A novel bis(triazole)furoxan energetic compound—graphene-like crystal structure for balancing energy and sensitivity

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Table S1. Crystallographic and refinement data of compound (12),(13)and(15).

Sample code	Compound(12)	Compound(13)	Compound(15)
Empirical formula	$C_{18}H_{28}N_{12}O_3$	$C_{16}H_{34}N_{20}O_{14}S$	$C_{10}H_{13}N_{13}O_7$
Formula weight	460.52	762.69	427.33
Temperature/K	296.15	193	193
Crystal system	triclinic	monoclinic	monoclinic
Space group	P-1	C2/c	P21/n
CCDC number	2358187	2358185	2358186
a/Å	10.205(2)	20.3798(6)	7.2046(2)
b/Å	10.512(2)	6.4900(2)	12.8063(3)
c/Å	11.184(3)	25.5179(7)	18.8900(5)
α/°	90.850(4)	90	90
β/°	94.688(4)	107.008(2)	100.014(2)
γ/°	96.027(4)	90	90
Volume/Å3	1188.8(5)	3227.51(17)	1716.32(8)
Z	2	4	4
ρ (g/cm3)	1.287	1.57	1.654
μ/mm-1	0.094	1.753	1.232
F(000)	488	1592	880
Crystal size/mm3	0.26 × 0.24 × 0.22	0.13 × 0.11 × 0.1	0.13 × 0.11 × 0.1
Radiation	MoKα (λ = 0.71073)	CuKα (λ = 1.54178)	CuKα (λ = 1.54178)
2Θ range for data collection/°	5.896 to 49.994	7.246 to 158.928	8.382 to 136.702
Index ranges	-12 ≤ h ≤ 8, -12 ≤ k ≤ 12, -10 ≤ l ≤ 13	-25 ≤ h ≤ 25, -7 ≤ k ≤ 8, -31 ≤ l ≤ 32	-8 ≤ h ≤ 7, -14 ≤ k ≤ 15, -22 ≤ l ≤ 22
Reflections collected	6023	18012	15195
Independent reflections	4139 [Rint = 0.0169, Rsigma = 0.0380]	3449 [Rint = 0.0560, Rsigma = 0.0346]	3148 [Rint = 0.0305, Rsigma = 0.0225]
Data/restraints/parameters	4139/10/332	3449/0/253	3148/0/284
Goodness-of-fit on F2	1.061	1.063	1.053
Final R indexes [I>=2σ (I)]	R1 = 0.0595, wR2 = 0.1679	R1 = 0.0375, wR2 = 0.0988	R1 = 0.0365, wR2 = 0.1001
Final R indexes [all data]	R1 = 0.0917, wR2 = 0.1821	R1 = 0.0476, wR2 = 0.1052	R1 = 0.0405, wR2 = 0.1034
Largest diff. peak/hole / e Å-3	0.75/-0.19	0.32/-0.42	0.32/-0.35

Table S2. Hydrogen bond parameters of compound 13

Bonds	Atoms	Symmetry code	Atoms2	Symmetry code	Distance
O5···H-N1[b]	O5	x, y, z	N1[b]	1-x, 1+y, 3/2-z	1.9605
O5···H-C1[b]	O5	x, y, z	C1[b]	1-x, 1+y, 3/2-z	2.5335
O5[a]···H-N1[c]	O5[a]	1-x, y, 3/2-z	N1[c]	x, 1+y, z	1.9605
O5[a]···H-O7	O5[a]	1-x, y, 3/2-z	O7	x, y, z	1.9593
O6[a]···H-N1[a]	O6[a]	1-x, y, 3/2-z	N1[a]	1-x, y, 3/2-z	1.9435
O6···H-N1	O6	x, y, z	N1	x, y, z	1.9435
O6···H-O4	O6	x, y, z	O4	x, y, z	1.9194
O3···H-N7[d]	O3	x, y, z	N7[d]	x, -1+y, z	2.0256
O6···H-O4	O6	x, y, z	O4	x, y, z	1.9530
O7···H-C1[c]	O7	x, y, z	C1c	x, 1+y, z	2.4761
Superscript: (a: 1-x, y, 3/2-z; b: 1-x, 1+y, 3/2-z; c: x, 1+y, z; d: x, -1+y, z). Unit: (Å)					

Table S3. Hydrogen bond parameters of compound 15.

Bonds	Atoms	Symmetry code	Atoms2	Symmetry code	Distance
O1···H-C6[a]	O1	x, y, z	C6[a]	-1/2+x, 3/2-y, -1/2+z	2.4174
O2···H-O7	O2	x, y, z	O7	x, y, z	2.6868
N13···H-O7	O7	x, y, z	N13	x, y, z	2.1967
O4[a]···H-C8	C8	x, y, z	O4[a]	-1/2+x, 3/2-y, -1/2+z	2.6890
O5[a]···H-O7[b]	O5[a]	-1/2+x, 3/2-y, -1/2+z	O7[b]	-1/2+x, 1/2-y, -1/2+z	2.0268
Superscript: (a: -1/2+x, 3/2-y, -1/2+z; b: -1/2+x, 1/2-y, -1/2+z). Unit: (Å)					

Table S4 Enrichment ratio for the crystal packing of compounds 13 and 15.

	Contacts%	Atoms	H^{in}	O	C	N
Compound13		H^{out}	23.2	28.4	5.0	22.1
		O	28.4	0.4	2.7	2.2
		C	5.0	2.7	2.6	6.2
		N	22.1	2.2	6.2	7.2
	Surface%		49.50	17.10	9.60	22.50
	Random Contacts%	Atoms	H^{in}	O	C	N
		H^{out}	24.50			
		O	16.92	2.92		
		C	9.50	3.28	0.92	
		N	22.27	7.69	4.32	5.06
	Enrichment Ratio	Atoms	H^{in}	O	C	N
		H^{out}	0.94			
		O	1.67	0.13		
		C	0.52	0.82	2.82	
		N	0.99	0.28	1.43	1.42
			H^{in}	O	C	N
Compound15		H^{out}	13.7	33.1	4.8	18.4
		O	33.1	4.5	7.4	8.7
		C	4.8	7.4	0.4	2.5
		N	18.4	8.7	2.5	6.6
	Surface%		41.85	29.10	7.75	21.20
	Random Contacts%	Atoms	H^{in}	O	C	N
		H^{out}	17.51			
		O	24.35	8.46		

		C	6.48	4.51	0.6	
		N	17.74	12.33	3.28	4.49
	Enrichment Ratio	Atoms	H^{in}	O	C	N
		H^{out}	0.78			
		O	1.35	0.53		
		C	0.74	1.64	0.66	
		N	1.03	0.70	0.76	1.46

Table S5 Topological parameters, energy properties calculated at the (3,-1) critical point of intermolecular interactions of compound 13 and compound 15

mark	$\rho(r_{BCP})$	$\nabla^2\rho$	V	G	H	$\left\|\frac{V}{G}\right\|$	λ_1	λ_2	λ_3	η(r)	$\|E_{int}\|$
Compound 15	（a.u.）	(a.u.)	(a.u.)	(a.u.)	(a.u.)						Kcal/mol
A	0.0351	0.0272	-0.0312	0.0272	-0.004	1.1470	-0.0550	-0.0531	0.2011	0.2734	7.2524
B	0.0152	0.0579	-0.0101	0.0123	0.0022	0.8211	-0.0167	-0.0158	0.0905	0.1856	2.7170
C	0.0358	0.0911	-0.0316	0.0271	-0.004	1.1660	-0.0565	-0.0547	0.2023	0.2792	7.4117
D	0.0320	0.0909	-0.0296	0.0261	-0.0035	1.1341	-0.0504	-0.0496	0.1908	0.2640	6.6281
E	0.0388	0.0981	-0.0357	0.0300	-0.0056	1.1900	-0.0634	-0.0619	0.2234	0.2839	8.0987
F	0.0379	0.0854	-0.0344	0.0280	-0.0067	1.2286	-0.0626	-0.0606	0.2085	0.3000	7.8985
G	0.0296	0.0877	-0.0251	0.0235	-0.0016	1.0680	-0.0453	-0.0432	0.1763	0.2570	6.0061
H	0.0330	0.0814	-0.0292	0.0248	-0.0045	1.1774	-0.0540	-0.0518	0.1871	0.2886	6.8165
I	0.0038	0.0127	0.0017	0.0024	0.0008	0.7083	0.0172	0.0023	0.0021	0.1470	0.1172
J	0.0019	0.0059	0.0007	0.0011	0.0004	0.6363	0.0011	0.0007	0.0077	0.1435	0.3094
Compound 17											
A	0.0107	0.0392	-0.0063	0.0081	0.0017	0.7778	-0.0109	-0.0103	0.0605	0.1803	1.6479
B	0.0113	0.0610	-0.0092	0.0122	0.0030	0.7541	-0.0107	-0.0045	0.0762	0.1406	1.7891
C	0.0152	0.0514	-0.0102	0.0115	0.0013	0.8870	-0.0180	-0.0175	0.0869	0.2071	2.6519
D	0.0096	0.0356	-0.0055	0.0072	0.0017	0.7638	-0.0101	-0.0089	0.0546	0.1847	1.4043
E	0.0211	0.0749	-0.0165	0.0176	0.0011	0.9375	-0.0294	-0.0262	0.1306	0.2253	3.9649

η(r) = |λ1(r)|/λ3(r) [27] E_{int}=-223.08*$\rho(r_{BCP})$+0.7423 (Neutral H-bonds).
One a.u. = 627.509 kcal/mol.

The methodology of Enthalpy of Formation:

The equations for the enthalpy of gas-phase formation, enthalpy of sublimation, and enthalpy of solid-phase formation of the energy-containing compounds based on the composition of the four elements of CHON are shown in (1)-(3):

$$\Delta H_{solid} = \Delta_f H(g) - \Delta H_{sub} \quad (1)$$

$$\Delta H_{sub} = 0.000267A^2 + 1.650087\left(v\sigma_{tot}^2\right)^{0.5} + 2.966078 \quad (2)$$

$$\Delta_f H(g) = H(C_aH_bO_cN_d) - aH(C)(g) - \frac{b}{2}H(H_2)(g) - \frac{c}{2}H(O_2)(g) - \frac{d}{2}H(N_2)(g) + aH_{vap}(graphite) \quad (3)$$

$H(C_aH_bO_cN_d)$, H(C)(g), $H(H_2)$(g), O_2(g), (N_2)(g), were calculated by the Gaussian16 software.

