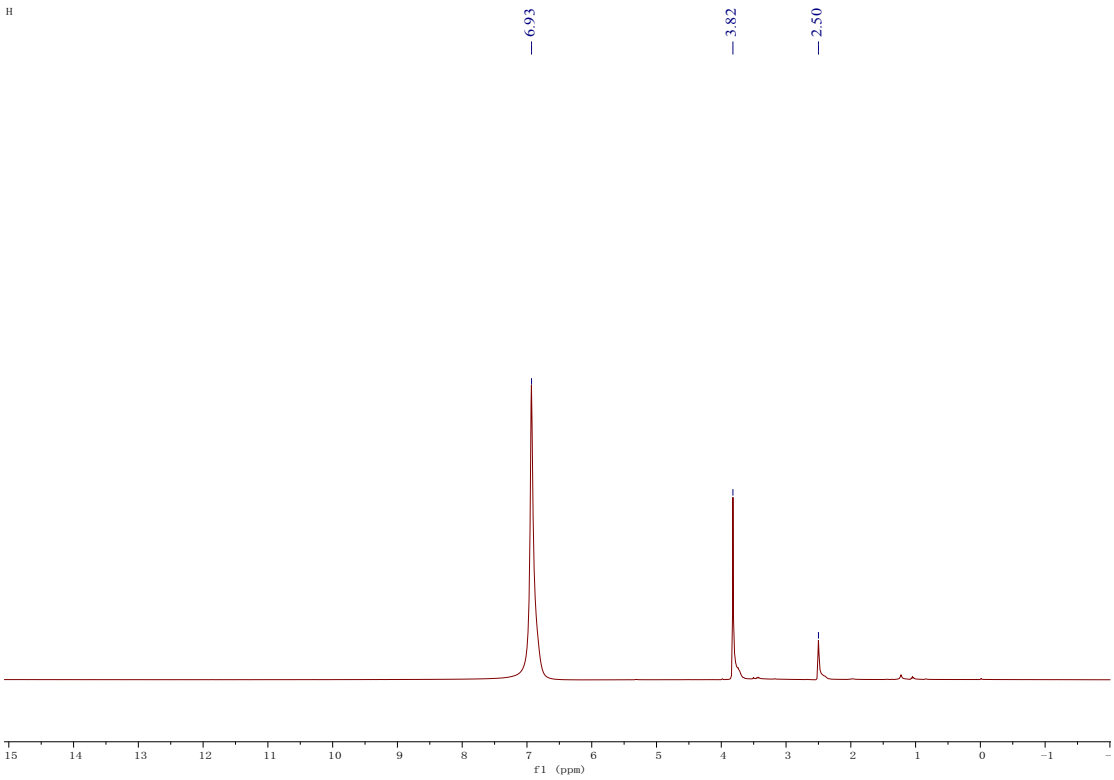


Figure S13 ^{1}H NMR spectrum of compound **6**.

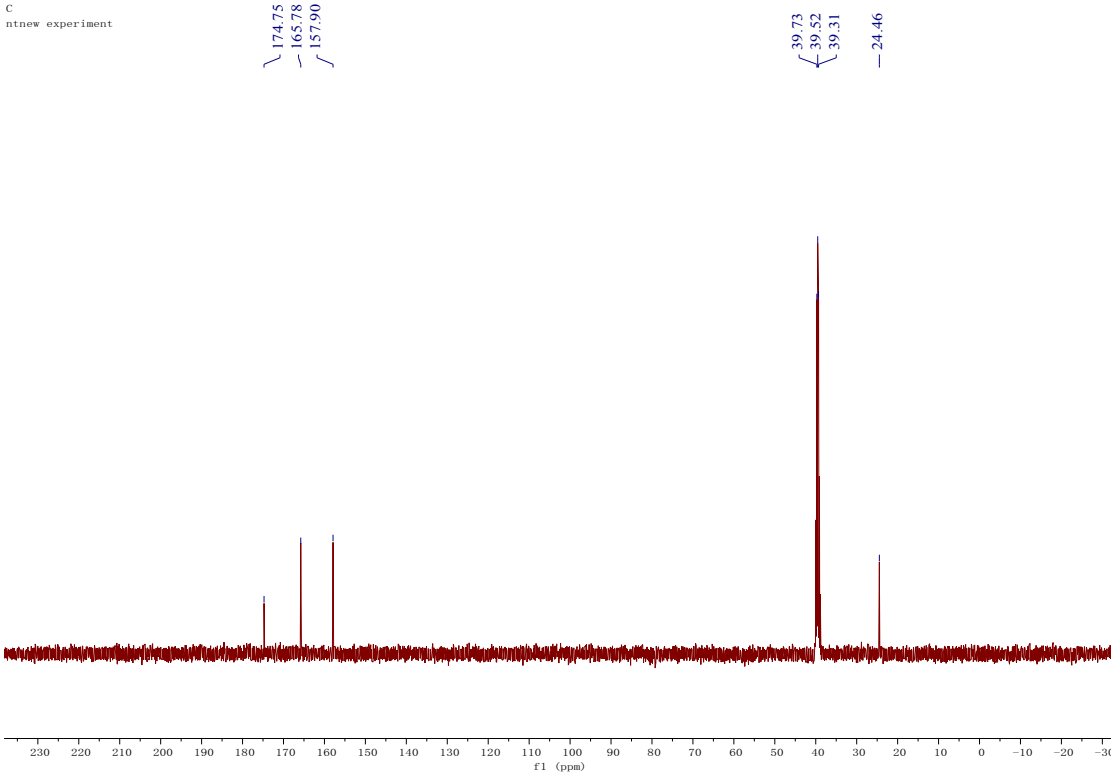


Figure S14 ^{13}C NMR spectrum of compound **6**.

