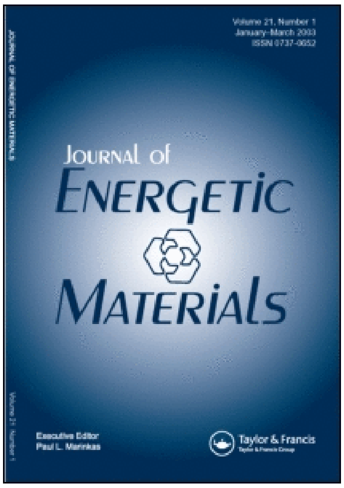

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Properties and impact sensitiveness of cyclic nitramine explosives containing nitroguanidine groups

Ian J. Dagley[a]; Michael Kony[a]; Gene Walker[b]
[a] Aeronautical and Maritime Research Laboratory (AMRL)-DSTO, Melbourne, Australia [b] Department of Chemistry, Australian Defence Force Academy, Canberra, Australia

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PROPERTIES AND IMPACT SENSITIVENESS OF CYCLIC NITRAMINE EXPLOSIVES CONTAINING NITROGUANIDINE GROUPS

Ian J. Dagley, Michael Kony

Aeronautical and Maritime Research Laboratory (AMRL)-DSTO, P.O. Box 4331,

Melbourne 3001, Australia

and

Gene Walker

Department of Chemistry, Australian Defence Force Academy, Northcott Drive,

Canberra 2600, Australia

ABSTRACT

Some properties of hexahydro-5-nitro-2-(nitroimino)-1,3,5-triazine **1**, octahydro-1,3-dinitro-5-(nitroimino)imidazo[4,5-*d*]imidazole **2**, dodecahydro-4,8-dinitro-2,6-bis(nitroimino)diimidazo[4,5-*b*:4',5'-*e*]pyrazine **3** and octahydro-2,5-bis(nitroimino)imidazo[4,5-*d*]imidazole **4** have been determined. They have acceptable thermal and hydrolytic stability. Compound **3** has greater friction sensitiveness than RDX, however the others are much less sensitive. Rotter impact

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test data indicate that the cyclic nitroguanidines **1-4** are more easy to initiate than RDX; however, the extent of propagation is less and depends on oxygen balance. Consideration of Hazard Index and Average Powder Explosiveness data indicates that US drop-weight tests, which measure a combination of ease of initiation and extent of propagation, would rank the cyclic nitroguanidines and reference explosives in the following order of increasing impact sensitiveness: NQ < **4** < NTO < **1** < RDX, **3** < **2**.

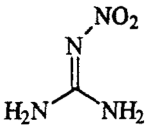
INTRODUCTION

It is difficult to formulate explosive compositions for use in ordnance that meet insensitive munitions criteria because of the limited performance/vulnerability trade-offs that can be achieved using existing explosive compounds. RDX provides the required performance but is sensitive, so compositions based on this explosive are often unacceptably vulnerable to hazardous stimuli. A number of less sensitive explosives, including nitroguanidine (NQ), have been proposed as alternatives; however, when these have been used, performance has been compromised.

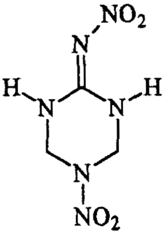
One possible approach to designing powerful and less sensitive cyclic nitramines is to combine some of the structural features of both RDX and NQ. Incorporation of a nitroguanidine group introduces the possibility of inter- and intra-molecular hydrogen bonding networks in the crystal structure[1-3].

Hydrogen bonding networks between amines and nitro groups contribute to the high thermal stability and density of some explosives[4,5] and may reduce sensitivity as breaking these bonds can absorb and help delocalise energy.[6,7]

Some explosive properties of one hybrid of RDX and NQ, hexahydro-5-nitro-2-(nitroimino)-1,3,5-triazine **1**, have been determined and it has been patented[8] as an insensitive explosive. Recently we have prepared two new cyclic nitramines of this type (**2** and **3**) and a related compound **4** containing a nitroguanidine group in each of two fused rings.[9,10] This paper describes an initial assessment of the properties of the new cyclic nitramine explosives **2-4**. The relative influence of nitroguanidine, secondary nitramine and dinitrourea groups in determining the impact sensitiveness of cyclic nitramines is also examined.



