Supporting Information

for

Assembly of three oxadiazole isomers toward versatile energetics

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1. GENERAL METHODS

All reagents were purchased from Energy Chemical of analytical grade and were used as supplied, if not stated otherwise. The melting points and decomposition temperatures were obtained on a differential scanning calorimeter (Mettler Toledo DSC823e) at a scan rate of 5 °C min^{-1} in closed Al containers with a nitrogen flow of 50 ml min^{-1}. Chemical shifts in the ^{1}H and ^{13}C NMR spectra were reported relative to Me_4Si as external standards. IR spectra were recorded using KBr pellets for solids on a Thermo Nicolet iS10 spectrometer. Elemental analyses were carried out on a vario EL III CHNOS elemental analyzer.

2. SAFETY PRECAUTIONS

Although none of the energetic materials described herein have exploded or detonated in the course of this research, these materials should be handled with extreme care using the best safety practices.

3. EXPERIMENTAL SECTION

3.1 Construction of the tricyclic backbone

3-Amino-4-aminoximinofurazan (1): Compounds **1** was prepared through previous method.[1] Light yellow solid (47.8 g, 76% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 10.50 6.29 6.20 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ154.93 144.48 140.52 ppm. IR (KBr): 3439 3387 3334 3203 1670 1618 1574 1531 1384 1186 914 797 cm^{-1}. Elemental analysis calcd for $C_3H_5N_5O_2$ (143.11): C 25.18, H 3.52, N 48.94 %; found C 25.38, H 3.41, N 48.82 %.

Ethyl 3-(4-amino-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazole-5-carboxylate (2): Ethyl oxalyl chloride (5 mL) was added slowly into the suspension of **1** (5.72 g, 40 mol) in acetonitrile. Anhydrous potassium carbonate (11.20 g, 81 mol) was added to the clear solution. After stirring for 12 h at 60 °C, the filtrate was concentrated and washed with excessive water to get **2** (3.89 g, 43% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 6.54 [4.51 4.49 4.48 4.46] [1.40 1.38 1.36]. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 167.89 160.18 155.89 153.65 137.21 64.14 14.21 ppm. IR (KBr): 3467 2987 1752 1729 1609 1507 1450 1371 1303 1214 1015 968 860 cm^{-1}. Elemental analysis calcd for $C_7H_7N_5O_4$ (225.05): C 37.34, H 3.13, N 31.10 %; found C 37.21, H 3.37, N 30.68 %.

3-(4-amino-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazole-5-carbohydrazide (3): 80% aqueous hydrazine solution (1.30 g, 20 mmol) was added into the suspension of **2** (2.25 g, 10 mmol) in ethanol (50 mL). After stirring for 2 h, the precipitate was filtered and washed with excessive water to get **3** (1.83 g, 75% yield).

^1H NMR (DMSO-d_6, 400 MHz): δ 6.71 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 170.14 159.33 156.20 151.25 136.73 ppm. IR (KBr): 3455 3112 1637 1559 1490 1346 1231 1183 1143 1016 971 921 759 637 584 cm^{-1}. Elemental analysis calcd for $C_5H_5N_7O_3$ (211.14): C 28.44, H 2.39, N, 46.44 %; found C 28.77, H 2.54, N 46.12 %.

4-(5-(5-amino-1,3,4-oxadiazol-2-yl)-1,2,4-oxadiazol-3-yl)-1,2,5-oxadiazol-3-amine (4): Potassium bicarbonate (1.40 g, 14 mmol) was added into the suspension of **3** (2.11 g, 10 mmol) in water. And then cyanogen bromide (1.28 g, 12 mmol) was added slowly to the mixture at 0-5 °C. After stirring for 24 h, the precipitate was filtered and washed with water and methanol to get **4** (1.089 g, 46% yield). ^1H NMR (DMSO-d_6, 400 MHz): δ 8.16, 6.54 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 165.95 164.13 159.96 155.89 146.99 137.14 ppm. IR (KBr): 3449 3329 3111 1660 1635 1601 1586 1566 1495 1379 1163 1129 1066 1007 964 908 cm^{-1}. Elemental analysis calcd for $C_6H_4N_8O_3$ (236.04): C 30.52, H 1.71, N 47.45 %; found C 30.32, H 1.89, N 47.29 %.

3.2 Controllable amino functionalization

5-(3-(4-nitro-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazol-5-yl)-1,3,4-oxadiazol-2-amine (5): 30% H_2O_2 (30 mL) was added in 98 % H_2SO_4 (30 mL) in an ice bath. **4** (2.5 g, 10.6 mmol) was added to the mixture with stirring for 0.5 h. The reaction was heated up to 25 °C and was stirred for 0.5 h. Then the reaction was heated up to 45 °C with stirring for another 2 h to get **5** (1.41 g, 63% yield). ^1H NMR (DMSO-d_6, 400 MHz): δ 8.17 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 165.98 164.68 160.20 157.87 146.87 140.85 ppm. IR (KBr): 3239 3160 1676 1664 1602 1565 1442 1402 1356 1308 1294 1176 1117 1093 1059 1029 1006 973 824 741 cm^{-1}. Elemental analysis calcd for $C_6H_2N_8O_5$ (266.01): C 27.08, H 0.76, N 42.11 %; found C 27.21, H 0.87, N 42.43, %.

Nitro (5-(3-(4-nitro-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazol-5-yl)-1,3,4-oxadiazol-2-yl) amide (6): **5** (0.5 g, 1.9 mmol) was added slowly in portions to HNO_3 (98 %, 5 mL) at -10 °C. After stirring for 3 h, the reaction mixture was warmed up to room temperature and stirred for 24 h. Then the reaction mixture was poured into ice water (80 mL). The precipitate was filtered and dried in air to get **6** (0.21 g, 36 % yield). ^1H NMR (400MHz, d_6-DMSO): δ 6.05 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 164.30 164.15 160.18 158.21 146.41 140.66 ppm. IR (KBr): 3439 2963 1592 1577 1544 1509 1360 1264 1165 1104 1035 970 823 802 727 cm^{-1}. Elemental analysis calcd for $C_6HN_9O_7$ (311.00): C 23.16, H 0.32, N 40.52 %; found C 23.43, H 0.55, N 40.75 %.

5-(3-(4-azido-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazol-5-yl)-1,3,4-oxadiazol-2-amine (7): $NaNO_2$ (0.35 g, 5mmol) was added in 98 % H_2SO_4 (25 mL) and cooled in an ice bath. **4** (0.78 g, 3.3 mmol) was added to the mixture until it was completely dissolved. And then, 98 % acetic acid (25 mL) and NaN_3 (0.86 g, 13.2 mmol) was respectively added to the mixture and the mixture was stirred for 12 h. After pouring the solution into ice water (150 mL), the precipitate was filtrated and dried in air to give product **7** (0.49 g, 50 % yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 8.16, 6.75 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 166.02, 164.65, 158.87, 153.59, 146.93, 139.42 ppm. IR (KBr): 3372 2149 1727 1675 1637 1604 1525 1436 1310 1200 1150 1092 1022 966 885 cm^{-1}. Elemental analysis calcd for $C_6H_2N_{10}O_3$ (262.03): C 27.49, H 0.77, N 53.43 %; found C 27.67, H 0.92, N 53.31 %.

N-(5-(3-(4-azido-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazol-5-yl)-1,3,4-oxadiazol-2-yl) nitramide (8): **4** (0.5 g, 1.9 mmol) was added slowly in portions to HNO_3 (98 %, 5 mL) with stirring for 3 h and cooling under -10 °C. The mixture was warmed up to room temperature and stirred for 24 h, and then the reaction mixture was poured into ice water (80 mL). The precipitated was collected by filtration and dried in air get **8** (0.37 g, 63% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 6.46 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 165.44 164.44 158.98 153.66 146.98 139.61 ppm. IR (KBr): 3458 2146 1664 1590 1543 1518 1339 1275 1200 1162 1104 968 774 717 cm^{-1}. Elemental analysis calcd for $C_6HN_{11}O_5$ (307.02): C 23.46, H 0.33, N 50.16 %; found C 23.21, H 0.24, N 50.37 %.

4-(5-(5-nitramino-1,3,4-oxadiazol-2-yl)-1,2,4-oxadiazol-3-yl)-1,2,5-oxadiazol-3-amine (9): **4** (0.40 g, 1.7 mmol) was added slowly in portions to HNO_3 (98 %, 4 mL) at -10 °C. The reaction mixture was stirred for 4h, and then the reaction mixture was poured into ice water (80 mL). The precipitate was filtered to get **9** (0.18 g, 38% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 8.14 5.92 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 165.88 164.26 159.56 153.20 146.93 142.59. IR (KBr): 3427 1720 1658 1534 1504 1385 1298 1168 1007 963 879 833 767 738 cm^{-1}. Elemental analysis calcd for $C_6H_3N_9O_5$ (281.03): C 25.63, H 1.08, N 44.84 %; found C 25.49, H 1.02, N 44.98 %.

N-(5-(3-(4-(nitroamino)-1,2,5-oxadiazol-3-yl)-1,2,4-oxadiazol-5-yl)-1,3,4-oxadiazol-2-yl) nitramide (10): **9** (0.4 g, 1.4 mmol) was added slowly in portions to HNO_3 (98 %, 4 mL) with stirring at -10 °C. The mixture was warmed up to room temperature and stirred for 24 h. Then the reaction mixture was poured into ice water (80 mL). The precipitated was collected by filtration and dried in air to get **10** (0.26 g, 58% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 7.20 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 164.16 163.85 160.09 154.17 146.51 142.37 ppm. IR (KBr): 3447 2146 1591 1543 1518 1459 1345 1277 1201 1162

1107 968 775 718 cm^{-1}. Elemental analysis calcd for $C_6H_2N_{10}O_7$ (326.01): C 20.10, H 0.62, N 42.95 %; found C 19.96, H 0.88, N 43.09 %.

3.3 Salt formation

***Dihydrazinium 4-(5-(5-nitramino-1,3,4-oxadiazol-2-yl)-1,2,4-oxadiazol-3-yl)-1,2,5-oxadiazol-3-nitramino (11)*:** A solution of 80% hydrazine hydrate (1 mmol) in 2 mL acetonitrile, was added drop wise to a suspension of **10** (0.28 g ,1 mmol) in 10mL acetonitrile. The precipitate was filtered and washed with acetonitrile to get **11** (0.29 g, 75% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 7.11 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 167.82 164.19 160.69 158.81 148.06 142.10 ppm. IR (KBr): 3445, 1635, 1511, 1426, 1309, 1262,1178, 1084, 971, 810, 775, 730 cm^{-1}. Elemental analysis calcd for $C_6H_{10}N_{14}O_7$ (388.07): C 18.56, H 2.08, N 50.51 %; found C 18.64, H 2.24, N 50.31 %.

Dihydroxylammonium 4-(5-(5-nitramino-1,3,4-oxadiazol-2-yl)-1,2,4-oxadiazol-3-yl)-1,2,5-oxadiazol-3-nitramino (12): A solution of 50% aqueous hydroxylamine (1 mmol) in 2 mL acetonitrile, was added drop wise to a suspension of **10** (0.28 g ,1 mmol) in 10mL acetonitrile. The precipitate was filtered and washed with acetonitrile to get **12** (0.32 g, 80% yield). ^{1}H NMR (DMSO-d_6, 400 MHz): δ 10.07 ppm. ^{13}C NMR (DMSO-d_6, 100 MHz): δ 167.80 164.18 160.67 158.75 148.03 142.09 ppm. IR (KBr): 3439, 3196, 1627, 1605, 1512, 1427, 1293, 1261, 1194, 1085, 1005, 972, 924, 880, 834, 770, 731 cm^{-1}. Elemental analysis calcd for $C_6H_8N_{12}O_9$ (392.05): C 18.37, H 2.06, N 42.86; found C 18.51, H 2.21, N 42.99 %.

4. SPECTRA ANALYSIS

4.1 ^{1}H and ^{13}C NMR

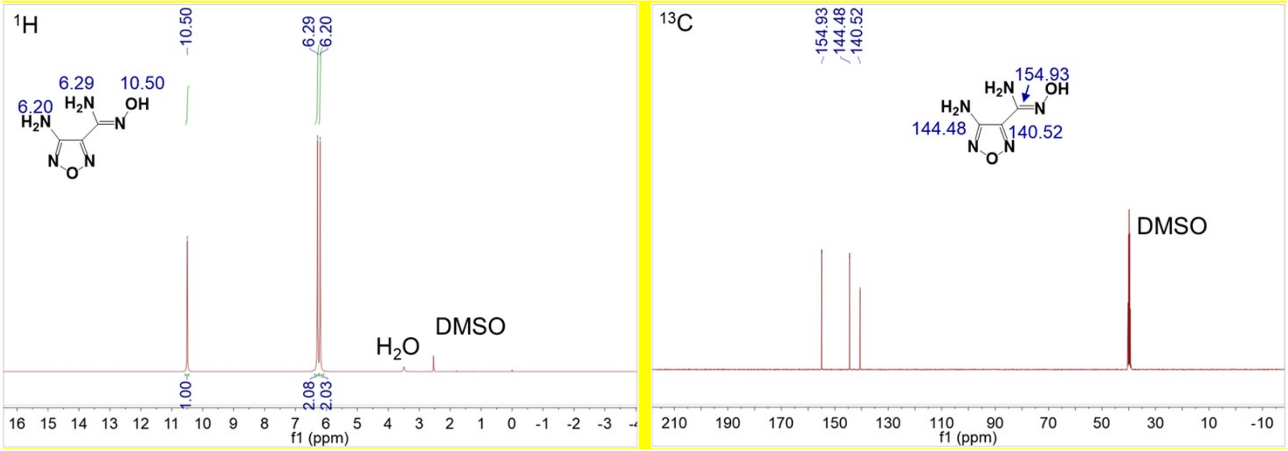


Figure S1. ^{1}H and ^{13}C NMR spectra of compound **1**.

