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ON THE SHOCK INSENSITIVITY OF TRIAMINOGUANIDINIUM AZOTETRAZOLATE (TAGZT)

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ABSTRACT

The high-nitrogen compound triaminoguanidinium azotetrazolate (TAGzT) belongs to a class of C, H and N compounds that are free of oxygen, but retain energetic material properties as a result of their high heat of formation. Its decomposition thus lacks secondary oxidation reactions of carbon and hydrogen. The extremely fast burning rate of TAGzT, as well as its high nitrogen and hydrogen content and low flame temperature makes it potentially useful as a propellant and explosive ingredient. It was found that TAGzT exhibits one of the fastest low pressure burning rates yet measured for an organic compound. Counter initiatively; TAGzT is completely shock insensitive up to diameters of at least 1 5/8", as demonstrated by the LANL large-scale gap test. Comparisons of this with similar high nitrogen compounds are made, and speculation into the mechanism of this behavior is given.

INTRODUCTION

New ingredients are being sought for propellants and explosives that will benefit formulations in varied metrics of performance including an increase in safety, faster burning rate, and reasonable impulse given a reduction in flame temperature, to name a few. Energetic materials that derive their energy from a positive heat of formation rather than oxidation of a carbon backbone have recently attracted attention as gas generants or propellants because a relatively high theoretical specific impulse can be achieved despite the relatively low decomposition temperature. The compound triaminoguanidium azotetrazolate (TAGzT) is particularly interesting because of its high heat of formation and high molar and volumetric gas production. The synthesis and physical properties of TAGzT were first reported by Tremblay, and later by Hiskey et al.[1, 2] The material is a bright yellow, needle-like, crystalline solid having a theoretical maximum density of 1.60 g/cm^3, a decomposition temperature of 195°C, and a heat of formation of +257 kcal/mol [1].

Because TAGzT contains no oxygen, the chemistry that dominates the ignition and combustion is more centered on reactions in the condensed phase rather than oxidation chemistry of the gas phase. In previous work, [3] the burning rate of TAGzT was determined, and it was found to be one of the fastest burning compounds known to exist (Table 1), comparable to the high-nitrogen compounds 3,3'-azobis(6-amino-1,2,4,5-tetrazine)-3.5 N-oxide ($DAATO_{3.5}$) and 3,6-bis(1H-1,2,3,4-tetrazol-5-ylamino)-s-tetrazine (BTATz).[4] However, despite it's remarkable combustion behavior and the fact that it has high calculated detonation performance, it has not been found to shock initiate to detonation even at diameters as large as 1 5/8". This insensitivity to shock initiation is all the more intriguing because of TAGzT's sensitivity to impact and friction initiation, which it's roughly similar to HMX, Table 1.

Table 1: Sensitivity and burning rate parameters for TAGzT, BTATz, $DAATO_{3.5}$ and HMX

Material	Impact Sensitivity (H_{50}, cm)	DSC onset, °C	Friction sensitivity, kg	Spark sensitivity, J	Burning rate @1000 psi, cm·s^{-1}	Burning rate exponent
TAGzT	25	195	10.0	0.312	4.89	0.67
BTATz[c]	32[a]	264[a]	>36[a]	<0.36[a]	4.59[a]	0.49[a]
$DAATO_{3.5}$[d]	50[a]	177[a]	2-14[a]	<0.36	5.39[a]	0.275[a]
HMX	25	249.9	13.6	>0.36	2.11[b]	0.84[b]

These sensitivities, however, are often mitigated when the mixture is formulated with a binder and other ingredients. For example, one formulation with 50% TAGzT, 30% Al powder and 20% GAP was found to be completely insensitive to impact, spark and friction.

RESULTS AND DISCUSSION

MATERIAL

The TAGzT used for these experiments was synthesized by the NSWC at Indian Head and recrystalized from boiling water before use. Prior to pressing, the material was also vacuum dried at 60°C. It was found that the material pressed well without binder, so all tests were run as the pure material. In all diameters, pellets were pressed to 95% TMD or greater.

