Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Supporting Information

Tunable 1,2,3-triazole-N-Oxides towards high energy density materials: theoretical insight into structure-property correlations

Yue Liu[1], Lishan Gong[1], Xiaoyi Yi[1], Piao He[1]*, Jianguo Zhang[2]

[1] *College of Chemistry and Chemical Engineering, Central South University, Changsha 410083, Hunan, P. R. China*

[2] *State Key Laboratory of Explosion Science and Technology, Beijing Institute of Technology, Beijing 100081 P. R. China*

Table of Contents

1. Optimized parameters
2. HOMO-LUMO orbitals and energy gap
3. Molecular electrostatic potential (ESP)
4. Thermodynamic parameters
5. Detonation parameters
6. References

1. Optimized parameters

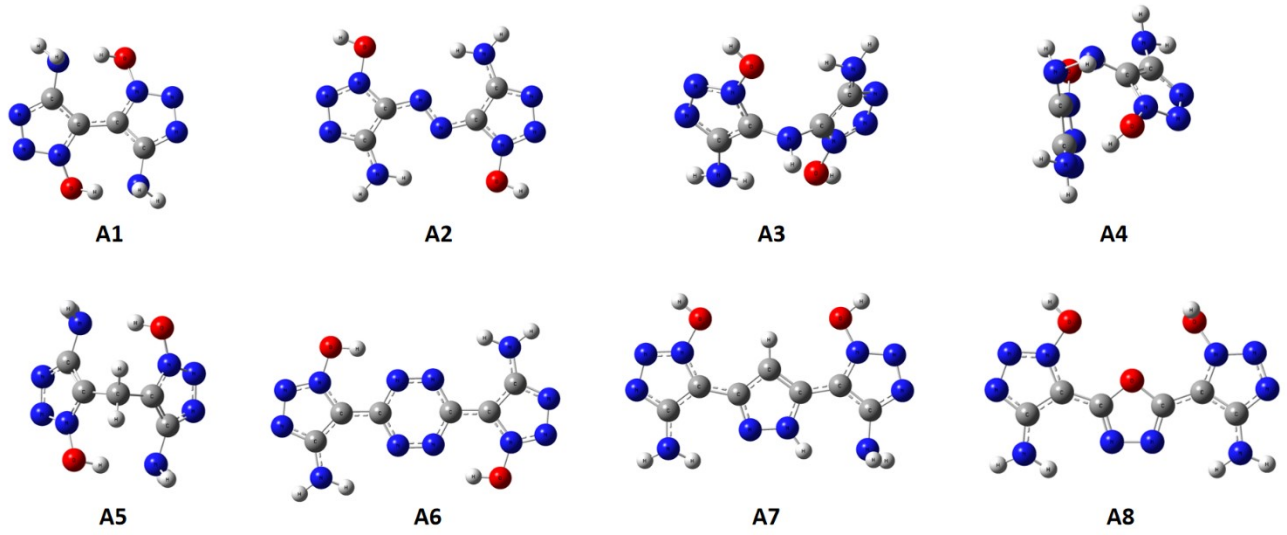

Figure S1 Optimized geometry of **A** series

*Corresponding authors: Piao He, E-mail: piaohe@csu.edu.cn

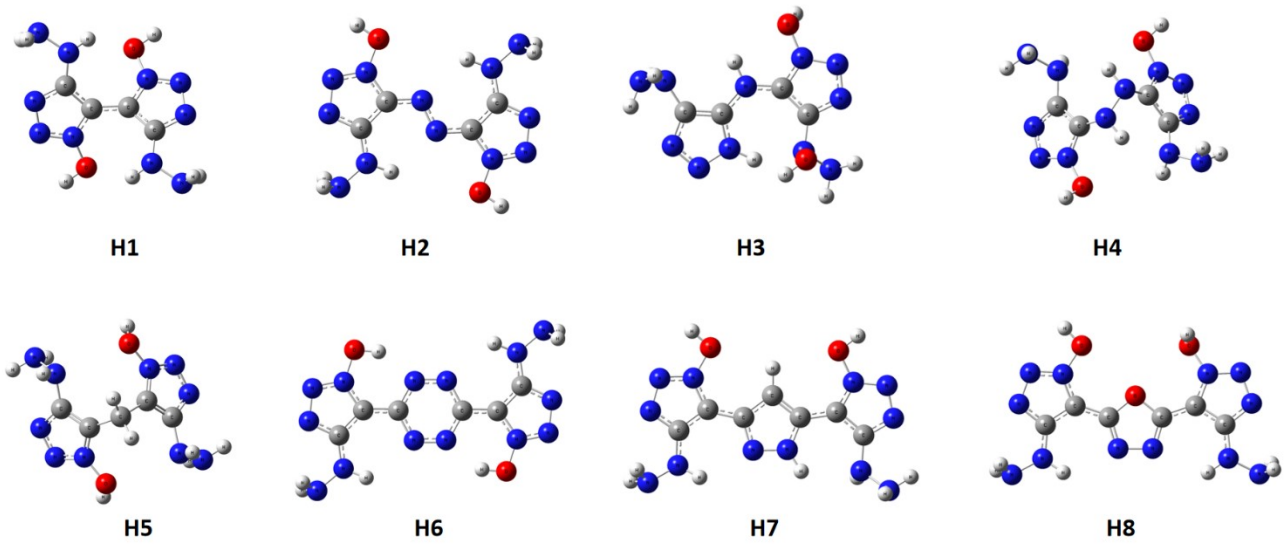

Figure **S2** Optimized geometry of **H** series