NQ



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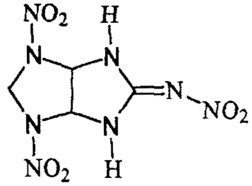

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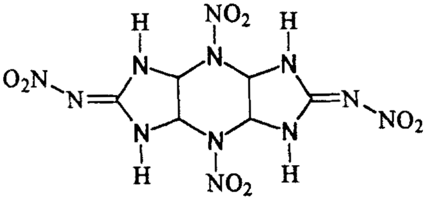

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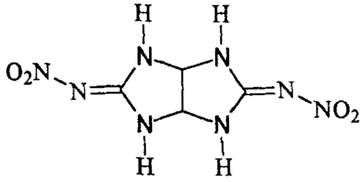


4

EXPERIMENTAL

Materials

Methods described in the literature were used to prepare and purify compounds **1**[8], **2**[9], **3**[9], and **4**[10] except compound **1** was recrystallized from acetone. Compounds **1a-4a**, **5** and **6** are discussed later in this paper and were prepared by literature methods[11-16]; the dinitroureas **1a-4a**, and **5** were purified by recrystallization from dry acetone/dry benzene.

Sensitivity to Atmospheric Moisture

The sensitivity of compound **2** to atmospheric moisture was assessed by exposing 160 mg samples to relative humidities of 95%, 79%, 58% and 31% at 20°C. These humidities were maintained in test chambers using saturated aqueous solutions of dibasic sodium phosphate, ammonium chloride, sodium bromide and calcium chloride in contact with an excess of the solute.[17] The exposed samples of the explosive were spread evenly over approximately 6 cm^2 on flat glass surfaces and were visually examined during an exposure period of 10 months. The ^1H NMR spectra of the samples were then recorded and an internal standard (4-methoxybenzonitrile) was used to determine the percentage of unchanged compound in each of the samples.

Vacuum Thermal Stability Test

Thermal stability of compounds **1-4** was assessed using a vacuum thermal stability test[18] conducted on duplicate 1 g samples heated at 120°C for 40 h. The average volume of evolved gas was used to estimate the volume of gas that would be evolved had the test been performed under standard conditions using duplicate 5 g samples.

BAM Friction Test

Sensitiveness to friction was determined using the BAM Friction Apparatus. The apparatus and standard test procedure followed are described in a UN Manual.[19] The result quoted is the Limiting Load defined as the lowest load at which at least one ignition is observed in a series of six trials.

Rotter Impact Tests

A Rotter Apparatus[20] fitted with a 5 kg drop-weight was used to determine the Figure of Insensitiveness (F of I), average powder explosiveness (X*) and Hazard Index (HI) of compounds by standard methods[21] but using 27 mg samples.

Impact sensitiveness was assessed from 25 drops in a test carried out in accordance with the Bruceton staircase procedure. The criterion for ignition was a 1 mL gas evolution or 0.5 mL accompanied by smoke or evidence of burning in the sample. The F of I value quoted is derived from the drop height $h_{50\%}$ for a 50% ignition probability. It is relative to RDX Grade F = 80 and rounded to the nearest 5 units. The average gas volume for positive results is also quoted.

Powder explosiveness (X) was usually assessed by conducting 20 drops at typically each of 0.75, 1.0, 1.5 and 2.0 times ($h_{50\%}$); however, to limit damage to the anvils it was occasionally necessary to perform only 10 drops at the greater heights. The number of ignitions and their average gas evolution (to 0.1 mL) were determined and, together with published data for the method standard (lead azide RD1343), were used in the calculations of X at each level, with the theoretical total gas volume evolved for complete ignition calculated using the NASA-Lewis code[22]. X is calculated from the mass (m) of explosive under test, average gas volume evolved per ignition (V_g) and the theoretical total gas volume evolved (V); the subscript s refers to data for the lead azide standard.

$$X = V_g m_s V_s / V_{gs} m V$$

X* is the average of all X values for the test levels greater than or equal to $h_{50\%}$. HI is the maximum value of X / height (m) for all the test levels.

Oxygen Balance

Oxygen balance was calculated using the expression:

$$OB_{100} = 100(2n_O - n_H - 2n_C - 2n_{COO})/MW$$

where n_O, n_H, and n_C represent the number of atoms of oxygen, hydrogen and carbon, respectively, in the molecule and n_{COO} represents the number of carboxyl groups. MW is the molecular weight of the compound.