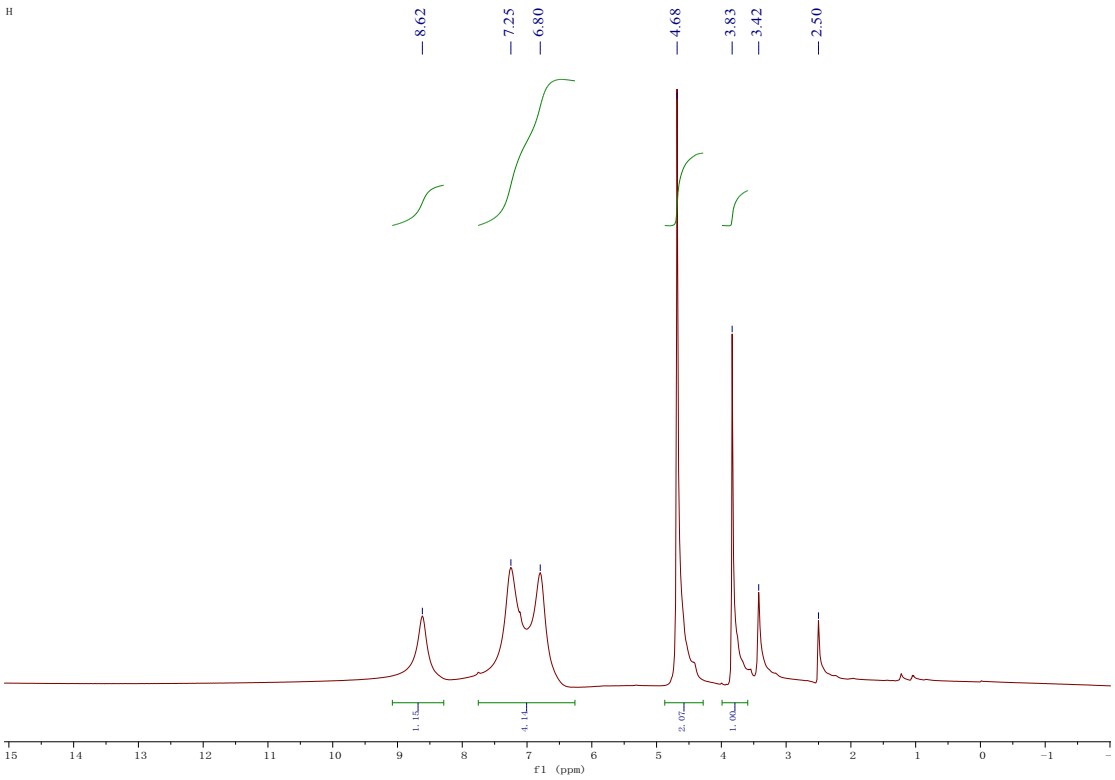


Figure S15 ^{1}H NMR spectrum of compound **7**.

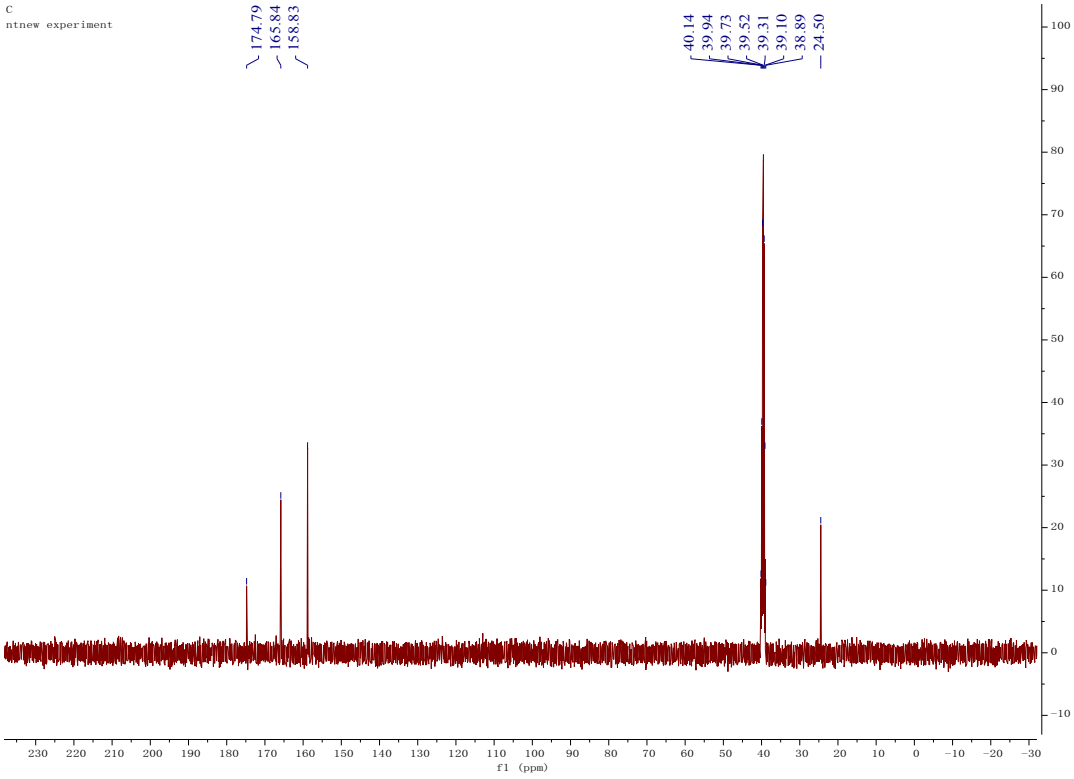


Figure S16 ^{13}C NMR spectrum of compound **7**.