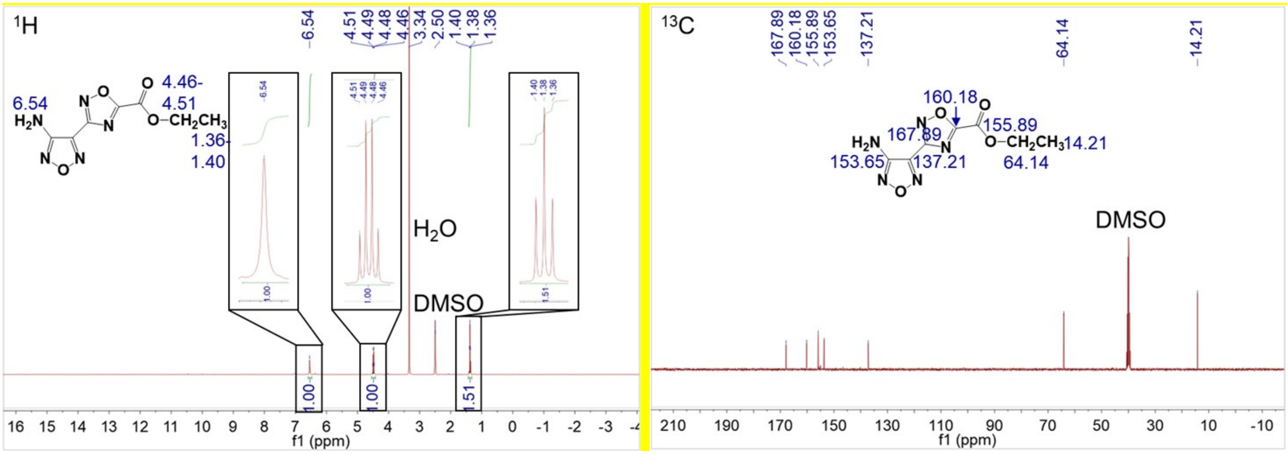


Figure S2. ^{1}H and ^{13}C NMR spectra of compound **2**.

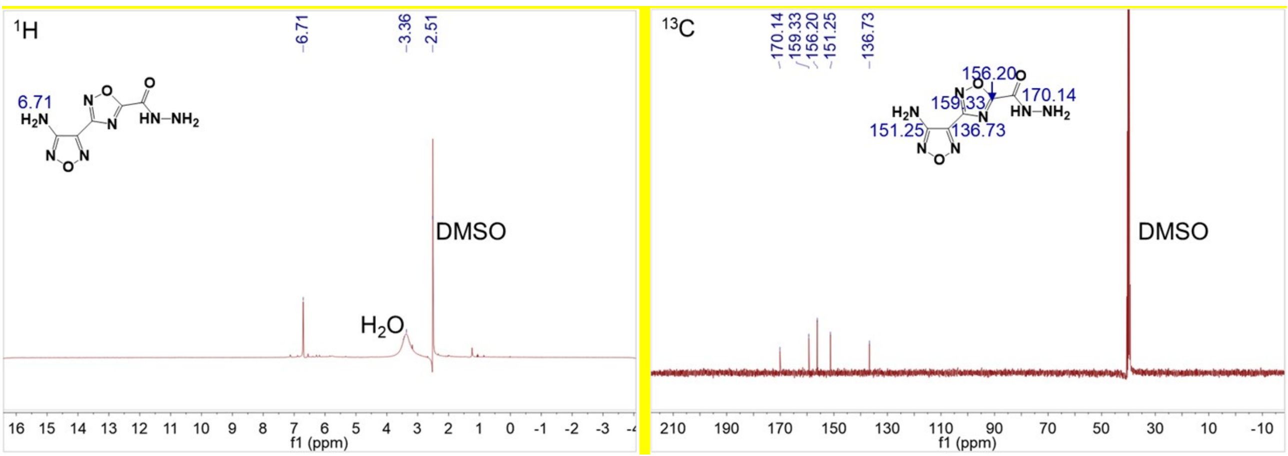


Figure S3. ^{1}H and ^{13}C NMR spectra of compound **3**.

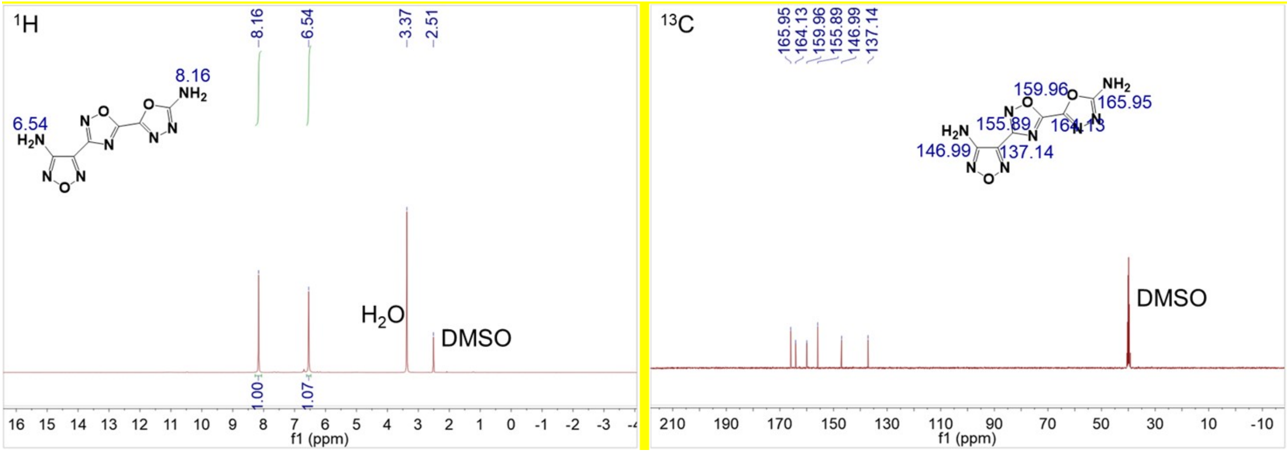


Figure S4. ^{1}H and ^{13}C NMR spectra of compound **4**.

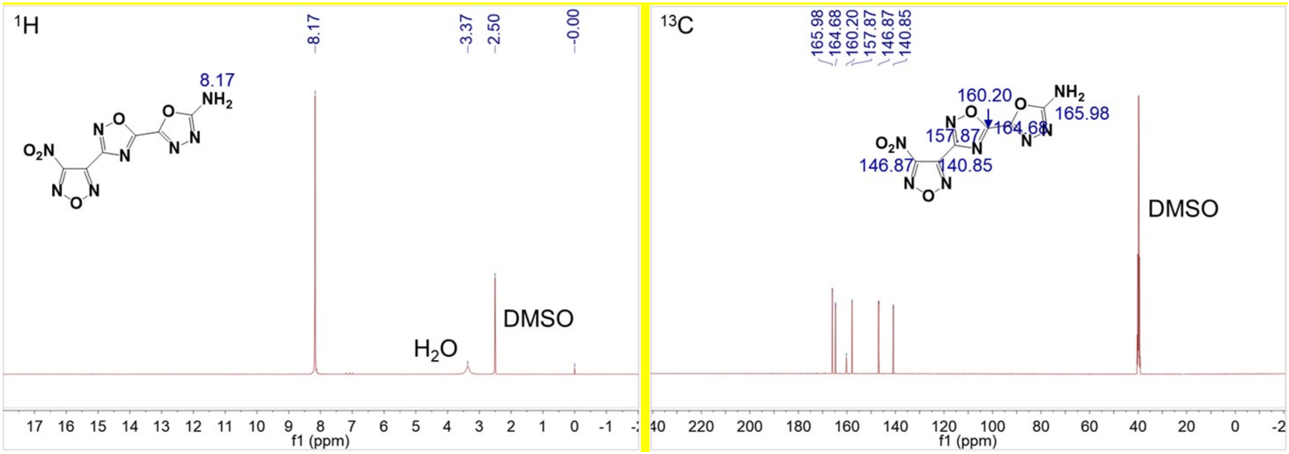


Figure S5. ^{1}H and ^{13}C NMR spectra of compound **5**.

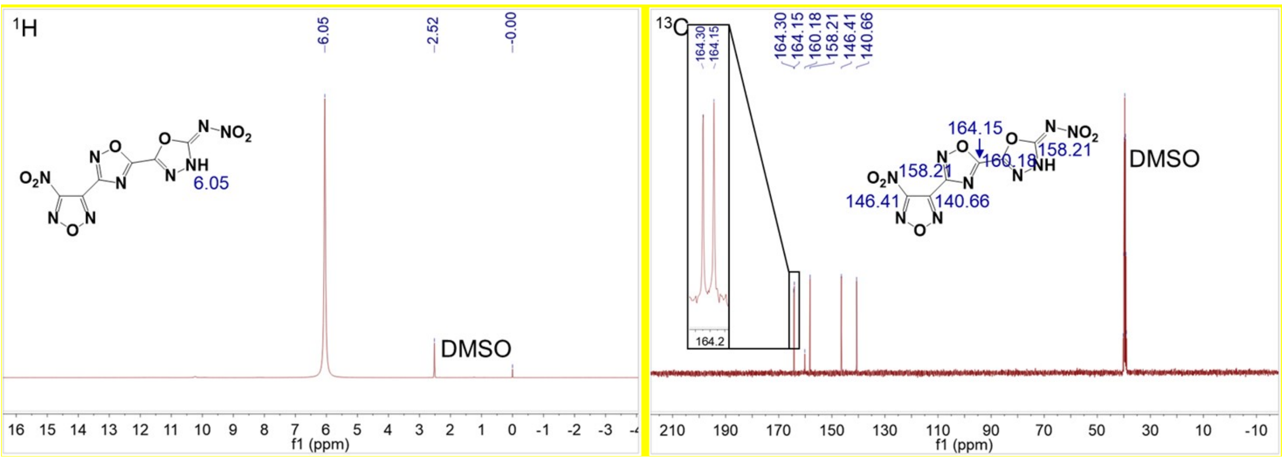


Figure S6. ^{1}H and ^{13}C NMR spectra of compound **6**.

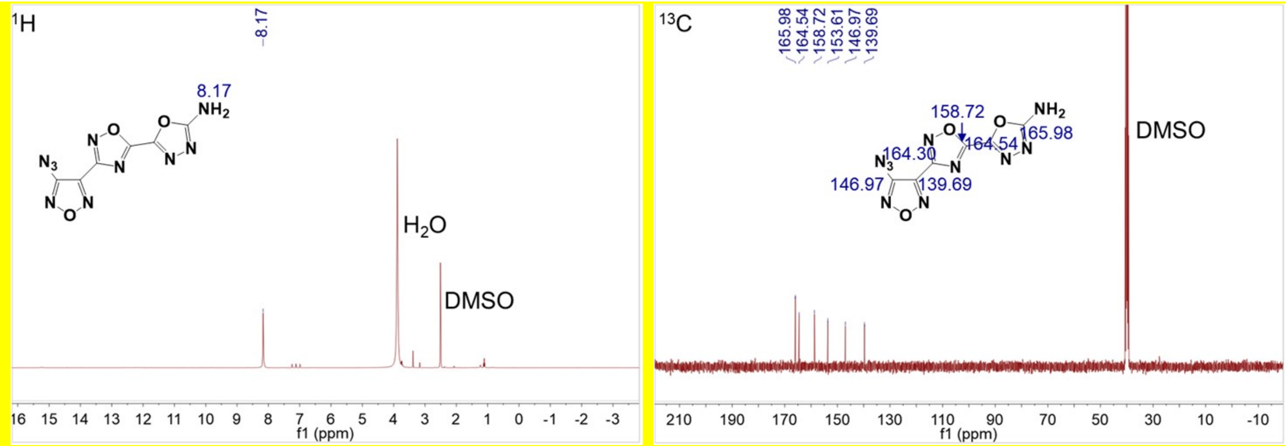


Figure S7. ^{1}H and ^{13}C NMR spectra of compound **7**.

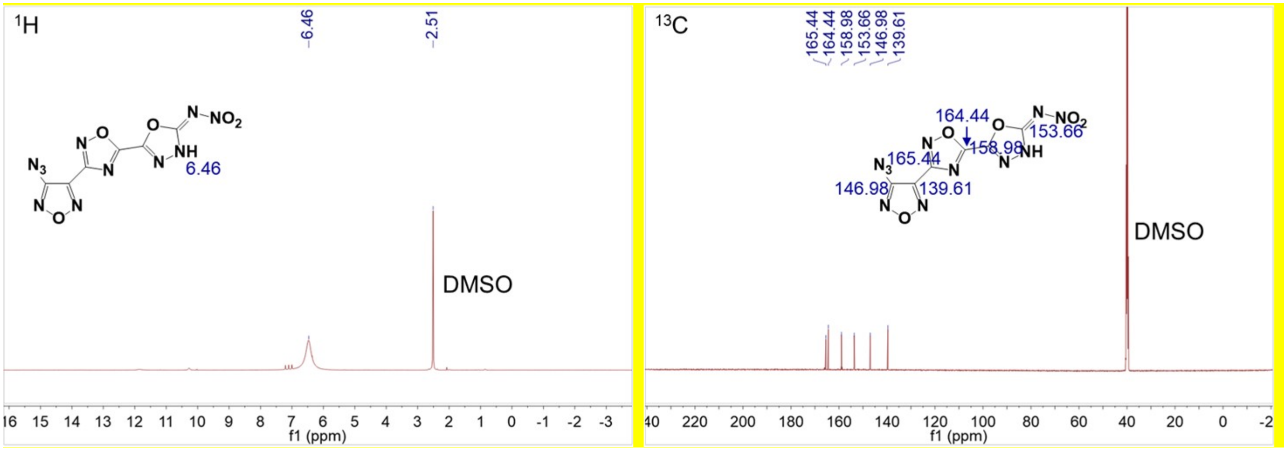


Figure S8. ^{1}H and ^{13}C NMR spectra of compound **8**.