The value of A^2 and $\left(v\sigma_{tot}^2\right)$ were obtained by the Multiwfn software

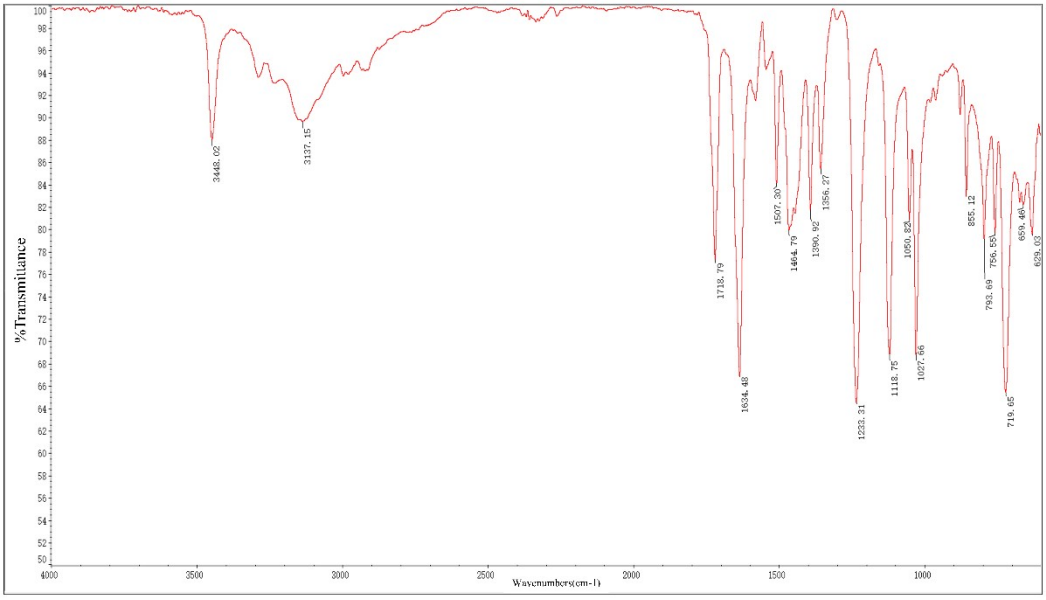


Figure S1. IR spectra of compound1 a)

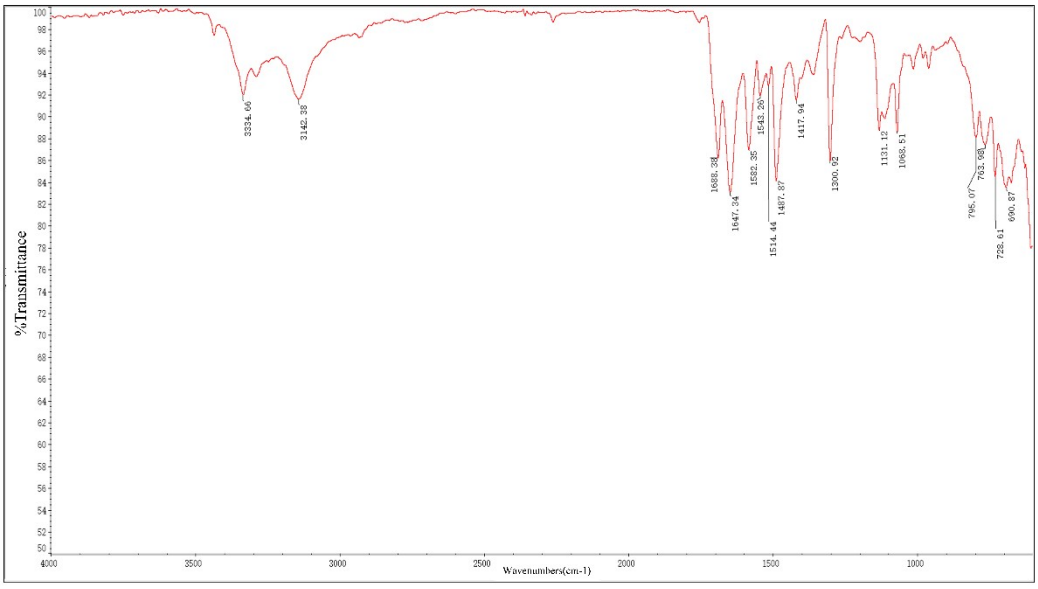


Figure S2. IR spectra of compound1 b)

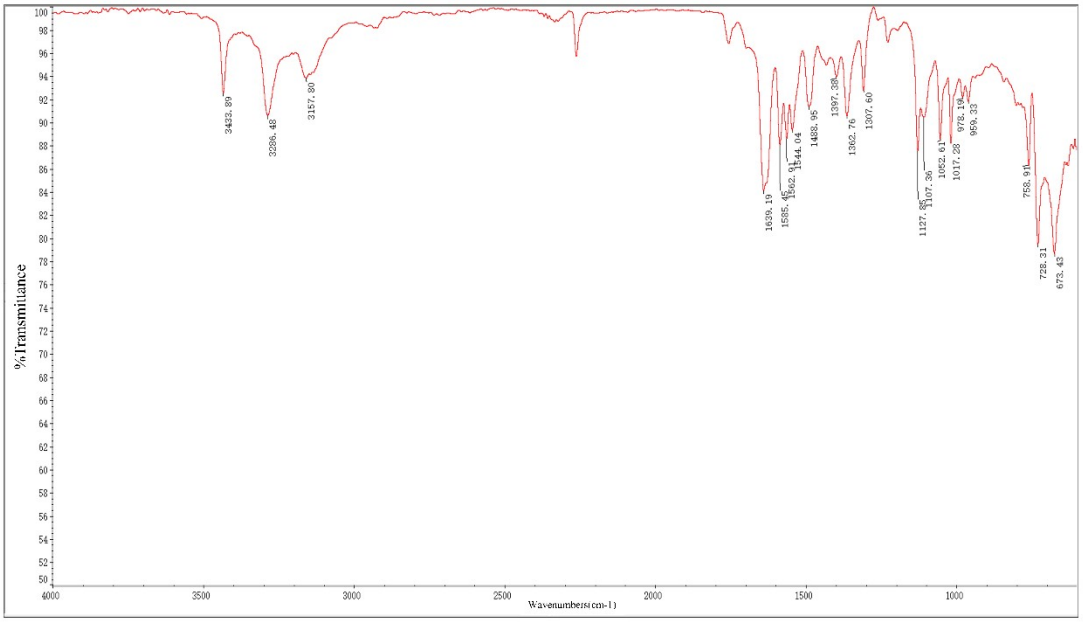


Figure S3. IR spectra of compound1 c)

