Supporting Information

High-Energy-Density Energetic Materials Based On 1-Nitramino-2,4-dinitroimidazole

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Table of contents

X-ray crystallography: Crystals of guanidinium 1-nitramino-2,4-dinitroimidazolate (NADNI-G) were removed from the flask and covered with a layer of hydrocarbon oil, respectively. A suitable crystal was selected, attached to a glass fiber, and placed in the low-temperature nitrogen stream. Data for NADNI-G was collected at 153(2) K using a Rigaku Saturn724 CCD diffractometer equipped with a graphite-monochromatized Mo*Kα* radiation (λ= 0.71073 Å) using omega scans. Data collection and reduction were performed and the unit cell was initially refined by using CrystalClear -SM Expert 2.0 r2[1] software. The reflection data were also corrected for *Lp* factors. The structure was solved by direct methods and refined by least squares method on F^2 using SHELXTL-97 system of programs[2]. Structure was solved in the space group *P*2(1)/c by analysis of systematic absences. In this all-light-atom structure the value of the Flack parameter did not allow the direction of polar axis to be determined and Friedel reflections were then merged for the final refinement. Band angles and dihedral angles of the data collection and refinement are given in Table S1.

Table S1. Band angles and dihedral angles for the structure of NADNI-G.

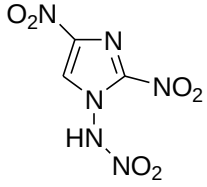
Band angle	degree/°	Band angle	degree/°
C1—N1—C2	106.06 (10)	C4—N8—H8B	121.1 (12)
C1—N1—N3	122.91 (10)	H8A—N8—H8B	116.5 (17)
C2—N1—N3	130.35 (10)	C4—N9—H9A	119.2 (11)
C2—N2—C3	102.65 (11)	C4—N9—H9B	118.3 (12)
N4—N3—N1	109.50 (9)	H9A—N9—H9B	121.6 (16)
O2—N4—O1	121.99 (11)	N1—C1—C3	104.53 (11)
O2—N4—N3	122.89 (11)	N1—C1—H1	127.7
O1—N4—N3	115.11 (10)	C3—C1—H1	127.7
O3—N5—O4	125.25 (11)	N2—C2—N1	113.66 (11)
O3—N5—C2	118.33 (11)	N2—C2—N5	122.43 (11)
O4—N5—C2	116.41 (11)	N1—C2—N5	123.84 (11)
O5—N6—O6	124.45 (12)	N2—C3—C1	113.09 (12)
O5—N6—C3	119.03 (12)	N2—C3—N6	120.95 (11)
O6—N6—C3	116.53 (11)	C1—C3—N6	125.95 (12)
C4—N7—H7A	119.0 (11)	N7—C4—N9	120.16 (13)
C4—N7—H7B	119.0 (11)	N7—C4—N8	120.09 (12)
H7A—N7—H7B	122.0 (16)	N9—C4—N8	119.73 (14)
C4—N8—H8A	122.4 (13)		
Dihedral angle	**degree/°**	**Dihedral angle**	**degree/°**
C1—N1—N3—N4	114.87 (13)	O3—N5—C2—N2	176.53 (11)
C2—N1—N3—N4	−75.95 (15)	O4—N5—C2—N2	−2.26 (17)
N1—N3—N4—O2	−0.52 (16)	O3—N5—C2—N1	−0.33 (17)
N1—N3—N4—O1	178.45 (10)	O4—N5—C2—N1	−179.12 (11)
C2—N1—C1—C3	−0.07 (13)	C2—N2—C3—C1	0.00 (14)

N3—N1—C1—C3	171.36 (10)	C2—N2—C3—N6	178.80 (11)
C3—N2—C2—N1	−0.05 (13)	N1—C1—C3—N2	0.05 (14)
C3—N2—C2—N5	−177.20 (11)	N1—C1—C3—N6	−178.68 (11)
C1—N1—C2—N2	0.08 (14)	O5—N6—C3—N2	4.12 (17)
N3—N1—C2—N2	−170.48 (11)	O6—N6—C3—N2	−175.71 (11)
C1—N1—C2—N5	177.19 (11)	O5—N6—C3—C1	−177.24 (12)
N3—N1—C2—N5	6.63 (19)	O6—N6—C3—C1	2.93 (18)

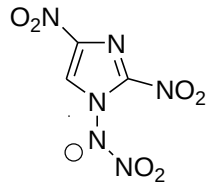
Theoretical study: Calculations were carried out by using the Gaussian 03 (Revision E.01) suite of programs.[3] The geometric optimization of the structures and frequency analyses were carried out by using the B3LYP functional with the 6-31+G** basis set,[4] and single-point energies were calculated at the MP2(full)/6-311++G** level. All of the optimized structures were characterized to be true local energy minima on the potential-energy surface without imaginary frequencies.

