Electronic Supplementary Information

Energetic salts of 4-nitramino-3-(5-dinitromethyl-1,2,4-oxadiazolyl)-furazan: Powerful alliance towards good thermal stability and high performances

Chunlin He, [a,b] Gregory H. Imler,[c] Damon A. Parrish,[c] and Jean'ne M. Shreeve*[b]

[a] School of Materials Science & Engineering, Beijing Institute of Technology, Beijing 100081, China.

[b] Department of Chemistry, University of Idaho, Moscow, Idaho 83844-2343, United States.

Email: jshreeve@uidaho.edu

[c] Naval Research Laboratory, 4555 Overlook Avenue, Washington, DC 20375, United States

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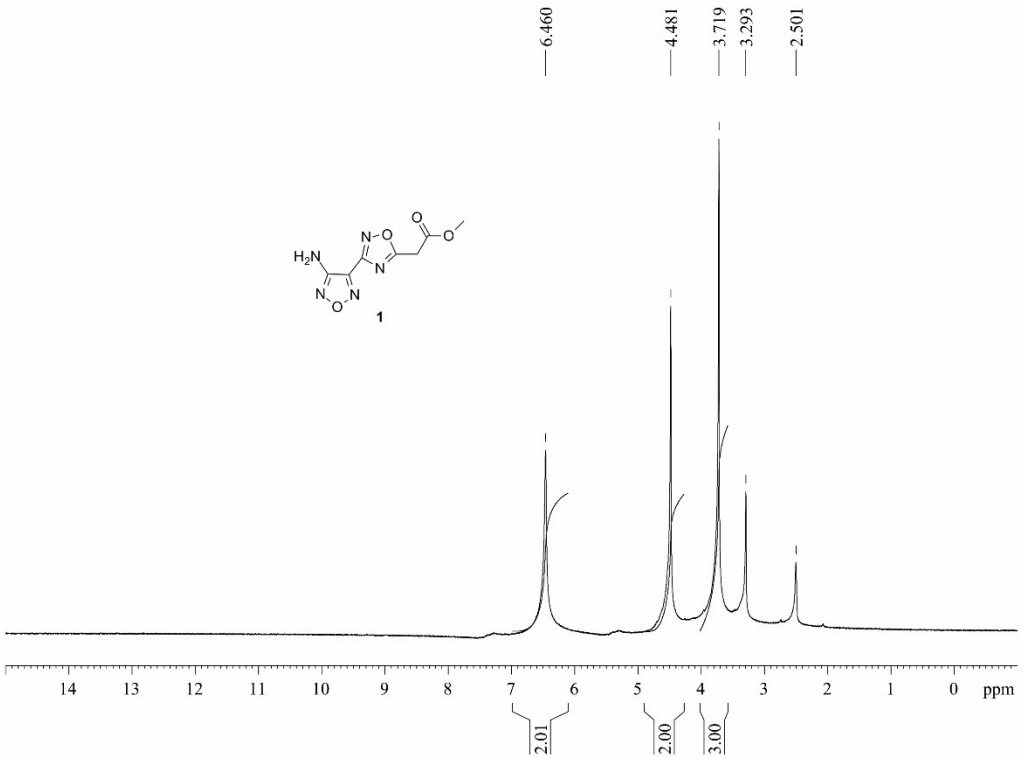


Figure S1 ^{1}H NMR spectra of **1**.