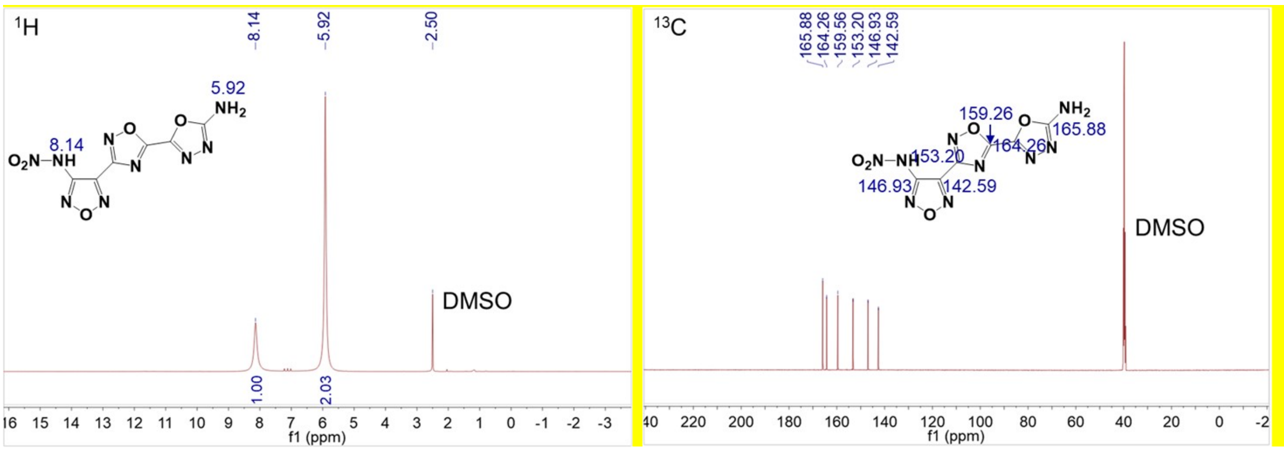


Figure S9. ^{1}H and ^{13}C NMR spectra of compound **9**.

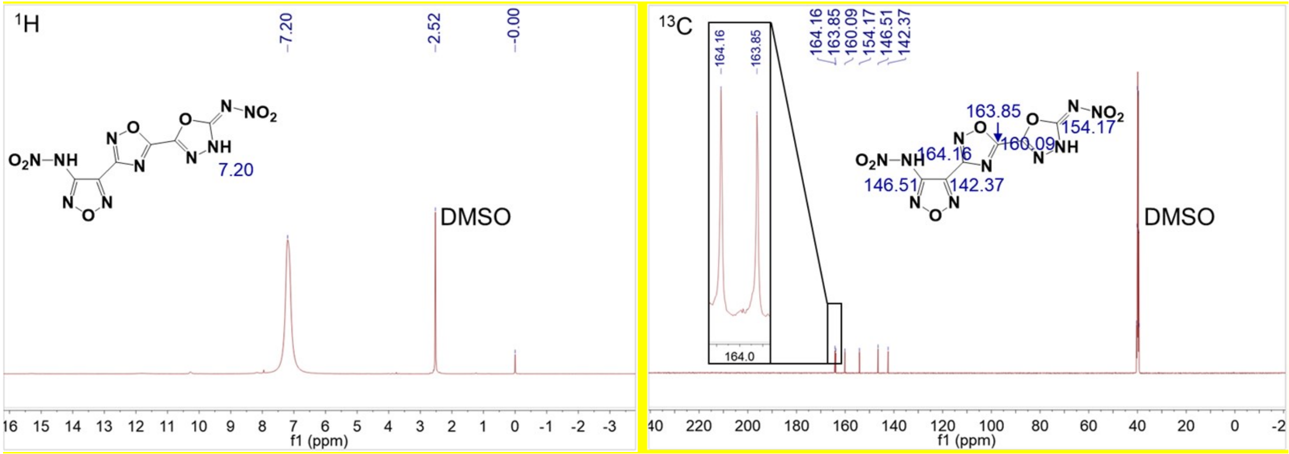


Figure S10. ^{1}H and ^{13}C NMR spectra of compound **10**.

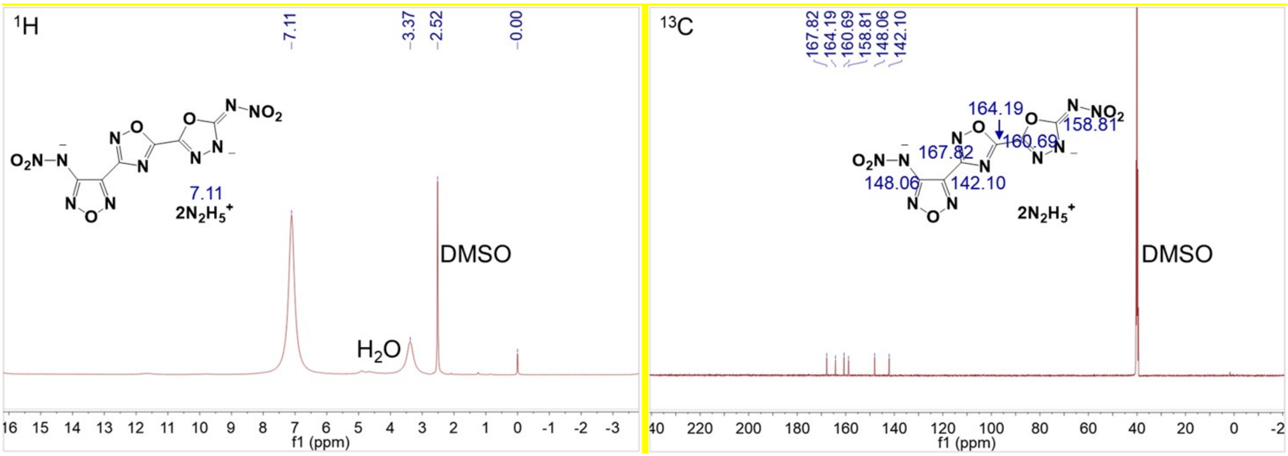


Figure S11. ^{1}H and ^{13}C NMR spectra of compound **11**.

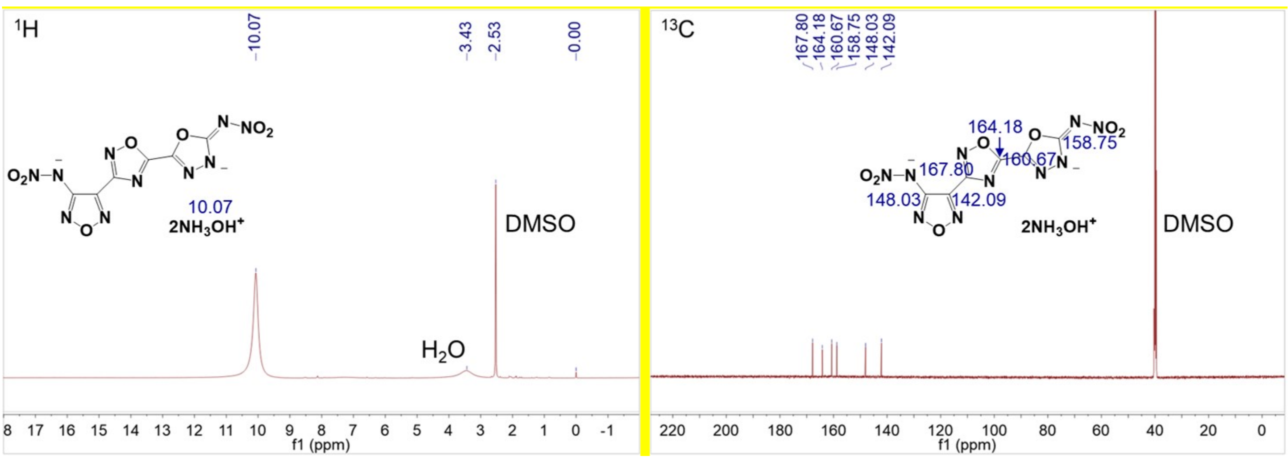


Figure S12. ^{1}H and ^{13}C NMR spectra of compound **12**.

4.2 IR spectra

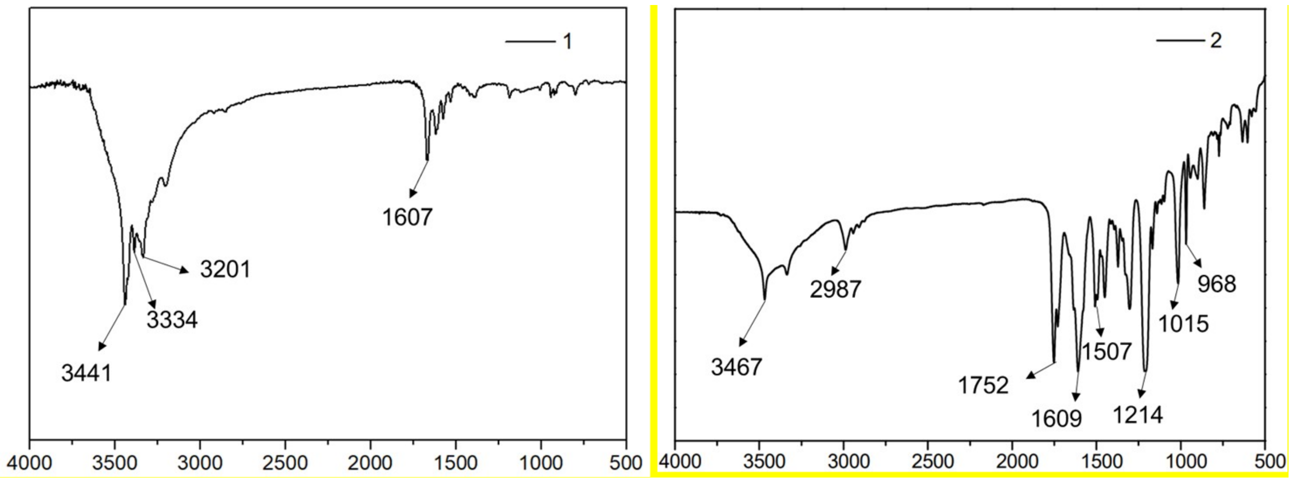


Figure S13. IR spectra of compounds **1 and 2**.

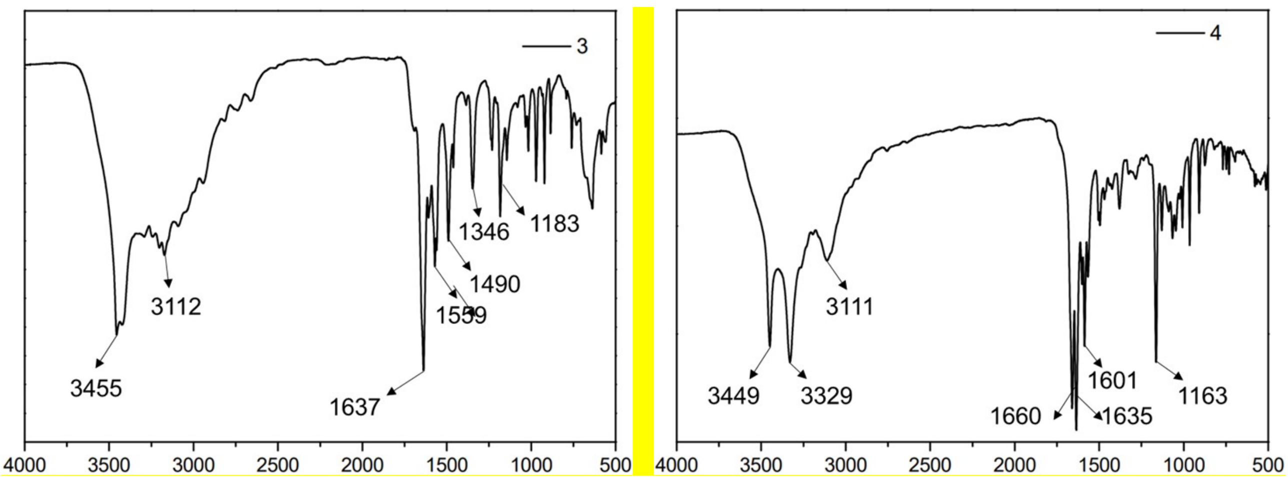


Figure S14. IR spectra of compounds **3 and 4**.

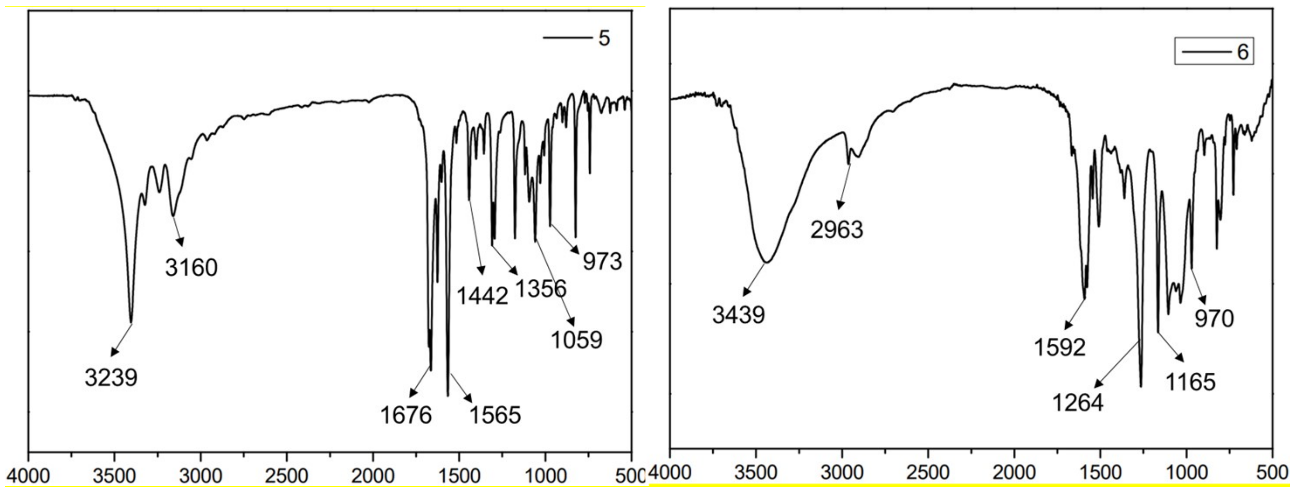


Figure S15. IR spectra of compounds **5 and 6**.