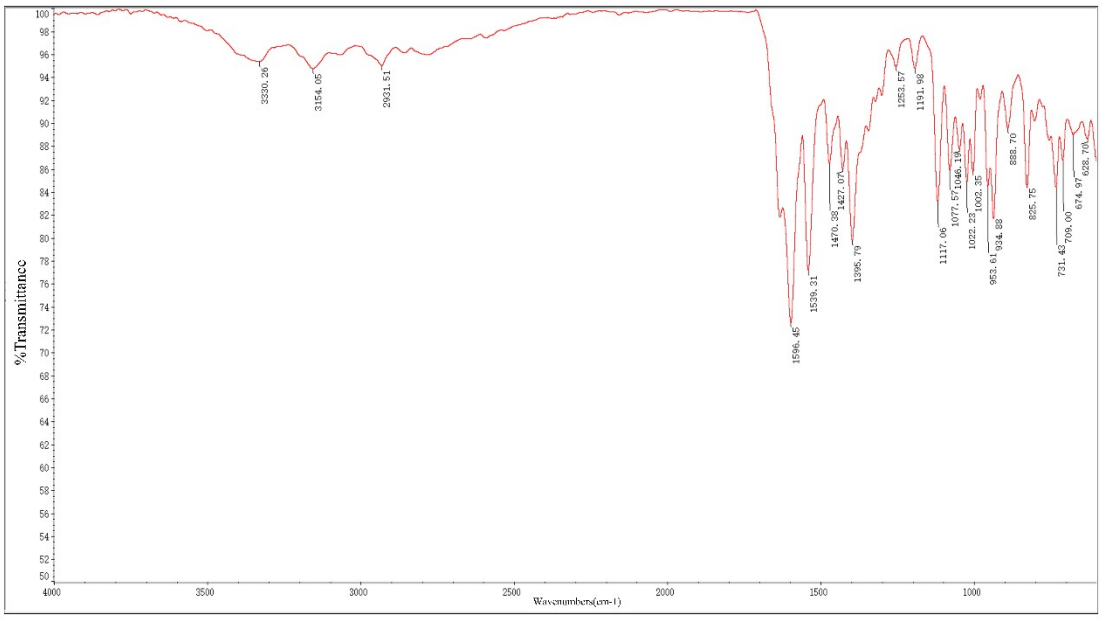


Figure S4. IR spectra of compound 11

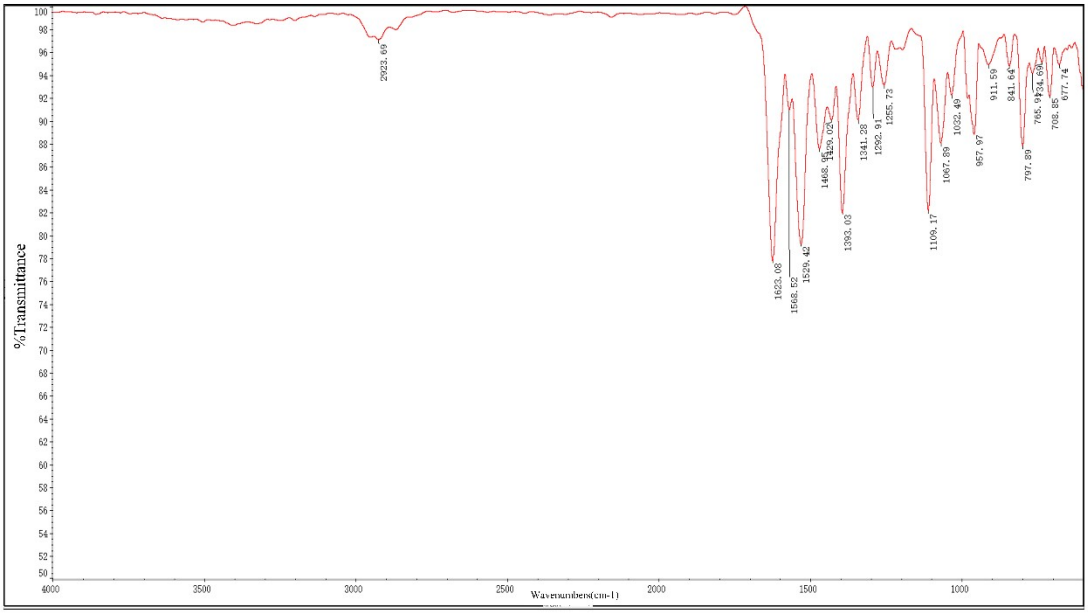


Figure S5. IR spectra of compound 12

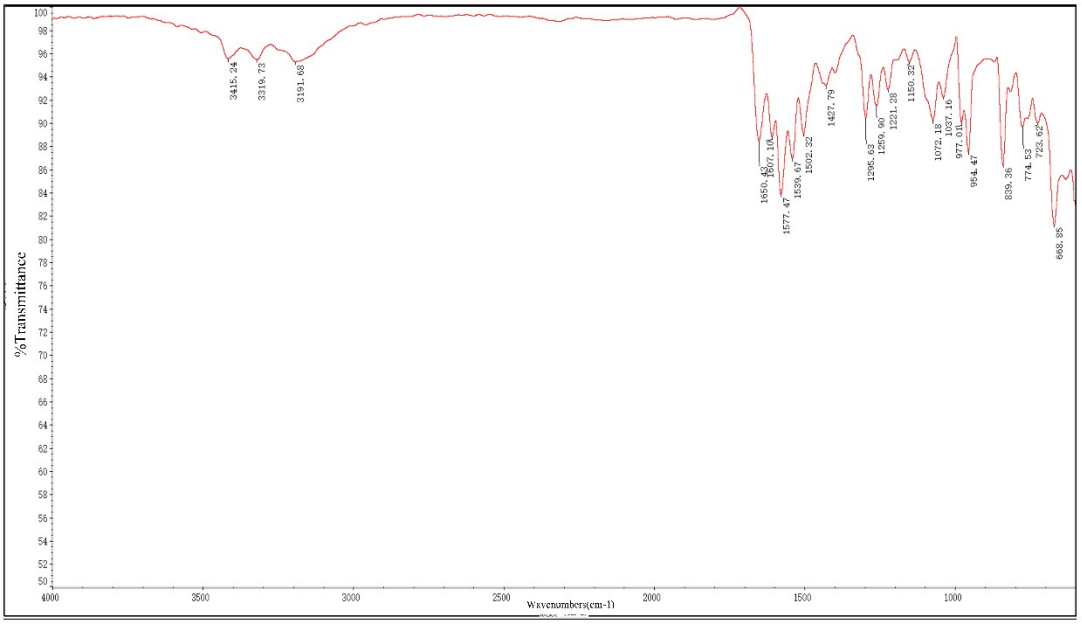


Figure S6. IR spectra of compound 13

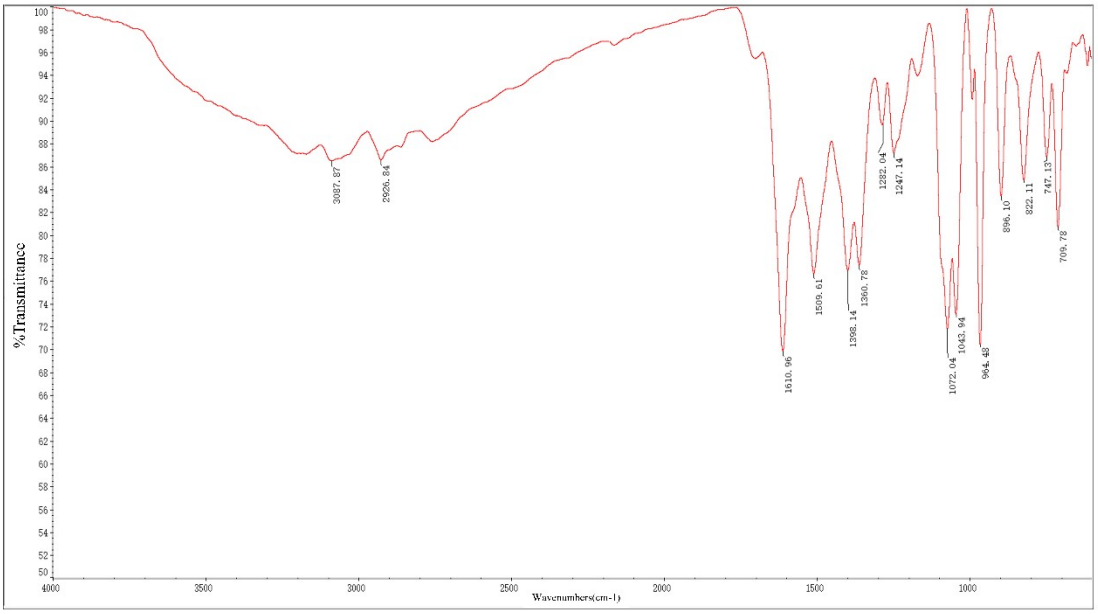


Figure S7. IR spectra of compound 14

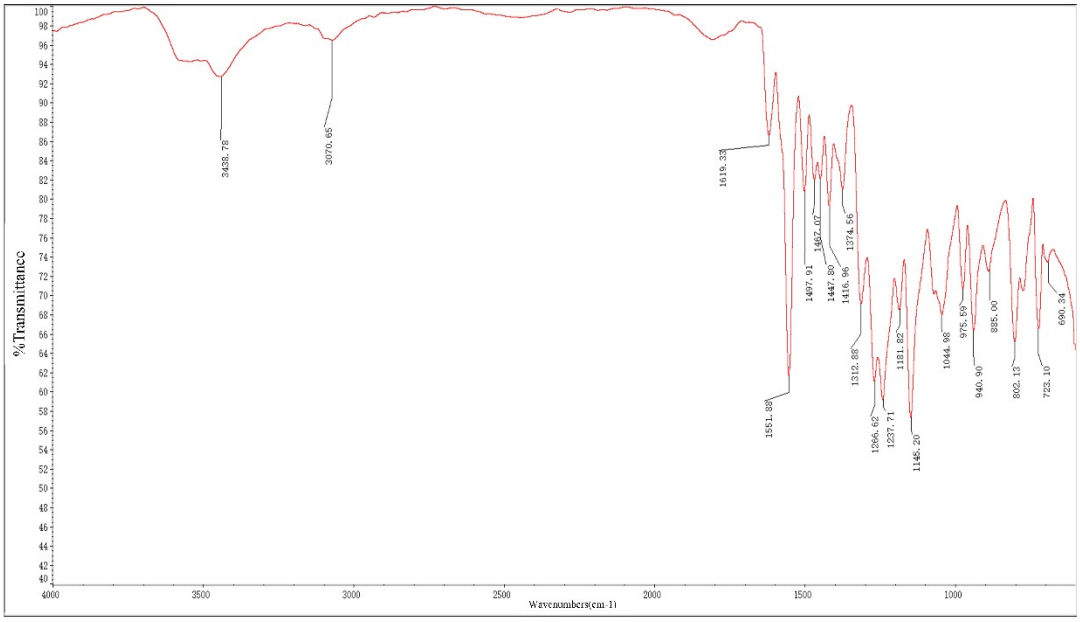


Figure S8. IR spectra of compound 15

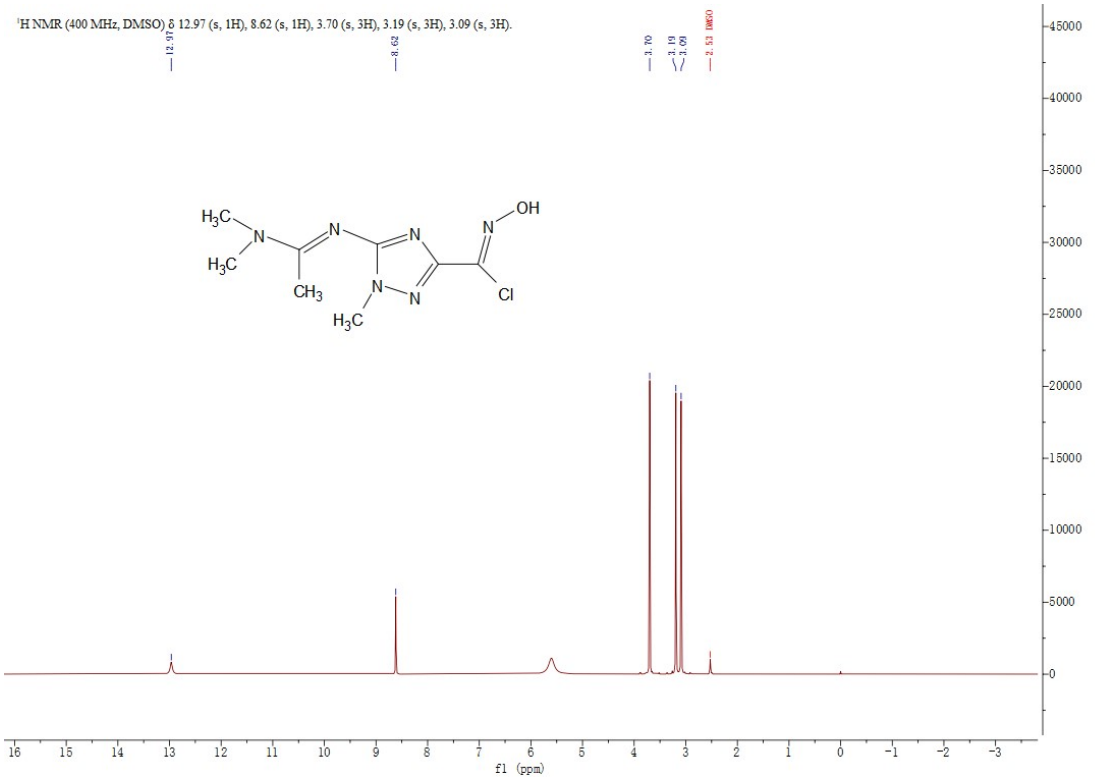


Figure S9. C NMR spectra of compound 11

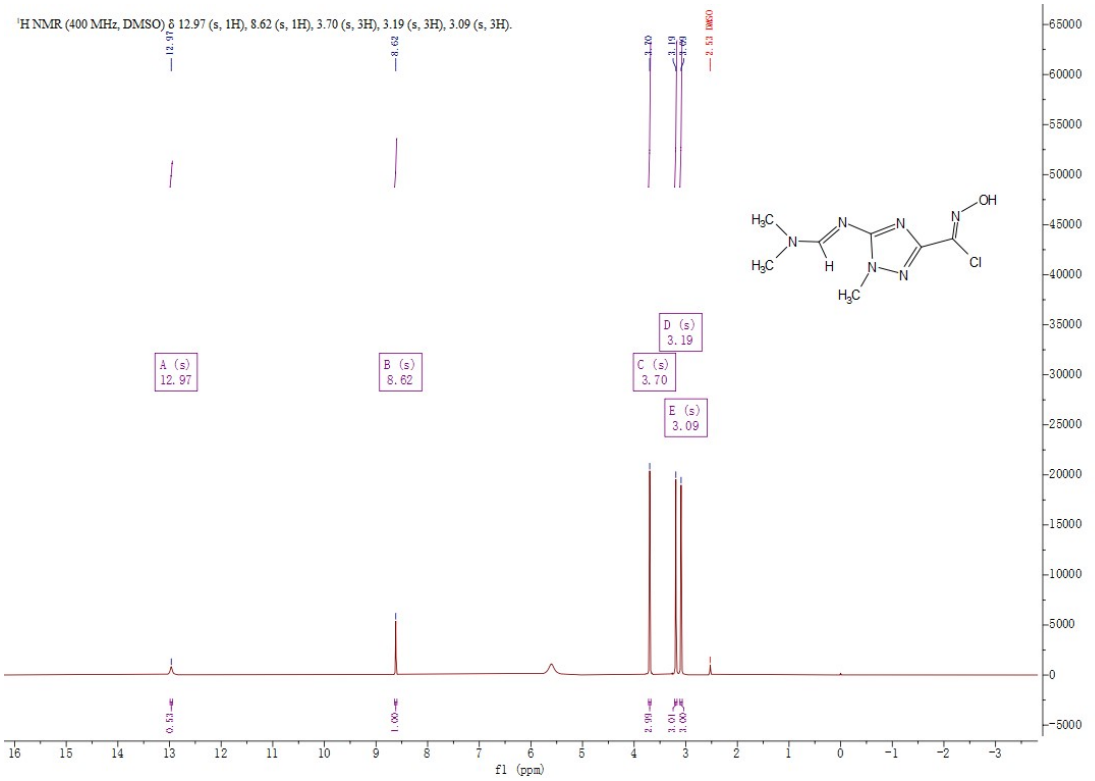