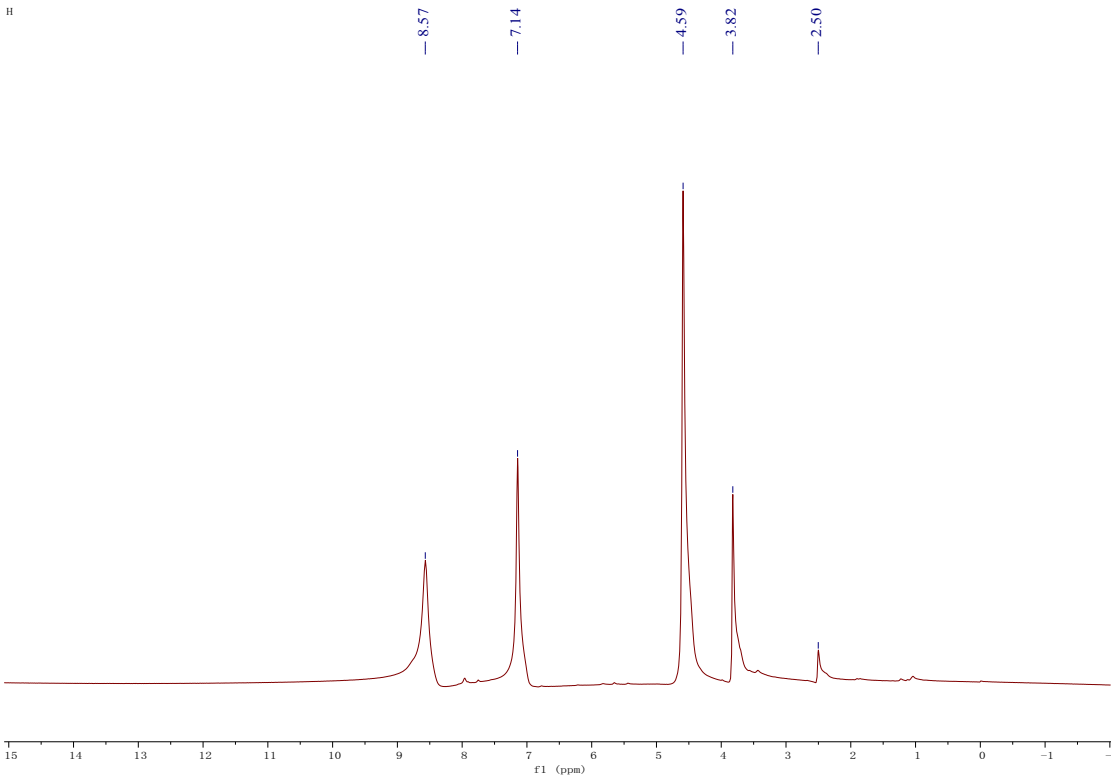


Figure S17 ^{1}H NMR spectrum of compound **8**.

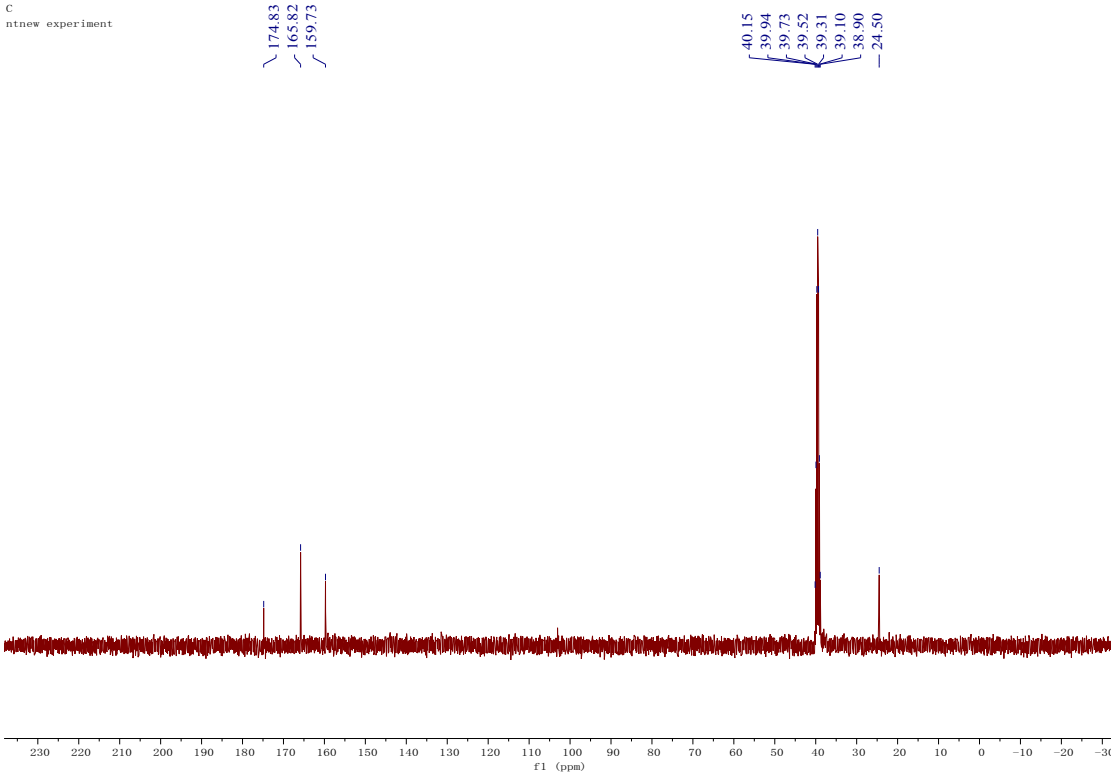


Figure S18 ^{13}C NMR spectrum of compound **8**.

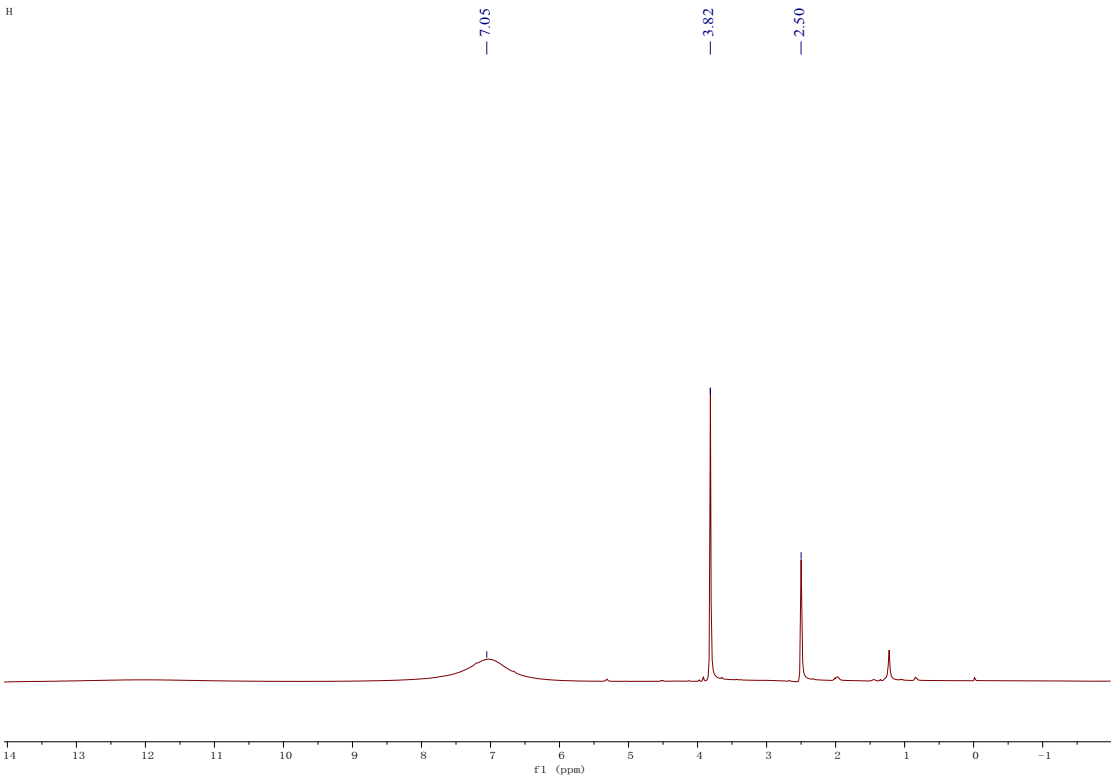


Figure S19 ^{1}H NMR spectrum of compound **9**.