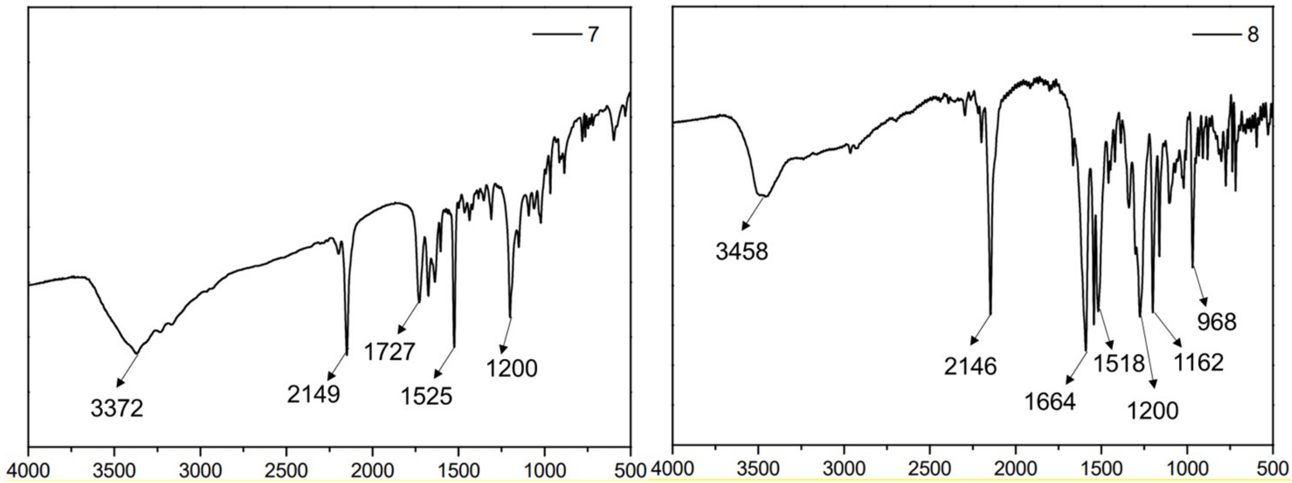


Figure S16. IR spectra of compounds **7 and 8**.

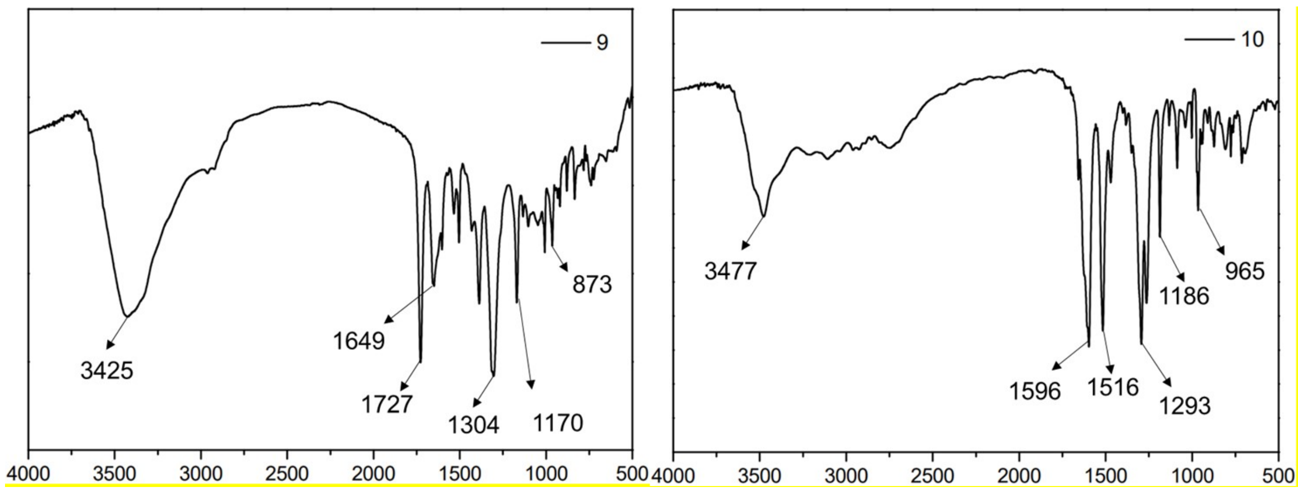


Figure S17. IR spectra of compounds **9 and 10**.

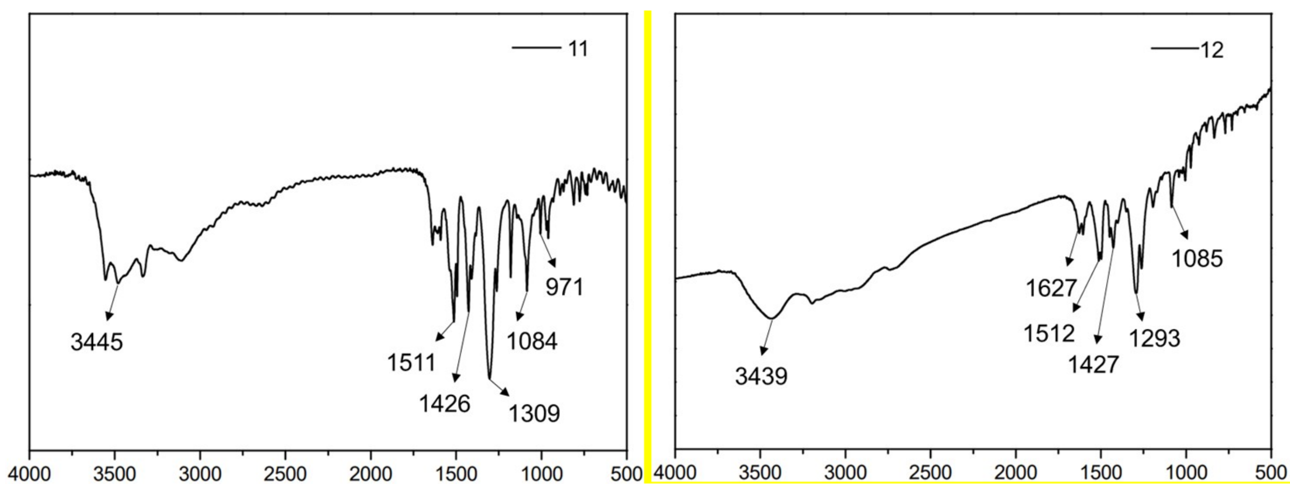


Figure S18. IR spectra of compounds **11 and 12**.

5. X-RAY CRYSTALLOGRAPHY DETAIL

An orange block crystal (**1**) of dimensions 0.12×0.10×0.08 mm^3, a colorless block crystal (**2**) of dimensions 0.10×0.10×0.10 mm^3, a colorless block crystal (**3**) of dimensions 0.21×0.11×0.08 mm^3, a colorless needle crystal (**4**.DMF) of dimensions 0.30×0.20×0.10 mm^3, a colorless plate crystal (**5**) of dimensions 0.10×0.06×0.03 mm^3, a colorless block crystal (**6**) of dimensions 0.22×0.20×0.18 mm^3, a colorless block crystal (**7**) of dimensions 0.12×0.10×0.08 mm^3, a colorless plate crystal (**8**) of dimensions 0.16×0.10×0.08 mm^3, and a colorless block crystal (**9**.$2H_2O$) of dimensions 0.20×0.10×0.05 mm^3, a colorless block crystal (**10**) of dimensions 0.12×0.08×0.03 mm^3 were mounted on an Enraf-Nonius CAD4 four-circle diffractometer using graphite-monochromated Mo Ka radiation (λ = 0.71073 Å) at 150, 170, and 296 K. Corrections for Lorentz and polarization effects and for absorption (ψ scan) were applied. The structure was solved by direct methods using SHELXS-97 and refined by full-matrix least-squares calculation on F2 with SHELXL-97. All non-hydrogen atoms were refined anisotropically. All hydrogen atoms were placed in calculated positions and were assigned fixed isotropic thermal parameters at 1.2 times the equivalent isotropic U of the atoms to which they were attached and allowed to ride on their respective parent atoms. The contributions of these hydrogen atoms were included in the structure-factor calculations.

Table S1. Crystallographic data for **1**, **2**, **3**, and **4**.

Compound	**1**	**2**	**3**	**4**.DMF
Formula	$C_3H_5N_5O_2$	$C_7H_7N_5O_4$	$C_5H_5N_7O_3$	$C_6H_4N_8O_3$.DMF
M_w	143.12	225.18	211.16	309.27
Crystal system	Monoclinic	Monoclinic	Orthorhombic	Triclinic
Space group	$P2_1/c$	$P2_1/n$	$Pna2_1$	P-1
a [Å]	7.654(6)	4.9648(16)	17.065(17)	6.1101(7)
b [Å]	11.714(10)	24.229(6)	12.243(12)	8.0905(10)
c [Å]	19.209(15)	8.225(2)	4.006(4)	13.8354(17)
α[°]	90	90	90	100.725(4)
β[°]	96.528(10)	106.449(12)	90	94.384(4)
γ[°]	90	90	90	99.483(4)
V [$Å^3$]	1711(2)	948.9(5)	837.0(14)	658.75(14)
Z	12	4	4	2
T [K]	296	150	296	150
λ [Å]	0.71073	1.34139	0.71073	1.34139
P_{calcd}[g cm^{-3}]	1.667	1.576	1.676	1.559
μ [mm^{-1}]	0.141	0.728	0.141	0.697
F(000)	888	464	432	320
θ range[°]	2.04-29.24	3.17-55.02	2.05-27.56	4.92-54.91
Index ranges	$-9 \le h \le 9$	$-5 \le h \le 6$	$-22 \le h \le 22$	$-7 \le h \le 7$
	$0 \le k \le 15$	$-29 \le k \le 29$	$-15 \le k \le 15$	$-9 \le k \le 9$
	$0 \le l \le 26$	$-10 \le l \le 10$	$-5 \le l \le 5$	$-16 \le l \le 16$
Data/restraints/ parameters	4151/3/275	1788/5/147	1922/1/144	2444/0/202
GOF on F2	1.074	1.092	1.038	1.078
R[F^2> 2σ(F^2)]	0.0713	0.0686	0.0508	0.0981
wR(F^2)	0.1889	0.2073	0.1015	0.2980

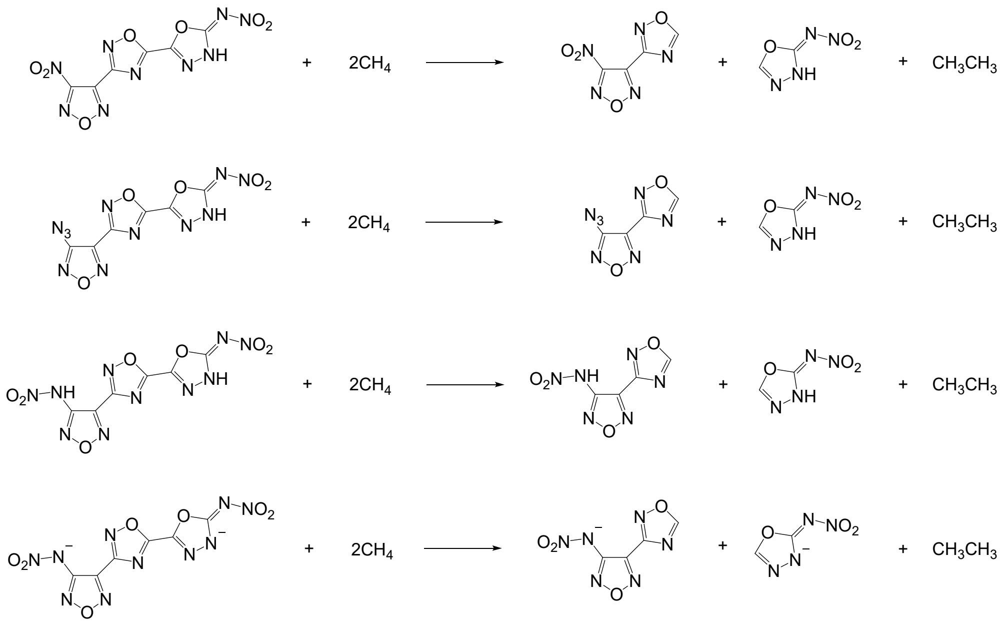
Table S2. Crystallographic data for **5, 6, 7, and 8.**

Compound	**5**	**6**	**7**	**8**.H_2O
Formula	$C_6H_2N_8O_5$	$C_6HN_9O_7$	$C_6H_2N_{10}O_3$	$2(C_6HN_{11}O_5).H_2O$
M_w	266.16	311.16	262.18	632.37
Crystal system	Monoclinic	Orthorhombic	Triclinic	Orthorhombic
Space group	$P2_1/n$	Pca_{21}	P-1	Pnna
a [Å]	7.2609(5)	6.404(2)	4.4199(13)	9.604(2)
b [Å]	18.6033(12)	21.152(7)	7.132(2)	40.625(8)
c [Å]	7.3808(5)	8.235(3)	16.288(5)	5.931(2)
α[°]	90	90	81.681(7)	90
β[°]	96.583(2)	90	87.434(7)	90
γ[°]	90	90	76.836(7)	90
V [$Å^3$]	990.4(12)	1115.5(7)	494.7(3)	2314.1(10)
Z	4	4	2	4
T [K]	296	150	296	150
λ [Å]	0.71073	1.34139	0.71073	1.34138
P_{calcd}[g cm^{-3}]	1.785	1.853	1.760	1.815
μ [mm^{-1}]	0.157	0.963	0.147	0.905
F(000)	536	624	264	1272
θ range[°]	2.99-27.49	3.64-53.58	2.96-25.12	3.79-55.08
Index ranges	$-8 \le h \le 9$	$-7 \le h \le 7$	$-5 \le h \le 5$	$-11 \le h \le 11$
	$-23 \le k \le 22$	$-25 \le k \le 25$	$-8 \le k \le 8$	$-49 \le k \le 49$
	$-7 \le l \le 9$	$-9 \le l \le 9$	$-19 \le l \le 19$	$-7 \le l \le 6$
Data/restraints/ parameters	2249/0/173	1811/58/218	1750/0/172	2190/0/204
GOF on F2	1.039	1.099	0.938	1.087
$R[F^2 > 2\sigma(F^2)]$	0.0425	0.1040	0.0582	0.0359
$wR(F^2)$	0.1045	0.1040	0.1717	0.1110

Table S3. Crystallographic data for **9** and **10.**

Compound	**9.**$2H_2O$	**10.**$2H_2O$
Formula	$C_6H_3N_9O_5.2H_2O$	$C_6H_2N_{10}O_7.2H_2O$
M_w	317.21	362.21
Crystal system	Monoclinic	Monoclinic
Space group	$P2_1$	$P2_1/c$
a [Å]	11.297(7)	23.945(3)
b [Å]	4.451(3)	4.5598(6)
c [Å]	12.821(8)	11.7638(17)
α[°]	90	90
β[°]	112.416(12)	90.12(1)
γ[°]	90	90
V [$Å^3$]	597.7(7)	1284.4(3)
Z	2	4
T [K]	296	170
λ [Å]	0.71073	0.71073
P_{calcd}[g cm^{-3}]	1.769	1.873
μ [mm^{-1}]	0.160	0.175
F(000)	324.0	736
θ range[°]	3.05-20.5	2.55-25.73
Index ranges	$-11 \le h \le 13$	$-29 \le h \le 29$
	$-5 \le k \le 5$	$-5 \le k \le 5$
	$-15 \le l \le 14$	$-14 \le l \le 14$
Data/restraints/parameters	1972/7/205	2648/0/234
GOF on F2	1.000	1.119
$R[F^2 > 2\sigma(F^2)]$	0.0707	0.0485
$wR(F^2)$	0.1845	0.1297

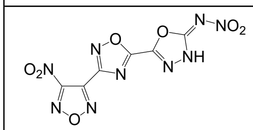
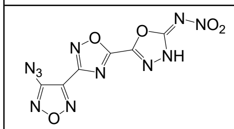
6. HEATS OF FORMATION

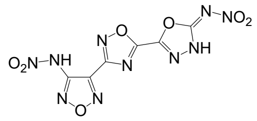
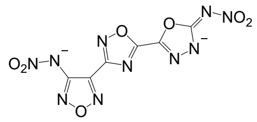
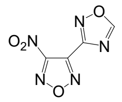
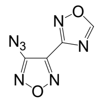
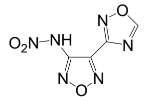
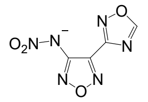
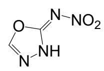
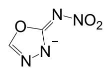


Scheme S1. Isodesmic reactions for calculating heats of formation.