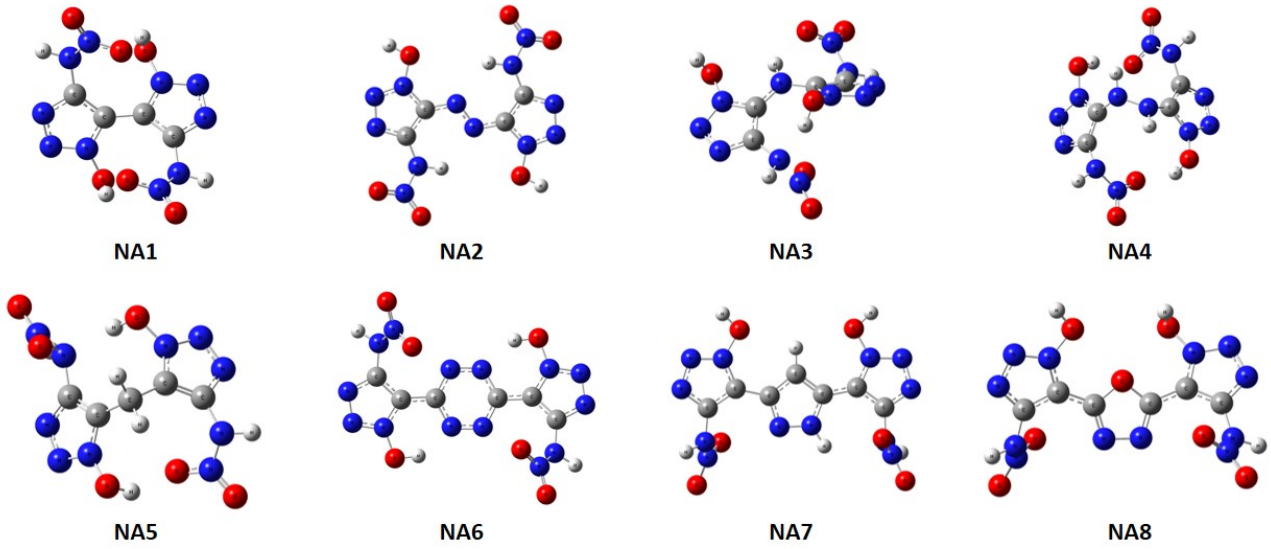

Figure S3 Optimized geometry of **NA** series

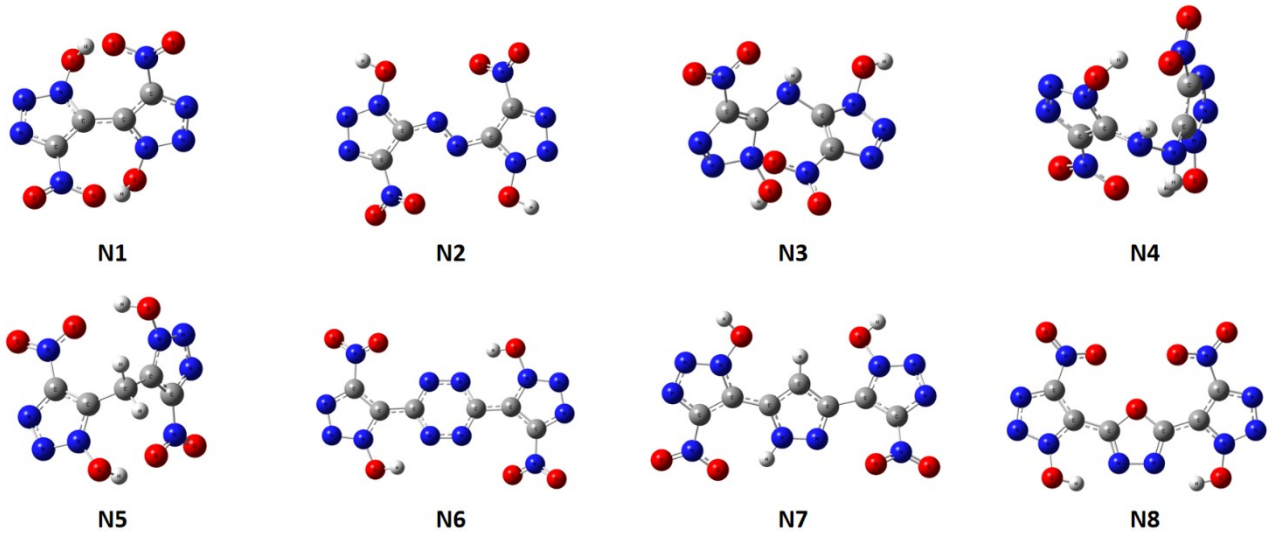

Figure S4 Optimized geometry of **N** series

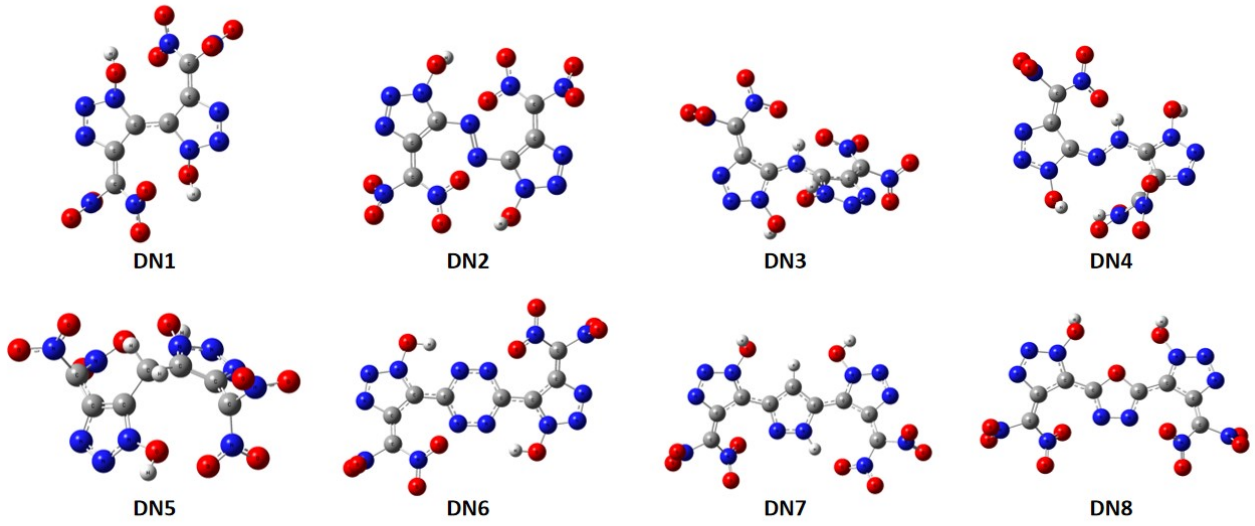

Figure S5 Optimized geometry of **DN** series

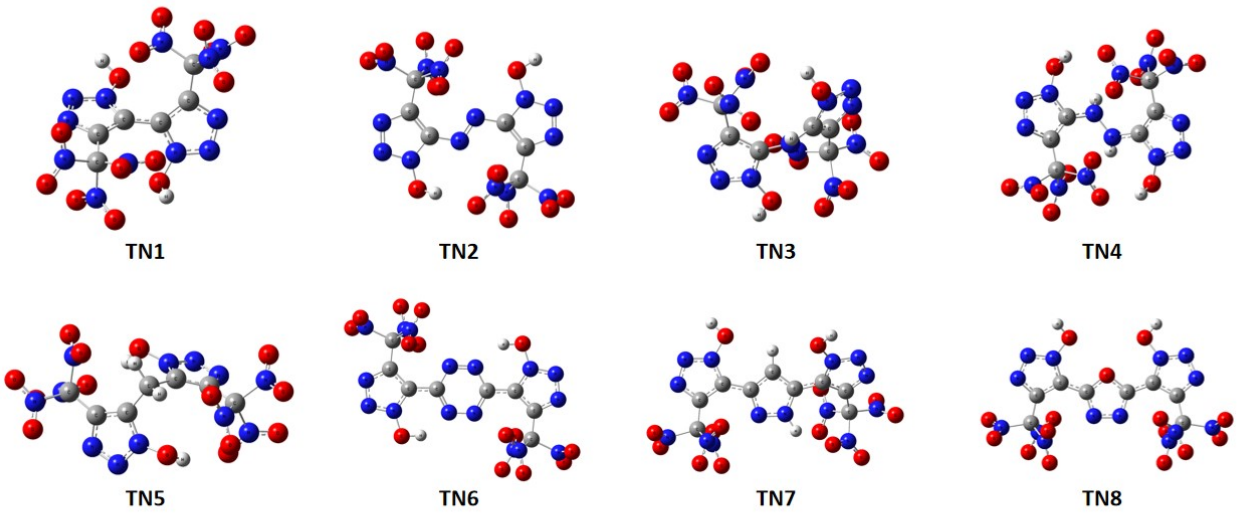

Figure S6 Optimized geometry of **TN** series

2. HOMO-LUMO orbitals and energy gap

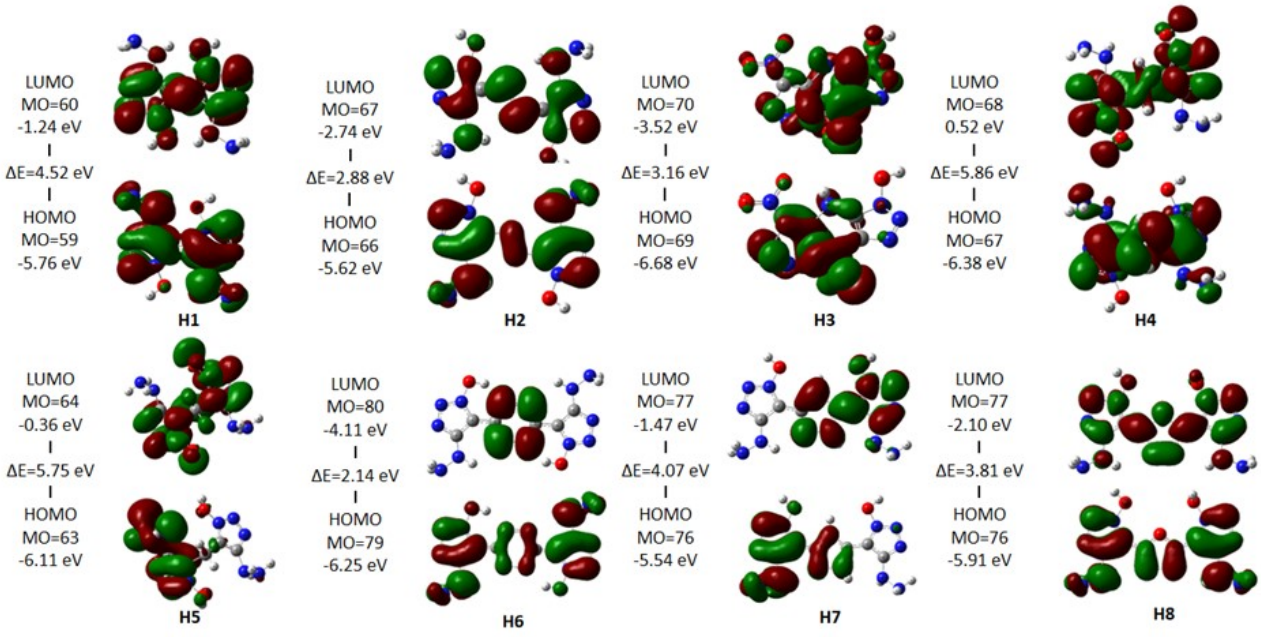

