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## Photoflash and laser ignition of select high-nitrogen materials

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## ABSTRACT

Gas-producing energetic materials that can be readily ignited with a photoflash are typically opaque sensitive primary explosives. In this study, we explore the photoactivity of select high-nitrogen (HiN) compounds that are much less sensitive than primary explosives. These HiN materials produce large amounts of gas upon decomposition and this makes them suitable for use in actuators, igniters, or micro-thrusters. This paper presents ignition experimental results using similar shaped pulses at two different wavelengths, specifically using a xenon photoflash and a single wavelength CO<sub>2</sub> laser. Several select HiN materials were tested for flash ignitability, and those that were found to be flash ignitable were further ignited with CO<sub>2</sub> laser heating. By comparing ignition behavior at various laser and flash intensities, some ignition mechanisms are suggested. Thermal heating, regardless of source, appears to be the dominant mechanism responsible for ignition and photochemical effects appear to be negligible in the ignition of the materials considered in this study. Higher laser and photoflash irradiance is shown to require less energy, and is therefore more efficient. The opacity of the material is an important consideration in ignitability, but not a sufficient criterion. We find that opaque materials that successfully propagate well in small capillary tubes are also more likely to successfully flash ignite. We suggest this is due to the higher burning rate of these materials and also in part due to the exothermic reaction occurring at or near the burning surface, allowing the reactions to proceed without quenching.

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## 1. Introduction

Remote, non-contact ignition of energetic materials, especially gas producing secondary explosives would have a wide variety of possible applications. Most photoflash-ignitable gas generating energetic materials that have been considered are primary explosives, such as azides and fulminates [1]; the sensitivity of these materials limits their applications. Secondary explosives that could be photoflash ignited may find use in a wider variety of applications such as igniters, gas generators in micro-thrusters and micro-actuators for microscale devices.

Some researchers have attempted to make gas generating secondary explosives photosensitive through the use of additives. One such attempt has been to add photosensitive carbon nanotubes into gas generating energetic materials, such as pentaerythritol tetranitrate (PETN), to create a flash ignitable mixture [2]. In part because it is a clear crystal, PETN will not flash ignite on its own. However, these flash ignitable PETN mixtures may have unacceptably high sensitivity for some non-explosive applications and the

use of carbon nanotubes and other additives could introduce uncertainty from batch to batch in processing and formulation. Such additives could also make the mixture less efficient and more fuel rich, possibly increasing the likelihood of a secondary reaction with air. Since the combustion products could be inhaled in many applications, avoiding any additives such as carbon nanotubes and metals is desired. Hence, a single compound that is flash ignitable, gas generating, and has acceptable sensitivity (not detonable) is desirable.

## 1.1. Ignition by irradiation

Non-contact ignition mechanisms can involve a wide range of electromagnetic radiation including visible light, infrared radiation (IR), microwaves, or even ionizing radiation. Research into ignition of explosives by radiation dates back to the 1950s. Eggert et al. published studies on the ignition of azides, acetylides, fulminates and perchlorates; detonations were also observed for some explosives under sufficiently high radiation intensities [3]. As far back as 1955, Nelson et al. discussed how heterogeneous flash ignition (ignition of a mixture of different substances, in this case in a mixture of fine light absorbing metallic particles suspended in polyethy-

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### Nomenclature

|                           |  |
|---------------------------|--|
| $I_{peak}$                | peak irradiance  |
| $E''$                     | energy flux deposited per unit area                            |
| $d$                       | diameter   |
| $A_p$                     | wall area  |
| $V$                       | volume   |
| $\delta$                  | flame thickness  |
| $\dot{q}''_{cond}$        | rate of heat conduction per unit area                          |
| $\dot{q}'''_{chem}$       | rate of heat generation per unit volume                        |
| $T$                       | temperature  |
| $\frac{d\varepsilon}{dt}$ | reaction rate, $\varepsilon$ is the reaction progress variable |
| $k_g$                     | gas phase thermal conductivity                                 |
| $\rho$                    | density  |
| $\alpha_g$                | gas phase thermal diffusivity                                  |
| $r_b$                     | burning rate   |
| $r$                       | reflectivity   |
| $Q$                       | heat release per unit mass                                     |
| $C_p$                     | heat capacity  |
| $m$                       | mass   |

lene) could result in very high but localized temperatures that can initiate reactions [4]. Early studies were also carried out by Bowden and Yoffee, who studied photodecomposition (direct decomposition of chemicals by absorption of light) of azides, nitrides, fulminates and perchlorates [1]. They also explored the photosensitization of silver azide by dyes and gold particles [1].