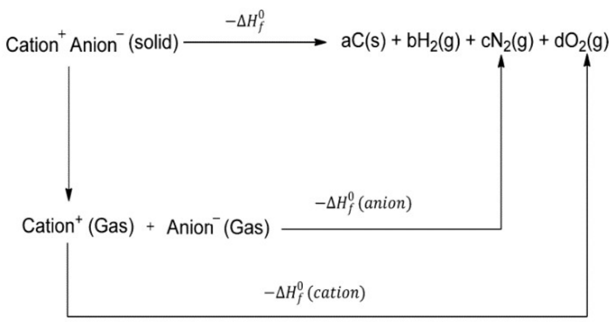
All the ab initio calculations involved in this work were carried out using the Gaussian 09 suite of programs. The geometric optimization and frequency analyses of the structures are based on available single-crystal structures and using the B3LYP functional with the 6-31++G(d,p) basis set. The geometrical were optimized with no constraints imposed under default convergence criteria. Total energy (E0) and zero-point energy (ZPE) were calculated at the MP2/6-311++g(d,p) level. The heats of formation were obtained by using the isodesmic reaction approach. Atomization energies were obtained by employing the G2 ab initio method. All of the optimized structures were characterized to be true local energy minima on the potential energy surface without imaginary frequencies.

Table S4 Calculated B3LYP/6-31++G(d,p)//MP2/6-311++g(d,p) zero point energy (ZPE), thermal correction (H_{corr}), and total energy (E_0) of the compounds.

Compound	ZPE/a.u.	H_{corr}/a.u.	E_0/a.u.
	0.120497	0.138315	-1245.44101
	0.121457	0.139272	-1204.593957

	0.135533	0.154569	-1300.56278
	0.11022	0.128828	-1299.582028
CH_4	0.044793	0.048605	-40.3796224
	0.074811	0.085324	-725.827622
	0.075772	0.086258	-684.9794947
	0.090462	0.102032	-780.9559035
	0.078068	0.089418	-780.4372687
	0.066072	0.073772	-520.7924862
	0.052138	0.059846	-520.2698824
CH_3CH_3	0.07461	0.079038	-79.5716305

For energetic salts, the solid-phase heat of formation is calculated on the basis of a Born-Haber energy cycle (Scheme S2). The number is simplified by equation 2:



Scheme S2. Born−Haber Cycle for the formation of energetic salts.

$\mathbf{\Delta H_f(salt,\ 298\ K) = \Delta H_f(cation,\ 298K) + \Delta H_f(anion,\ 298K) - \Delta H_L}$ (1)

in which $\triangle H_L$ can be predicted by using the formula suggested by Jenkins, etal.(equation 2):

$$\Delta H_L = U_{pot} + [p(n_M/2 - 2) + q(n_X/2 - 2)]RT \qquad (2)$$

In this equation, n_M and n_X depend on the nature of the ions M_p^+ and X_q^-, respectively.

The equation for lattice potential energy U_{pot} (equation 3) has the form:

$$U_{POT}\ [\mathrm{kJ\ mol^{-1}}] = \gamma(\rho_m/M_m)^{1/3} + \delta \qquad (3)$$

where ρ_m [g cm^{-3}] is the density of the salt, M_m is the chemical formula mass of the ionic material, and values for g and the coefficients γ ($kJmol^{-1}cm$) and δ ($kJmol^{-1}$) are assigned literature values.

7. MOLECULAR STRUCTURES

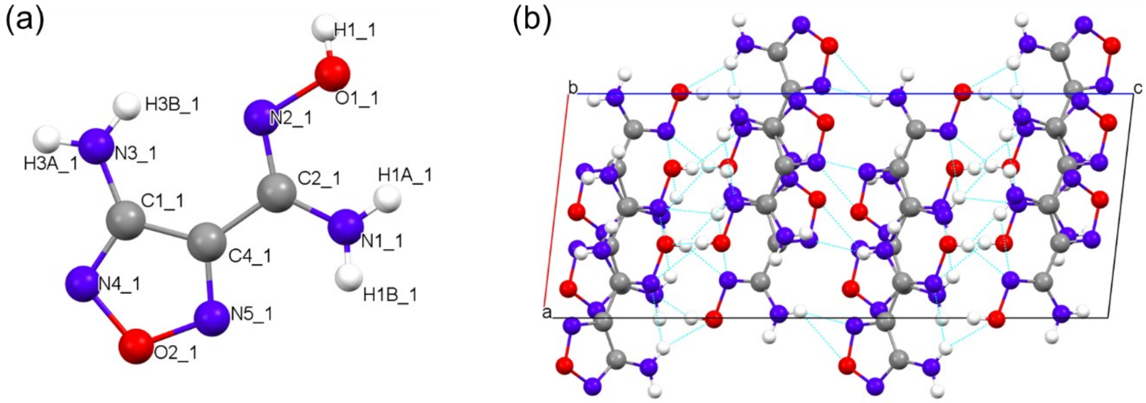


Figure S19. (a) Crystal structure of **1**; (b) Packing diagram of **1**

Table S5. Bond lengths (Å) and bond angles (°) for **1**.

Bond lengths		
C1_1 N4_1 1.302(4)	C1_2 N4_2 1.312(4)	C1_3 N3_3 1.376(4)
C1_1 N3_1 1.363(4)	C1_2 N3_2 1.354(4)	C1_3 C3_3 1.432(4)
C1_1 C4_1 1.438(4)	C1_2 C4_2 1.434(4)	N1_3 C2_3 1.339(4)
N1_1 C2_1 1.329(4)	N1_2 C2_2 1.333(4)	O1_3 N2_3 1.432(3)
C2_1 N2_1 1.306(3)	C2_2 N2_2 1.298(3)	C2_3 N2_3 1.298(4)
C2_1 C4_1 1.469(4)	C2_2 C4_2 1.479(4)	C2_3 C3_3 1.470(4)
N2_1 O1_1 1.429(3)	N2_2 O1_2 1.435(3)	O2_3 N5_3 1.385(4)
O2_1 N5_1 1.384(4)	O2_2 N5_2 1.382(4)	O2_3 N4_3 1.394(3)
O2_1 N4_1 1.398(3)	O2_2 N4_2 1.402(3)	C3_3 N5_3 1.295(4)
C4_1 N5_1 1.298(4)	C4_2 N5_2 1.297(4)	C1_3 N4_3 1.300(4)
Bond angles		
N4_1 C1_1 N3_1 123.8(3)	C4_1 N5_1 O2_1 106.2(2)	N4_3 C1_3 N3_3 122.7(3)
N4_1 C1_1 C4_1 109.0(2)	N4_2 C1_2 N3_2 124.4(3)	N4_3 C1_3 C3_3 109.2(2)
N3_1 C1_1 C4_1 127.1(3)	N4_2 C1_2 C4_2 108.5(3)	N3_3 C1_3 C3_3 128.0(3)
N2_1 C2_1 N1_1 127.8(3)	N3_2 C1_2 C4_2 127.0(3)	N2_3 C2_3 N1_3 127.2(3)
N2_1 C2_1 C4_1 113.4(2)	N2_2 C2_2 N1_2 128.2(3)	N2_3 C2_3 C3_3 114.8(3)
N1_1 C2_1 C4_1 118.9(2)	N2_2 C2_2 C4_2 112.8(2)	N1_3 C2_3 C3_3 118.1(3)

C2_1 N2_1 O1_1 109.7(2)	N1_2 C2_2 C4_2 119.0(2)	C2_3 N2_3 O1_3 108.9(2)
N5_1 O2_1 N4_1 110.3(2)	C2_2 N2_2 O1_2 109.8(2)	N5_3 O2_3 N4_3 110.4(2)
N5_1 C4_1 C1_1 108.9(3)	N5_2 O2_2 N4_2 110.7(2)	N5_3 C3_3 C1_3 109.0(3)
N5_1 C4_1 C2_1 121.0(3)	N5_2 C4_2 C1_2 109.8(3)	N5_3 C3_3 C2_3 120.5(3)
C1_1 C4_1 C2_1 130.0(2)	N5_2 C4_2 C2_2 120.9(2)	C1_3 C3_3 C2_3 130.5(2)
C1_1 N4_1 O2_1 105.5(2)	C1_2 C4_2 C2_2 129.2(3)	C1_3 N4_3 O2_3 105.4(2)
C1_2 N4_2 O2_2 105.3(2)	C4_2 N5_2 O2_2 105.7(2)	C3_3 N5_3 O2_3 106.0(2)

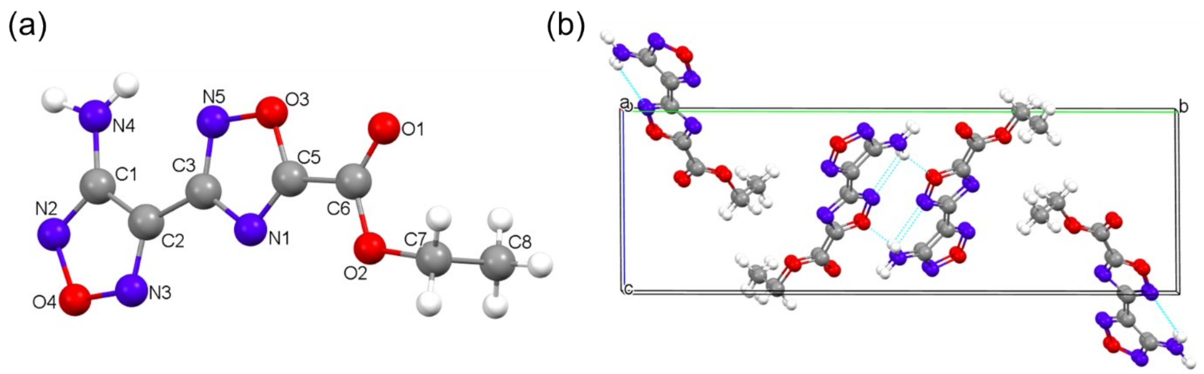


Figure S20. (a) Crystal structure of **2**; (b) Packing diagram of **2**.

Table S6. Bond lengths (Å) and bond angles (°) for **2**.

Bond lengths		
O3 N5 1.402(2)	O1 C6 1.196(3)	C7 H7B 0.9900
N4 H1N 0.8791	N1 C3 1.378(3)	C7 C8 1.454(5)
N4 H2N 0.9833	N1 C5 1.292(3)	C8 H8A 0.9800
N4 C1 1.338(3)	O2 C6 1.321(3)	C8 H8B 0.9800
C1 C2 1.438(3)	O2 C7 1.469(3)	C8 H8C 0.9800
C2 C3 1.454(3)	O4 N2 1.409(3)	N3 C2 1.303(3)
C3 N5 1.310(3)	O4 N3 1.365(3)	O3 C5 1.343(3)
C5 C6 1.498(3)	N2 C1 1.312(3)	C7 H7A 0.9900
Bond angles		
C5 N1 C3 101.1(2)	O1 C6 C5 121.7(2)	N4 C1 C2 127.7(2)
C6 O2 C7 116.5(2)	O2 C6 C5 111.0(2)	N3 C2 C1 109.9(2)

N3 O4 N2 111.16(17)	O2 C7 H7A 109.5	N3 C2 C3 121.7(2)
C1 N2 O4 105.19(18)	O2 C7 H7B 109.5	C1 C2 C3 128.4(2)
C2 N3 O4 105.7(2)	H7A C7 H7B 108.1	N1 C3 C2 125.4(2)
C5 O3 N5 105.85(17)	C8 C7 O2 110.6(2)	N5 C3 N1 115.3(2)
H1N N4 H2N 120.5	C8 C7 H7A 109.5	N5 C3 C2 119.3(2)
C1 N4 H1N 122.7	C8 C7 H7B 109.5	N1 C5 O3 114.6(2)
C1 N4 H2N 116.6	C7 C8 H8A 109.5	N1 C5 C6 130.5(2)
N2 C1 N4 124.2(2)	C7 C8 H8B 109.5	O3 C5 C6 114.8(2)
N2 C1 C2 108.1(2)	C7 C8 H8C 109.5	C3 N5 O3 103.01(18)
H8A C8 H8B 109.5	H8B C8 H8C 109.5	O1 C6 O2 127.2(2)
H8A C8 H8C 109.5		

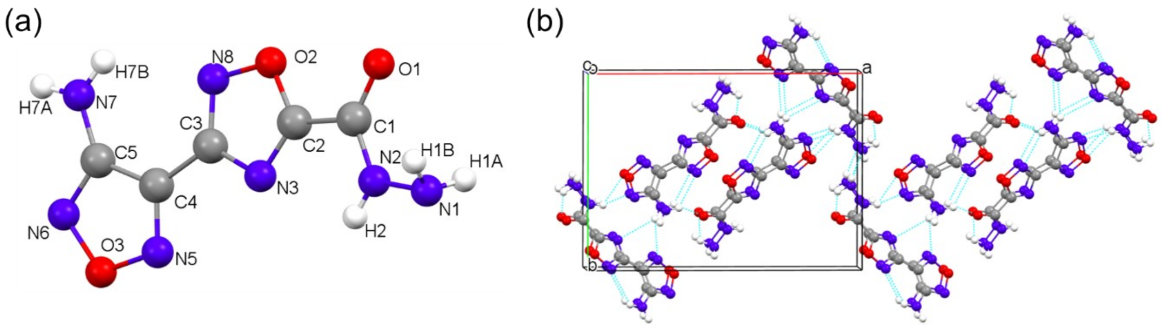


Figure S21. (a) Crystal structure of **3**; (b) Packing diagram of **3**.