Geometry coordinates

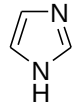
The optimized structure of the following structure.



C	-1.720781	-0.139667	-0.109970
C	-0.782365	-1.046034	-0.553632
N	0.391233	-0.342194	-0.562347
C	0.083388	0.937494	-0.103218
N	-1.185457	1.077139	0.154636
N	1.607142	-0.922906	-0.869800
N	1.061249	2.002007	-0.028836
O	0.738051	3.037322	0.522986
O	2.166223	1.762652	-0.548063
N	2.322426	-1.411754	0.329407
O	1.650834	-1.647793	1.311825
O	3.507898	-1.581226	0.138207
N	-3.137740	-0.417495	0.054992
O	-3.858964	0.483517	0.458378
O	-3.489575	-1.565405	-0.234081
H	-0.839949	-2.082933	-0.842833
H	2.230786	-0.233946	-1.296620

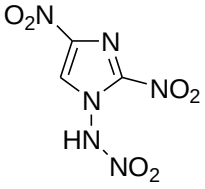
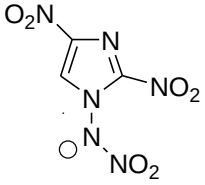
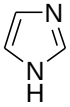


C	1.766250	-0.185589	-0.052914
C	0.838891	-1.186756	-0.317225
N	-0.359826	-0.559161	-0.380638
C	-0.092352	0.781467	-0.143621
N	1.181963	1.027954	0.062579
N	-1.522650	-1.249013	-0.743486
N	3.181671	-0.376470	0.079900
O	3.896036	0.593997	0.333862
O	3.601525	-1.537382	-0.071941
N	-1.084475	1.835381	-0.180867
O	-0.775935	2.911379	0.332463
O	-2.147950	1.596426	-0.752484
N	-2.423262	-1.196634	0.265066
O	-2.187129	-0.583040	1.334865
O	-3.488788	-1.803590	0.050303
H	0.927263	-2.251455	-0.451850



C	-1.143441	-0.265354	0.000128
N	-0.153596	-1.224728	-0.000110
C	0.981373	-0.557101	0.000021
N	0.770415	0.795060	0.000060
C	-0.595949	0.995734	-0.000110
H	-2.189372	-0.536971	0.000202
H	1.972293	-0.988861	0.000013
H	1.480730	1.511773	0.000094
H	-1.033281	1.982060	-0.000189

Table S2. Ab Initio computional data (B3LYP/6-31+G(d,p)/MP2(full)/6-311++G**)

	E_0 (hartree)	ZPE (hartree)	H_T (hartree)	HOF ^{Exp} (kJ mol^{-1})	HOF ^{Calcd} (kJ mol^{-1})
	-893.385741	0.094361	0.013303	-	278.5
	-892.8927874	0.081101	0.012969	-	7.1
	-225.7346277	0.071152	0.005713	-	132.9
NH_3	-56.43462[2]	0.034377	0.003818	-45.9[a]	-
CH_4	-40.39849[2]	0.044791	0.003812	-74.6[a]	-
CH_3NO_2	-244.5543604	0.049857	0.005272	-74.3[b]	-
NH_2NO_2	-260.5693535	0.039259	0.004652	-6.1[a]	-
NH_2NH_2	-111.63188	0.053399	0.004202	95.4[a]	-

[a] Eur. J. Inorg. Chem. **2008**, 2560-2568; [b] New J. Chem., **2008**, 32, 317-322

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(2) G. M. Sheldrick, SHELXTL-97, Structure Determination Software Suite. Bruker AXS, Madison WI, **2008**.

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