More recently, research into photoflash ignition of different explosive mixtures, with a wide variety of additives, has been carried out [2]. Single-walled carbon nanotubes (SWNTs) have been extensively studied because of their high absorptivity and their unique ignition behavior when irradiated with a photoflash [5–7]. As discussed above, Maana et al. demonstrated that single wall carbon nanotubes mixed with PETN could ignite at remarkably low flash irradiance [2]. Braidy et al. experimentally showed that iron (Fe) nanoparticles embedded in SWNTs will flash ignite to produce oxides of iron [8]. Carbon nanotubes have also been added to fuel/air mixtures to allow for photoflash ignition [9]. However metals and carbon nanotube additives may be undesirably toxic for some applications so alternatives are of interest.

High energy ball milling has also been shown to produce flash ignitable energetic materials. When micron size aluminum particles and poly-carbon monofluoride (PMF) are mechanically activated via high-energy ball milling, the resulting Al/PMF composite particles are ignitable by an optical flash [10]. This is thought to be due to the nanoscale features produced in the milling process, similar to what is observed with nanoscale aluminum. Other additives considered to make explosives more sensitive to ignition by photoflash and lasers include gold and silver nanoparticles [11,12].

Non-thermal ignition mechanisms for photoflash ignition have been hypothesized. Such mechanisms can involve direct photoexcitation of electrons within molecules at only certain wavelengths of light, which get preferentially absorbed [13]. This photothermal effect can directly lead to chemical breakup or the energy can degrade to heat [13]. Abboud et al. suggests that localized surface plasmon resonance enhances heating in flash irradiated aluminum nanoparticles (nAl), which in turn leads to ignition [14]. Greenfield et al. examined the photoactivity of a tetrazine derivative of PETN known as *pentaerythritol trinitrate chlorotetrazine* (PetrinTzCl) [15]. The derivative PetrinTzCl exhibited a much higher level of photoactivity than PETN when irradiated with a 532 nm wavelength laser. It was reasoned that since tetrazine has a high heat of formation and is a good chromophore (can contribute to color, and hence absorption in the visible spectrum), tetrazine derivatives such as

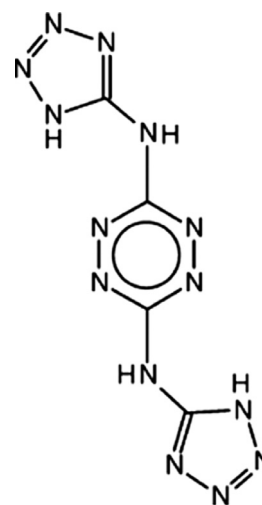


Fig. 1. BTATz molecule [22].

PetrinTzCl could possibly display direct photochemical decomposition under visible light. Other researchers reported on the photoactivity of nanoparticles of gold and other metals to UV and visible radiation due to plasmon resonance, with potential applications in engineering of metals and semiconductors [11]. Flash irradiation has also been researched as a cheap, chemical-free method for the deoxygenation reaction of graphite oxide in the bulk production of graphene based materials [16]. It is theorized that the flash creates strong photothermal and photoacoustic effects in order to start the decomposition [16].

### 1.2. High-nitrogen compounds

High-nitrogen (HiN) materials are a class of energetic compounds that derive most of their energy from the high positive heat of formation, released by decomposition, rather than from oxidation reactions of carbon or hydrogen, as is the case with most other energetic materials [17]. HiN materials usually contain significant amounts of nitrogen in their molecular structure, and release large amounts of gaseous nitrogen upon decomposition, along with other gases like hydrogen [18]. With a few notable exceptions such as azides, hydrazines and tetrazines, HiN compounds are usually aromatic heterocyclic compounds with a wide variety of substituents. Research into HiN materials dates back to the late 1800s, followed by research on tetrazine and tetrazole rings by the 1950s and 1960s. Over the decades, a number of new (mostly nitrogen) derivatives based on these rings and other new structures have been synthesized and tested [19–21].

Derivatives of the tetrazine and tetrazole rings are among the most important HiN materials that have been studied. These include 6,6-bis(1H-1,2,3,4-tetrazole-5-ylamino)-s-tetrazine (BTATz), 3,6-dihydrazino-1,2,4,5-tetrazine (DHT) and mixed N-oxides of 3,3'-zeo-bis(6-amino-1,2,4,5-tetrazine) [22,18]. The tetrazole and tetrazine ring and their high nitrogen derivatives yield relatively clean and less toxic products [23,24]. DAATO3.5, which is an N-oxide of 3,3'-zeo-bis(6-amino-1,2,4,5-tetrazine) has one of the highest burning rates of organic compounds. Azides, which are molecules containing the azide R-N<sub>3</sub> functionality or salts with the (N<sub>3</sub><sup>-</sup>) anion, are widely used HiN materials and have been used as airbag gas generators and as detonators. However, many azides are primary explosives. Another class of HiN materials consists of furazan derivatives of 3,4-diaminofurazan (which was first synthesized in 1968). One of these derivatives includes 3,3'-diamino-4,4'-azoxyfurazan (DAAF). Figures 1 and 2 show the molecular structure of two important HiN materials, BTATz and DAATO3.5.

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