RESULTS AND DISCUSSION

Properties

The compounds **2-4** can be purified by crystallisation from dilute nitric acid heated to 70°C. Heating above this temperature caused hydrolysis; similar behaviour under these conditions has been previously observed with other substituted nitroguanidines.[23] The crystals of **4** obtained by this method are the most suitable for explosive processing and those of **3** the least satisfactory since they are fine needles with a large length to width ratio.

The new cyclic nitroguanidines exhibit good stability when exposed to atmospheric moisture. One of the compounds, the bicyclic trinitro compound **2**, was tested at length by exposing samples of the compound to a range of relative humidities for 10 months then examining them in solution using ^1H NMR spectroscopy. Within the limits of the accuracy of this technique, no decomposition of compound 2 was detected.

Some physical and explosive properties of the cyclic nitroguanidines **2-4** determined in this study are listed in Table 1 where they are compared to data for RDX and the insensitive explosives **1**, NQ, and 3-nitro-1,2,4-triazol-5-one (NTO).

The densities of the new cyclic nitroguanidines **2-4** are very similar, ranging from 1.83-1.85 Mg/m^3, and significantly higher than the values reported for the other cyclic nitroguanidine **1** (1.75 Mg/m^3), NQ (1.771 Mg/m^3) and RDX (1.806 Mg/m^3). Using these measured densities and heats of formation (estimated for compounds **2-4** from AM1 calculations[27]), the detonation pressures predicted by the method of Kamlet and Jacobs[29] were determined. The bicyclic bis(nitroguanidine) **4** is predicted to have a detonation pressure slightly higher than those of NQ and the monocyclic nitroguanidine **1**. The compounds **2** and **3**, that contain nitrimine and secondary nitramine groups, are predicted to have higher

TABLE 1

Some Physical and Explosive Properties of Nitroguanidines, NTO and RDX

	1	2	3	4	NQ	NTO	RDX
Density (Mg/m^3)	1.75[8]	1.85[a]	1.83[a]	1.840[24]	1.771[1]	1.93[25]	1.806[26]
Heat of Formation (kcal/mol)	16.3[8]	51.4[b]	97.4[b]	53.0[b]	-22.1[28]	-14.30[25]	14.71[28]
Calculated[29] Detonation Pressure (Gpa)	28.6	33.8	32.3	29.7	27.0	31.5	34.5
Decomposition Point (°C)	207[8]	207[9]	299[9]	326[10]	250[30]	258[31]	216[31]
Vacuum Thermal Stability Test (mL/5g, 120°C, 40h)	3.8[c]	5.5[c]	0.3[c]	0.0[c]	0.1[30]	0.0[31]	0.2-0.6[31]
BAM Friction Test (N)	>360	>360	24	>360	>360	>360	108

[a] Densities determined in this study using a gas pycnometer at 20 ± 2°C.

[b] Estimated from AM1 calculations using the method in reference 27.

[c] These values were estimated from the results of duplicate tests using 1g of the test explosive.

detonation performance, intermediate between that of the insensitive explosive, NTO, and RDX.

The bicyclic nitramine **2** undergoes explosive decomposition at 207°C when heated in open capillary tubes using a melting point apparatus, and a similar decomposition point has been reported for the monocyclic nitramine **1** and RDX. The other cyclic nitroguanidines undergo decomposition at much higher temperatures; the tricyclic compound **3** decomposes at 299°C while the decomposition point of bis(nitroguanidine) **4**, 326°C, is unusually high for an explosive. This is attributed to the large number of intramolecular hydrogen bonds (eight to any one molecule) revealed in the crystal structure[24] of this compound. Thermograms obtained by differential scanning calorimetry show that the cyclic nitroguanidines **1-4** all decompose without melting.

The thermal stability of the cyclic nitroguanidines **1-4** was assessed using a vacuum thermal stability test conducted at 120°C for 40 h and the results are compared in Table 1 with some literature values for NQ, NTO and RDX. All these results are quoted as the volume of gas evolved (in mL) for 5 g of the test sample (for the cyclic nitroguanidines they represent estimates from duplicate tests conducted using 1 g of the test sample). The two cyclic nitroguanidines, **3** and **4**, with the high melting points have very good thermal stability; compound **4** produced no gas and compound **3** gave a very low evolved gas volume. The thermal stability values estimated for the other cyclic nitroguanidines **1** and **2** are 3.8 and 5.5 mL which represent less than 0.3% decomposition[32]. These compounds, **1** and **2**, are clearly less thermally stable than RDX which registers comparatively low evolved gas volumes (0.2-0.6 mL) under the same test conditions. The US Navy assess the stability of military explosives in a test conducted on 0.2 g of sample at 100°C for 48 h; the result is quoted as the mL of gas evolved per gram of sample. As the limit set for this test is 2.0 mL/g [33], the nitroguanidines **1** and **2** would be expected to pass this test and be deemed to have adequate thermal stability.