LARGE-SCALE GAP TEST

Two attempts were made to determine the detonation velocity of TAGzT, first at 0.5" diameter, then at 1" diameter, initiated with SE-1 detonators and boosted with PBX9501, both of which failed to detonate. It was then decided that the next higher diameter to determine the detonation velocity would also be fired in the configuration of the LANL Large-Scale Gap Test so that in the event of a failing detonation a "no-go" result would be directly comparable to previously characterized explosives. In the large-scale gap test, the donor charge is 1 5/8" diameter by 4" PBX9205 (92% RDX, 6% polystyrene, 2% dioctyl phalate) that is driven in the 1 5/8" diameter by 4" acceptor charge of test material, Fig. 1. In this case no spacer was used at the "gap", but in a normal series to determine the shock sensitivity of a material Dural spacers that attenuate the shock strength of the PBX9205 (about 25 GPa) are used and their thicknesses are varied to deliver lower pressures until a threshold pressure is determined. In addition to the standard LANL Large-Scale Gap Test closure switches were added to determine the shock speed in the TAGzT. The test resulted in a "no-go". This showed conclusively that TAGzT is exceptionally shock insensitive and has an abnormally high critical diameter. Figure 2 below shows the high speed video record of the TAGzT portion of this test, and Fig. 3 gives the shock velocity as recorded by the closure switches, which was 6.18 mm/μs at the first switch (overdriven portion) and 1.65 mm/μs at the last switch. The witness plate showed no dent, and in fact visual traces of unreacted TAGzT were found.

Figure 1. LANL large-scale gap test with TAGzT (yellow) fired with PBX9205 (white)

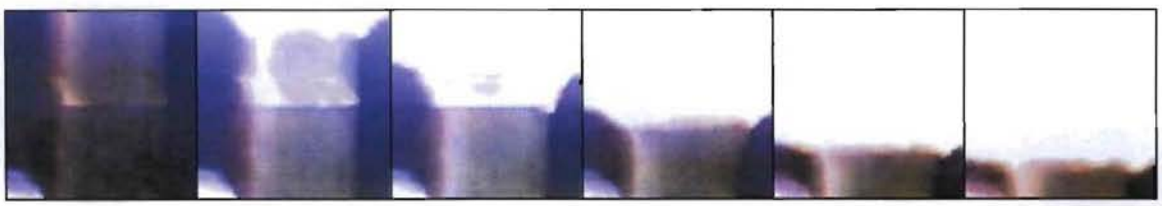

Figure 2. Image sequence from LANL large-scale gap test with TAGzT showing a decaying shock velocity to about ~1.7 mm/μs at end of cylinder.

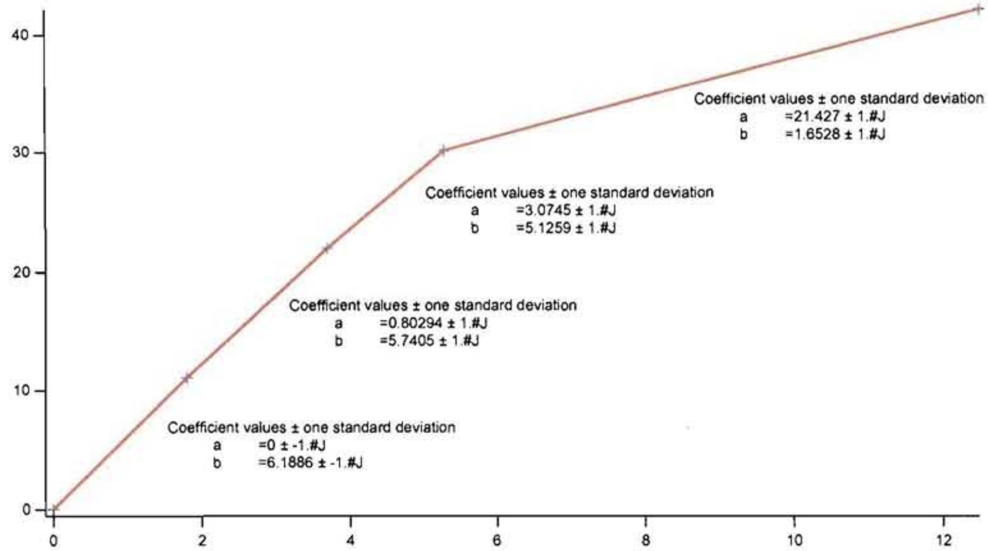

Figure 3. Velocity trace from LANL large-scale gap test with TAGzT showing a decaying shock velocity to about ~1.7 mm/µs at end of cylinder.

SUMMARY AND CONCLUSIONS

While these results maybe interpreted as negative when one is considering TAGzT as a stand-alone explosive, it should be noted that this is an extremely intriguing and positive result in terms of insensitive propellant design. TAGzT has not only proven useful in TBX formulations, but is also of great interest for modern propellant formulations, where shock insensitivity is of great interest.

FUTURE WORK

A heavily confined 1 5/8" TAGzT shot will be performed in order to determine the detonation velocity at theoretical infinite confinement. We are also characterizing the detonation performance of TAGzT/RDX mixtures at 0.5" diameters in order to determine the minimum addition of RDX to produce detonation and to determine the "phantom" detonation velocity of TAGzT.

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