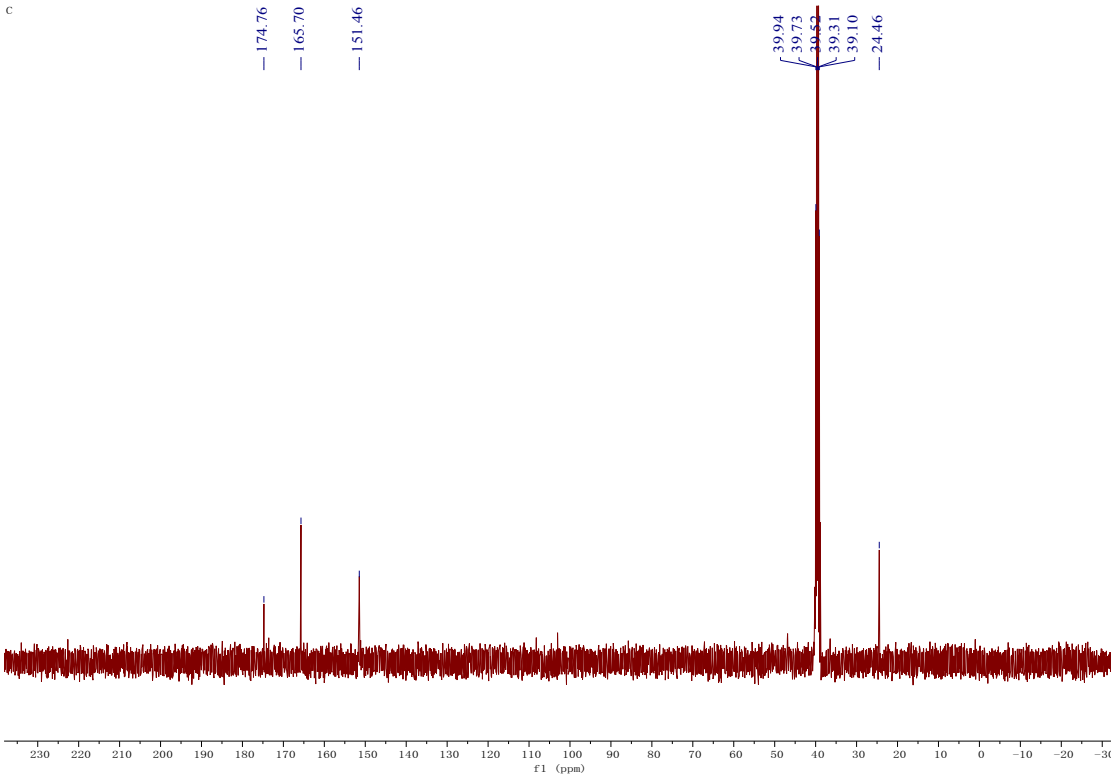


Figure S20 ^{13}C NMR spectrum of compound **9**.

2. DSC data

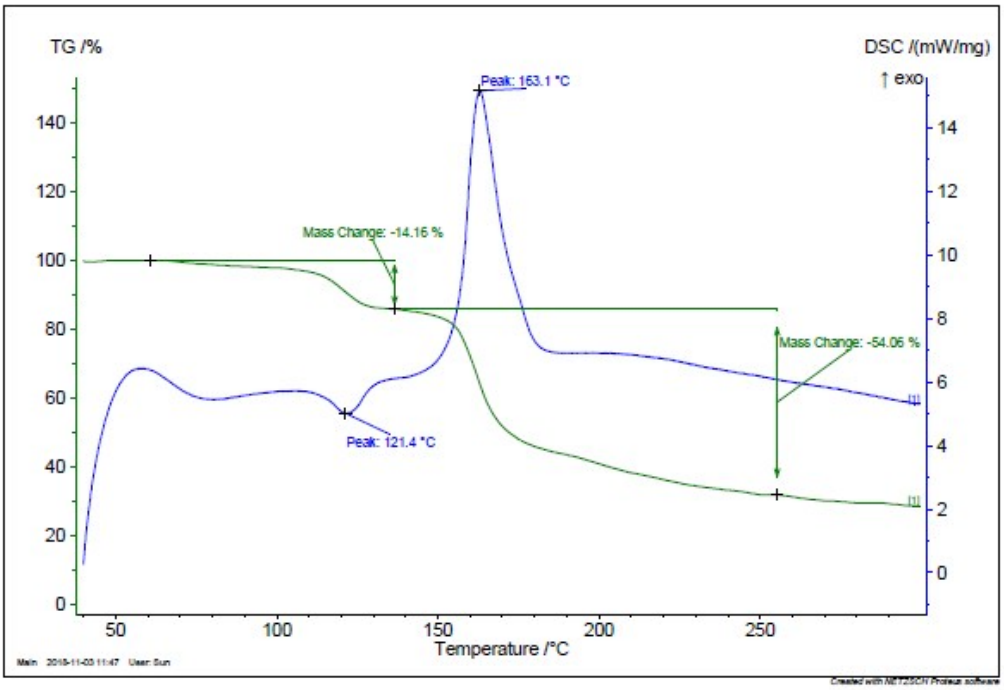


Figure S21 DSC curve of compound **3** with a heating rate of 5 K min^{-1}.