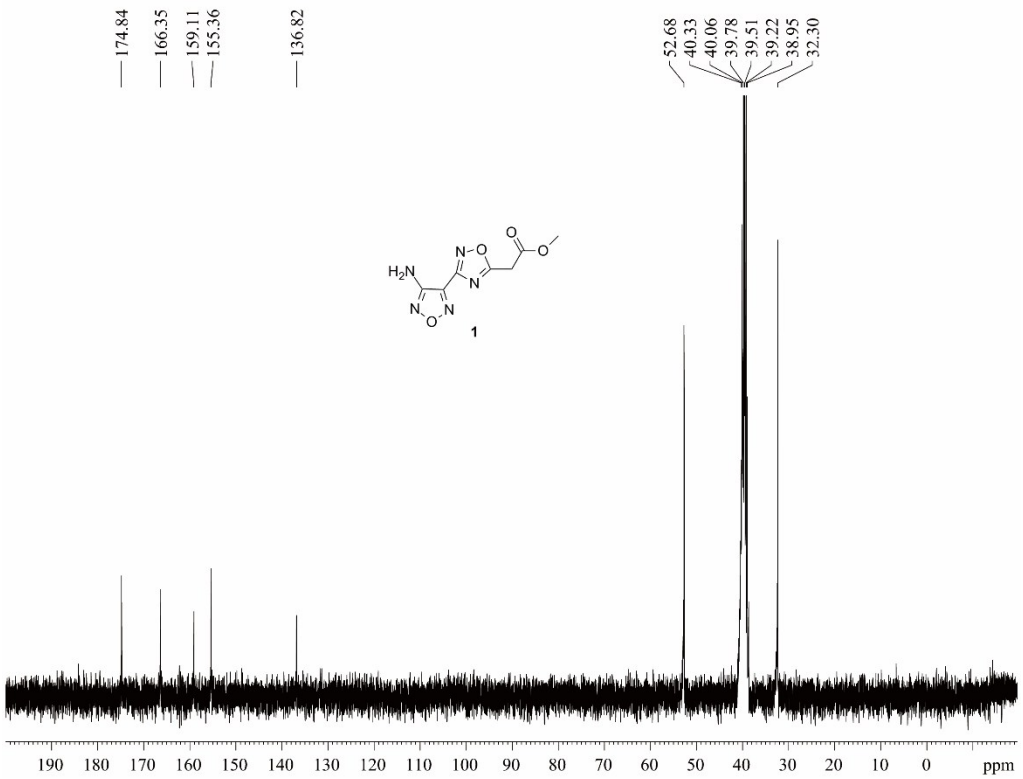


Figure S2 ^{13}C NMR spectra of **1**.

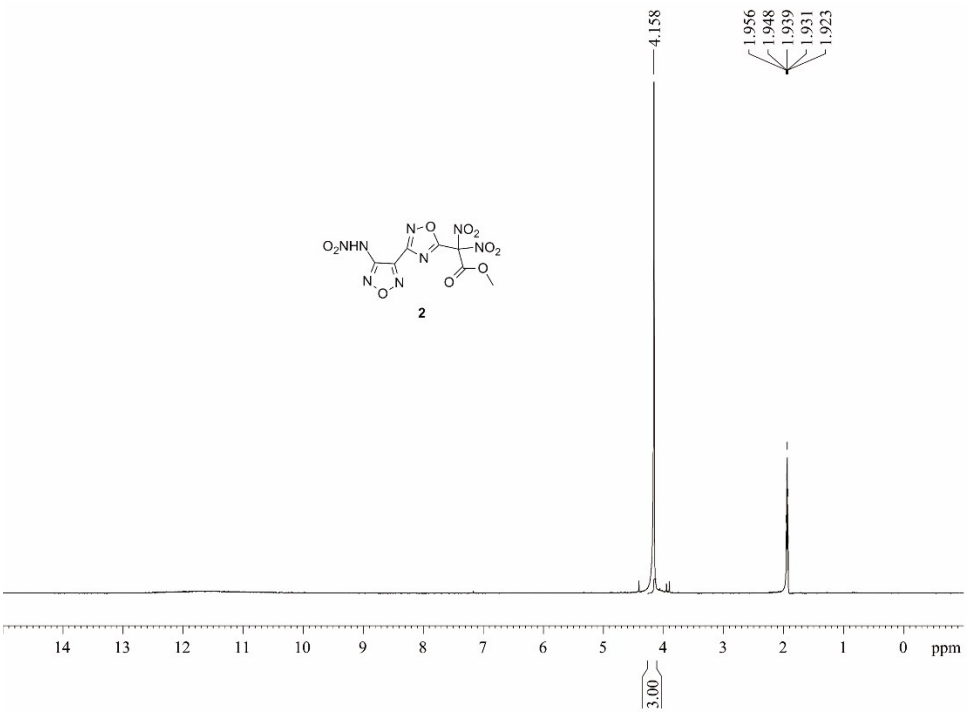


Figure S3 ^{1}H NMR spectra of **2**.