Figure S7 HOMO-LUMO energy and HOMO-LUMO gap of **H** series

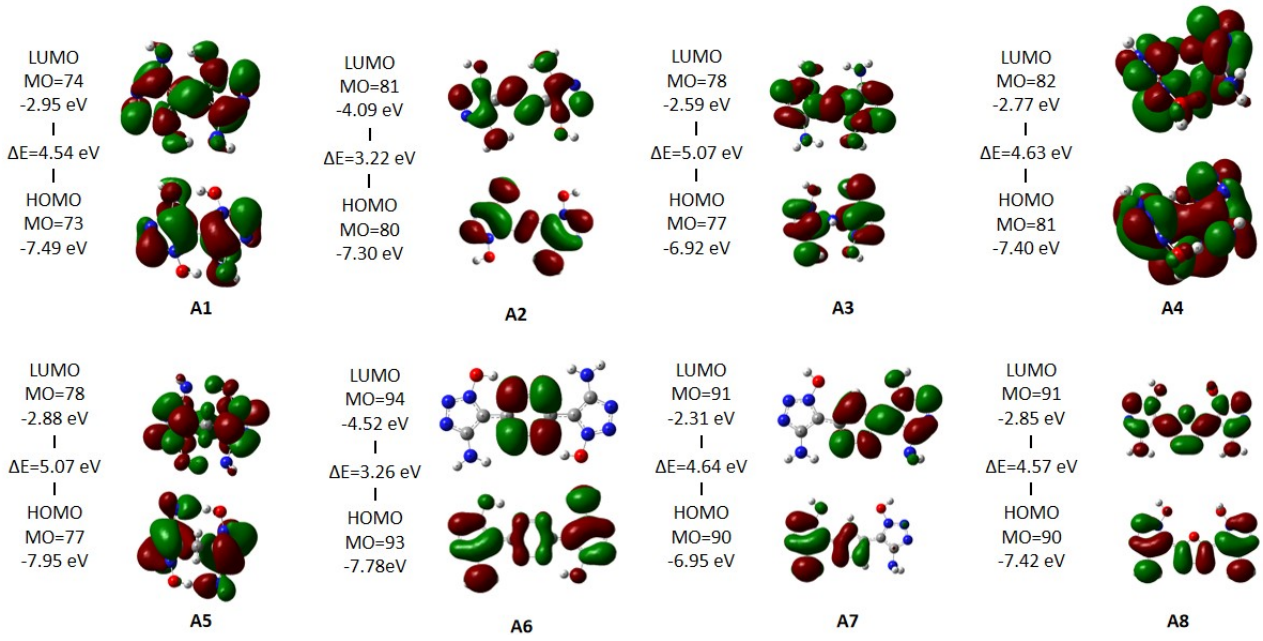

Figure S8 HOMO-LUMO energy and HOMO-LUMO gap of **A** series

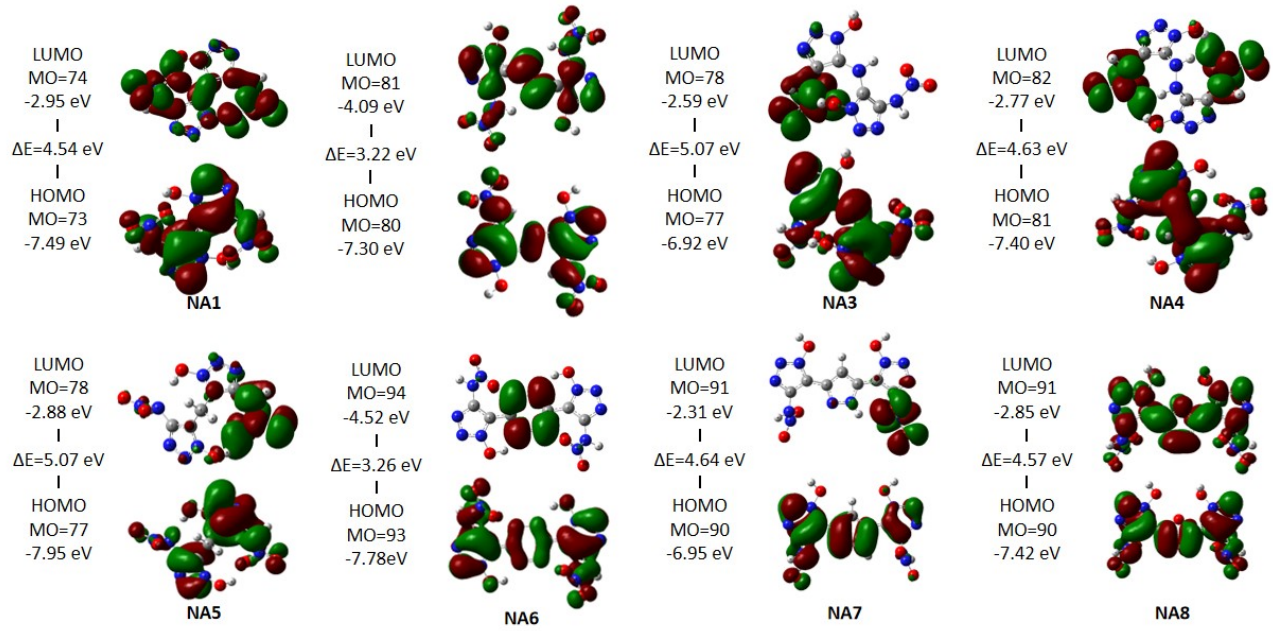

Figure S9 HOMO-LUMO energy and HOMO-LUMO gap of **NA** series

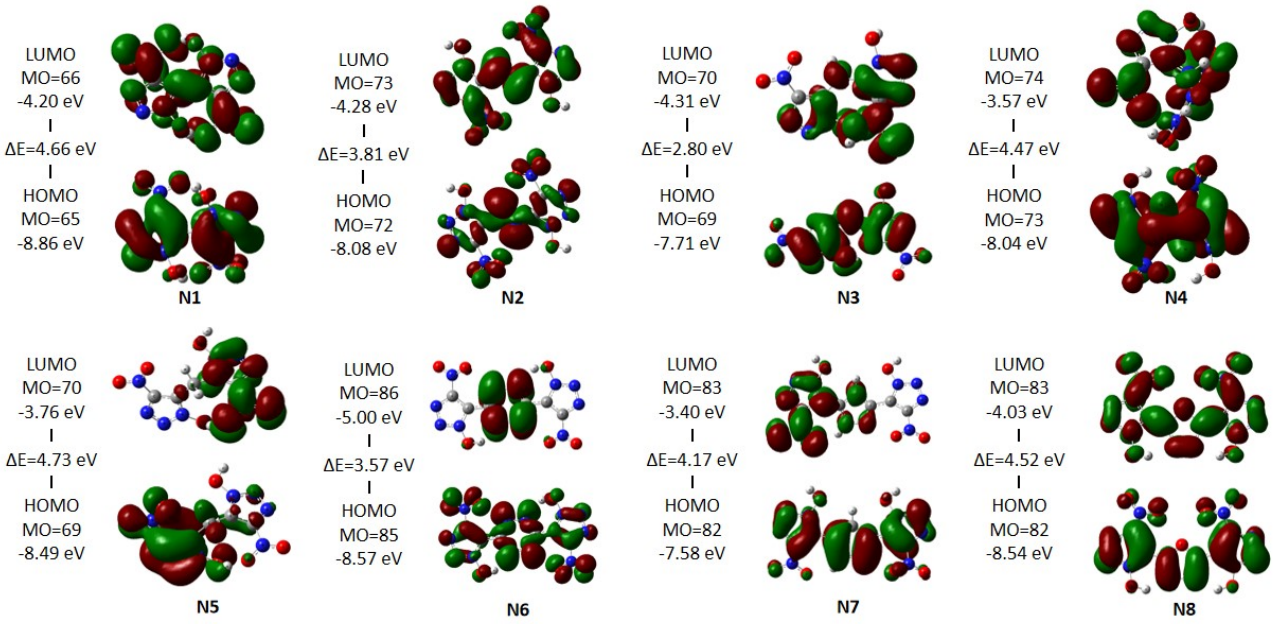

Figure S10 HOMO-LUMO energy and HOMO-LUMO gap of **N** series

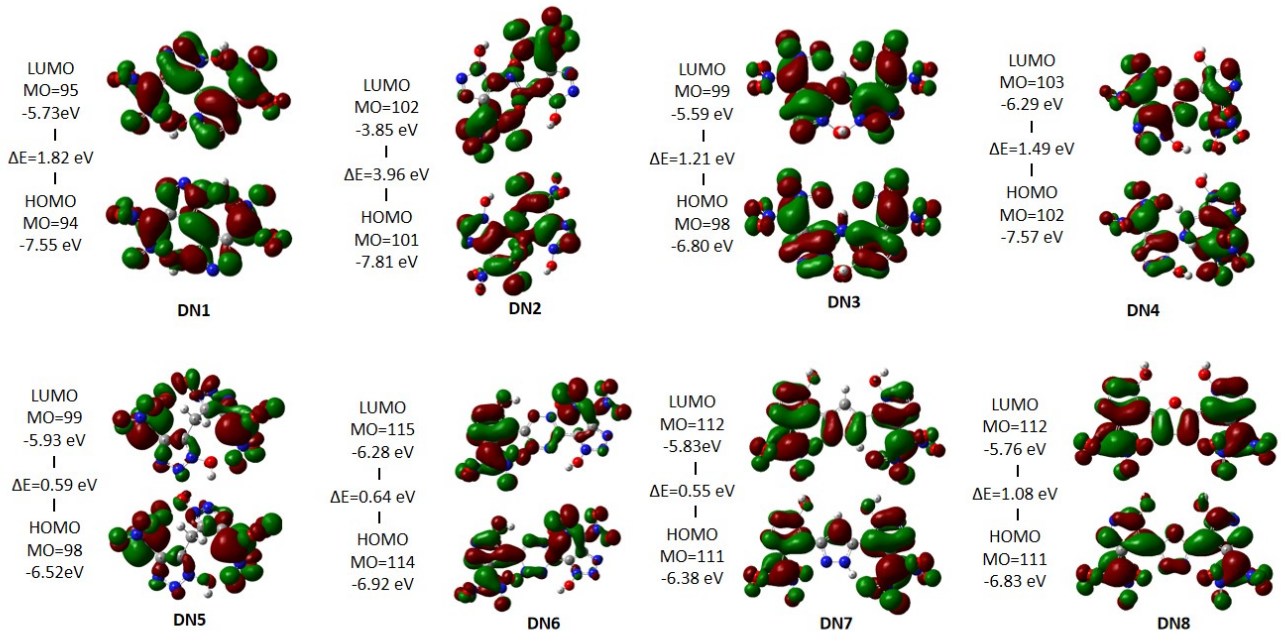

Figure S11 HOMO-LUMO energy and HOMO-LUMO gap of **DN** series

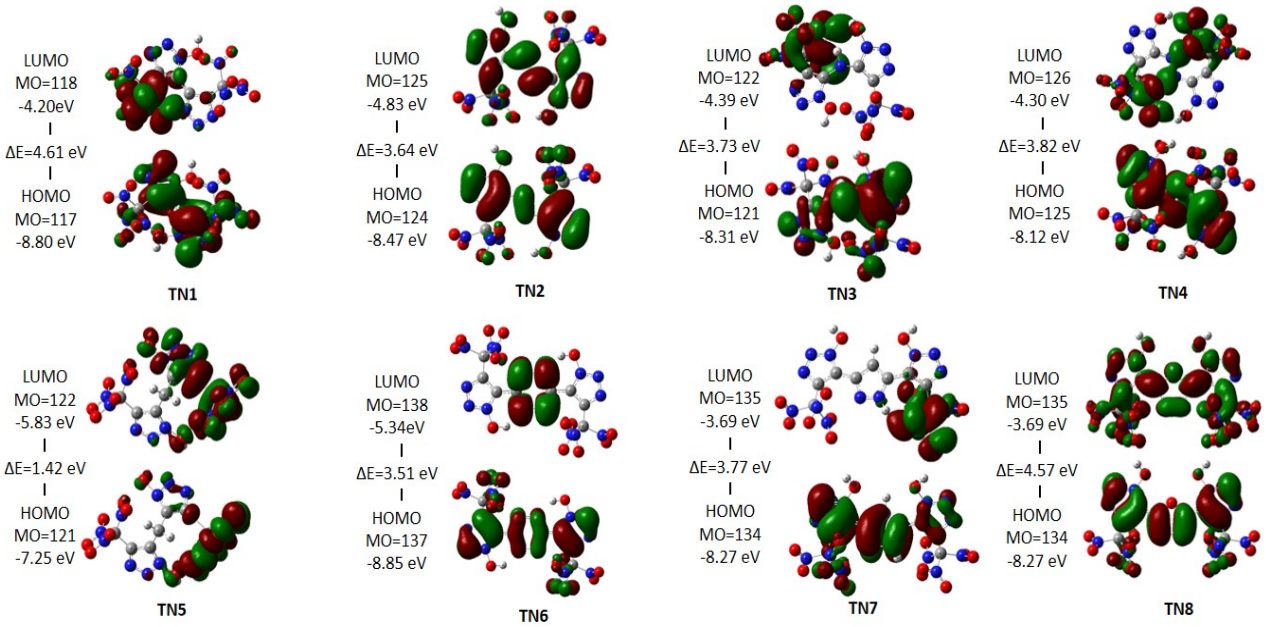