The sensitiveness to friction was assessed using the BAM friction test. Three of the cyclic nitroguanidines **1**, **2** and **4** were ranked much less sensitive than RDX (>360 N cf. 108 N). The tricyclic compound **3** was ranked much more sensitive (24 N) and accordingly should be handled with particular care.

Rotter Impact Tests

The Rotter drop-weight apparatus was used to determine three rankings of hazard assessment (F of I, HI and X*) for the cyclic nitroguanidines. The results are compared with data for NQ, NTO and RDX in Table 2.

The Figure of Insensitiveness (F of I) which is typically quoted from the Rotter tests is derived from the height for a 50% probability of initiation ($h_{50\%}$) and is a measure of the ease of initiation of the compound; the evolved gas volume is also noted and is a measure of the extent of propagation.[20] The F of I results show that the cyclic nitramines **1-4** are all more easy to initiate than RDX; the tricyclic compound **3** is the most sensitive (F of I = 20), compounds **2** and **4** have F of I values of 30 and 35, and compound **1** (F of I = 55) is the least sensitive of

TABLE 2

Impact Test Results and OB_{100} Values for Nitroguanidines, NTO and RDX

	1	2	3	4	NQ	NTO	RDX
OB_{100}	-2.10	-0.76	-1.06	-2.61	-1.92	0	0
Rotter Apparatus Tests							
F of I	55	30	20	35	100-105[30]	80-90[31]	80[34,a]
Gas Evolution (mL)	1.6	9	4	1.7	1-2[30]	3-5[31]	18[34,a]
Hazard Index (HI, m^{-1})	64.2	184.6	83	13.9	3.8[34]	14.9[34,b]	88.5[34,a]
Average Powder Explosiveness (X*)	39.4	68.9	58.2	11.7	3.8[34]	25.3[34,b]	51[34a]
ERL-LASL Type 12 (cm)					>320[35]	>280[25]	23-28[35]
Bureau of Mines Apparatus, 2 kg (cm)	80[8]				47[36]		32-42[37]

[a] Grade A.

[b] Particle size less than 850 μm.

these cyclic nitroguanidines. Despite their relative ease of initiation, the cyclic nitroguanidines (particularly 1 and 4) give comparatively low evolved gas volumes (1.6-9 mL cf. 18 mL for RDX) indicating that they propagate poorly and therefore have low explosiveness.

Spear and Montelli[34] found that some explosives ranked as relatively insensitive by US drop-weight tests give unexpectedly low F of I values (ie indicating high impact sensitiveness). Examples are NTO and NQ which are ranked much less sensitive than RDX by the ERL test, while only slightly less sensitive by the Rotter Test (Table 2). The insensitive explosive 1 is a further example of this type, being ranked more sensitive than RDX from the F of I values and much less sensitive by the Bureau of Mines test. These differences arise primarily because the F of I values are only a measure of the ease of initiation. The results of the US tests are influenced by both ease of initiation and the extent of propagation, since the criteria for determining a positive result (a go) requires a greater extent of propagation in the sample under test.

Rankings that correspond more closely to those derived from US impact tests can be obtained by using the Rotter apparatus to determine X* and HI which are measures of impact explosiveness. X* and HI values for compounds 1-4 have been determined and are compared in Table 2 to published values for the reference explosives. The results suggest that in US impact tests the sensitiveness of compound 4 would be ranked greater than that of NQ and similar to, though possibly slightly less than, that of NTO; compound 1 would be ranked more sensitive than NTO and less sensitive than RDX; the sensitiveness of compound **3** would be ranked similar to that of RDX; and compound **2** would be ranked considerably more sensitive than RDX.