Figure S10. H NMR spectra of compound 11

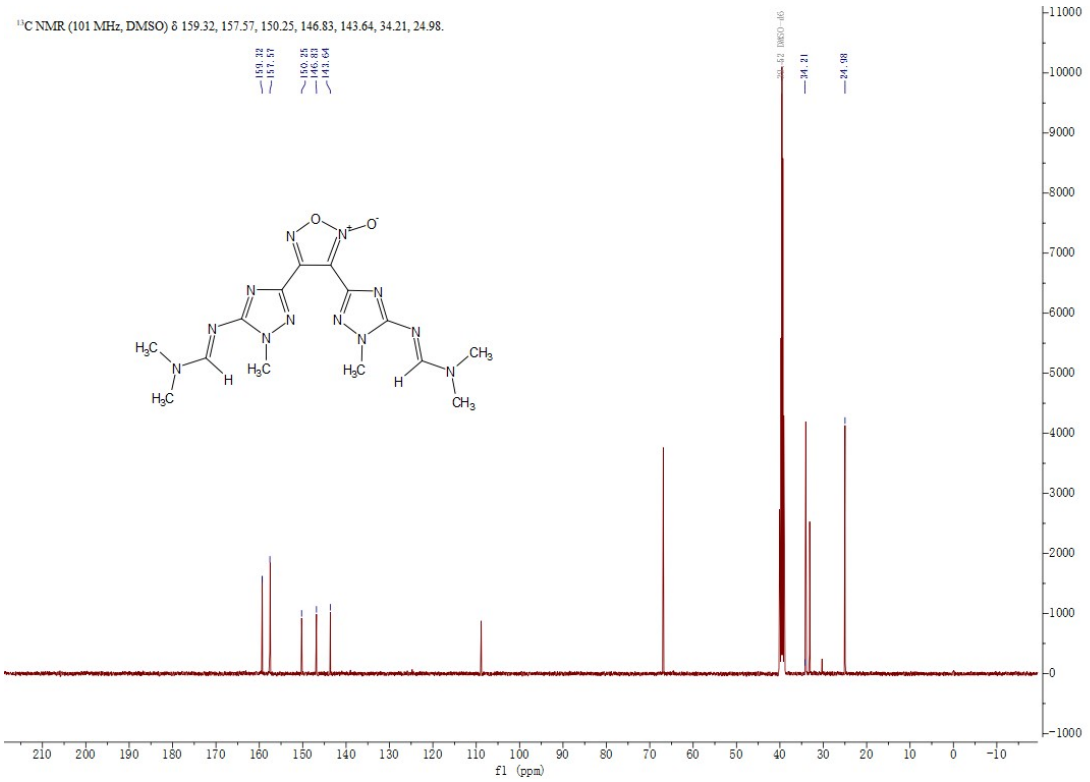


Figure S11. C NMR spectra of compound 12

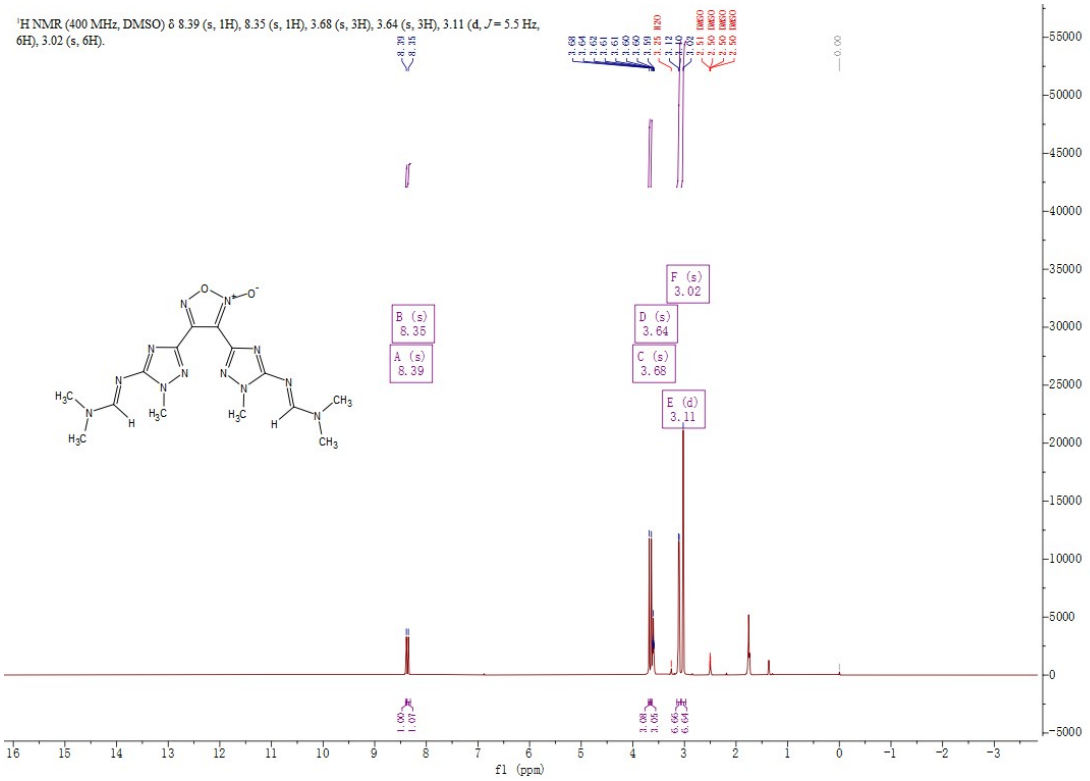


Figure S12. H NMR spectra of compound 12

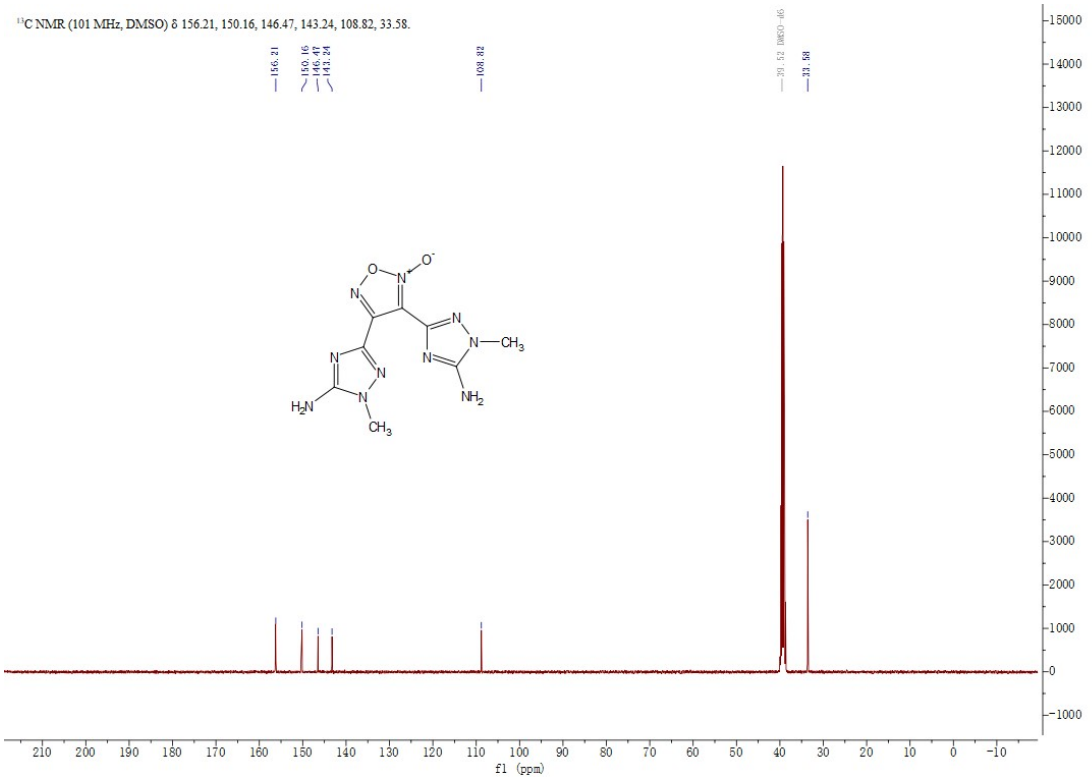


Figure S13. C NMR spectra of compound 13

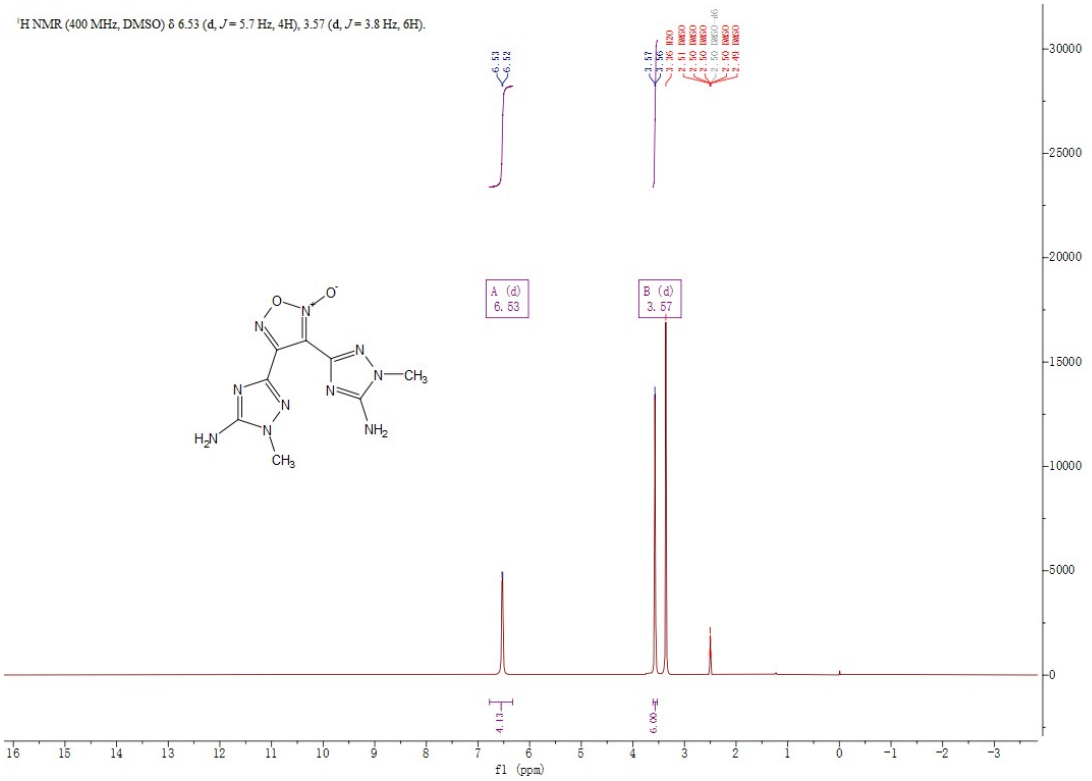


Figure S14. H NMR spectra of compound 13

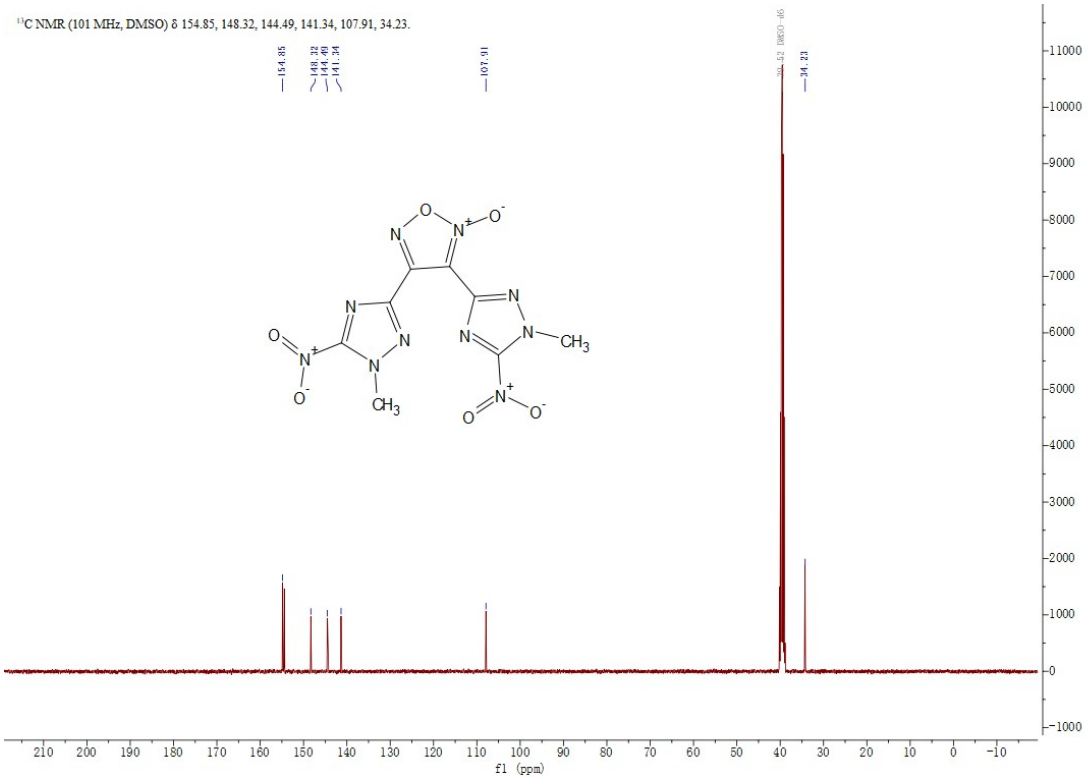


Figure S15. C NMR spectra of compound 14

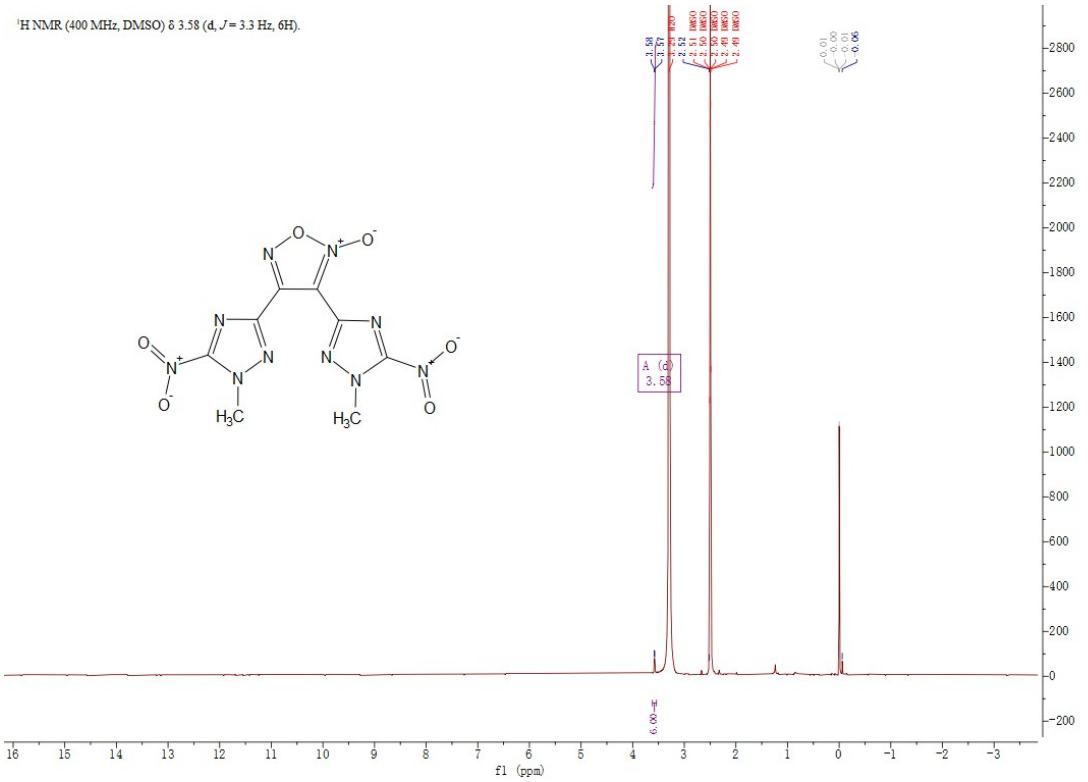