Figure S12 HOMO-LUMO energy and HOMO-LUMO gap of **TN** series

3. Molecular electrostatic potential (ESP)

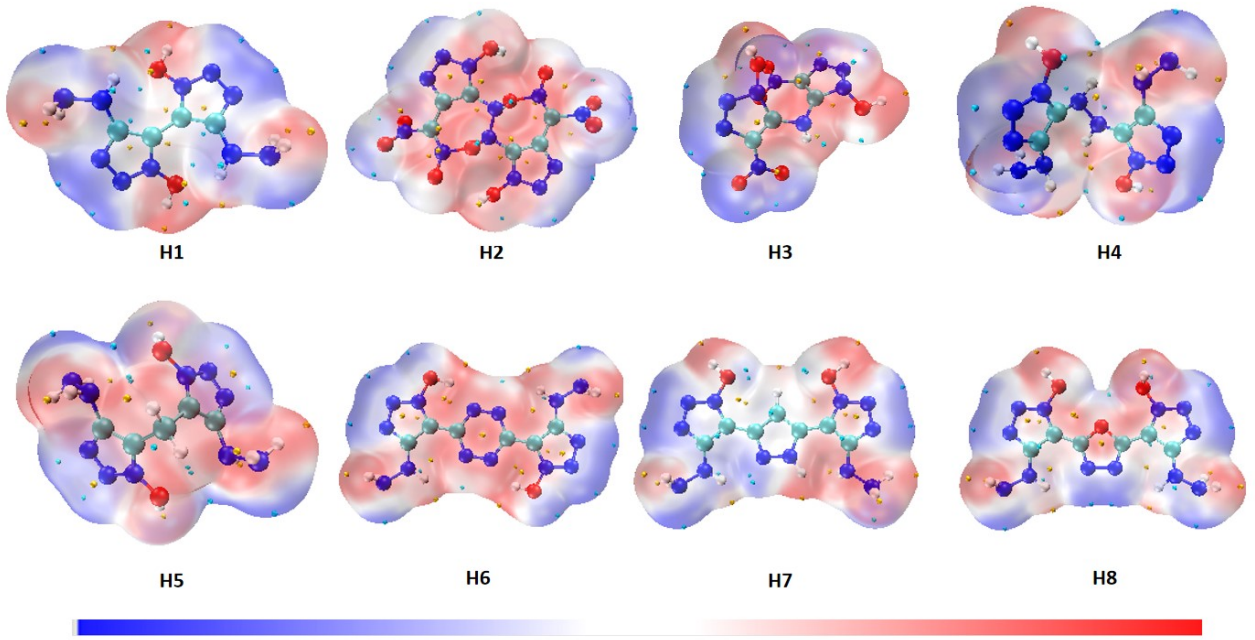

Figure S13 Molecular electrostatic potential of -$NHNH_2$**(H)** series ranging from -0.03 a.u.(blue) to 0.03 a.u.(red), isosurface=0.001

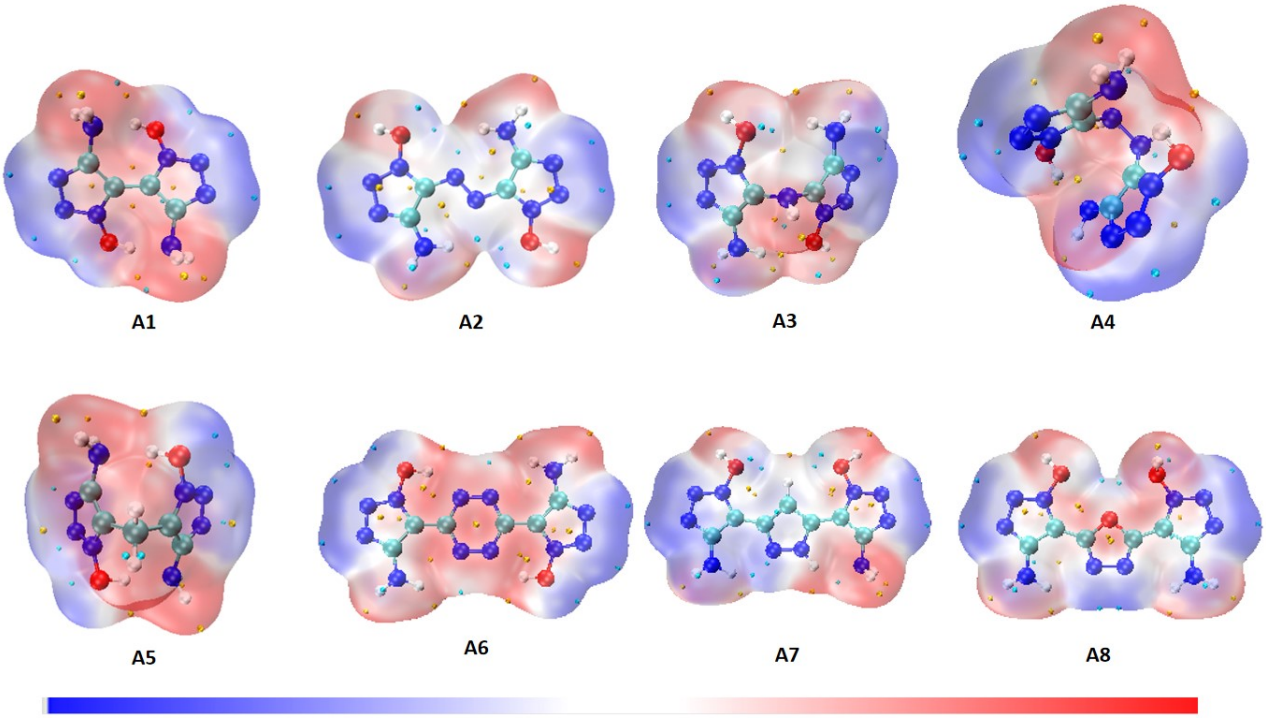

Figure S14 Molecular electrostatic potential of –NH_2**(A)** series ranging from -0.03 a.u.(blue) to 0.03 a.u.(red), isosurface=0.001