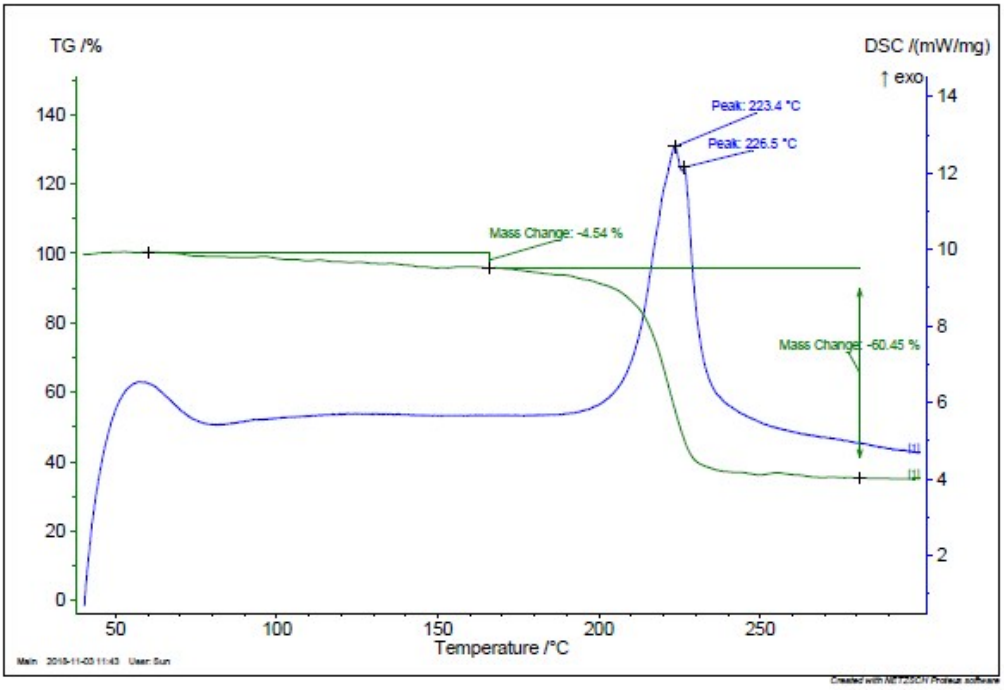


Figure S22 DSC curve of compound **4** with a heating rate of 5 K min^{-1}.

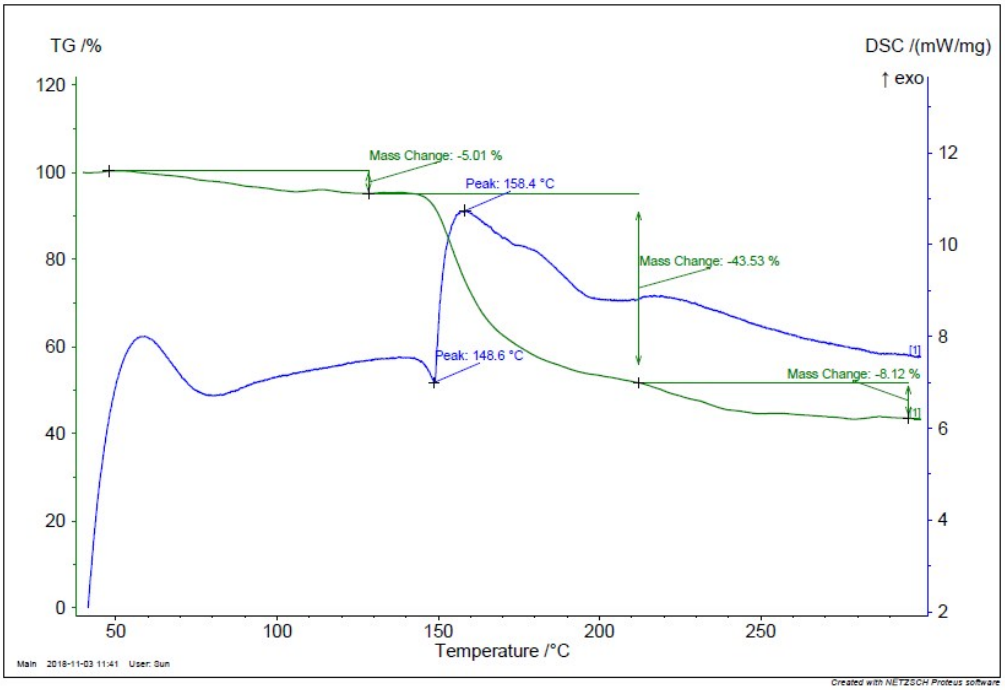


Figure S23 DSC curve of compound **5** with a heating rate of 5 K min^{-1}.

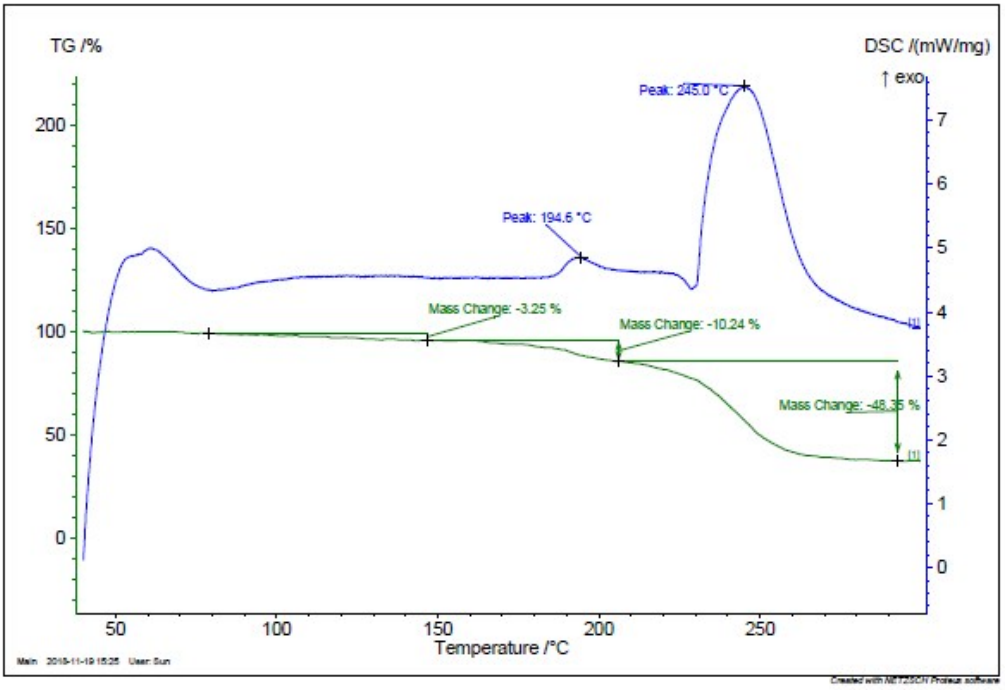


Figure S24 DSC curve of compound **6** with a heating rate of 5 K min^{-1}.