Figure S16. H NMR spectra of compound 14

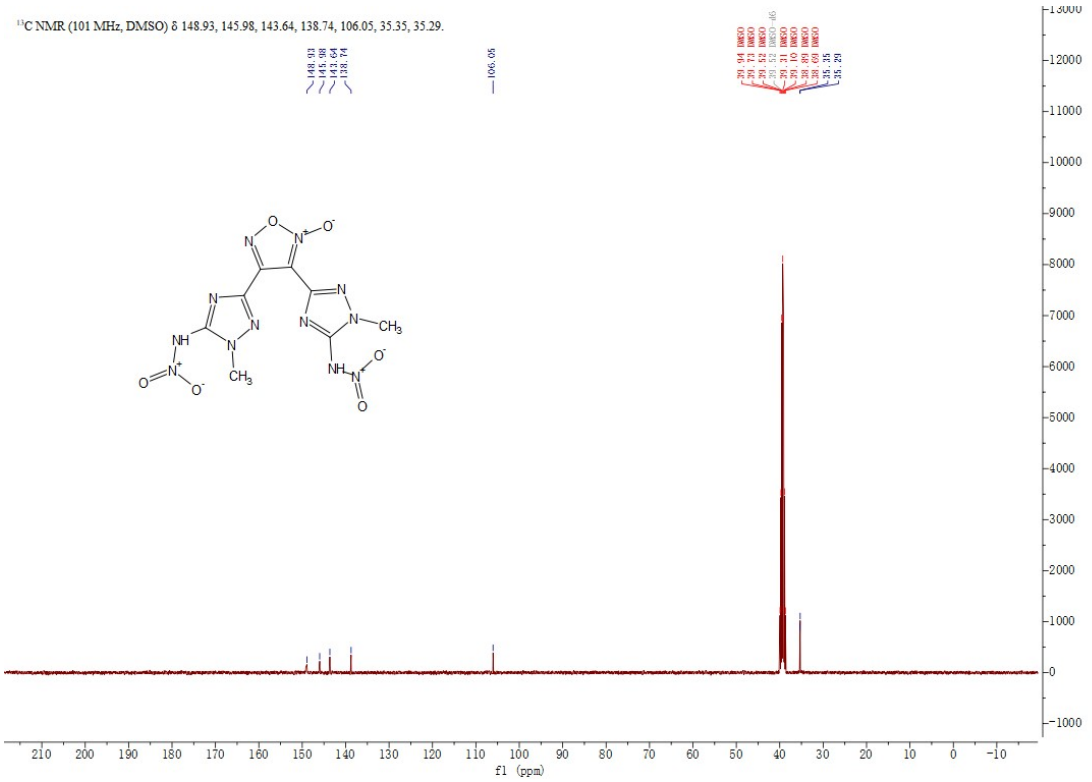


Figure S17. C NMR spectra of compound 15

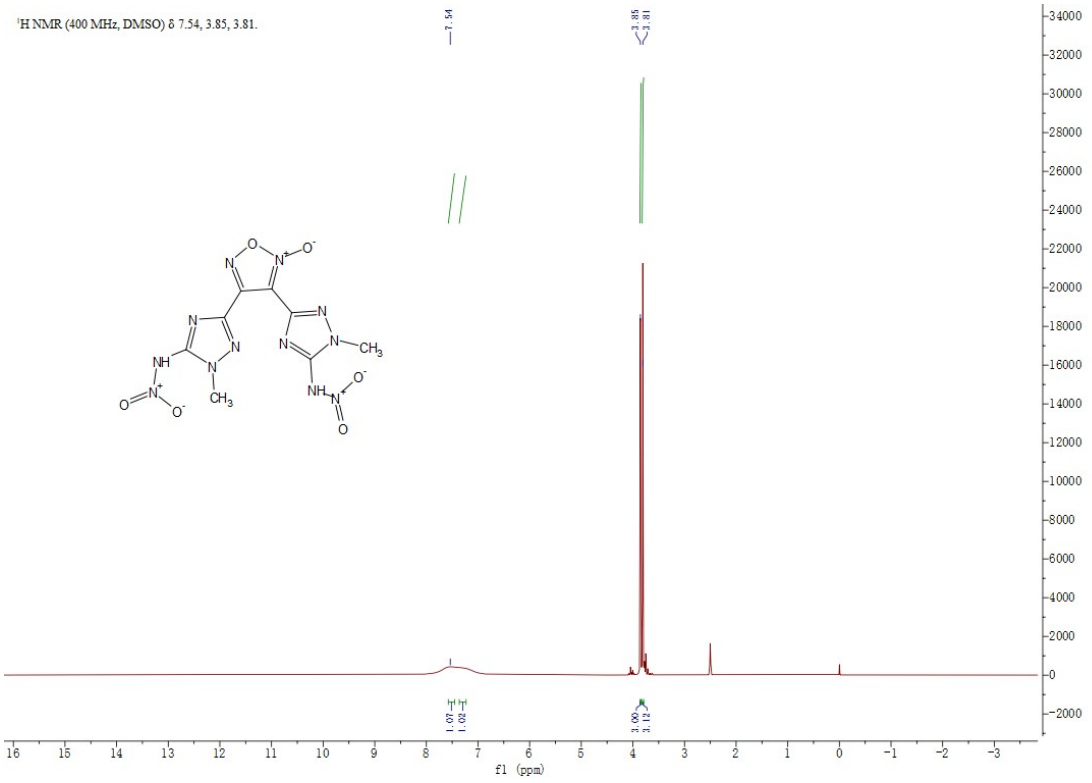


Figure S18. H NMR spectra of compound 15

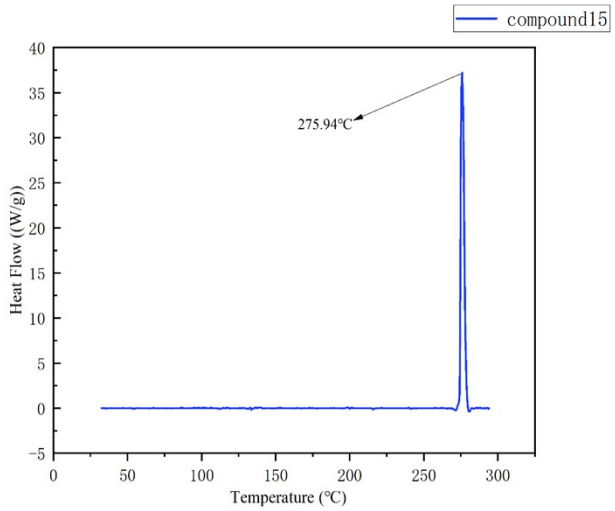


Figure S19. The exothermic temperature of compound 13

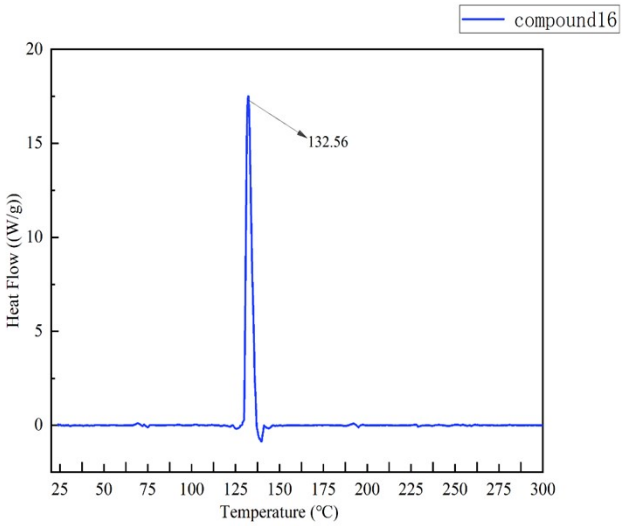


Figure S20. The exothermic temperature of compound 14

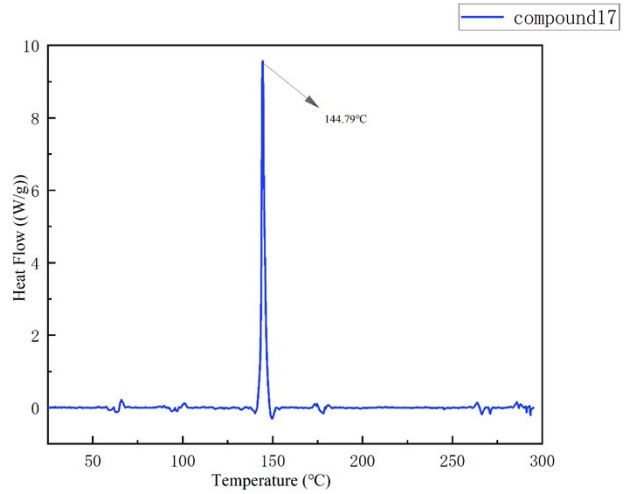


Figure S21. The exothermic temperature of compound 15