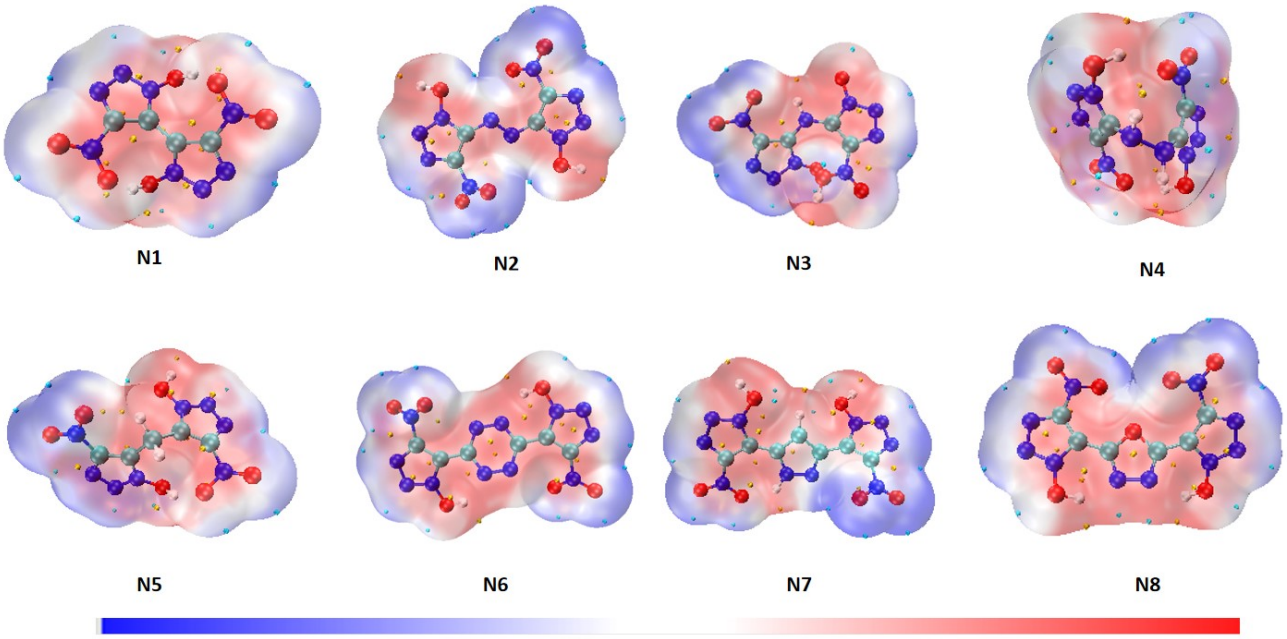

Figure S15 Molecular electrostatic potential of $–NO_2$**(N)** series ranging from -0.03 a.u.(blue) to 0.03 a.u.(red), isosurface=0.001

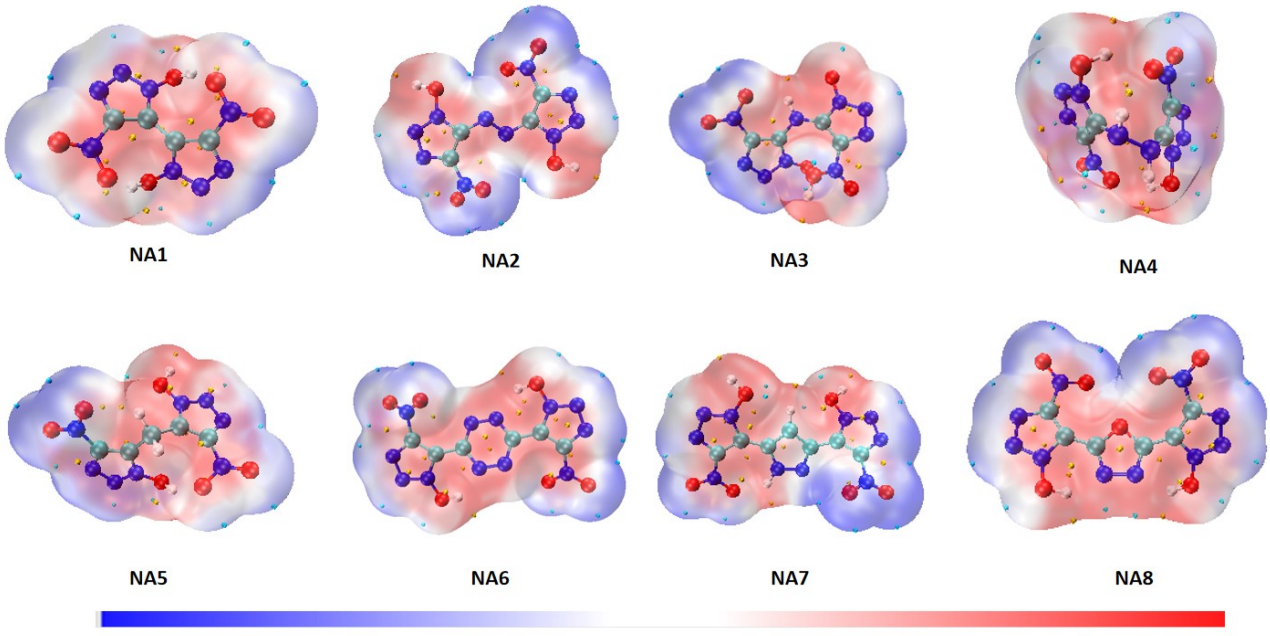

Figure S16 Molecular electrostatic potential of $-NHNO_2$**(NA)** series ranging from -0.03 a.u.(blue) to 0.03 a.u.(red), isosurface=0.001

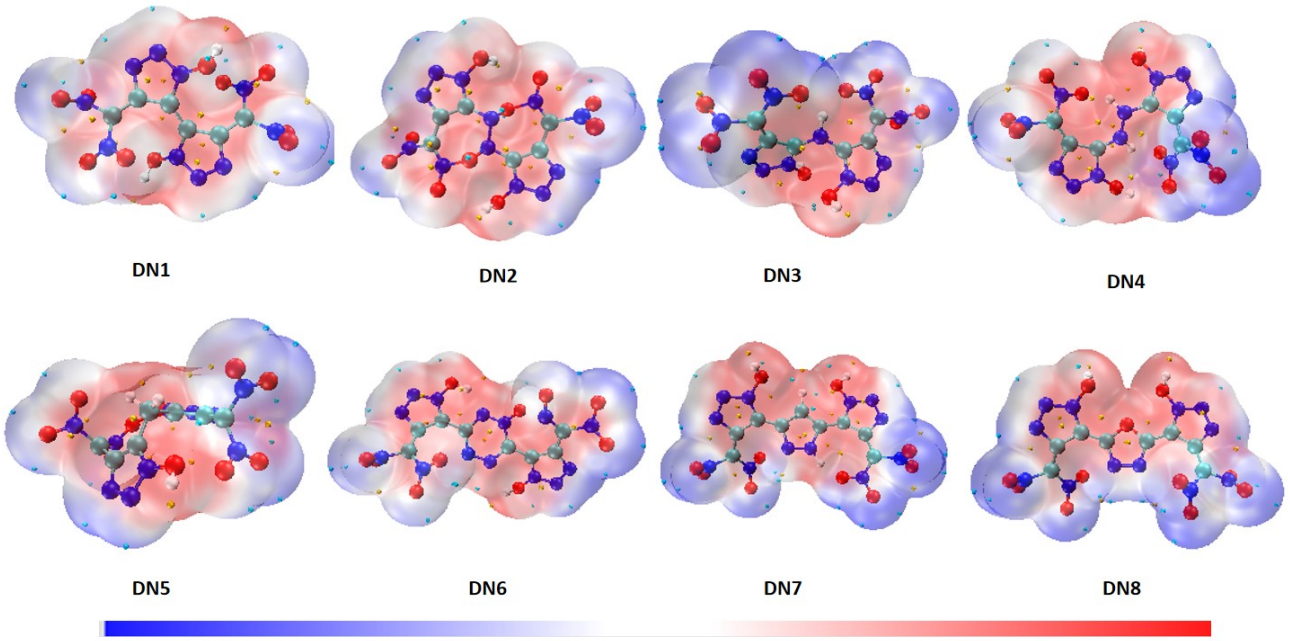

Figure S17 Molecular electrostatic potential of $–C(NO_2)_2$ **(DN)** series ranging from -0.03 a.u.(blue) to 0.03 a.u.(red), isosurface=0.001

