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A Less Sensitive Explosive (U)



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Kien-Yin Lee
Michael D. Coburn

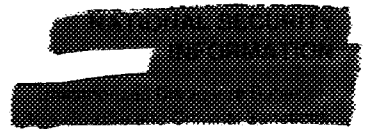
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Los Alamos National Laboratory.
LESS SENSITIVE EXPLOSIVE (U). Kien-Yin Lee
and Michael D. Coburn. Feb. 1985. 9 p.

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3-NITRO-1,2,4-TRIAZOL-5-ONE,
A LESS SENSITIVE EXPLOSIVE ()

by

Kien-Yin Lee and Michael D. Coburn

ABSTRACT ()

A new, less sensitive explosive has been prepared and subjected to preliminary characterization tests. The compound, 3-nitro-1,2,4-triazol-5-one (NTO) has a crystal density of 1.93 g/cm³ and calculated detonation velocity and pressure equivalent to those of RDX. Results from initial small-scale sensitivity tests indicate that NTO is less sensitive than RDX and HMX in all respects. A 4.13-cm-diam, unconfined plate-dent test at 92% of crystal density gave the detonation pressure predicted for NTO by the BKW calculation. Finally, NTO can be prepared in high yield from inexpensive starting materials.

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I. INTRODUCTION

In the past, the common explosives RDX, HMX, and TNT were considered adequate for all weapons applications. Due to the many catastrophic explosions resulting from unintentional initiation of munitions by either impact or shock aboard ships, aircraft carriers, and ammunition trains, these explosives have become less attractive.

Triaminotrinitrobenzene (TATB) is noted for its insensitivity and is currently employed for insensitive high explosive (IHE) applications in nuclear weapons. Unfortunately, TATB does not produce sufficient performance to replace HMX in some applications. The need, therefore, exists for explosives that are powerful yet resistant to accidental and sympathetic initiation.

This report describes a new candidate energetic explosive, 3-nitro-1,2,4-triazol-5-one (NTO). The chemical, physical, and explosive properties of NTO



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are reported together with the procedure for its preparation. Results from initial performance are also included.

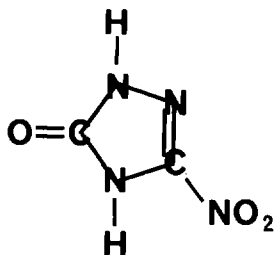
II. PROPERTIES OF NTO

NTO is a white crystalline compound, moderately soluble in water to give yellow solutions. The chemical properties of NTO are given in Table I. The CO-balanced explosive was found to have a crystal density of 1.93 g/cm^3 by x-ray crystallography.

Results of small-scale screening tests of NTO are tabulated in Table II. For comparison, data from RDX are also listed. It can be seen that NTO is less sensitive and more stable than RDX in all the tests.

TABLE I
CHEMICAL PROPERTIES OF NTO

Structural Formula:



$\text{C}_2\text{H}_2\text{N}_4\text{O}_3$

Molecular Weight: 130

Acidity: $\text{pka} = 3.67^1$

NMR Spectrum:

^1H NMR ($\text{DMSO}-d_6$): 12.7 ppm (>NH)

^{13}C NMR ($\text{DMSO}-d_6$): 148.0 ppm (>C- NO_2), 154.4 ppm (>C=O)

Elemental Analysis:

Calc: C, 18.46; H, 1.54; N, 43.08

Found: C, 18.78; H, 1.92; N, 43.47

III. PREPARATION OF NTO

NTO is prepared by a modification of the procedure reported in the Russian literature.² It consists of preparing the intermediate 1,2,4-triazol-5-one (TO) followed by nitration with 90% nitric acid.

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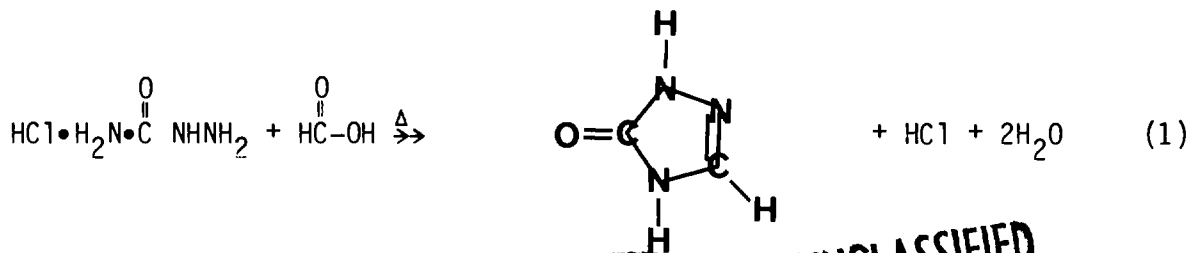
TABLE II

PHYSICAL AND EXPLOSIVE PROPERTIES

	NTO	RDX
Crystal Density (g/cm ³)	1.93	1.806
DTA Exotherm (°C)	>236	210
Heat of Formation (kcal/mole)	-14.30	+14.71
Henkin Critical Temp (°C) (0.64-mm size)	237	219.6
Impact Sensitivity (cm)		
Type 12	291	22
Type 12B	293	41
Spark Sensitivity (J)		
3 mil	0.91	0.22
10 mil	3.40	0.55
Vacuum Stability		
(ml/g/48 h at 100°C)	0.2	
(ml/g/48 h at 120°C)	0.3	0.12-0.9

A. Preparation of T0 (2 mol)

T0 is obtained by condensing semicarbazide hydrochloride (SC) and formic acid, Eq. 1. Thus, to 230 ml of formic acid (85%) in a 1-L round-bottom flask is added 223 g SC slowly with stirring. The mixture is then heated under reflux for 4 h. The reaction mixture is concentrated by distillation until crystallization begins to occur (about 2 h). Water is added and distillation is continued until the mixture is near dryness. After cooling in the refrigerator overnight, the solid is collected by filtration, washed with a small amount of ice water to remove excess formic acid and dried in an oven at 100°C overnight. The yield of T0 is 96-98%. The purity of T0 is determined by NMR spectroscopy.