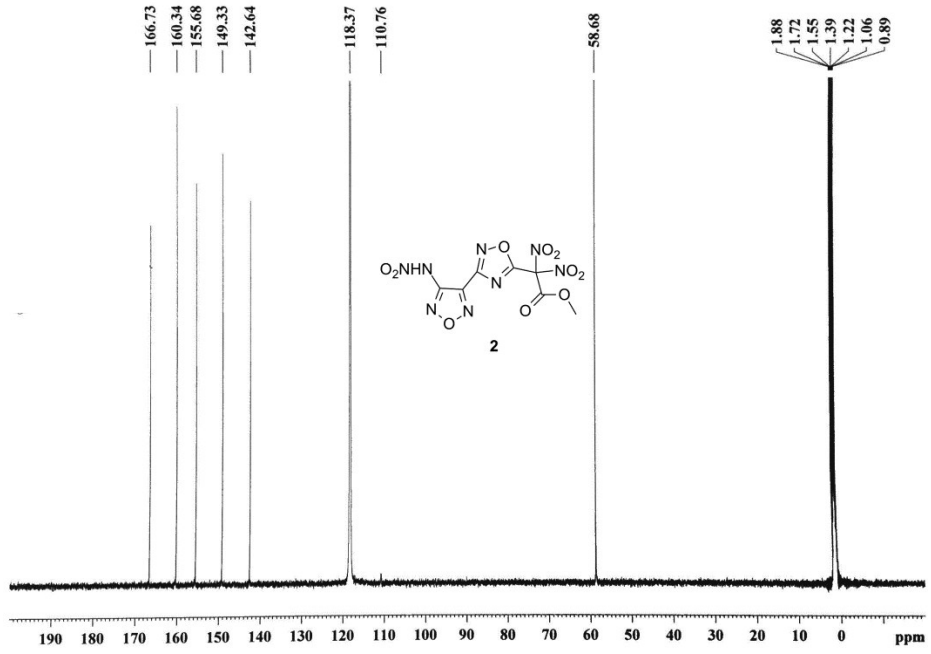


Figure S4 ^{13}C NMR spectra of **2.**

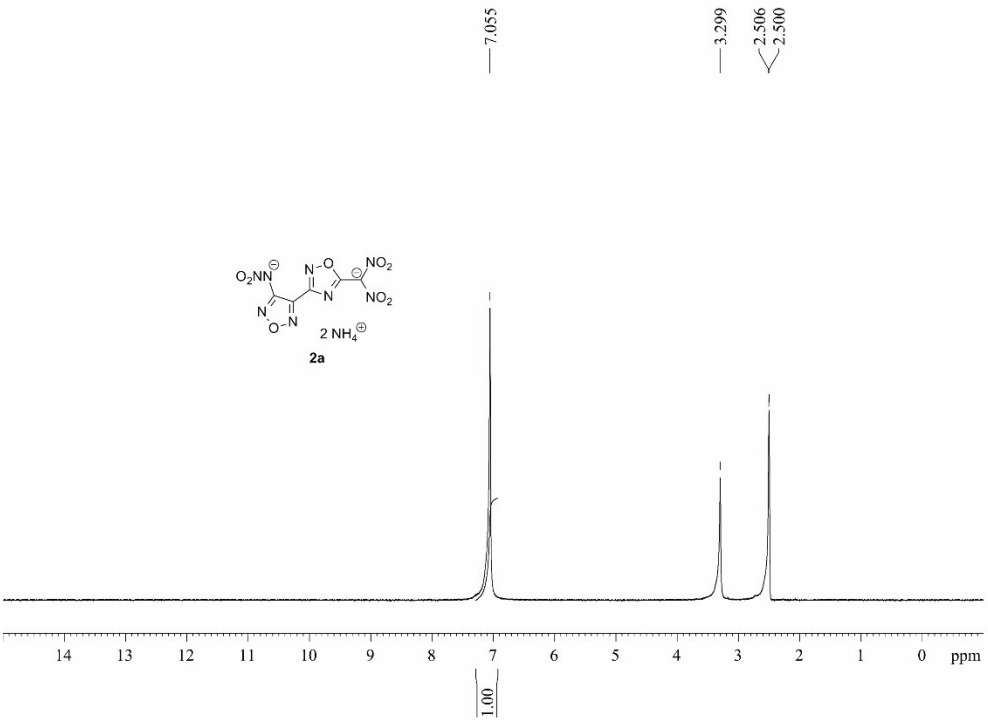


Figure S5 ^{1}H NMR spectra of **2a**.