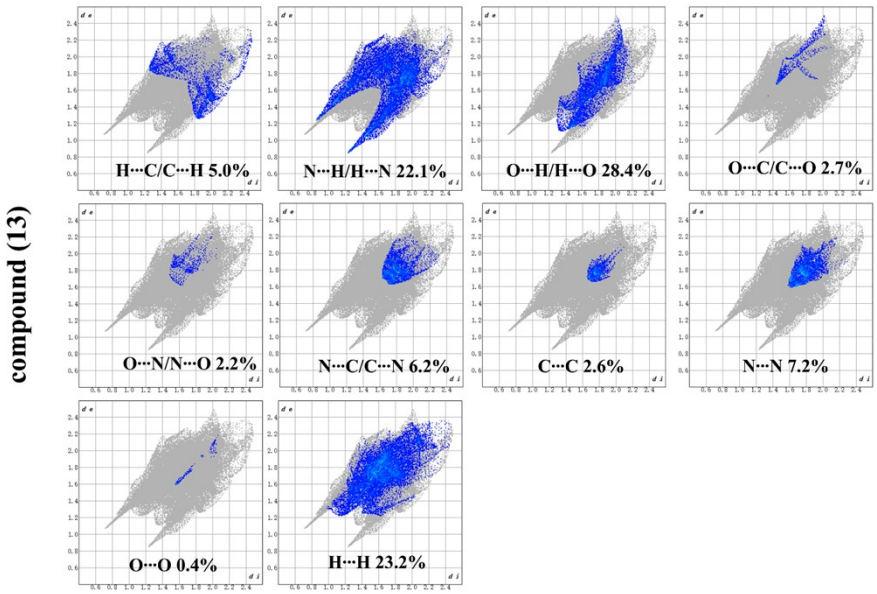


Figure S22. Decomposed fingerprint plots of compound (13).

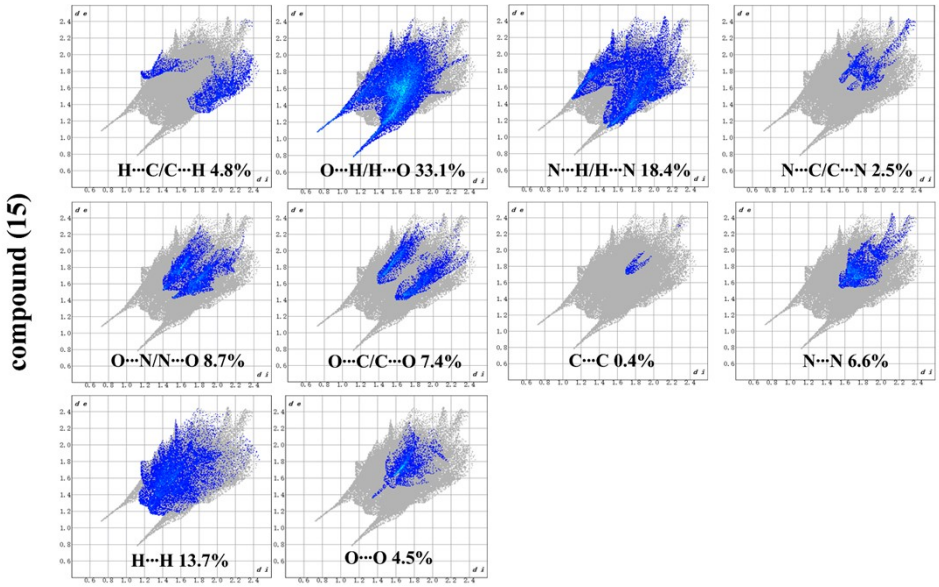


Figure S23. Decomposed fingerprint plots of compound (14).

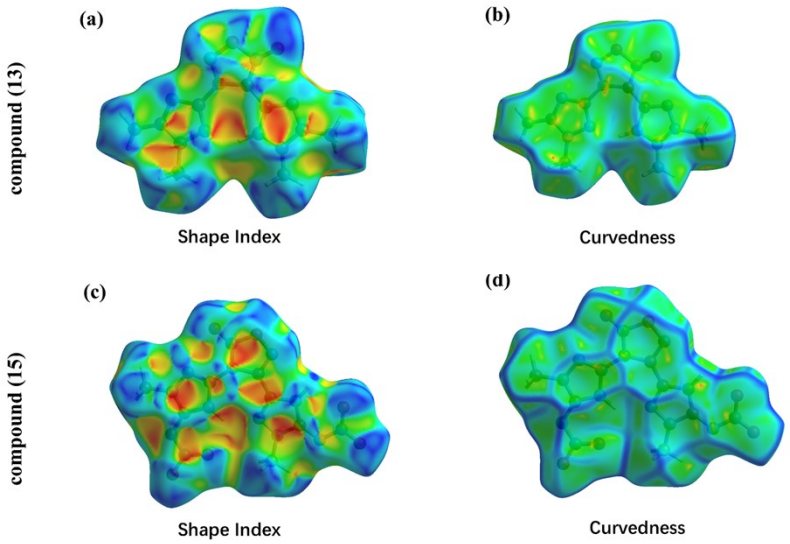
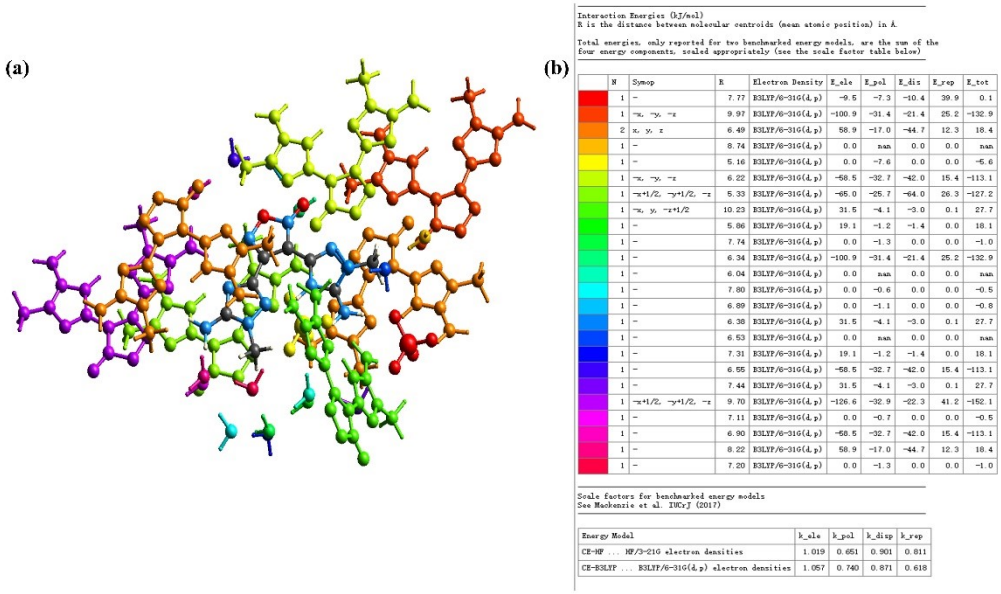


Figure S24. Shape index and Curvedness mapped in Hirshfeld face of compound (13) and compound (15)



Interaction Energies (kJ/mol)
R is the distance between molecular centroids (mean atomic position) in Å.