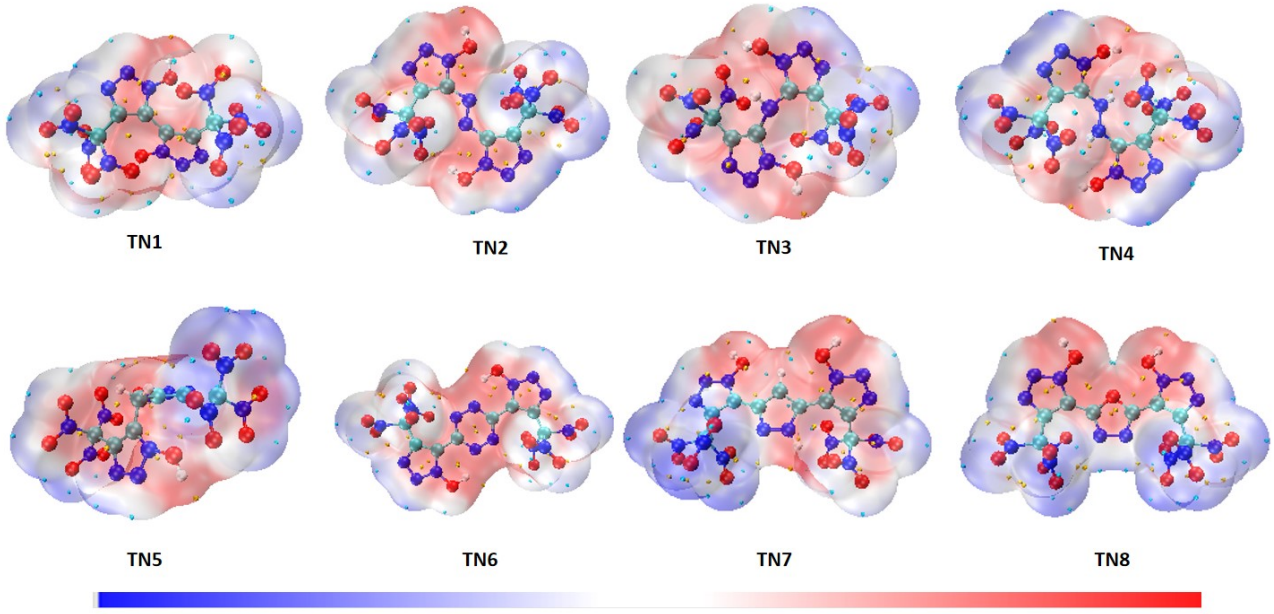

Figure S18 Molecular electrostatic potential of $–C(NO_2)_3$ **(TN)** series ranging from -0.03 a.u.(blue) to 0.03 a.u.(red), isosurface=0.001

4. Thermodynamic parameters

Table. S1 48 substituted 1,2,3-triazole-N-oxides, their sum of electronic and zero-point energy (E_0), thermal correction from 0K to 298K (H_T), gas-phase heat of formation under 0K ($HOF_{(g,0K)}$), gas-phase heat of formation under 298K ($HOF_{(g,298K)}$), condensed-phase heat of formation under 298K ($HOF_{(c,298K)}$), compared with HMX and RDX.

No.	Formula	E_0 (a.u.) 6-311G(d,p)	H_T (a.u.)	$HOF_{(g,0K)}$ (kJ/mol)	$HOF_{(g,298K)}$ (kJ/mol)	$HOF_{(c,298K)}$ (kJ/mol)
A1	$C_4H_6N_8O_2$	-744.3836	0.0121	556.58	515.31	407.13
A2	$C_4H_6N_{10}O_2$	-853.8553	0.0159	777.03	737.05	609.78
A3	$C_4H_7N_9O_2$	-799.7129	0.0154	643.38	602.22	480.74
A4	$C_4H_8N_{10}O_2$	-855.0447	0.0140	723.37	670.04	548.65
A5	$C_4H_8N_8O_2$	-783.6766	0.0133	563.86	516.30	401.28
A6	$C_6H_6N_{12}O_2$	-1039.5681	0.0176	1030.74	984.59	847.35
A7	$C_7H_8N_{10}O_2$	-969.4000	0.0180	789.61	743.49	597.12
A8	$C_6H_6N_{10}O_3$	-1005.3304	0.0178	667.02	625.67	487.56
H1	$C_4H_6N_{10}O_2$	-1153.4359	0.0174	693.33	648.38	527.47
H2	$C_4H_6N_{12}O_2$	-1262.8989	0.0204	936.72	891.02	753.75
H3	$C_4H_9N_{11}O_2$	-1208.7895	0.0188	716.29	666.57	531.21
H4	$C_4H_{10}N_{12}O_2$	-1264.1121	0.0195	820.53	764.02	620.97
H5	$C_5H_6N_{10}O_2$	-1192.7469	0.0183	653.29	601.25	468.40
H6	$C_6H_8N_{14}O_2$	-1448.6179	0.0216	1174.01	1120.58	971.04
H7	$C_7H_{10}N_{12}O_2$	-1378.4630	0.0225	898.25	846.48	686.23
H8	$C_6H_8N_{12}O_3$	-1414.3777	0.0223	816.92	769.60	616.58
N1	$C_4H_2N_8O_6$	-1042.7257	0.0146	655.74	620.66	520.77
N2	$C_4H_2N_{10}O_6$	-1152.1600	0.0181	974.29	939.62	808.31
N3	$C_4H_3N_9O_6$	-1098.0263	0.0175	817.55	781.46	657.05
N4	$C_4H_4N_{10}O_6$	-1153.3958	0.0168	798.51	752.09	634.49
N5	$C_5H_4N_8O_6$	-1082.0204	0.0168	658.47	619.55	493.11
N6	$C_6H_2N_{12}O_6$	-1337.8724	0.0196	1228.76	1187.26	1041.45
N7	$C_7H_4N_{10}O_6$	-1267.7344	0.0200	908.88	867.70	710.86
N8	$C_6H_2N_{10}O_7$	-1303.6514	0.0189	821.32	782.35	640.72