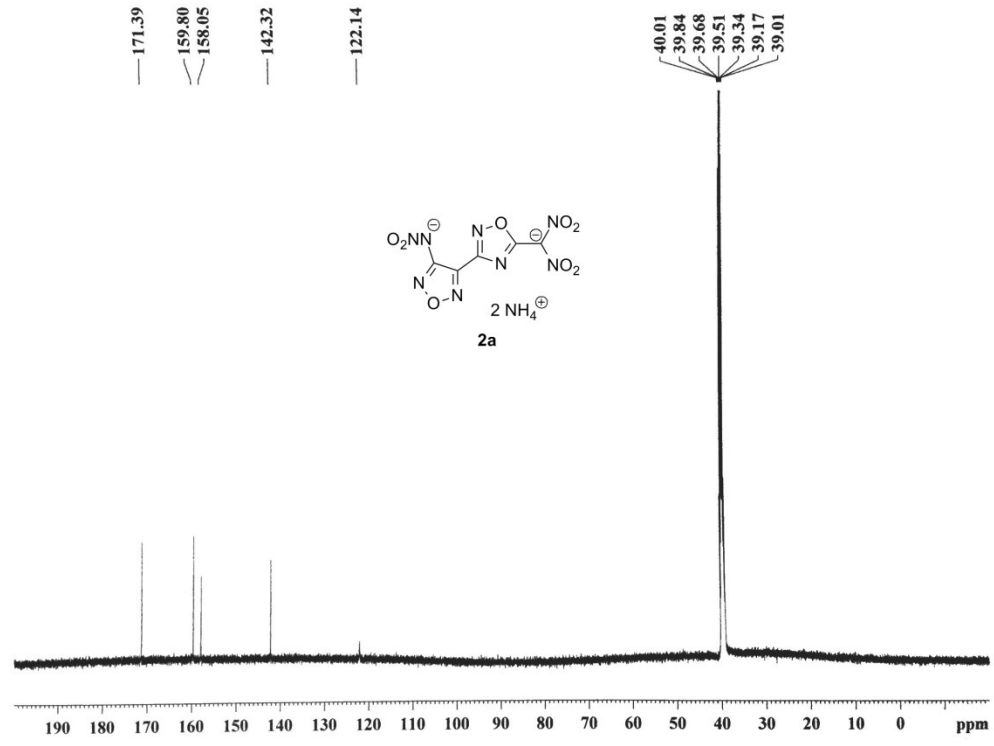


Figure S6 ^{13}C NMR spectra of **2a.**

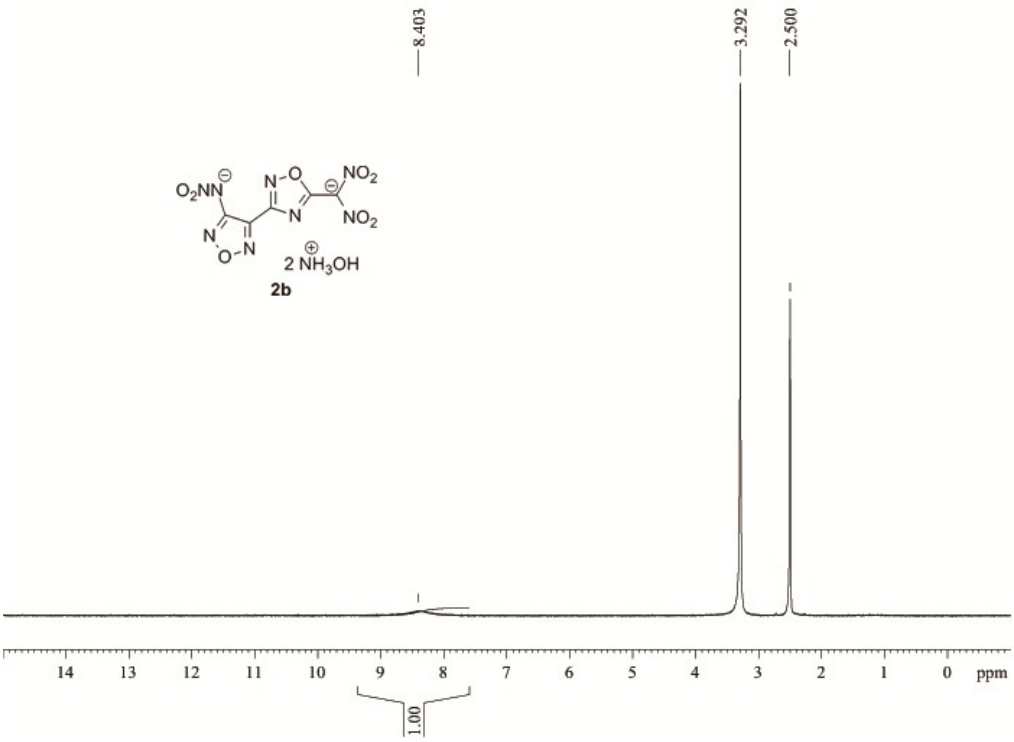


Figure S7 ^{1}H NMR spectra of **2b**.