Consideration of both the Rotter impact sensitiveness (F of I) and powder explosiveness (X*, HI) tests on the new explosives provides a more complete indication of the relative risk of these cyclic nitroguanidine explosives. Unlike the accepted insensitive explosives NQ and NTO, the cyclic nitroguanidines **2-4** all have F of I values much lower than that of RDX; this comparative ease of ignition cautions against fully accepting the relative rankings suggested from the HI and X* data. If further development of any of these explosives were to be pursued it would be necessary to perform charge hazard tests to obtain a more accurate assessment of the risks associated with their use.

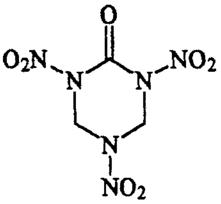
Effect of Type of Nitramine Trigger Linkage on Ease of Impact Initiation

Studies on impact sensitiveness data obtained using the NSWC-ERL drop-weight test showed that the observed ranking of explosives is determined by both the nature of the 'trigger linkage' (defined as the weakest bond) and the oxygen balance of the explosive.[38,39] More recently Wilson and coworkers[40] have suggested that oxygen balance is probably more relevant to propagation than initiation. If this is the case the Rotter impact F of I values should be much more strongly influenced by the nature of the trigger linkage than results from US tests. The results for the cyclic nitroguanidines in Table 2 and those obtained for structurally related compounds **1a-4a** (see below), which contain dinitrourea groups instead of nitroguanidine groups, indicate that this is the case and allow the relative sensitiveness of different nitramine groups in various cyclic structures to be assessed.

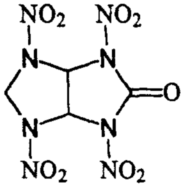
The only energetic group in compound **4** is a nitroguanidine forming part of a fused 5-membered ring and this compound has an F of I of 35. Compounds **2** and **3** also contain secondary nitramine groups and have less negative oxygen balances (-0.76 and -1.06 cf. -2.61) but give similar F of I values (30 and 20) suggesting that in these three compounds it is the nitroguanidine in the fused 5-membered rings that is the common trigger linkage determining the ease of initiation. For this series of compounds the evolved gas volume increases as the oxygen balance becomes less negative, indicating that it is the oxygen balance that is determining the extent of propagation.

F of I values for the analogues **2a**-**4a**, where these nitroguanidine groups have been replaced by dinitrourea groups, have been determined and are all 20 even though the oxygen balance varies over a wide range (+1.95 to +3.10). The common trigger linkage in this series is contained in the dinitrourea group in the fused 5-membered rings and, from comparison with the results obtained for compounds **2**-**4**, this generally confers slightly greater sensitiveness to ignition than a nitroguanidine in a fused 5-membered ring. It can also be concluded that secondary nitramine groups in **2**-**4** and **2a**-**4a** are less sensitive trigger linkages than the nitroguanidine groups or the dinitrourea groups since they are not the determinants of the F of I values observed for compounds.

Replacement of a nitroguanidine group with a dinitrourea group markedly increases the oxygen balance and the similar high evolved gas volumes recorded for compounds **1a**-**4a**, which have positive oxygen balances, indicate a common high extent of propagation after ignition.

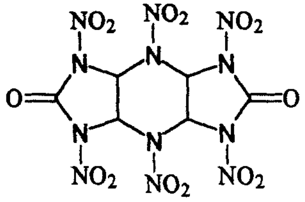


1a

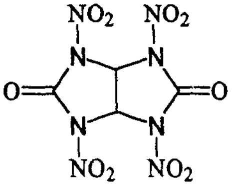


2a

F of I	60	20
Evolved gas (mL)	14	14
OB_{100}	+1.69	+1.95



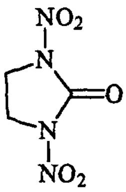
3a

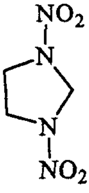


4a

F of I	20	20
Evolved gas (mL)	15	13
OB_{100}	+2.56	+3.10

The incorporation of nitroguanidine or dinitrourea groups into a 6-membered ring, rather than a fused 5-membered ring, appears to reduce their sensitiveness with **1** and **1a** having higher F of I values than **2**-**4** and **2a**-**4a**. The F of I values of **1** and **1a** are lower than that of RDX, and the F of I of compound **5** is lower than that of compound **6**, indicating that, in cyclic nitramines, secondary nitramine groups bonded to two methylenes are comparatively insensitive nitramine trigger linkages. In general, the impact sensitiveness of cyclic compounds containing only secondary nitramines is very dependent on the molecular conformation in the crystalline state, and a range of impact sensitivities has been observed for different polymorphic forms of certain nitramines.[41,42]