Table S7. Bond lengths (Å) and bond angles (°) for **3**.

Bond lengths		
O1 C1 1.217(5)	N6 C5 1.313(5)	N1 N2 1.412(5)
O2 C2 1.327(5)	N7 C5 1.329(6)	N1 H1A 0.86(5)
O2 N8 1.408(4)	N7 H7A 0.8600	N1 H1B 0.98(5)
O3 N5 1.367(4)	N7 H7B 0.8600	N2 C1 1.316(6)
O3 N6 1.401(5)	N8 C3 1.299(6)	N2 H2 0.8600
C3 C4 1.461(6)	C1 C2 1.492(6)	N3 C2 1.290(6)
C4 C5 1.429(6)	N5 C4 1.286(6)	N3 C3 1.368(5)
Bond angles		

C5 N6 O3 104.9(4)	C2 O2 N8 106.4(3)	O2 C2 C1 117.0(4)
C5 N7 H7A 120.0	N5 O3 N6 111.1(3)	N8 C3 N3 115.7(4)
C5 N7 H7B 120.0	N2 N1 H1A 103(3)	N8 C3 C4 120.9(4)
H7A N7 H7B 120.0	N2 N1 H1B 106(3)	N3 C3 C4 123.4(4)
C3 N8 O2 102.3(4)	H1A N1 H1B 117(5)	N5 C4 C5 110.3(4)
O1 C1 N2 125.8(4)	C1 N2 N1 122.5(4)	N5 C4 C3 120.7(4)
O1 C1 C2 121.9(5)	C1 N2 H2 118.8	C5 C4 C3 129.0(4)
N2 C1 C2 112.3(4)	N1 N2 H2 118.8	N6 C5 N7 123.7(4)
N3 C2 O2 114.2(4)	C2 N3 C3 101.4(4)	N6 C5 C4 108.0(4)
N3 C2 C1 128.8(5)	C4 N5 O3 105.7(4)	N7 C5 C4 128.3(4)

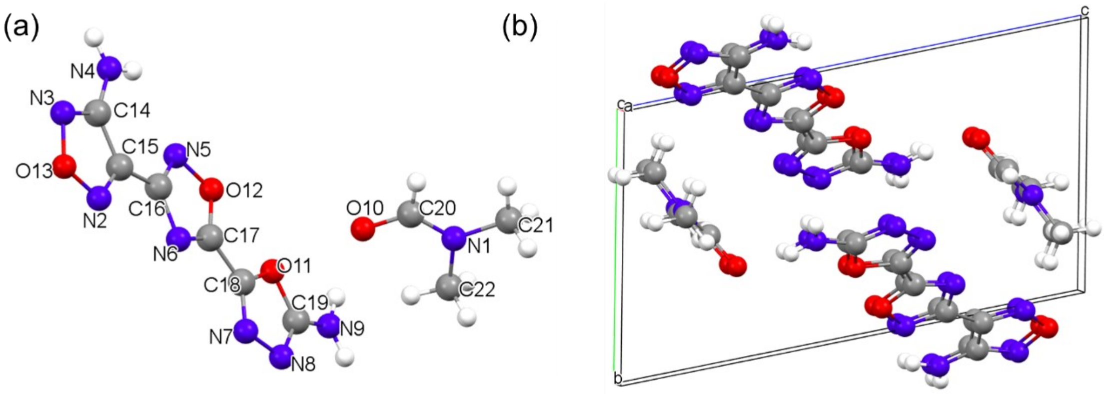


Figure S22. (a) Crystal structure of **4**; (b) packing diagram of **4**.

Table S8. Bond lengths (Å) and bond angles (°) for **4**.

Bond lengths		
N1 C20 1.323(9)	N5 O12 1.411(6)	N8 N7 1.397(7)
N1 C21 1.464(9)	N5 C16 1.300(8)	N8 C19 1.303(8)
N1 C22 1.435(9)	N6 C16 1.378(8)	N7 C18 1.274(9)
N2 O13 1.361(7)	N6 C17 1.304(8)	N9 C19 1.328(8)
N2 C15 1.302(9)	C14 C15 1.438(9)	O10 C20 1.244(8)
N3 O13 1.407(7)	C15 C16 1.454(9)	O11 C18 1.376(8)
N3 C14 1.325(8)	C17 C18 1.453(9)	O11 C19 1.364(7)
N4 C14 1.336(9)	O12 C17 1.333(7)	
Bond angles		

C20 N1 C21 121.6(6)	C19 O11 C18 101.4(5)	C19 N8 N7 106.7(5)
C20 N1 C22 121.2(6)	C17 O12 N5 105.6(5)	C18 N7 N8 105.9(5)
C22 N1 C21 117.0(6)	N2 O13 N3 111.4(5)	N2 C15 C16 122.2(6)
C15 N2 O13 106.1(5)	N3 C14 N4 123.3(6)	C14 C15 C16 128.0(6)
C14 N3 O13 104.9(5)	N3 C14 C15 107.9(6)	N5 C16 N6 116.0(6)
C16 N5 O12 103.0(5)	N4 C14 C15 128.8(6)	N5 C16 C15 120.8(6)
C17 N6 C16 100.4(5)	N2 C15 C14 109.8(6)	N6 C16 C15 123.2(6)
O12 C17 C18 118.0(5)	N8 C19 O11 112.1(5)	N6 C17 O12 115.0(5)
N7 C18 O11 113.8(5)	N9 C19 O11 118.6(5)	N6 C17 C18 127.0(6)
N7 C18 C17 129.0(6)	O10 C20 N1 125.5(6)	N8 C19 N9 129.2(6)
O11 C18 C17 117.2(6)		

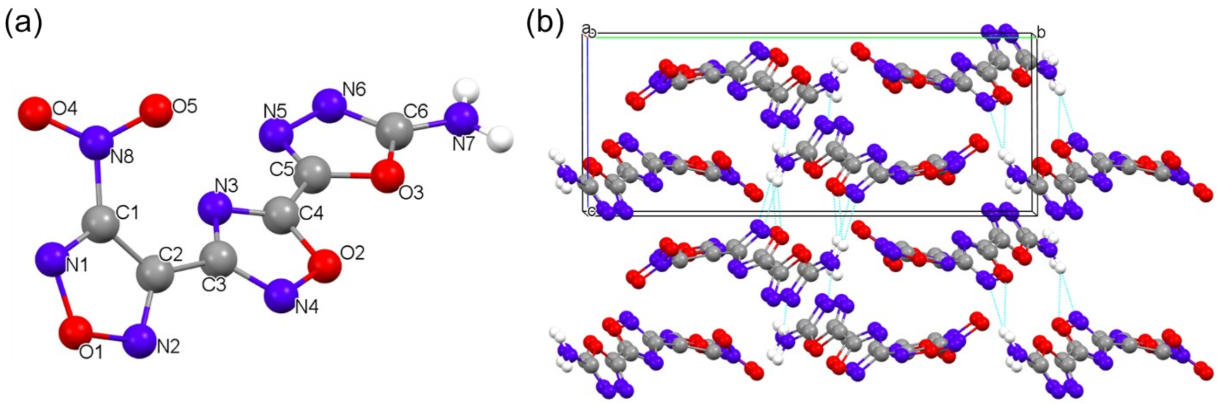


Figure S23. (a) Crystal structure of **5**; (b) Packing diagram of **5**.

Table S9. Bond lengths (Å) and bond angles (°) for **5**.

Bond lengths		
C4 C5 1.443(2)	C1 N1 1.282(3)	N4 O2 1.3967(18)
C5 N5 1.279(2)	C1 C2 1.412(3)	N5 N6 1.390(2)
C5 O3 1.362(2)	C1 N8 1.449(3)	N7 H7A 0.8600
C6 N7 1.307(2)	C2 N2 1.295(2)	N7 H7B 0.8600
C6 N6 1.311(2)	C2 C3 1.464(2)	N8 O4 1.209(2)
C6 O3 1.3576(19)	C3 N4 1.298(2)	N8 O5 1.216(2)

N1 O1 1.364(2) N2 O1 1.382(2)	C3 N3 1.364(2) C4 O2 1.3357(19)	C4 N3 1.292(2)
Bond angles		
N7 C6 N6 128.70(16) N7 C6 O3 119.23(15) N6 C6 O3 112.07(15) C1 N1 O1 104.31(17) C2 N2 O1 105.16(17) C4 N3 C3 101.19(14) C3 N4 O2 102.90(13) C5 N5 N6 106.28(14) C6 N6 N5 106.13(13) C6 N7 H7A 120.0 C6 O3 C5 102.00(12)	N1 C1 C2 111.05(18) N1 C1 N8 120.27(19) C2 C1 N8 128.58(17) N2 C2 C1 107.92(17) N2 C2 C3 121.88(18) C1 C2 C3 130.16(16) N4 C3 N3 115.77(16) N4 C3 C2 121.36(16) N3 C3 C2 122.87(16) N3 C4 O2 114.26(14) C4 O2 N4 105.88(13)	N3 C4 C5 127.57(15) O2 C4 C5 118.17(15) N5 C5 O3 113.51(14) N5 C5 C4 126.99(16) O3 C5 C4 119.50(14) C6 N7 H7B 120.0 H7A N7 H7B 120.0 O4 N8 O5 126.4(2) O4 N8 C1 118.0(2) O5 N8 C1 115.50(18) N1 O1 N2 111.56(14)

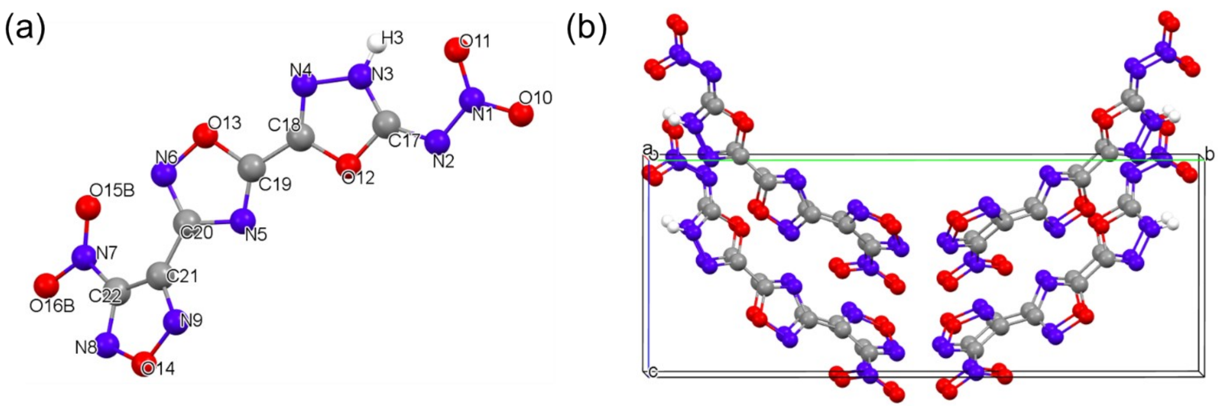


Figure S24. (a) Crystal structure of **6**; (b) Packing diagram of **6**.

Table S10. Bond lengths (Å) and bond angles (°) for **6**.

Bond lengths		
N8 C22 1.30(2) N8 O14 1.43(2) N9 C21 1.32(2) N9 O14 1.344(17) O12 C18 1.369(17)	O16A N7 1.193(18) N1 O11 1.247(14) N1 O10 1.256(14) N1 N2 1.340(15) N2 C17 1.338(18)	N7 O15A 1.224(18) N7 O16B 1.24(2) N7 C22 1.50(2) C18 C19 1.431(18) C20 C21 1.43(2)

O12 C17 1.376(14)	N3 C17 1.308(16)	C21 C22 1.43(2)
O13 C19 1.398(19)	N3 N4 1.411(16)	N5 C20 1.404(19)
N7 O15B 1.20(2)	N3 H3 0.8800	N6 C20 1.32(2)
N5 C19 1.282(16)	N4 C18 1.264(16)	N6 O13 1.403(15)
Bond angles		
O11 N1 O10 121.3(12)	O15B N7 O16A 117(3)	O15A N7 C22 113.1(12)
O11 N1 N2 122.9(12)	O15B N7 O15A 38(3)	O16B N7 C22 111.5(16)
O10 N1 N2 115.8(11)	O16A N7 O15A 132(2)	N3 C17 N2 137.8(11)
C17 N2 N1 112.8(10)	O15B N7 O16B 130(3)	N3 C17 O12 106.7(11)
C17 N3 N4 111.8(10)	O16A N7 O16B 19(4)	N2 C17 O12 115.6(10)
C17 N3 H3 124.1	O15A N7 O16B 131(3)	N4 C18 O12 115.3(12)
N4 N3 H3 124.1	O15B N7 C22 114.7(17)	N4 C18 C19 125.7(13)
C18 N4 N3 102.2(11)	O16A N7 C22 115.0(16)	C21 N9 O14 106.6(15)
C19 N5 C20 102.4(13)	O12 C18 C19 119.0(11)	N5 C19 O13 113.3(14)
C20 N6 O13 103.5(11)	C18 O12 C17 104.0(10)	N5 C19 C18 131.0(15)
C22 N8 O14 102.4(13)	C19 O13 N6 105.8(10)	O13 C19 C18 115.6(11)
N6 C20 N5 114.8(13)	N9 O14 N8 112.2(13)	N5 C20 C21 121.3(13)
N6 C20 C21 123.8(14)	N9 C21 C22 107.0(12)	N9 C21 C20 120.7(14)
N8 C22 C21 111.7(16)	C20 C21 C22 132.2(14)	N8 C22 N7 117.3(14)
C21 C22 N7 130.9(12)		

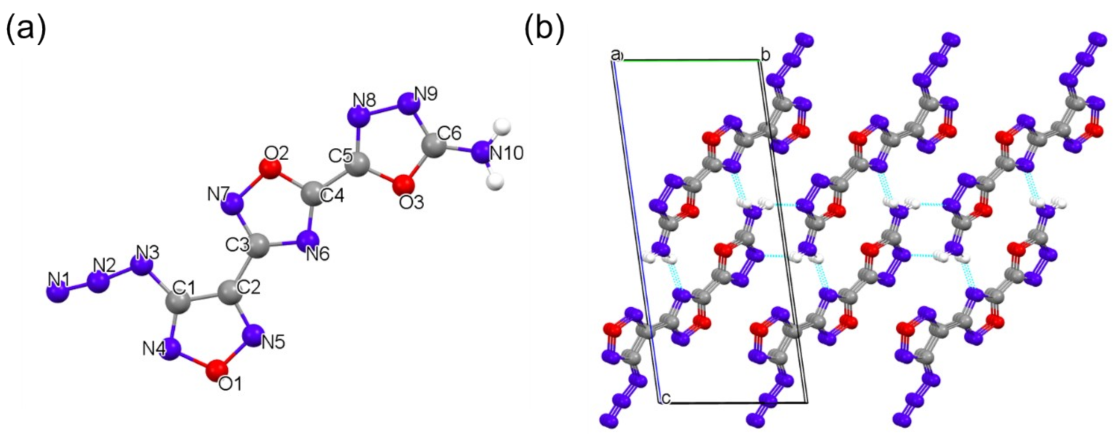


Figure S25. (a) Crystal structure of **7**; (b) Packing diagram of **7**.