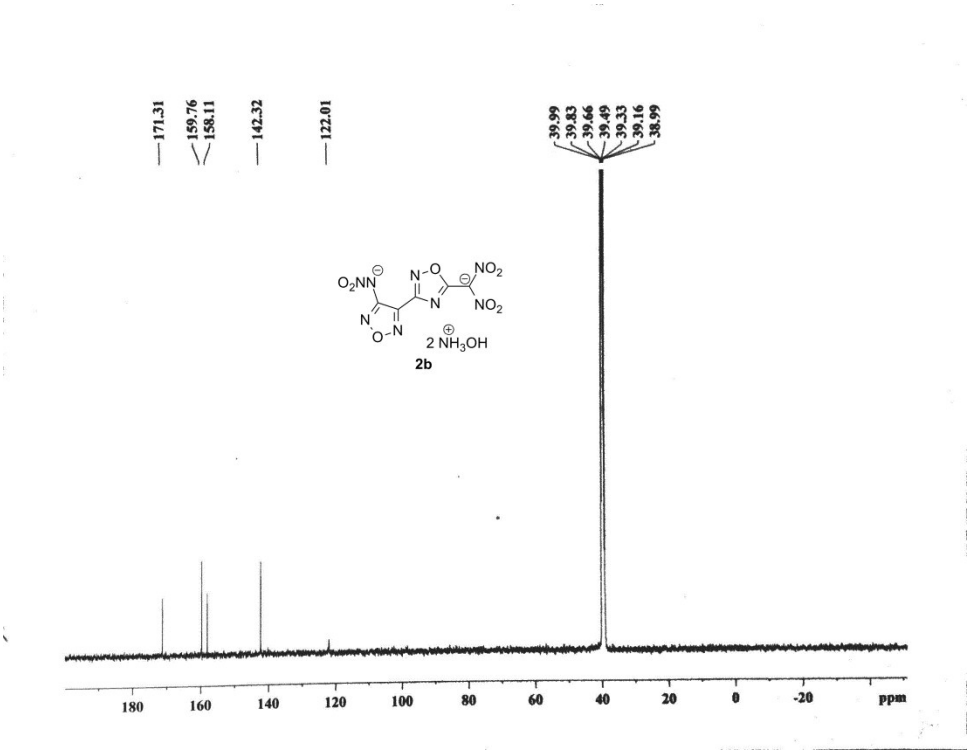


Figure S8 ^{13}C NMR spectra of **2b.**

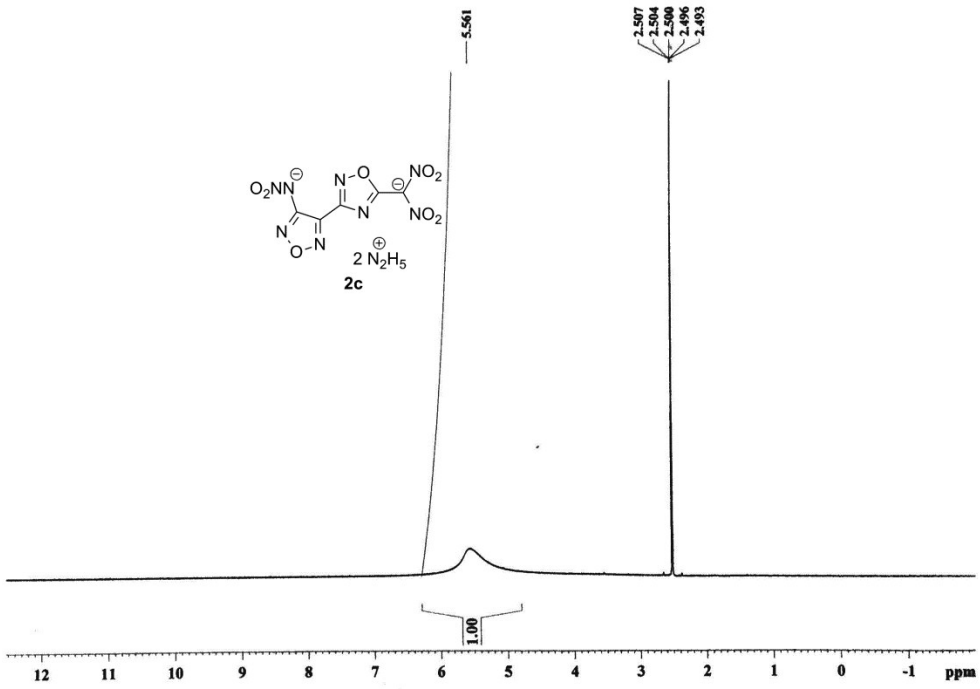


Figure S9 ^{1}H NMR spectra of **2c**.

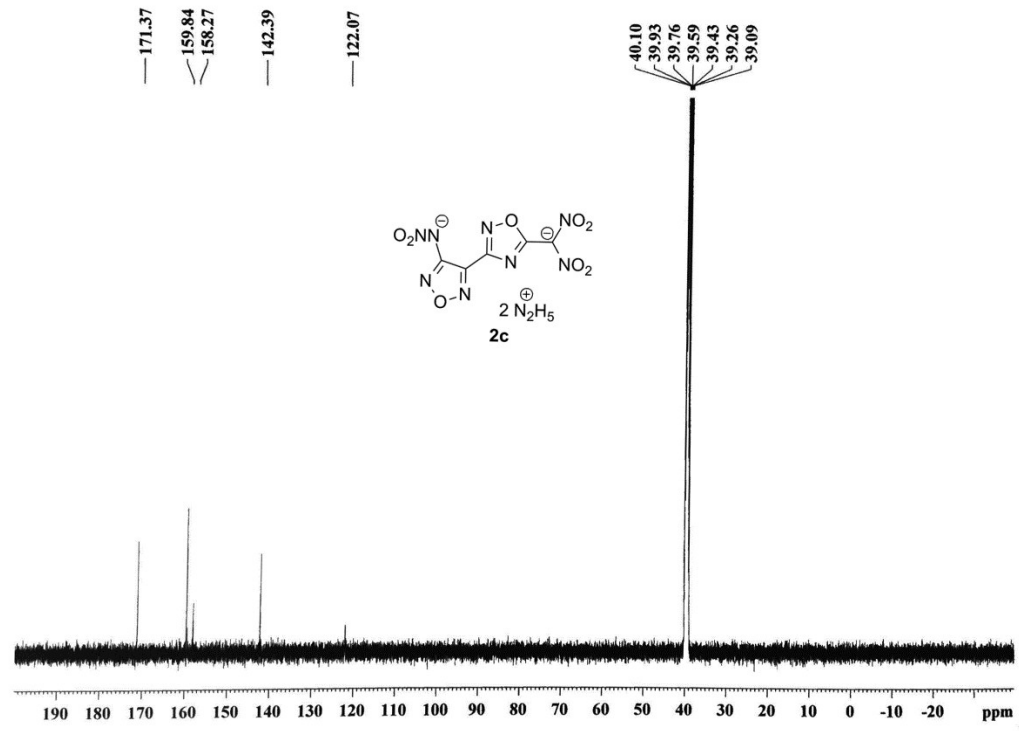


Figure S10 ^{13}C NMR spectra of **2c.**

Table S1. Crystal data and structure refinement for **2a**.