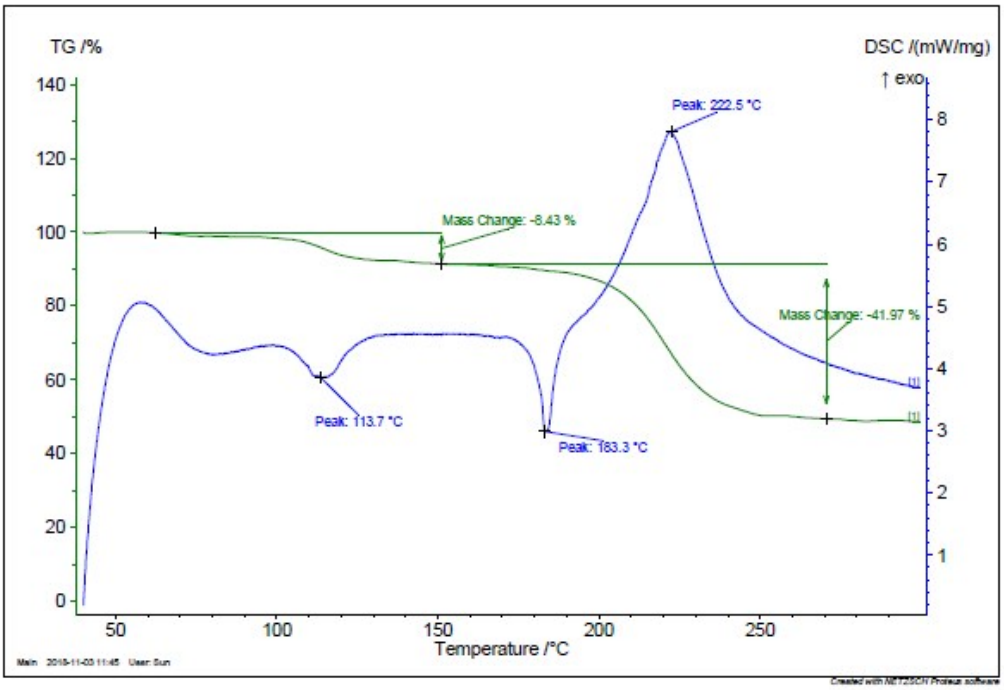


Figure S25 DSC curve of compound **7** with a heating rate of 5 K min^{-1}.

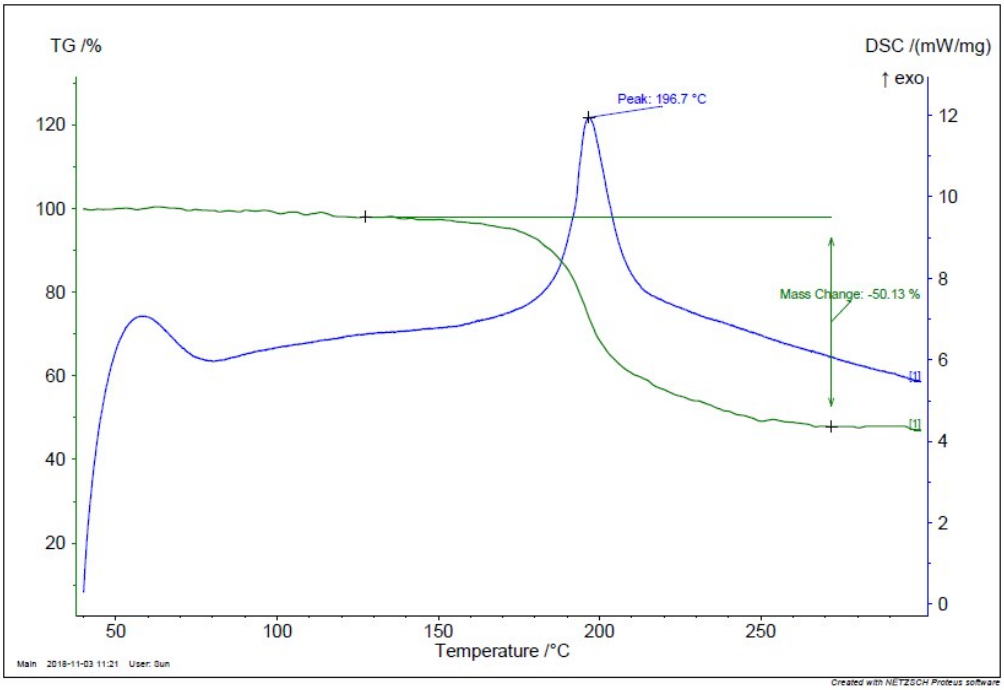


Figure S26 DSC curve of compound **8** with a heating rate of 5 K min^{-1}.