NA1	$C_4H_4N_{10}O_6$	-1153.4045	0.0174	775.74	730.78	615.81
NA2	$C_4H_4N_{12}O_6$	-1262.8634	0.0204	1029.84	984.14	831.56
NA3	$C_4H_5N_{11}O_6$	-1208.7606	0.0188	792.04	742.32	608.54
NA4	$C_4H_6N_{12}O_6$	-1264.0762	0.0195	914.58	858.07	719.27
NA5	$C_5H_6N_{10}O_6$	-1192.7150	0.0183	737.20	685.16	554.53
NA6	$C_6H_4N_{14}O_6$	-1448.5791	0.0216	1275.71	1222.28	1062.66
NA7	$C_7H_6N_{12}O_6$	-1378.4245	0.0225	999.41	947.64	776.84
NA8	$C_6H_4N_{12}O_7$	-1414.3378	0.0223	921.55	874.23	706.55
DN1	$C_6H_2N_{10}O_{10}$	-1529.0888	0.0231	1085.80	1044.72	905.36
DN2	$C_6H_2N_{12}O_{10}$	-1638.5073	0.0239	1445.58	1397.89	1246.01
DN3	$C_6H_3N_{11}O_{10}$	-1584.4582	0.0248	1067.12	1022.09	854.93
DN4	$C_6H_4N_{12}O_{10}$	-1639.8157	0.0260	1079.75	1029.21	850.16
DN5	$C_7H_4N_{10}O_{10}$	-1568.3879	0.0253	1076.93	1032.23	872.36
DN6	$C_8H_2N_{14}O_{10}$	-1824.2383	0.0269	1651.52	1600.90	1399.51
DN7	$C_9H_4N_{12}O_{10}$	-1754.1045	0.0279	1320.59	1272.03	1064.54
DN8	$C_8H_2N_{12}O_{11}$	-1790.0139	0.0278	1253.03	1209.27	993.13
TN1	$C_4H_2N_8O_6$	-1939.4254	0.0293	919.72	868.90	702.26
TN2	$C_4H_2N_{10}O_6$	-2048.8958	0.0308	1143.63	1088.15	901.14
TN3	$C_4H_3N_9O_6$	-1994.7749	0.0302	953.40	896.34	722.76
TN4	$C_4H_4N_{10}O_6$	-2050.1105	0.0307	1023.38	959.12	777.53
TN5	$C_5H_4N_8O_6$	-1978.7337	0.0295	886.84	827.18	659.67
TN6	$C_6H_2N_{12}O_6$	-2234.6024	0.0328	1413.49	1352.40	1131.52
TN7	$C_7H_4N_{10}O_6$	-2164.4533	0.0334	1122.38	1062.07	829.69
TN8	$C_6H_2N_{10}O_7$	-2200.3644	0.0332	1050.46	994.70	771.95
RDX	$C_3H_6N_6O_6$	---	---	---	192.00	79.00[1]
HMX	$C_4H_8N_8O_8$	---	---	---	---	102.41[1]

Table. S2 48 substituted 1,2,3-triazole derivatives, their predicted values of density ρ, oxygen balance OB(based on CO_2), detonation energy Q, detonation velocity D, detonation pressure P, and ΔV_{Truazl}, predicted values of impact sensitivities. h50, compared with HMX, CL-20 and RDX.

No.	ρ (g/cm^3)	OB (%)	Q (kcal/g)	D (km/s)	P (GPa)	ΔV_{Truazl} (cm^3)	h50 (cm)
A1	1.73	-72.73	1.13	6.69	19.34	240.99	103.94
A2	1.74	-63.72	1.20	6.66	19.26	272.96	58.47
A3	1.71	-71.36	1.16	6.80	19.92	253.64	105.69
A4	1.75	-70.18	1.18	7.02	21.46	263.03	107.24
A5	1.67	-90.57	1.10	6.64	18.64	205.22	268.83
A6	1.74	-74.82	1.18	6.20	16.71	236.80	64.78
A7	1.71	-96.97	1.06	6.27	16.92	170.35	221.75
A8	1.75	-72.18	1.09	7.31	23.29	223.29	78.19
H1	1.67	-70.18	1.15	8.58	31.20	257.07	107.24
H2	1.68	-62.50	1.24	9.08	35.03	289.46	64.31
H3	1.69	-69.14	1.11	8.61	31.59	248.74	108.61
H4	1.67	-68.22	1.15	8.71	32.22	262.75	109.84
H5	1.64	-85.95	1.07	7.96	26.53	210.70	246.10
H6	1.68	-72.73	1.20	8.95	34.11	249.81	69.42
H7	1.67	-92.52	1.06	8.00	27.15	183.22	210.29
H8	1.71	-70.27	1.12	8.51	31.13	239.36	82.44
N1	1.85	-18.60	1.62	8.84	35.28	469.50	92.37
N2	1.85	-16.78	1.70	9.03	36.83	491.45	59.30
N3	1.95	-20.51	1.67	9.34	40.56	481.57	94.17
N4	1.86	-22.22	1.58	8.95	36.26	457.03	95.81
N5	1.84	-35.29	1.55	8.60	33.24	424.80	194.75
N6	1.85	-33.14	1.60	8.63	33.58	427.35	64.35
N7	1.86	-49.38	1.46	8.29	31.10	365.59	175.23
N8	1.88	-29.45	1.51	8.60	33.69	415.26	75.02
NA1	1.82	-22.22	1.57	8.78	34.41	452.87	95.81

NA2	1.85	-20.25	1.59	8.97	36.28	459.65	63.95
NA3	1.82	-23.76	1.50	8.86	35.39	434.20	97.31
NA4	1.82	-25.16	1.53	8.84	34.84	441.08	98.70
NA5	1.77	-37.09	1.48	8.40	31.01	406.25	187.26
NA6	1.82	-34.78	1.52	8.51	32.36	404.91	68.20
NA7	1.79	-49.72	1.41	8.12	29.16	355.61	170.98
NA8	1.85	-31.46	1.46	8.55	32.96	401.12	78.64
DN1	1.87	-12.83	1.86	9.21	38.45	548.60	43.11
DN2	1.92	-11.94	1.94	9.59	42.32	567.17	33.17
DN3	1.90	-14.40	1.78	9.25	39.16	523.25	45.01
DN4	1.88	-15.84	1.72	9.29	39.35	507.02	46.83
DN5	1.88	-24.74	1.80	9.10	37.72	513.52	74.77
DN6	1.89	-24.67	1.80	9.05	37.39	499.52	37.68
DN7	1.88	-36.36	1.70	8.74	34.74	455.70	77.46
DN8	1.85	-21.72	1.73	8.95	36.60	491.75	41.58
TN1	1.92	3.43	1.80	9.63	42.80	565.65	20.51
TN2	1.92	3.24	1.79	9.64	42.77	561.41	17.28
TN3	1.93	1.66	1.76	9.65	43.02	554.07	21.73
TN4	1.93	0.00	1.75	9.69	43.43	547.62	22.95
TN5	1.89	-6.67	1.74	9.27	39.26	535.52	32.71
TN6	1.91	-8.79	1.72	9.21	38.91	514.73	20.44
TN7	1.90	-18.05	1.65	9.01	37.17	482.88	36.52
TN8	1.92	-5.99	1.69	9.22	39.17	514.26	21.88
RDX	1.80	-21.62	1.50	8.75	34.70	465.00[2] 475.00[2] 480.00[2]	37.5
HMX	1.90	-21.62	1.498	9.10	39.30	450.00[2]	37.0
CL-20	2.04	-10.93	1.36	9.46	46.7	372.39	13.00

6. References

1. J. S. P. Politzer; Murray, *Central European Journal of Energetic Materials*, 2011, 209-220.
2. M. K. Kamalvand, M. H.; Jafari, M., *Propellants, Explosives, Pyrotechnics*, 2015, **40**, 551-557.