Table S11. Bond lengths (Å) and bond angles (°) for **7**.

Bond lengths		
C1 N4 1.291(5)	C2 C3 1.447(5)	C4 C5 1.423(5)
C1 N3 1.390(5)	C3 N7 1.297(5)	C5 N8 1.289(5)
C1 C2 1.419(5)	C3 N6 1.370(5)	C5 O3 1.362(4)
C2 N5 1.308(5)	C4 N6 1.293(5)	C6 N9 1.304(5)
C6 O3 1.359(4)	C4 O2 1.343(4)	C6 N10 1.312(5)
N1 N2 1.114(5)	N8 N9 1.384(5)	N5 O1 1.365(4)
N2 N3 1.251(5)	N10 H10A 0.8600	N7 O2 1.407(4)
N4 O1 1.393(4)	N10 H10B 0.8600	
Bond angles		
N4 C1 N3 125.4(4)	N9 C6 N10 128.4(4)	O3 C5 C4 121.1(3)
N4 C1 C2 110.1(4)	N9 C6 O3 112.1(3)	C3 N7 O2 102.9(3)
N3 C1 C2 124.4(4)	N10 C6 O3 119.5(4)	C5 N8 N9 106.4(3)
N5 C2 C1 108.4(4)	N1 N2 N3 171.9(5)	C6 N9 N8 106.4(3)
N5 C2 C3 121.8(4)	N2 N3 C1 112.6(4)	C6 N10 H10A 120.0
C1 C2 C3 129.7(4)	C1 N4 O1 104.5(3)	C6 N10 H10B 120.0
N7 C3 N6 115.5(4)	C2 N5 O1 105.6(3)	H10A N10 H10B 120.0
N7 C3 C2 119.7(4)	C4 N6 C3 101.9(3)	N5 O1 N4 111.3(3)
N6 C3 C2 124.8(4)	N8 C5 C4 126.1(4)	C4 O2 N7 106.1(3)
N6 C4 O2 113.6(3)	C6 O3 C5 102.2(3)	N8 C5 O3 112.8(3)
N6 C4 C5 130.8(4)	O2 C4 C5 115.6(3)	

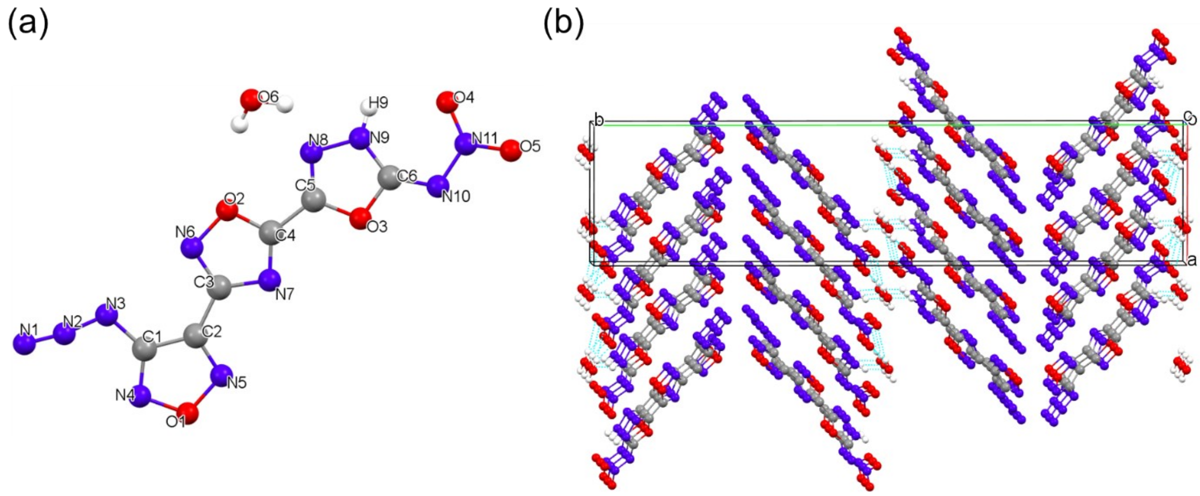


Figure S26. (a) Crystal structure of **8**; (b) Packing diagram of **8**.

Table S12. Bond lengths (Å) and bond angles (°) for **8**.

Bond lengths		
O1 N4 1.387(2)	N7 C3 1.380(2)	O4 N11 1.2330(19)
O1 N5 1.3727(18)	N7 C4 1.291(2)	O5 N11 1.2354(19)
O2 N6 1.4005(19)	N8 N9 1.371(2)	N1 N2 1.121(2)
O2 C4 1.340(2)	N8 C5 1.281(2)	N2 N3 1.259(2)
O3 C5 1.364(2)	N9 H9 0.8616	N3 C1 1.392(2)
O3 C6 1.3604(19)	N9 C6 1.329(2)	N4 C1 1.298(2)
C4 C5 1.450(2)	N10 N11 1.365(2)	N5 C2 1.304(2)
O6 H6 0.9179	N10 C6 1.315(2)	N6 C3 1.306(2)
C2 C3 1.458(2)	C1 C2 1.429(2)	
Bond angles		
N5 O1 N4 111.34(12)	C4 O2 N6 105.57(13)	N8 C5 O3 114.44(15)
C6 N10 N11 113.22(14)	C6 O3 C5 103.97(12)	N8 C5 C4 125.30(16)
O4 N11 O5 122.13(14)	N1 N2 N3 170.24(17)	N9 C6 O3 107.04(14)
O4 N11 N10 122.71(14)	N2 N3 C1 115.00(14)	N10 C6 O3 115.64(14)
O5 N11 N10 115.17(14)	C1 N4 O1 105.00(14)	N10 C6 N9 137.31(16)
N3 C1 C2 123.77(15)	C2 N5 O1 105.51(14)	N6 C3 C2 120.32(15)
N4 C1 N3 126.83(16)	C3 N6 O2 103.18(14)	N7 C3 C2 124.30(15)

N4 C1 C2 109.39(15)	C4 N7 C3 100.74(14)	O2 C4 C5 114.68(14)
N5 C2 C1 108.76(15)	C5 N8 N9 103.29(14)	N7 C4 O2 115.13(15)
N5 C2 C3 122.12(15)	N8 N9 H9 121.6	N7 C4 C5 130.12(16)
C1 C2 C3 129.08(16)	C6 N9 N8 111.25(13)	O3 C5 C4 120.23(14)
N6 C3 N7 115.37(15)	C6 N9 H9 127.1	

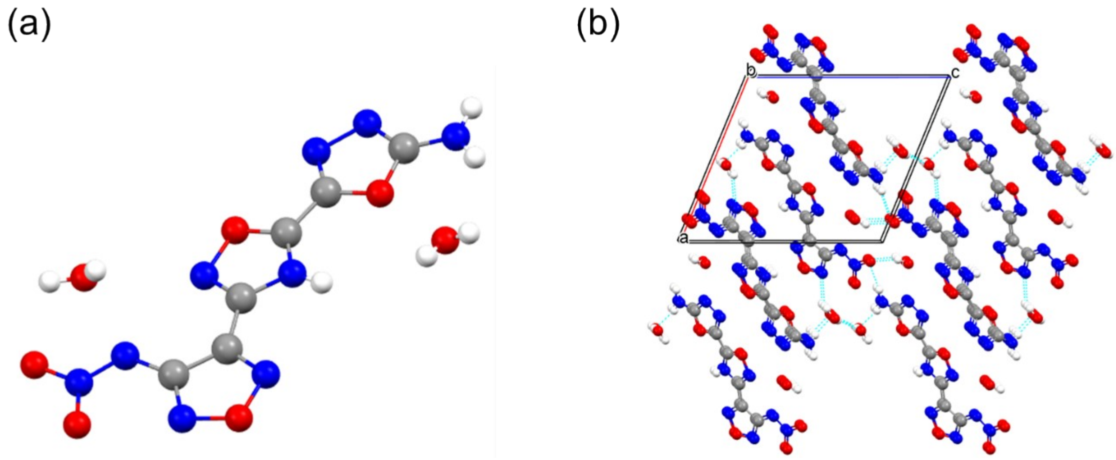


Figure S27. (a) Crystal structure of **9**; (b) Packing diagram of **9**.

Table S13. Bond lengths (Å) and bond angles (°) for **9**.

Bond lengths		
C1 N3 1.330(11)	C3 N6 1.304(12)	C6 N8 1.306(11)
C1 N2 1.369(10)	C3 N5 1.355(10)	C6 O5 1.360(9)
C1 C2 1.426(12)	C4 N5 1.281(10)	N1 O2 1.255(9)
C2 N4 1.272(11)	C4 O4 1.337(10)	N1 O1 1.271(9)
C2 C3 1.482(11)	C4 C5 1.429(11)	N1 N2 1.306(9)
C6 N9 1.290(11)	C5 N7 1.282(11)	N3 O3 1.382(9)
N4 O3 1.377(9)	C5 O5 1.370(11)	N7 N8 1.386(9)
N6 O4 1.429(9)		
Bond angles		
N3 C1 N2 130.1(8)	N5 C3 C2 123.8(8)	N8 C6 O5 110.0(8)
N3 C1 C2 106.1(8)	N5 C4 O4 113.6(7)	O2 N1 O1 119.8(8)
N2 C1 C2 123.8(8)	N5 C4 C5 130.0(8)	O2 N1 N2 123.8(9)

N4 C2 C1 112.4(8)	O4 C4 C5 116.3(8)	O1 N1 N2 116.4(8)
N4 C2 C3 119.3(8)	N7 C5 O5 114.0(8)	N1 N2 C1 117.0(7)
C1 C2 C3 128.3(8)	N7 C5 C4 126.2(9)	C1 N3 O3 105.5(7)
N6 C3 N5 115.3(8)	O5 C5 C4 119.7(8)	C2 N4 O3 104.5(7)
N6 C3 C2 120.8(8)	N9 C6 N8 131.2(8)	C4 N5 C3 103.2(7)
C4 O4 N6 105.7(6)	N9 C6 O5 118.8(9)	C3 N6 O4 102.2(7)
C6 O5 C5 102.5(7)	N4 O3 N3 111.5(7)	C5 N7 N8 104.3(8)
C6 N8 N7 109.1(7)		

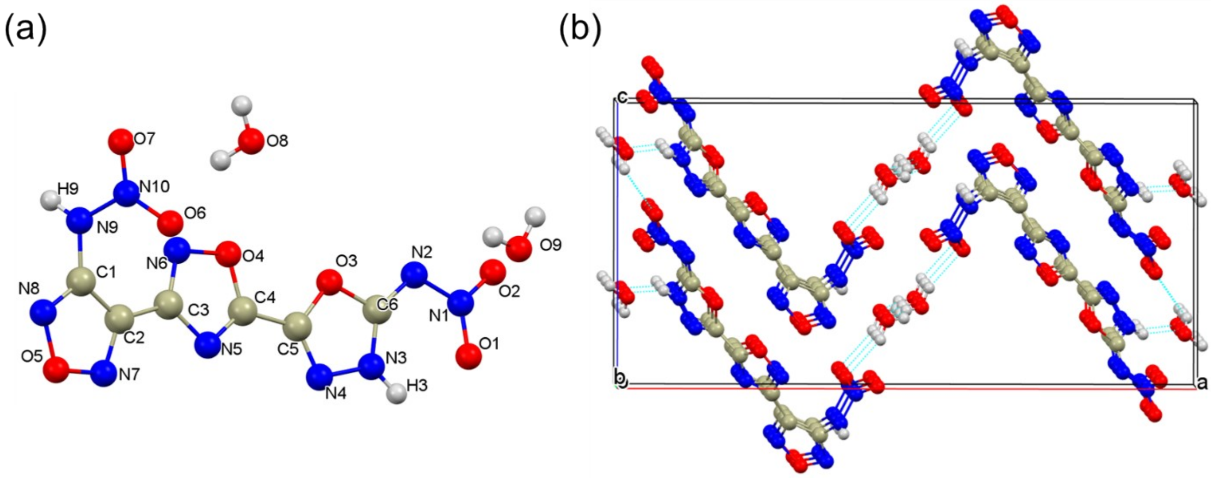


Figure S28. (a) Crystal structure of **10**; (b) Packing diagram of **10**.

Table S14. Bond lengths (Å) and bond angles (°) for **10**.