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B. Preparation of NTO

In a 2-L beaker containing a mixture of 222.5 ml of 90% nitric acid and 118.8 ml of water is gradually added 112.45 g (1.32 mol) of TO with the temperature controlled at about 40°C. The mixture is heated to boiling until brown fumes start to evolve, then it is removed from the heat and set aside. At this point, the reaction becomes exothermic and large quantities of brown fumes are evolved. NTO is precipitated as white solid that is collected by filtration, washed with ice water to remove excess nitric acid, and dried in an oven at 100°C for 24 h. The yield is 142.11 g (83%).

IV. PERFORMANCE TESTS OF NTO

We have evaluated the performance of NTO by conducting unconfined plate-dent tests at various charge diameters and pressed densities. We recrystallized NTO from water and its purity was analyzed by both ^1H NMR and ^{13}C NMR spectroscopy before pressing. Results of these tests are compiled in Table III. For comparison, data from RDX and TATB are also listed.

V. PREPARATION AND PROPERTIES OF AMINE SALTS OF NTO

NTO is relatively acidic ($\text{pK}_a = 3.67$), and forms stable salts with mono- or bivalent metals. The potassium, sodium, and lithium salts of NTO have been reported.² We have studied the formation of amine salts with NTO and have prepared, for the first time, the ammonium and ethylenediaminium salts of NTO (ANTO and ENT0, respectively). Both salts were prepared by the addition of the corresponding base to a methanol solution of NTO, followed by evaporation of the solvent in the case of ANTO or by filtration in the case of ENT0. If NTO is treated with ammonium hydroxide solution, the salt thus obtained is a hydrate. We have characterized and evaluated both salts and found they could be explosives of interest. Some of the physiochemical properties of ANTO and ENT0 are reported in Table IV. In earlier studies,³ we have demonstrated that both ammonium and ethylenediaminium salts of 5-nitrotetrazole form low-melting eutectics with ammonium nitrate (AN) and the eutectics behave like a monomolecular explosive. We have investigated the formation of the eutectics ANTO/AN and ENT0/AN. Preliminary results indicate that both salts form eutectics with AN and the eutectic temperatures of both eutectics are below 100°C.

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TABLE III
 DETONATION PROPERTIES

Explosive	Charge Density (g/cm ³)	Charge Diameter (cm)	P _{CJ} (kbar)	
			Measured	BKW
NTO	1.93 (100% TMD)	--	--	349
	1.781 (92.2% TMD)	4.13	278	284
	1.782 (92.3% TMD)	2.54	240	284
	1.855 (96.1% TMD)	2.54	Failed	316
	1.759 (91.1% TMD)	1.27	250	271
RDX	1.767 (97.8% TMD)	4.12	338	348
TATB	1.87 (96.5%)	4.12	277	313

TABLE IV
 PHYSIOCHEMICAL PROPERTIES OF
 AMINE SALTS OF NTO

	ANTO	ENTO
Structural Formula:	C ₂ H ₅ N ₅ O ₃ •H ₂ O	C ₆ H ₁₂ N ₁₀ O ₆
Molecular Weight:	165	320
Melting Point (°C):	190	221
NMR Spectrum:		
¹³ C NMR (DMSO-d ₆):	164.6 ppm (>C=O)	164.4 ppm (>C=O)
¹³ C NMR (DMSO-d ₆):	159.4 ppm (>C-NO ₂)	159.1 ppm (>C-NO ₂)
Elemental Analysis:		
Calc:	C, 14.55; H, 4.27; N, 42.42	C, 22.50; H, 3.75; N, 43.75
Found:	C, 15.07; H, 4.11; N, 42.42	C, 23.09; H, 3.78; N, 44.12

VI. CONCLUSIONS AND FUTURE WORK

We have prepared an explosive compound that could be a candidate for future nuclear and conventional weapons applications. Initial small-scale sensitivity tests of NTO indicate that it is much less sensitive than RDX and TNT in all respects, but somewhat more sensitive than TATB. Results from a 4.13-cm-diam unconfined plate-dent test of NTO at 92.2% of crystal density gave the detonation pressure predicted for NTO by the BKW calculation, which is equal to that measured for TATB at 96.5% of crystal density. We are in the process of scaling up the production of NTO so that a full-scale gap test can be performed to determine the shock sensitivity of NTO. NTO can be easily pressed without a

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binder to the desired charge diameters and densities. We are going to study and characterize PBXs of NTO with different binders and plasticizers. We also plan to continue to characterize and evaluate the two eutectics, ANTO/AN and ENTO/AN.

ACKNOWLEDGMENTS

The authors are grateful to T. M. Benziger for his valuable discussions. Appreciation for their technical contribution is also given to J. F. Baytos, M. L. Clancy, H. H. Cady, C. E. Hannaford, J. L. Janney, J. O. Martinez, C. C. Maxwell, R. M. Montoya, M. J. Naranjo, J. P. Ritchie, J. R. Stine, M. M. Stineciper, L. A. Stretz, H. Wasserman, and M. J. Urizar.

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