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Supporting Information

Super-High-Energy Materials Based on Bis(2,2-dinitroethyl)nitramine

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X-ray crystallography: Crystals of bis(2,2-dinitroethyl)nitramine(BDNENA) was 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 bis(2,2-dinitroethyl)nitramine was collected at 113(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 C2/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 and S2.

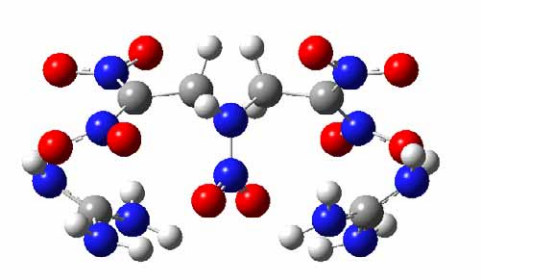
Table S1. Band angles and dihedral angles for the structure of BDNENA.

$O1^i$—N1—O1	126.1 (2)	N2—C1—C2	114.31 (14)
$O1^i$—N1—N2	116.96 (12)	N2—C1—H1A	108.7
O1—N1—N2	116.96 (12)	C2—C1—H1A	108.7
N1—N2—C1	117.53 (10)	N2—C1—H1B	108.7
N1—N2—$C1^i$	117.53 (10)	C2—C1—H1B	108.7
C1—N2—$C1^i$	124.9 (2)	H1A—C1—H1B	107.6
O3—N3—O2	125.50 (18)	N3—C2—N4	106.95 (14)
O3—N3—C2	117.81 (16)	N3—C2—C1	106.58 (15)
O2—N3—C2	116.54 (18)	N4—C2—C1	113.86 (15)
O4—N4—O5	125.55 (16)	N3—C2—H2	109.8
O4—N4—C2	116.64 (16)	N4—C2—H2	109.8
O5—N4—C2	117.69 (15)	C1—C2—H2	109.8
$O1^i$—N1—N2—C1	−171.64 (10)	O3—N3—C2—C1	75.19 (18)
O1—N1—N2—C1	8.36 (10)	O2—N3—C2—C1	−100.55 (18)
$O1^i$—N1—N2—$C1^i$	8.36 (10)	O4—N4—C2—N3	−44.21 (19)
O1—N1—N2—$C1^i$	−171.64 (10)	O5—N4—C2—N3	139.51 (16)
N1—N2—C1—C2	−72.79 (14)	O4—N4—C2—C1	−161.66 (15)
$C1^i$—N2—C1—C2	107.21 (14)	O5—N4—C2—C1	22.1 (2)
O3—N3—C2—N4	−46.94 (19)	N2—C1—C2—N3	170.01 (15)
O2—N3—C2—N4	137.32 (16)	N2—C1—C2—N4	−72.3 (2)

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.

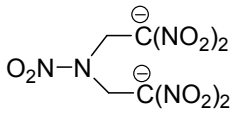
Geometry coordinates

The optimized structure of the following structure [bis(guanidinium) BDNA salt].



O	0.526541	1.351967	0.959596
O	4.191760	0.068133	1.394855
O	2.549239	-0.120180	2.806123
O	4.077373	-1.781073	-0.525592
O	2.033119	-2.190125	-1.159778
N	0.000000	0.743506	0.000007
N	0.000001	-0.575624	-0.000001
N	3.051881	-0.380806	1.705234
N	2.853237	-1.712898	-0.340113
C	0.827729	-1.285331	1.020257
H	0.507269	-2.320579	0.946509
H	0.574701	-0.906126	2.005197
C	2.292846	-1.116163	0.794353
C	3.246069	1.628359	-1.182106
N	2.351695	1.808201	-2.171101
H	2.352872	1.164556	-2.947034
H	1.452304	2.217971	-1.955521
N	4.332167	0.875636	-1.370077
H	4.697252	0.425411	-0.525342
H	4.368116	0.278043	-2.183251
N	3.099433	2.283298	-0.022992
H	2.153085	2.510695	0.261864
H	3.679716	1.937483	0.740290

O	-0.526541	1.351978	-0.959576
O	-4.191760	0.068139	-1.394855
O	-2.549236	-0.120152	-2.806122
O	-4.077369	-1.781083	0.525576
O	-2.033115	-2.190144	1.159755
N	-3.051878	-0.380792	-1.705237
N	-2.853233	-1.712905	0.340096
C	-0.827726	-1.285321	-1.020267
H	-0.507264	-2.320569	-0.946531
H	-0.574698	-0.906104	-2.005203
C	-2.292843	-1.116157	-0.794362
C	-3.246075	1.628348	1.182118
N	-4.332170	0.875620	1.370080
H	-4.697252	0.425399	0.525342
H	-4.368120	0.278021	2.183251
N	-3.099439	2.283296	0.023010
H	-2.153091	2.510697	-0.261844
H	-3.679721	1.937487	-0.740276
N	-2.351702	1.808185	2.171116
H	-2.352878	1.164533	2.947043
H	-1.452314	2.217961	1.955541



C	-0.256402	0.052053	-2.210914
C	-0.781103	-0.100195	-0.809468
N	0.181679	-0.613437	0.189583
C	0.781038	0.227307	1.248937
C	0.282134	-0.020352	2.646089
N	1.193606	-0.379892	3.648890
O	0.816267	-0.807965	4.760123
O	2.422729	-0.251009	3.383519
N	0.530706	-1.913975	0.131896
O	1.347673	-2.335285	0.971140
O	0.014989	-2.616515	-0.757513
N	-0.888377	-0.629631	-3.260511
O	-2.011387	-1.154544	-3.013412
O	-0.371972	-0.717747	-4.394497

N	0.719789	1.006728	-2.461274
O	1.161311	1.619566	-1.438480
O	1.130261	1.295448	-3.608777
N	-1.034611	0.294183	2.953285
O	-1.745464	0.688942	1.975814
O	-1.509232	0.240833	4.111073
H	-1.634286	-0.773938	-0.845141
H	-1.107788	0.852554	-0.393600
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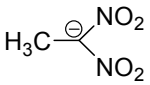
H	1.857368	0.070130	1.247483



C	-7.001058	-0.246088	-0.426835
C	-5.537562	-0.089098	-0.154223
N	-4.890407	1.058571	-0.603753
O	-3.662625	1.251237	-0.465626
O	-5.628028	1.924879	-1.175400
N	-4.891229	-1.052007	0.615898
O	-3.663500	-1.029311	0.852142
O	-5.629531	-1.979524	1.080630
H	-7.624260	0.207893	0.358539
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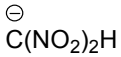
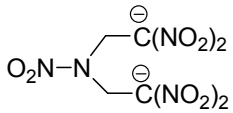
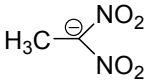
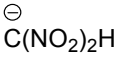
H	-7.251788	-1.307409	-0.469083



C	-6.568211	0.113431	0.245351
N	-5.759917	1.225093	0.122244
O	-6.351687	2.339781	0.277897
O	-4.536838	1.159207	-0.123328
N	-6.177358	-1.204282	0.122242
O	-7.107273	-2.057497	0.277908
O	-5.002440	-1.550446	-0.123330
H	-7.605684	0.291699	0.457303

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})
	-1232.9160043	0.136771	0.018912	-	26.0
	-487.381106	0.067130	0.008621	-	-94.8
	-448.1640711	0.039724	0.006879	-	-222.8
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_3CH_3	-79.6068548	0.074609	0.00443	-84.68[b]	-
CH_3NH_2	-95.6318759	0.064032	0.004369	-23.0[a]	-

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

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