Empirical formula	$C_5H_8N_{10}O_8$	
Formula weight	336.21	
Temperature	150(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 18.9552(14) Å	α= 90°.
	b = 11.1084(8) Å	β= 126.1840(10)°.
	c = 14.2841(18) Å	γ = 90°.
Volume	2427.6(4) $Å^3$	
Z	8	
Density (-123°C)	1.840 Mg/m^3	
Density (20°C)	1.795 Mg/m^3	
Absorption coefficient	0.171 mm^{-1}	
F(000)	1376	
Crystal size	0.219 x 0.219 x 0.070 mm^3	
Theta range for data collection	2.266 to 29.970°.	
Index ranges	-26<=h<=25, -15<=k<=15, -17<=l<=20	
Reflections collected	16212	
Independent reflections	3432 [R_{int} = 0.0223]	
Completeness to theta = 25.242°	100.0 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7460 and 0.6997	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	3432 / 8 / 232	
Goodness-of-fit on F^2	1.028	
Final R indices [I>2sigma(I)]	R_1 = 0.0331, wR_2 = 0.0857	
R indices (all data)	R_1 = 0.0421, wR_2 = 0.0911	
Largest diff. peak and hole	0.418 and -0.275 e.$Å^{-3}$	

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

	x	y	z	U(eq)
O(1)	6132(1)	4071(1)	3130(1)	26(1)
O(2)	5542(1)	4702(1)	1368(1)	22(1)
N(3)	5885(1)	4926(1)	2411(1)	17(1)
N(4)	6021(1)	6011(1)	2854(1)	18(1)
C(5)	5769(1)	6959(1)	2099(1)	16(1)
N(6)	5399(1)	6997(1)	975(1)	22(1)
O(7)	5287(1)	8231(1)	703(1)	24(1)
N(8)	5592(1)	8931(1)	1667(1)	20(1)
C(9)	5890(1)	8176(1)	2521(1)	17(1)
C(10)	6254(1)	8609(1)	3690(1)	16(1)
N(11)	6452(1)	7885(1)	4535(1)	21(1)
O(12)	6720(1)	8686(1)	5460(1)	20(1)
C(13)	6654(1)	9806(1)	5048(1)	16(1)
N(14)	6373(1)	9811(1)	3954(1)	18(1)
C(15)	6891(1)	10833(1)	5779(1)	17(1)
N(16)	7307(1)	10768(1)	6962(1)	16(1)
O(17)	7431(1)	9736(1)	7402(1)	22(1)
O(18)	7522(1)	11693(1)	7571(1)	22(1)
N(19)	6694(1)	11997(1)	5253(1)	18(1)
O(20)	7209(1)	12836(1)	5764(1)	26(1)
O(21)	5993(1)	12103(1)	4276(1)	24(1)
N(22)	6329(1)	5236(1)	5097(1)	20(1)
N(23)	5994(1)	1781(1)	2159(1)	21(1)

Table S3. Bond lengths [Å] and angles [°] for **2a**.

O(1)-N(3)	1.2686(12)	O(2)-N(3)	1.2500(12)
N(3)-N(4)	1.3135(13)	N(4)-C(5)	1.3750(14)
C(5)-N(6)	1.3200(14)	C(5)-C(9)	1.4419(15)
N(6)-O(7)	1.4057(13)	O(7)-N(8)	1.3764(12)
N(8)-C(9)	1.3034(14)	C(9)-C(10)	1.4608(14)
C(10)-N(11)	1.3076(14)	C(10)-N(14)	1.3699(14)
N(11)-O(12)	1.4141(12)	O(12)-C(13)	1.3500(13)
C(13)-N(14)	1.3198(14)	C(13)-C(15)	1.4288(14)
C(15)-N(16)	1.3812(13)	C(15)-N(19)	1.4296(13)
N(16)-O(18)	1.2488(12)	N(16)-O(17)	1.2601(12)
N(19)-O(20)	1.2302(12)	N(19)-O(21)	1.2398(12)
N(22)-H(22A)	0.898(9)	N(22)-H(22B)	0.900(9)
N(22)-H(22C)	0.890(9)	N(22)-H(22D)	0.902(9)
N(23)-H(23A)	0.903(9)	N(23)-H(23B)	0.905(9)
N(23)-H(23C)	0.895(9)	N(23)-H(23D)	0.898(9)
O(2)-N(3)-O(1)	119.94(9)	O(2)-N(3)-N(4)	124.98(10)
O(1)-N(3)-N(4)	115.08(9)	N(3)-N(4)-C(5)	116.53(9)
N(6)-C(5)-N(4)	131.86(10)	N(6)-C(5)-C(9)	108.51(9)
N(4)-C(5)-C(9)	119.62(9)	C(5)-N(6)-O(7)	104.63(9)
N(8)-O(7)-N(6)	111.66(8)	C(9)-N(8)-O(7)	105.44(9)
N(8)-C(9)-C(5)	109.76(9)	N(8)-C(9)-C(10)	120.60(10)
C(5)-C(9)-C(10)	129.62(10)	N(11)-C(10)-N(14)	115.71(9)
N(11)-C(10)-C(9)	122.48(10)	N(14)-C(10)-C(9)	121.74(10)
C(10)-N(11)-O(12)	102.98(8)	C(13)-O(12)-N(11)	106.46(8)
N(14)-C(13)-O(12)	112.85(9)	N(14)-C(13)-C(15)	126.52(10)
O(12)-C(13)-C(15)	120.62(9)	C(13)-N(14)-C(10)	101.99(9)
N(16)-C(15)-C(13)	123.94(9)	N(16)-C(15)-N(19)	118.28(9)
C(13)-C(15)-N(19)	117.77(9)	O(18)-N(16)-O(17)	120.84(9)
O(18)-N(16)-C(15)	121.64(9)	O(17)-N(16)-C(15)	117.46(9)
O(20)-N(19)-O(21)	122.93(9)	O(20)-N(19)-C(15)	120.24(9)
O(21)-N(19)-C(15)	116.83(9)	H(22A)-N(22)-H(22B)	112.1(14)
H(22A)-N(22)-H(22C)	114.6(15)	H(22B)-N(22)-H(22C)	109.6(15)
H(22A)-N(22)-H(22D)	108.1(14)	H(22B)-N(22)-H(22D)	107.1(14)
H(22C)-N(22)-H(22D)	104.7(14)	H(23A)-N(23)-H(23B)	112.1(14)
H(23A)-N(23)-H(23C)	108.1(14)	H(23B)-N(23)-H(23C)	107.9(14)
H(23A)-N(23)-H(23D)	110.4(14)	H(23B)-N(23)-H(23D)	108.3(14)
H(23C)-N(23)-H(23D)	109.9(15)		