Bond lengths		
O1 N1 1.243(4)	O2 N1 1.224(4)	N1 N2 1.369(4)
N7 C2 1.310(5)	O3 C5 1.372(4)	N2 C6 1.314(5)
N8 C1 1.295(5)	O3 C6 1.358(4)	N3 H3 0.8800
N9 H9 0.8800	O4 N6 1.408(4)	N3 N4 1.394(4)
N9 N10 1.390(5)	O4 C4 1.343(4)	N3 C6 1.332(4)
N9 C1 1.389(5)	O5 N7 1.381(4)	N4 C5 1.265(4)
C1 C2 1.434(5)	O5 N8 1.373(4)	N5 C3 1.370(4)
C2 C3 1.457(5)	O6 N10 1.234(5)	N5 C4 1.300(5)
C4 C5 1.461(5)	O7 N10 1.208(5)	N6 C3 1.305(5)

O8 H8A 0.8601 O8 H8B 0.8594	O9 H9A 0.8700	O9 H9B 0.8698
Bond angles		
C6 O3 C5 104.4(3)	O1 N1 N2 121.0(3)	C1 C2 C3 130.1(3)
C4 O4 N6 105.3(3)	O2 N1 O1 123.3(3)	N5 C3 C2 123.8(3)
N8 O5 N7 111.3(3)	O2 N1 N2 115.7(3)	N6 C3 N5 116.4(3)
C1 N9 H9 122.0	C6 N2 N1 114.6(3)	N6 C3 C2 119.8(3)
C1 N9 N10 116.0(3)	N4 N3 H3 124.5	O4 C4 C5 116.5(3)
O6 N10 N9 118.0(3)	C6 N3 H3 124.5	N5 C4 O4 115.1(3)
O7 N10 O6 126.0(4)	C6 N3 N4 111.0(3)	N5 C4 C5 128.3(3)
O7 N10 N9 116.0(4)	C5 N4 N3 103.1(3)	O3 C5 C4 118.1(3)
N8 C1 N9 121.2(3)	C4 N5 C3 100.4(3)	N4 C5 O3 114.8(3)
N8 C1 C2 109.9(3)	C3 N6 O4 102.8(3)	N4 C5 C4 127.1(3)
N9 C1 C2 128.8(3)	C2 N7 O5 105.8(3)	N2 C6 O3 114.7(3)
N7 C2 C1 107.6(3)	C1 N8 O5 105.3(3)	N2 C6 N3 138.6(3)
N7 C2 C3 122.3(3)	N10 N9 H9 122.0	H9A O9 H9B 104.5
N3 C6 O3 106.7(3)	H8A O8 H8B 109.4	

8. HYDROGEN-BONDING INTERACTIONS

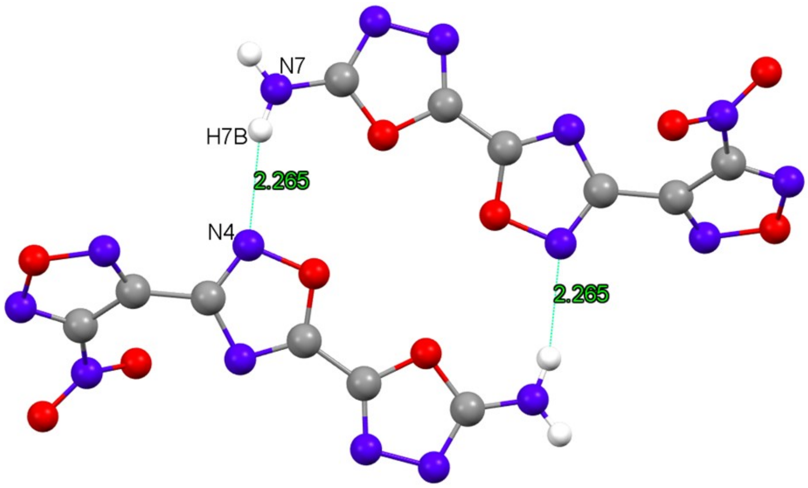


Figure S29. Hydrogen bonds for **5**.

Table S15. Hydrogen bonds for **5**

D-H···A	D-H/Å	H···A/Å	D···A/Å	D-H···A/°
N7-H7B···N4’	0.860	2.265	3.102	164.25

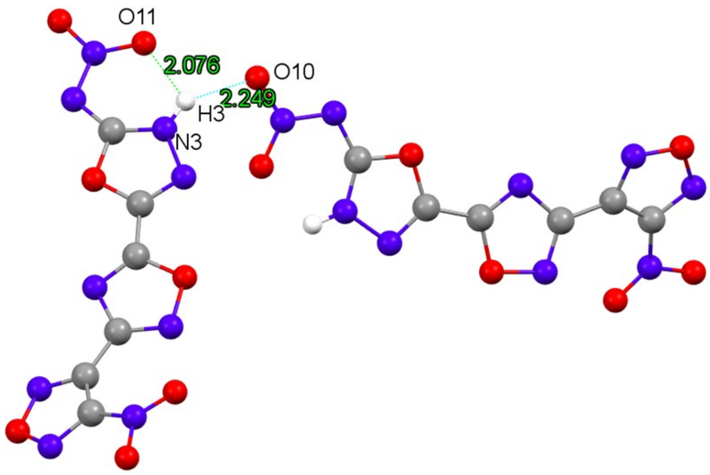


Figure S30. Hydrogen bonds for **6**.

Table S16. Hydrogen bonds for **6**.

D-H···A	D-H/Å	H···A/Å	D···A/Å	D-H···A/°
N3-H3···O10	0.880	2.249	2.996	142.43
N3-H3···O11	0.880	2.076	2.541	112.06

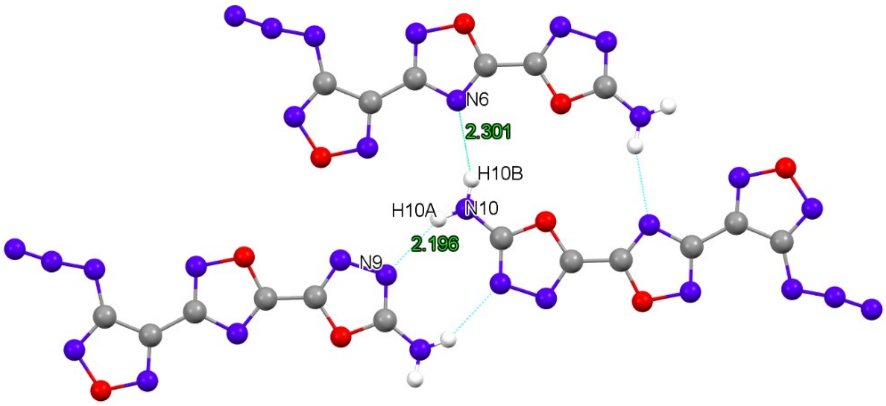


Figure S31. Hydrogen bonds for **7**.

Table S17. Hydrogen bonds for **7**.

D-H···A	D-H/Å	H···A/Å	D···A/Å	D-H···A/°
N10-H10A···N9	0.860	2.196	3.123	152.38
N10-H10B···N6	0.860	2.301	2.984	159.85

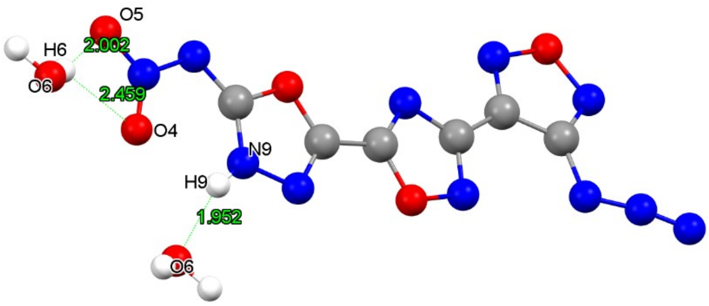


Figure S32. Hydrogen bonds for **8.H$_2$O**.

Table S18. Hydrogen bonds for **8.H$_2$O**.

D-H···A	D-H/Å	H···A/Å	D···A/Å	D-H···A/°
O6-H6···O5	0.918	2.002	2.883	160.35
O6-H6···O4	0.918	2.459	3.152	132.48
N9-H9···O6’	0.861	1.952	2.771	158.40

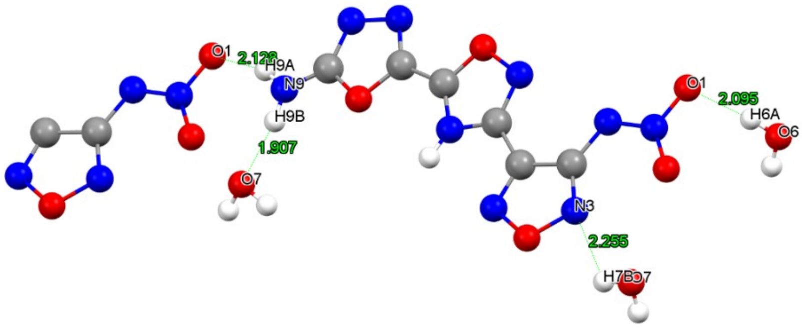


Figure S33. Hydrogen bonds for **9**.2H_2O.

Table S19. Hydrogen bonds for **9**.2H_2O.

D-H···A	D-H/Å	H···A/Å	D···A/Å	D-H···A/°
O6-H6A···O1	0.836	2.095	2.890	158.84
O7-H7B···N3	0.828	2.255	2.882	126.99
N9-H9B···O7	0.860	1.907	2.750	166.24
N9-H9B···O1	0.861	2.128	2.940	157.22

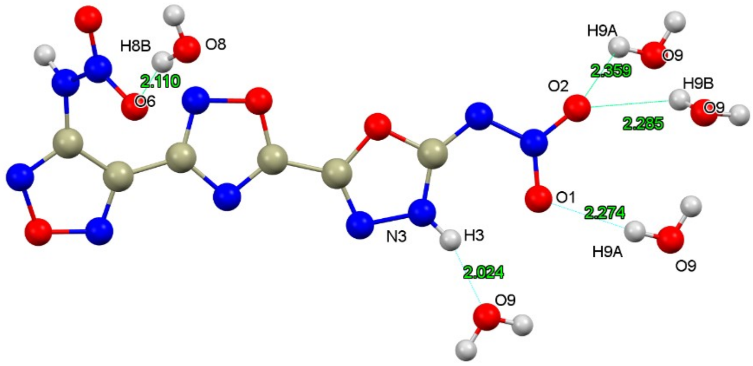


Figure S34. Hydrogen bonds for **10.**2H_2O.

Table S20. Hydrogen bonds for **10.**2H_2O.

D-H···A	D-H/Å	H···A/Å	D···A/Å	D-H···A/°
O8-H8B···O6	0.859	2.110	2.923	157.61
N3-H3···O9	0.870	2.359	2.902	120.77
O9-H9A···O2	0.870	2.285	2.885	120.58
O9-H9B···O2	0.870	2.274	3.043	147.46
O9-H9A···O1	0.880	2.024	2.851	156.25

9. Reasons for enhanced performance of 10

In this study, compound **10** exhibits much enhanced thermal stability than **10a** and **10c** (no data valuable for **10b**) and slightly higher density than **10c** (no data valuable for **10a** and **10c**). As can be seen in Figure S35, this is mainly due to the hydrogen transfer of the nitroamino group linked to 1,3,4-oxadiazole in compound 10, which changed "-NH-NO_2" to "-N=NO_2", generating intramolecular H-bond to increase thermal stability. At the same time, hydrogen transfer also puts 1,3,4-oxadiazole in a plane with the nitroamino group, which in turn makes compound **10** somewhat more planar than compounds **10a**, **10b**, and **10c**, which is more favorable for molecular stacking, and thus for density. In recent years, many studies have shown that the intramolecular H-bond and enhanced planarity contributes to intramolecular conjugation as well as intermolecular stacking, which in turn improves stability and energy. Thus, compound **10** can have better thermal stability and density.

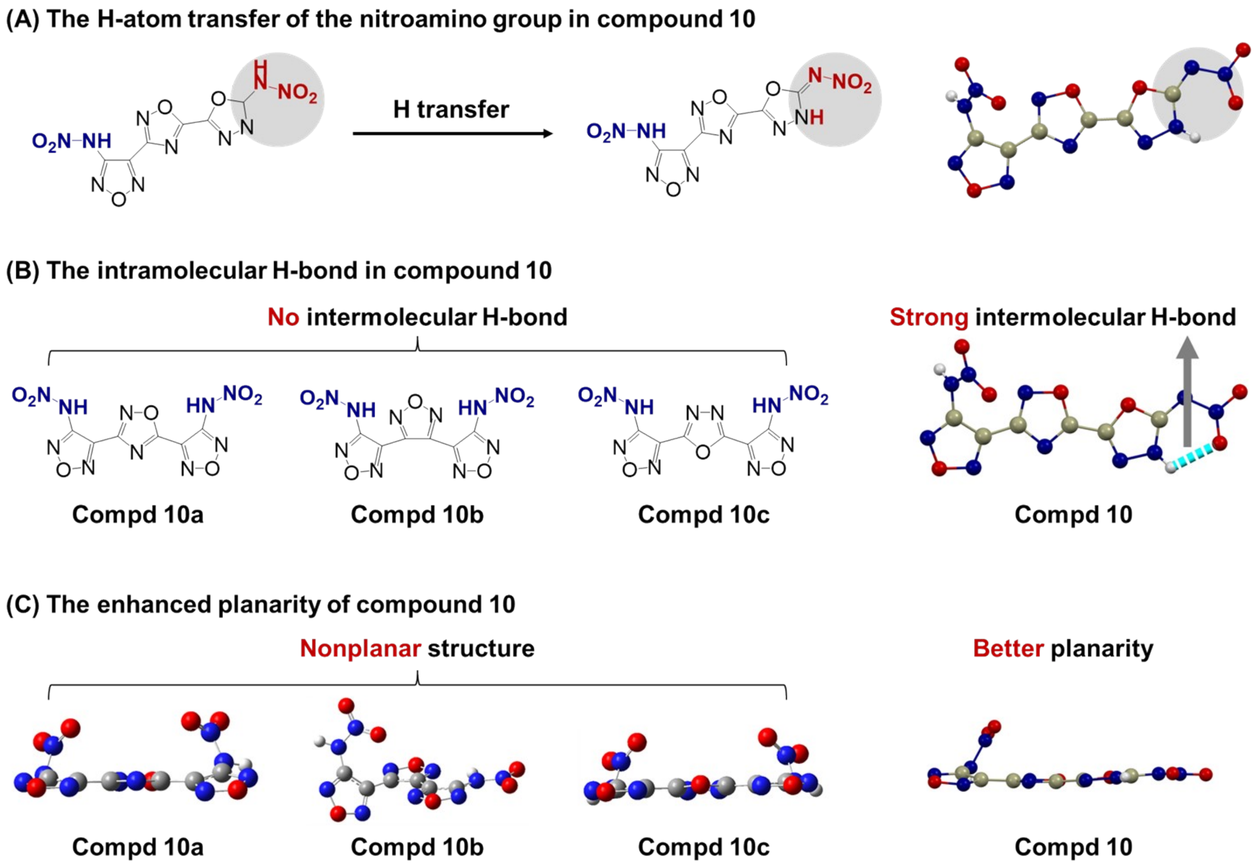


Figure S35. The reasons for compound **10** with enhanced performance.