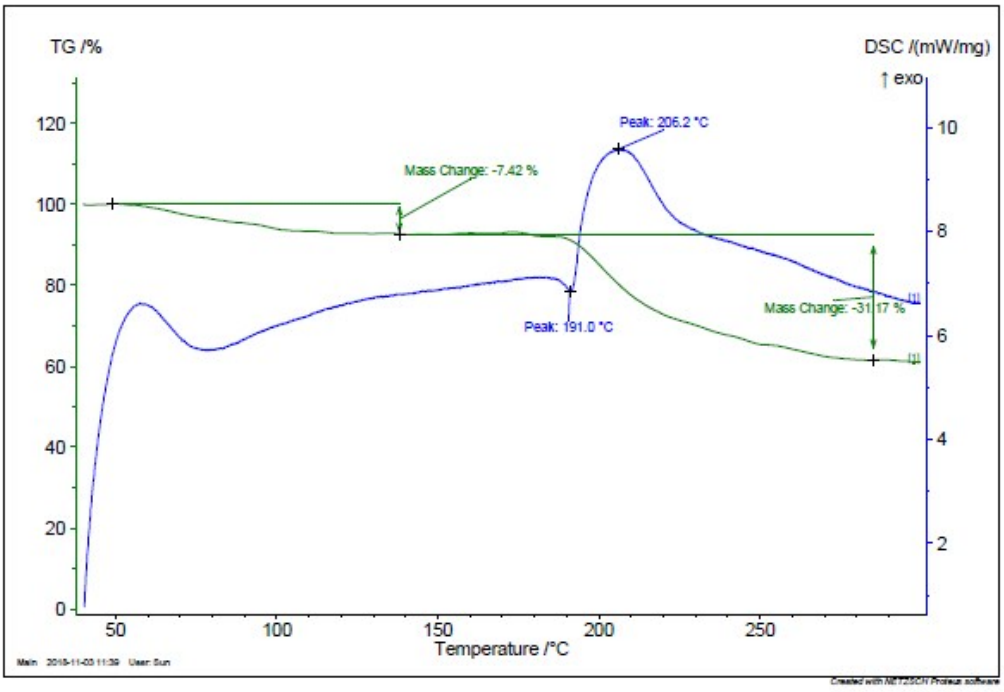
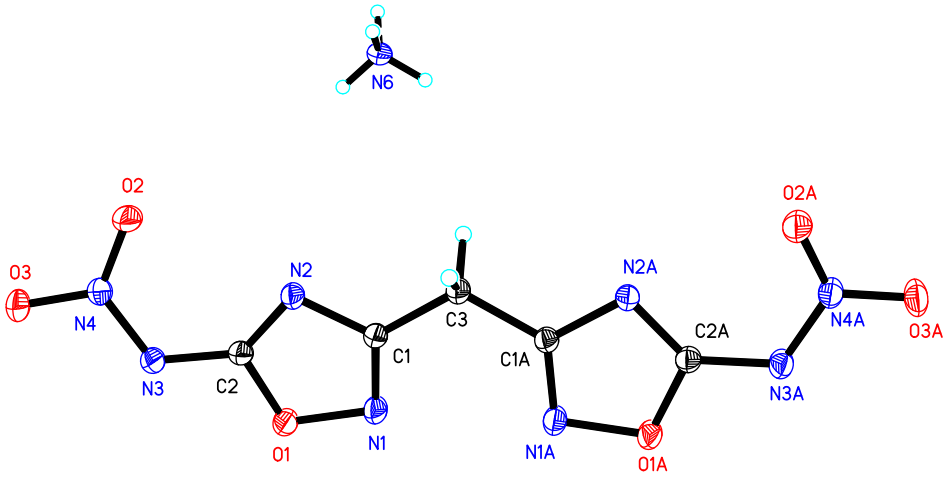
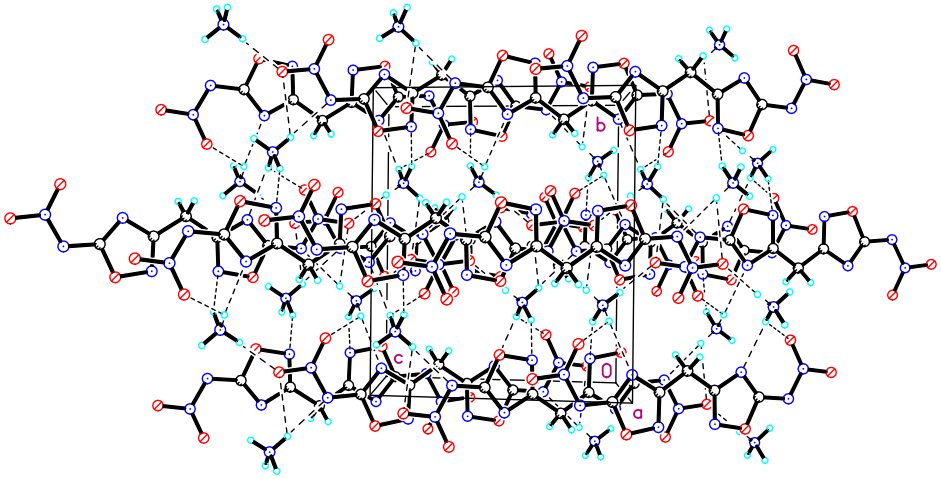


Figure S27 DSC curve of compound **9** with a heating rate of 5 K min^{-1}.

3. Crystallographic data for compound 4



(a)



(b)

Figure S28: (a) Displacement ellipsoid plot (30%) of salt **4**. (b) Ball-and-stick packing diagram of salt **4** viewed down the a-axis. The dashed lines indicate hydrogen bonding.

Table S1. Crystal data and structure refinement for compound **4**.

Identification code	mo_d8v18858_0m	
Empirical formula	C5 H10 N10 O6	
Formula weight	306.23	
Temperature	296(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	C 2/c	
Unit cell dimensions	a = 11.0337(7) Å	α= 90°.
	b = 11.2495(6) Å	β= 105.107(2)°.
	c = 9.7684(6) Å	γ = 90°.
Volume	1170.59(12) $Å^3$	
Z	4	
Density (calculated)	1.738 Mg/m^3	
Absorption coefficient	0.155 mm^{-1}	
F(000)	632	
Crystal size	0.180 x 0.150 x 0.120 mm^3	
Theta range for data collection	2.633 to 25.980°.	
Index ranges	-13<=h<=13, -13<=k<=13, -12<=l<=12	
Reflections collected	10651	

Independent reflections	1134 [R(int) = 0.0318]
Completeness to theta = 25.242°	98.5 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.7456 and 0.6680
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	1134 / 0 / 117
Goodness-of-fit on F^2	1.082
Final R indices [I>2sigma(I)]	R1 = 0.0295, wR2 = 0.0778
R indices (all data)	R1 = 0.0317, wR2 = 0.0796
Extinction coefficient	0.027(7)
Largest diff. peak and hole	0.237 and -0.177 e.$Å^{-3}$

Table S2. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for compound **4**. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
N(1)	991(1)	6057(1)	3699(1)	35(1)
N(2)	1626(1)	4311(1)	4786(1)	28(1)
N(3)	3066(1)	5265(1)	6860(1)	32(1)
N(4)	3518(1)	4245(1)	7505(1)	30(1)
N(6)	5842(1)	2947(1)	910(1)	36(1)
O(1)	1909(1)	6257(1)	5001(1)	37(1)
O(2)	3303(1)	3255(1)	6955(1)	40(1)
O(3)	4204(1)	4381(1)	8730(1)	43(1)
C(1)	875(1)	4906(1)	3658(1)	25(1)
C(2)	2232(1)	5177(1)	5574(1)	26(1)
C(3)	0	4224(2)	2500	29(1)

Table S3. Bond lengths [Å] and angles [°] for compound **4**.