Table S4. Anisotropic displacement parameters ($Å^2 x\ 10^3$) for **2a**. 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}
O(1)	41(1)	16(1)	22(1)	1(1)	19(1)	4(1)
O(2)	25(1)	24(1)	15(1)	-6(1)	11(1)	-2(1)
N(3)	19(1)	18(1)	16(1)	-2(1)	11(1)	0(1)
N(4)	24(1)	16(1)	16(1)	-2(1)	12(1)	-1(1)
C(5)	17(1)	19(1)	16(1)	-3(1)	10(1)	-3(1)
N(6)	29(1)	20(1)	17(1)	-2(1)	14(1)	-4(1)
O(7)	32(1)	23(1)	17(1)	0(1)	14(1)	-3(1)
N(8)	23(1)	21(1)	17(1)	-2(1)	12(1)	-4(1)
C(9)	17(1)	18(1)	17(1)	-2(1)	11(1)	-3(1)
C(10)	17(1)	15(1)	17(1)	-2(1)	10(1)	-2(1)
N(11)	28(1)	16(1)	15(1)	-4(1)	11(1)	-2(1)
O(12)	29(1)	13(1)	15(1)	-2(1)	11(1)	-2(1)
C(13)	17(1)	14(1)	17(1)	-1(1)	10(1)	-1(1)
N(14)	22(1)	15(1)	17(1)	-3(1)	11(1)	-4(1)
C(15)	20(1)	14(1)	14(1)	-1(1)	9(1)	-1(1)
N(16)	16(1)	17(1)	15(1)	-2(1)	8(1)	-2(1)
O(17)	25(1)	18(1)	17(1)	2(1)	9(1)	-1(1)
O(18)	28(1)	20(1)	18(1)	-6(1)	13(1)	-5(1)
N(19)	23(1)	16(1)	17(1)	0(1)	12(1)	0(1)
O(20)	33(1)	16(1)	26(1)	-2(1)	15(1)	-5(1)
O(21)	25(1)	25(1)	18(1)	4(1)	10(1)	4(1)
N(22)	22(1)	21(1)	15(1)	1(1)	10(1)	3(1)
N(23)	22(1)	18(1)	19(1)	-2(1)	11(1)	0(1)

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

	x	y	z	U(eq)
H(22A)	6747(9)	5048(14)	5841(9)	30
H(22B)	6320(11)	4721(12)	4603(12)	30
H(22C)	6350(10)	5989(9)	4903(13)	30
H(22D)	5806(7)	5177(14)	4976(14)	30
H(23A)	5473(7)	1664(14)	1469(10)	32
H(23B)	6026(10)	2508(10)	2469(13)	32
H(23C)	6060(10)	1213(12)	2650(12)	32
H(23D)	6430(8)	1732(14)	2081(14)	32

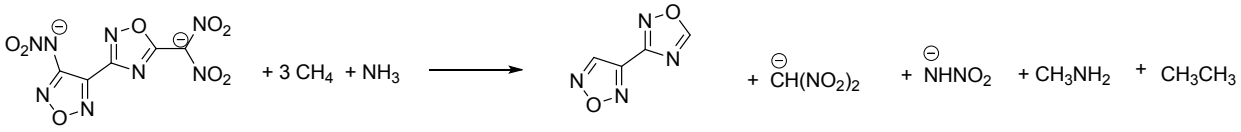
Table S6. Torsion angles [°] for **2a**.