	5	**6**
F of I	45	130
Evolved gas (mL)	12	14
OB_{100}	0	-2.47

CONCLUSIONS

The cyclic nitramine explosives **1-4** exhibit good stability when exposed to atmospheric moisture. Results from a vacuum thermal stability test indicate that the thermal stability of the nitramines **1** and **2**, which have the lowest decomposition points, is acceptable; thermal stability increases with decomposition point and the bis(nitroguanidine) **4** which decomposes at 326°C has very good thermal stability. Three of the cyclic nitramines are much less sensitive to friction than RDX, however the tricyclic nitramine **3** is much more sensitive and accordingly should be handled with particular care.

Data from Rotter impact tests indicate that the cyclic nitroguanidines, and particularly **2-4** where this group is in fused 5-membered rings, are more easy to initiate than RDX; however, the extent of propagation is less and dependent on oxygen balance. Consideration of Hazard Index and Average Powder Explosiveness data lead to the conclusion that US drop-weight tests, which measure a combination of ease of initiation and extent of propagation, would rank the cyclic nitroguanidines and reference explosives in the following order of increasing impact sensitiveness: NQ < **4** < NTO < **1** < RDX, **3** < **2**. The F of I values determined using the Rotter Apparatus are more strongly influenced by the ease of initiation than results obtained using US drop-weight tests and so F of I rankings better reflect the sensitiveness of the trigger linkage in the compounds. F of I results for the nitroguanidines **1-4** and their analogues containing dinitrourea groups **1a-4a** indicate that nitroguanidine and dinitrourea groups confer similar sensitiveness and they are more sensitive trigger linkages than secondary nitramines.

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REFERENCES

1. J.H. Bryden, L.A. Burkardt, E.W. Hughes and J. Donohue, Acta Cryst. *9*, 573 (1956).
2. C.S. Choi, Acta Cryst. *B37*, 1955 (1981).
3. S. Nordenson, Acta Cryst. *B37*, 1774 (1981).
4. H.H Cady and A.C. Larson, Acta Cryst. *18*, 485 (1965).
5. D.A. Cichra, J.R. Holden and C. Dickinson, "Estimation of the "Normal" Densities of Explosives from Empirical Atomic Volumes," NSWC TR 79-273, Naval Surface Warfare Center, Silver Spring, MD, 1980.
6. J.W. Rogers, Jr., H.C. Peebles, R.R. Rye, J.E.Houston and J.S. Binkley, J. Phys. Chem. *80*, 4513 (1984).
7. J.P. Ritchie, D.T. Cromer, R.R. Ryan, R.F. Stewart and H.J. Wasserman, in "Proceedings Eighth Symposium (International) on Detonation," NSWC MP 86-194, Naval Surface Warfare Center, Silver Spring, MD, 1986, p. 839.
8. D. Huang and R.R. Rindone, US Patent 4,937,340 (1990).
9. I.J. Dagley and J.L. Flippen-Anderson, Aust. J. Chem., in press.

10. M. Kony and I.J. Dagley, Heterocycles, *38*, 595 (1994).

11. H. Bergman, H. Ostmark and A. Langlet, in "Paper Summaries Tenth International Detonation Symposium," NSWC DD/MP-92/456, Naval Surface Warfare Center, Silver Spring, MD, 1993, p. 302.

12. Li, W., Hua, G., and Chen, M., in "Proceedings of the Symposium on Pyrotechnics and Explosives," China Academic Publishers, Beijing, 1987, p. 187.

13. M. Vedachalam, V.T. Ramakrishnan, J.H. Boyer, I.J. Dagley, K.A. Nelson, H.G. Adolph, R. Gilardi, C. George and J.L. Flippen-Anderson, J. Org. Chem. *56*, 3413 (1991).

14. Z. Peng and D. Wan, Acta Armamentarii *3*, 23 (1980).

15. T. Urbanski, "Chemistry and Technology of Explosives," Vol. 3, Pergamon, NY, 1967; p 19.

16. L. Goodman, J. Am. Chem. Soc. *75*, 3019 (1953).

17. M. Windholz, Ed., "The Merck Index," 10th edn, Merck and Co.,Inc., New Jersey, 1983, p. MISC-103.

18. "Methods of Test for Explosives and Related Materials", DEF (AUST) 5604, Method 3/80, Australian Defence Standard, Department of Defence, Melbourne, 1980.