Total energies, only reported for two benchmarked energy models, are the sum of the four energy components, scaled appropriately (see the scale factor table below)

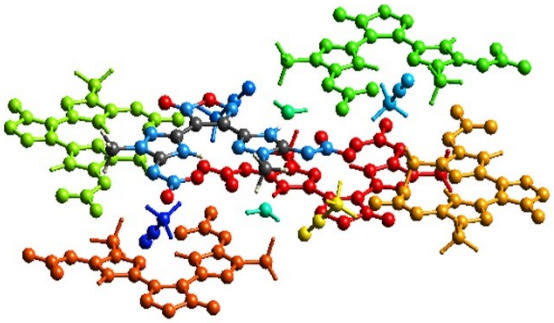
N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
1	-	7.77	B3LYP/6-31G(d,p)	-9.5	-7.3	-10.4	39.9	0.1
1	-x, -y, -z	9.97	B3LYP/6-31G(d,p)	-100.9	-31.4	-21.4	25.2	-132.9
2	x, y, z	6.49	B3LYP/6-31G(d,p)	58.9	-17.0	-44.7	12.3	18.4
1	-	8.74	B3LYP/6-31G(d,p)	0.0	nan	0.0	0.0	nan
1	-	5.16	B3LYP/6-31G(d,p)	0.0	-7.6	0.0	0.0	-6.6
1	-x, -y, -z	6.22	B3LYP/6-31G(d,p)	-58.5	-32.7	-42.0	15.4	-113.1
1	-x+1/2, -y+1/2, -z	5.33	B3LYP/6-31G(d,p)	-65.0	-25.7	-64.0	26.3	-127.2
1	-x, y, -z+1/2	10.23	B3LYP/6-31G(d,p)	31.5	-4.1	-3.0	0.1	27.7
1	-	5.86	B3LYP/6-31G(d,p)	19.1	-1.2	-1.4	0.0	18.1
1	-	7.74	B3LYP/6-31G(d,p)	0.0	-1.3	0.0	0.0	-1.0
1	-	6.34	B3LYP/6-31G(d,p)	-100.9	-31.4	-21.4	25.2	-132.9
1	-	6.04	B3LYP/6-31G(d,p)	0.0	nan	0.0	0.0	nan
1	-	7.80	B3LYP/6-31G(d,p)	0.0	-0.6	0.0	0.0	-0.5
1	-	6.89	B3LYP/6-31G(d,p)	0.0	-1.1	0.0	0.0	-0.8
1	-	6.38	B3LYP/6-31G(d,p)	31.5	-4.1	-3.0	0.1	27.7
1	-	6.53	B3LYP/6-31G(d,p)	0.0	nan	0.0	0.0	nan
1	-	7.31	B3LYP/6-31G(d,p)	19.1	-1.2	-1.4	0.0	18.1
1	-	6.55	B3LYP/6-31G(d,p)	-58.5	-32.7	-42.0	15.4	-113.1
1	-	7.44	B3LYP/6-31G(d,p)	31.5	-4.1	-3.0	0.1	27.7
1	-x+1/2, -y+1/2, -z	9.70	B3LYP/6-31G(d,p)	-126.6	-32.9	-22.3	41.2	-152.1
1	-	7.11	B3LYP/6-31G(d,p)	0.0	-0.7	0.0	0.0	-0.5
1	-	6.90	B3LYP/6-31G(d,p)	-58.5	-32.7	-42.0	15.4	-113.1
1	-	8.22	B3LYP/6-31G(d,p)	58.9	-17.0	-44.7	12.3	18.4
1	-	7.20	B3LYP/6-31G(d,p)	0.0	-1.3	0.0	0.0	-1.0

Scale factors for benchmarked energy models
See Mackenzie et al. IMCxJ (2017)

Energy Model	k_ele	k_pol	k_disp	k_rep
CE-HF ... HF/3-21G electron densities	1.019	0.651	0.901	0.811
CE-B3LYP ... B3LYP/6-31G(d,p) electron densities	1.057	0.740	0.871	0.618

Figure S25. Energy framework of cluster compound (13) calculated for B3LYP/6-31G(**).

(a)



(b)

Interaction Energies (kJ/mol)
R is the distance between molecular centroids (mean atomic position) in Å

Total energies, only reported for two benchmarked energy models, are the sum of the four energy components, scaled appropriately (see the scale factor table below)

	N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
	1	-x, -y, -z	7.38	B3LYP/6-31G(d,p)	-29.0	-11.3	-49.1	34.2	-60.6
	1	-x, -y, -z	7.39	B3LYP/6-31G(d,p)	-43.3	-10.7	-43.7	30.2	-73.1
	2	-x+1/2, y+1/2, -z+1/2	12.71	B3LYP/6-31G(d,p)	-7.0	-1.7	-3.9	0.0	-12.0
	1	-	6.36	B3LYP/6-31G(d,p)	0.0	nan	0.0	0.0	nan
	1	-x, -y, -z	7.31	B3LYP/6-31G(d,p)	-0.4	-4.0	-33.1	15.3	-22.8
	2	x+1/2, -y+1/2, z+1/2	9.51	B3LYP/6-31G(d,p)	-11.1	-3.0	-16.4	14.7	-19.2
	2	x, y, z	12.81	B3LYP/6-31G(d,p)	6.7	-0.9	-2.4	0.0	4.4
	1	-x, -y, -z	7.32	B3LYP/6-31G(d,p)	-8.2	-8.4	-59.5	27.4	-49.7
	1	-	5.12	B3LYP/6-31G(d,p)	0.0	nan	0.0	0.0	nan
	1	-	3.84	B3LYP/6-31G(d,p)	-29.0	-11.3	-49.1	34.2	-60.6
	1	-	4.97	B3LYP/6-31G(d,p)	5.6	-1.0	-5.0	0.0	0.8
	1	-	9.37	B3LYP/6-31G(d,p)	-7.0	-1.7	-3.9	0.0	-12.0
	1	-	6.58	B3LYP/6-31G(d,p)	-29.0	-11.3	-49.1	34.2	-60.6
	1	-	6.01	B3LYP/6-31G(d,p)	-11.1	-3.0	-16.4	14.7	-19.2
	1	-	7.69	B3LYP/6-31G(d,p)	5.6	-0.9	-2.4	0.0	3.1
	1	-	8.69	B3LYP/6-31G(d,p)	-43.3	-10.7	-43.7	30.2	-73.1
	1	-	7.46	B3LYP/6-31G(d,p)	-10.1	-1.7	-3.9	0.0	-15.3
	2	-x+1/2, y+1/2, -z+1/2	12.47	B3LYP/6-31G(d,p)	3.0	-0.7	-5.7	0.0	-2.3
	1	-	3.82	B3LYP/6-31G(d,p)	-0.4	-4.0	-33.1	15.3	-22.8
	1	-	6.98	B3LYP/6-31G(d,p)	-4.7	-0.2	-1.0	0.0	-6.0

Scale factors for benchmarked energy models
See Mackenzie et al. IUCrJ (2017)

Energy Model	k_ele	k_pol	k_disp	k_rep
CE-HF ... HF/3-21G electron densities	1.019	0.651	0.901	0.811
CE-B3LYP ... B3LYP/6-31G(d,p) electron densities	1.057	0.740	0.871	0.618

Figure S26. Energy framework of cluster compound (15) calculated for B3LYP/6-31G(**).