O(2)-N(3)-N(4)-C(5)	-0.61(16)
O(1)-N(3)-N(4)-C(5)	179.28(10)
N(3)-N(4)-C(5)-N(6)	0.59(18)
N(3)-N(4)-C(5)-C(9)	179.35(9)
N(4)-C(5)-N(6)-O(7)	178.55(11)
C(9)-C(5)-N(6)-O(7)	-0.32(12)
C(5)-N(6)-O(7)-N(8)	0.09(12)
N(6)-O(7)-N(8)-C(9)	0.20(12)
O(7)-N(8)-C(9)-C(5)	-0.39(12)
O(7)-N(8)-C(9)-C(10)	-178.82(9)
N(6)-C(5)-C(9)-N(8)	0.47(13)
N(4)-C(5)-C(9)-N(8)	-178.56(10)
N(6)-C(5)-C(9)-C(10)	178.72(11)
N(4)-C(5)-C(9)-C(10)	-0.31(18)
N(8)-C(9)-C(10)-N(11)	170.64(11)
C(5)-C(9)-C(10)-N(11)	-7.45(18)
N(8)-C(9)-C(10)-N(14)	-6.24(16)
C(5)-C(9)-C(10)-N(14)	175.67(11)
N(14)-C(10)-N(11)-O(12)	0.77(13)
C(9)-C(10)-N(11)-O(12)	-176.29(9)
C(10)-N(11)-O(12)-C(13)	-0.33(11)
N(11)-O(12)-C(13)-N(14)	-0.19(12)
N(11)-O(12)-C(13)-C(15)	-179.18(10)
O(12)-C(13)-N(14)-C(10)	0.61(12)
C(15)-C(13)-N(14)-C(10)	179.52(11)
N(11)-C(10)-N(14)-C(13)	-0.88(13)
C(9)-C(10)-N(14)-C(13)	176.20(10)
N(14)-C(13)-C(15)-N(16)	-169.63(11)
O(12)-C(13)-C(15)-N(16)	9.20(17)
N(14)-C(13)-C(15)-N(19)	9.60(17)
O(12)-C(13)-C(15)-N(19)	-171.57(10)
C(13)-C(15)-N(16)-O(18)	177.98(10)
N(19)-C(15)-N(16)-O(18)	-1.25(16)
C(13)-C(15)-N(16)-O(17)	-4.71(16)
N(19)-C(15)-N(16)-O(17)	176.06(9)
N(16)-C(15)-N(19)-O(20)	37.48(15)
C(13)-C(15)-N(19)-O(20)	-141.79(11)
N(16)-C(15)-N(19)-O(21)	-142.82(10)
C(13)-C(15)-N(19)-O(21)	37.90(14)

Table S7. Hydrogen bonds for **2a** [Å and °].

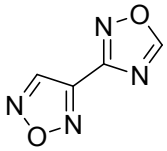
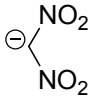
D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(22)-H(22A)...O(17)#1	0.898(9)	2.057(9)	2.9467(13)	170.7(15)
N(22)-H(22B)...O(1)	0.900(9)	2.047(10)	2.9100(13)	160.2(15)
N(22)-H(22C)...N(11)	0.890(9)	2.207(9)	3.0958(14)	176.5(14)
N(22)-H(22D)...O(2)#2	0.902(9)	2.168(12)	2.9226(13)	140.8(13)
N(22)-H(22D)...O(2)#3	0.902(9)	2.323(14)	2.9535(13)	126.8(13)
N(23)-H(23A)...O(21)#5	0.903(9)	2.366(13)	3.0654(14)	134.3(13)
N(23)-H(23B)...O(1)	0.905(9)	1.929(9)	2.8345(13)	177.8(15)
N(23)-H(23C)...N(14)#6	0.895(9)	2.223(9)	3.1146(14)	174.0(14)
N(23)-H(23D)...O(17)#7	0.898(9)	2.334(13)	3.0441(14)	135.9(13)

Symmetry transformations used to generate equivalent atoms:
#1 -x+3/2,y-1/2,-z+3/2 #2 -x+1,y,-z+1/2 #3 x,-y+1,z+1/2
#4 -x+1,-y+1,-z #5 -x+1,y-1,-z+1/2 #6 x,y-1,z
#7 x,-y+1,z-1/2 #8 -x+3/2,-y+3/2,-z+1



Scheme S1 Isodesmic reaction

Table S8. Calculated (B3LYP/6-31+G**// MP2/6-311++G**) total energy(E_0), zero-point energy (*ZPE*), values of the correction (H_r), and heats of formation (HOF) for anion of **2.**

	ZPE	H_r	E_0	Corrected E_0	HOF (kJ mol^{-1})
CH_4	0.044793	0.048605	-40.3796224	-40.33281	-74.6[a]
NH_3	0.034375	0.038193	-56.4154635	-56.37865	-45.9[a]
	0.072749	0.080577	-521.7442786	-521.66661	+306.2[b]
	0.039727	0.046607	-448.0309267	-447.98591	-233.0[b]
	0.026168	0.030444	-259.936099	-259.90670	-6.7[b]
CH_3NH_2	0.06403	0.06840	-95.59384	-95.52800	-23.0[a]
CH_3CH_3	0.07461	0.079038	-79.5716305	-79.49558	-84[a]
2(Anion)	0.097858	0.115533	-1227.33333	-1227.22171	+45.4

[a] Data from NIST, [b] Calculated from G2

Table S9. Calculated heat of formation for **2a -2c**.

Compound	ΔH_L (kJ/mol)	ΔH_f^{Cation} (kJ/mol)	ΔH_f^{Anion} (kJ/mol)	ΔH_f^{298} (kJ/mol)
2a	1294.979233	626.4	45.4	145.0
2b	1266.289433	669.5	45.4	259.9
2c	1260.303684	770	45.4	466.9