19. "Recommendations on the Transport of Dangerous Goods, Tests and Criteria", second edition, ST/SG/AC.10/11/Rev.1, United Nations, New York, 1990.

20. H.N. Mortlock and J. Wilby, Explosivstoffe *14*, 49, 1966.

21. Royal Armament Research and Development Establishment, "Sensitiveness Collaboration Comittee, Manual of Tests," Test 1A and 1D, 1988.

22. S. Gordon and B. McBride, "Computer Program for the Calculation of Complex Chemical Equlibrium Compositions, Rocket Performance, Incident and Reflected Shocks, and Chapman-Jouguet Detonations," NASA Report SP-273, National Aeronautics and Space Administration, 1976.

23. A.F. McKay, Chem. Rev., *51*, 301 (1952).

24. J.L. Flippen-Anderson, M. Kony and I.J. Dagley, Acta Cryst. *C50*, 974 (1994).

25. K-Y. Lee, L.B. Chapman and M.D. Coburn, J. Energetic Materials, *5*, 27 (1987).

26. C.S. Choi and E. Prince, Acta Cryst. *B28*, 2857 (1972).

27. J-L.G. De Paz and J. Ciller, Propellants Explos. Pyrotech. *18*, 33 (1993).

28. D.R. Stull, E.F. Westrum, Jr. and G.C. Sinke, "The Chemical Thermodynamics of Organic Compounds," Wiley, New York, 1969.

29. M.J. Kamlet and S.J. Jacobs, J. Phys. Chem. *48*, 23 (1968).

30. R.J. Spear, L.D. Redman and R. Barrow, "Conversion of Propellant Grade Picrite to Spherical Nitroguanidine, an Insensitive Filler for Melt-Cast TNT Formulations," MRL-TR-91-33, Materials Research Lab., Maribyrnong, Australia, 1991.

31. R.J. Spear, C.N. Louey and M.G. Wolfson, "A Preliminary Assessment of 3-Nitro-1,2,4-triazol-5-one (NTO) as an Insensitive High Explosive," MRL-TR-89-18, Materials Research Lab., Maribyrnong, Australia, 1989.

32. B.M. Dobratz and P.C. Crawford, "LLNL Explosives Handbook. Properties of Chemical Explosives and Explosive Simulants," LLNL Report UCRL-52997, Lawrence Livermore National Laboratory, University of California, Livermore, CA, 1985.

33. H.T. Simmons, Sr., "Vacuum Thermal Stability Testing for Explosives," NOLTR 70-142, Naval Ordnance Laboratory, Silver Spring, MD, 1970.

34. R.J. Spear and L. Montelli, "An Investigation of the Material Hazard of Some Insensitive High Explosives," MRL-TR-91-22, Materials Research Lab., Maribyrnong, Australia, 1991.

35. T.R. Gibbs and A Popolato, "LASL Explosive Property Data," University of California Press, Berkeley, 1980, pp. 448-449.

36. "Encyclopedia of Explosives and Related Items," Vol. 6, B.T. Fedoroff and O.E. Sheffield, eds., Picatinny Arsenal, Dover, NJ, 1974, p. G 155.

37. "Encyclopedia of Explosives and Related Items," Vol. 3, B.T. Fedoroff and O.E. Sheffield, eds., Picatinny Arsenal, Dover, NJ, 1966, p. C 618.

38. M. Kamlet, in "Proceedings Sixth Symposium (International) on Detonation," ONR Report ACR 221, 1976, p. 312.

39. M.J. Kamlet and H.G. Adolph, Propellants Explos. *4*, 30 (1979).

40. D.E. Bliss, S.L. Christian and W.S.Wilson, J. Energetic Materials, *9*, 319 (1991).

41. Y. Kohno, K. Maekawa, T. Tsuchioka, T. Hasizume aand A. Imamura, Combust. Flame, *96*, 343 (1994).

42. R.B. Wardle, J.C. Hinshaw, P. Braithwaite, G. Johnstone, R. Jones, K. Poush, V.A. Lyon and S. Collignon, In "Proceedings International Symposium on Energetic Materials Technology," American Defence Preparedness Association, March, 1994, p. 259.