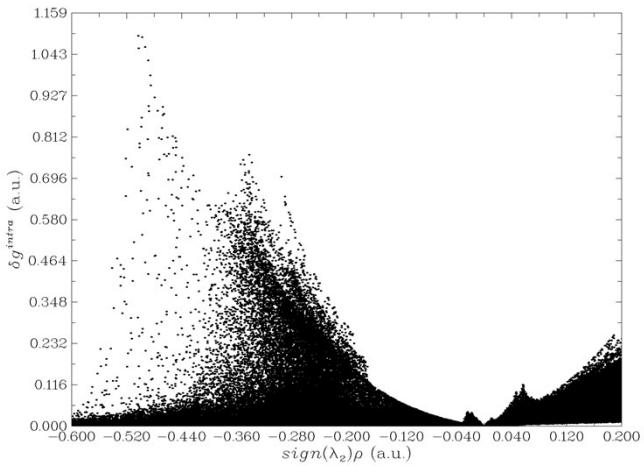


Figure S27. The Scattered graph of IGMH of the cluster of compound 13

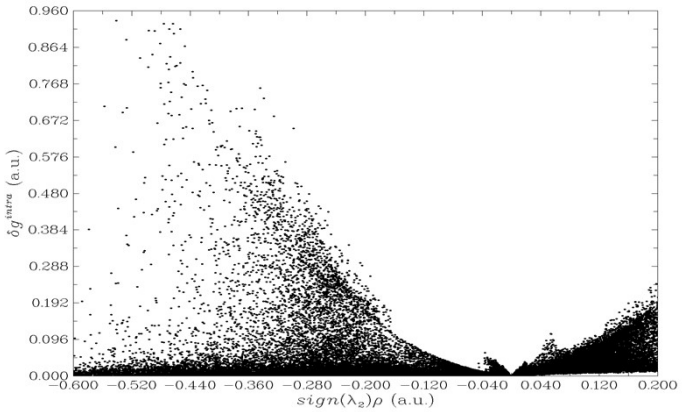


Figure S28. The Scattered graph of IGMH of the cluster of compound 15

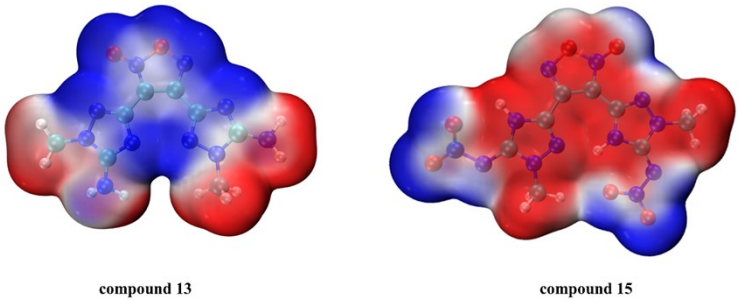


Figure S29. The electrostatic potential diagram of the compound 13 and 15

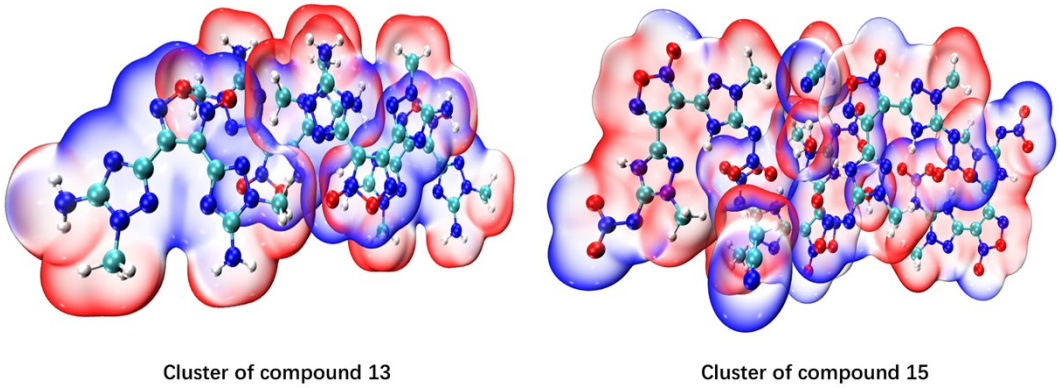


Figure S30. The electrostatic potential diagram of the cluster

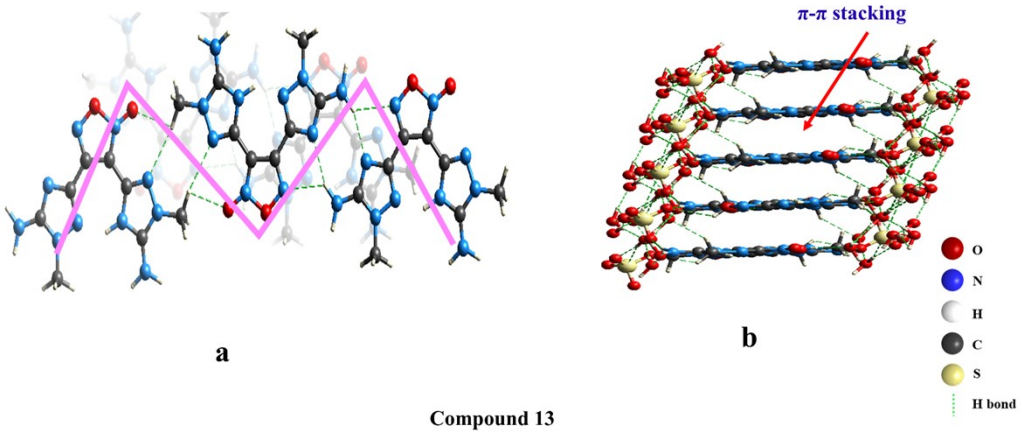


Figure S31. Crystal stacking of 2D to 3D for compound13

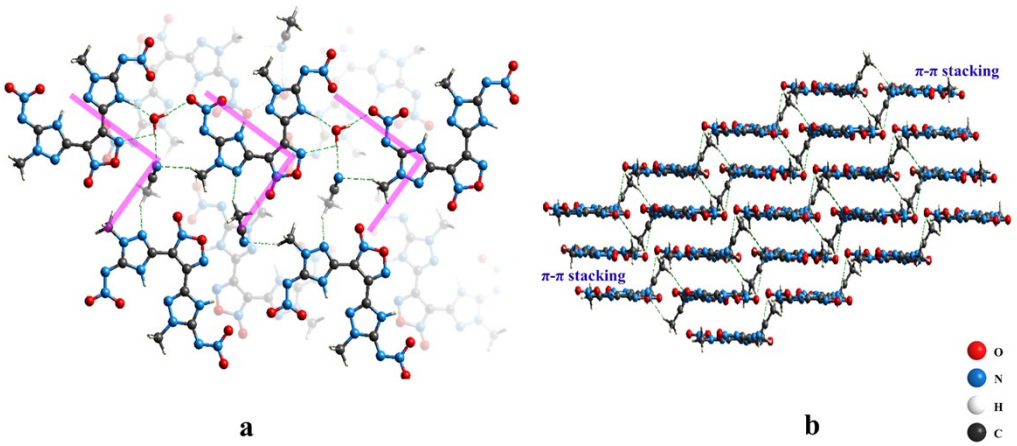


Figure S32. Crystal stacking of 2D to 3D for compound15

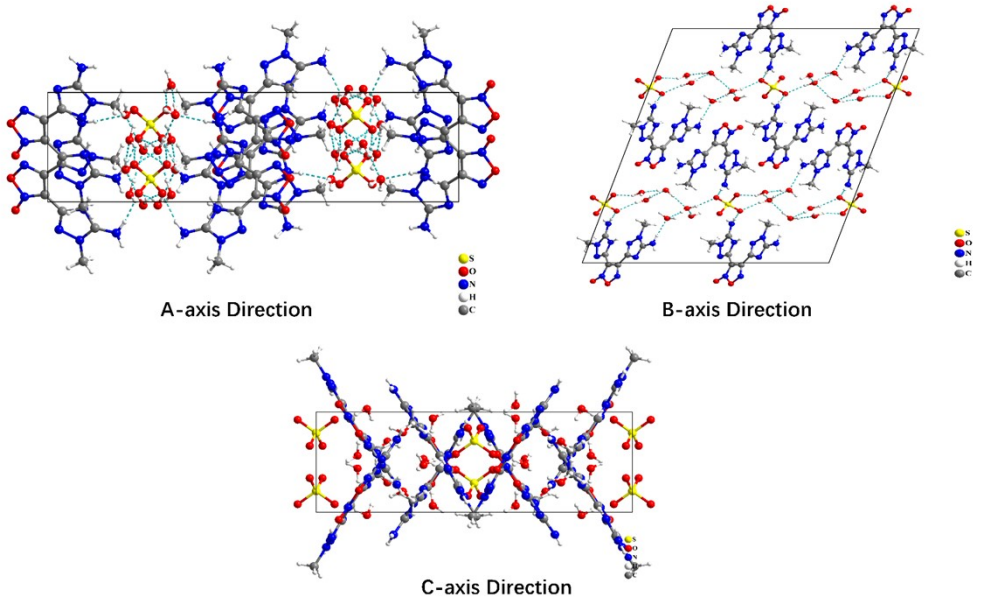


Figure S33. Supramolecular aggregations of compound 13 dirgams from three axial (a,b,c) directions.

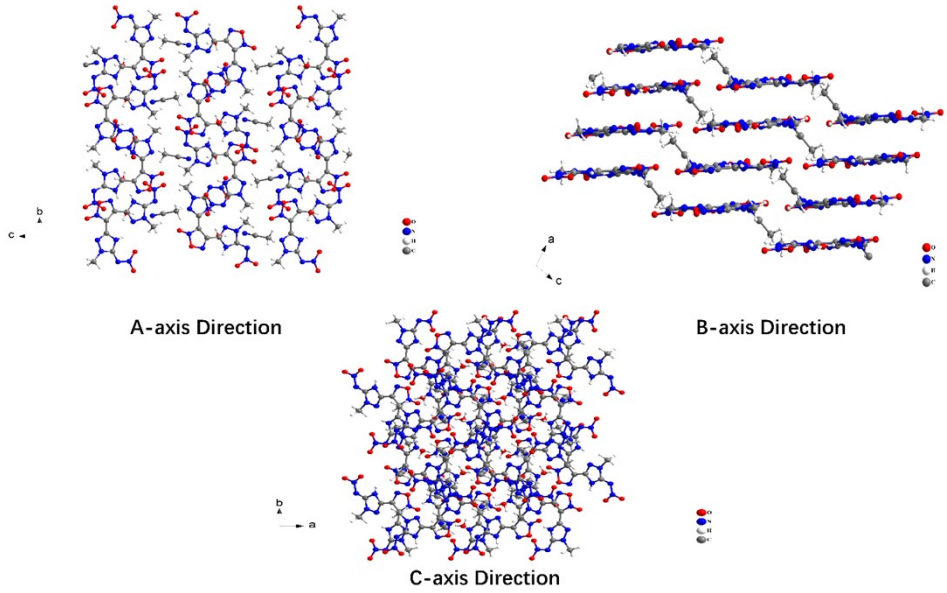


Figure S34. Supramolecular aggregations of compound 15 dirgams from three axial (a,b,c) of directions.

The energy framework of compound **15** is shown in Figure S35. The solid red, green, and blue lines represent the Coulomb interaction, the dispersive interaction, and the total interaction, respectively. The Coulomb interaction causes attraction when its value is positive and repulsion when it is negative. The dispersive interaction describes the electron long-range correlation and acts as an attractor. The total interaction represents the molecules in terms of energy. The Coulomb attraction is between the acetonitrile molecules and part of the two molecules in the upper and lower layers. The dispersion effect embodies the mutual attraction between the layers. The total energy represents the attraction between the acetonitrile molecules and the surrounding molecules. This interaction creates the crystal stacking of the graphene structure, which can be visualised as a regular stacking of shapes like forks from the top left to the bottom right. The energy framework calculation analysis of compounds **13** and **15** is provided in Figure S25 and Figure S26.

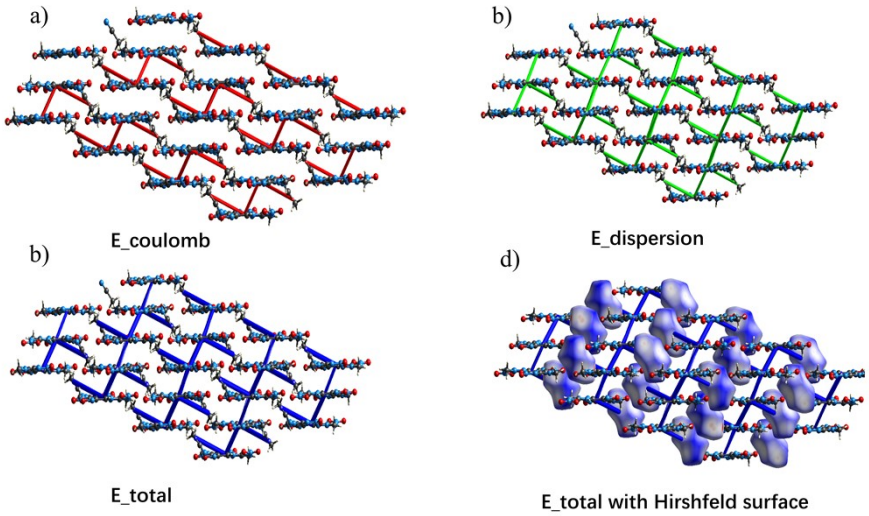


Figure S35 Coulomb energy(a); dispersion energy(b); total energy(c); Total energy plus Hirshfeld surface of acetonitrile(d);