N(1)-C(1)	1.3000(15)
N(1)-O(1)	1.4223(13)
N(2)-C(2)	1.3122(15)
N(2)-C(1)	1.3690(15)
N(3)-N(4)	1.3407(14)

N(3)-C(2)	1.3537(15)
N(4)-O(2)	1.2323(13)
N(4)-O(3)	1.2471(14)
N(6)-H(6A)	0.92(2)
N(6)-H(6B)	0.88(2)
N(6)-H(6C)	0.99(2)
N(6)-H(6D)	0.84(3)
O(1)-C(2)	1.3455(14)
C(1)-C(3)	1.4936(14)
C(3)-C(1)#1	1.4936(14)
C(3)-H(3)	0.968(15)

C(1)-N(1)-O(1)	103.09(9)
C(2)-N(2)-C(1)	102.57(9)
N(4)-N(3)-C(2)	116.97(10)
O(2)-N(4)-O(3)	121.94(10)
O(2)-N(4)-N(3)	124.22(10)
O(3)-N(4)-N(3)	113.83(10)
H(6A)-N(6)-H(6B)	107.7(19)
H(6A)-N(6)-H(6C)	106.1(16)
H(6B)-N(6)-H(6C)	109.9(16)
H(6A)-N(6)-H(6D)	119(2)
H(6B)-N(6)-H(6D)	117(2)
H(6C)-N(6)-H(6D)	96(2)
C(2)-O(1)-N(1)	106.20(9)
N(1)-C(1)-N(2)	115.35(10)
N(1)-C(1)-C(3)	125.03(11)
N(2)-C(1)-C(3)	119.62(10)
N(2)-C(2)-O(1)	112.77(10)
N(2)-C(2)-N(3)	135.87(11)
O(1)-C(2)-N(3)	111.27(10)
C(1)-C(3)-C(1)#1	118.15(14)
C(1)-C(3)-H(3)	106.7(9)
C(1)#1-C(3)-H(3)	108.2(9)

Symmetry transformations used to generate equivalent atoms:

#1 -x,y,-z+1/2

Table S4. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for compound **4**. The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2\ a^{*2}U^{11} + ...\ + 2\ h\ k\ a^*\ b^*\ U^{12}\ ]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
N(1)	37(1)	26(1)	31(1)	-2(1)	-8(1)	0(1)
N(2)	31(1)	23(1)	25(1)	0(1)	-1(1)	-1(1)
N(3)	36(1)	25(1)	29(1)	-3(1)	-3(1)	2(1)
N(4)	28(1)	30(1)	28(1)	-3(1)	0(1)	2(1)
N(6)	41(1)	25(1)	37(1)	-2(1)	3(1)	0(1)
O(1)	43(1)	22(1)	35(1)	-2(1)	-10(1)	0(1)
O(2)	45(1)	26(1)	42(1)	-4(1)	-5(1)	4(1)
O(3)	47(1)	42(1)	29(1)	-2(1)	-10(1)	6(1)
C(1)	25(1)	23(1)	24(1)	0(1)	3(1)	1(1)
C(2)	27(1)	23(1)	27(1)	-2(1)	4(1)	1(1)
C(3)	32(1)	23(1)	27(1)	0	-2(1)	0

Table S5. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10^3) for compound **4**.

	x	y	z	U(eq)
H(3)	516(15)	3721(14)	2077(16)	41(4)
H(6A)	5300(20)	3369(19)	210(20)	75(6)
H(6B)	6203(19)	3450(20)	1590(20)	70(6)
H(6C)	5324(19)	2380(20)	1270(20)	74(6)
H(6D)	6310(20)	2440(20)	680(20)	91(8)

Table S6. Torsion angles [°] for compound **4**.

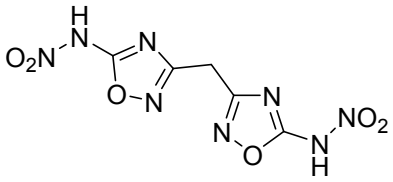
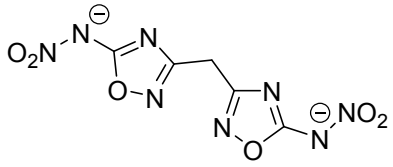
C(2)-N(3)-N(4)-O(2)	6.27(18)
C(2)-N(3)-N(4)-O(3)	-174.72(11)
C(1)-N(1)-O(1)-C(2)	0.33(13)
O(1)-N(1)-C(1)-N(2)	-0.63(14)
O(1)-N(1)-C(1)-C(3)	179.09(9)
C(2)-N(2)-C(1)-N(1)	0.66(14)
C(2)-N(2)-C(1)-C(3)	-179.07(9)
C(1)-N(2)-C(2)-O(1)	-0.40(13)
C(1)-N(2)-C(2)-N(3)	175.83(14)
N(1)-O(1)-C(2)-N(2)	0.06(14)
N(1)-O(1)-C(2)-N(3)	-177.12(9)
N(4)-N(3)-C(2)-N(2)	7.5(2)
N(4)-N(3)-C(2)-O(1)	-176.21(10)
N(1)-C(1)-C(3)-C(1)#1	-4.30(9)
N(2)-C(1)-C(3)-C(1)#1	175.41(11)

Symmetry transformations used to generate equivalent atoms:

#1 -x,y,-z+1/2

4 Ab Initio computational data

Table S7. Ab Initio computational data

Species	E_0 (Hartree)	H_{corr} (Hartree)	ZPE (Hartree)	HOF (kJ/mol)
	-1079.9555336	0.156665	0.139739	115.9468485
	-1078.8589014	0.129729	0.113645	18.44134449
CH_4	-40.3984857	0.048605	0.044793	-74.6[1]
NH_3	-56.4341763	0.038190	0.034372	-45.9[2]
	-261.5728129	0.050912	0.046541	75[2]
NH_2NO_2	-260.5478787	0.042613	0.039257	-6.1[3]
$^{-}NHNO_2$	-260.0119632	0.030444	0.026168	-84.0[4]
CH_3CH_3	-79.6068548	0.079039	0.074609	-84.68[5]
CH_3NH_2	-95.6318759	0.068401	0.064032	-23.0[2]

[a] Total energy (E_0) calculated by B3LYP/6-31+G**/MP2/6-311++G** method (Hartree/Particle); [b] Values of thermal correction (HT) (Hartree/Particle); [c] Zero-point correction (ZPE) (Hartree/Particle); [d] Heat of formation (HOF) (kJ/mol).

5 Detonation performances calculation

Detonation pressure (P) and detonation velocity (D) were calculation according to the Kamlet-Jacobs equations[6].

$$D = 1.01\left(N\bar{M}^{1/2}Q^{1/2}\right)^{1/2}(1 + 1.30\rho) \quad (4)$$

$$P = 1.558\rho^2\bar{M}^{1/2}Q^{1/2} \quad (5)$$

where each term in eqs 4 and 5 is defined as follows: D, the detonation velocity (km s^{-1}); P, the detonation pressure (GPa); N, the moles of detonation gases per gram explosive; M , the average molecular weight of these gases (g mol^{-1}); Q, the heat of detonation (J g^{-1}); and ρ, the loaded density of explosives (g cm^{-3}). The measured density was used for the calculation here.

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