


A new cocrystal explosive trinitrotoluene (TNT):1-amino-4-bromonaphthalene with reduced sensitivity

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Abstract: A 1:1 cocrystal of TNT and 1-amino-4-bromonaphthalene was prepared via solvent evaporation. The structure of the cocrystal was determined via single-crystal and powder X-ray diffraction. Crystal structure determination studies indicated that the main mechanism of cocrystallization originates from the intermolecular hydrogen bonding (amino-nitro), halogen bonding, and $\pi - \pi$ stacking. Investigation of intermolecular interactions by Hirshfeld surface and fingerprint plot analysis revealed that the structures were stabilized by H...H, O...H, C...C ($\pi - \pi$) and Br...O interactions. Hirshfeld surfaces showed that these weak interactions were significant for crystal packing. We analyzed the crystal packing and showed how this may influence sensitivity to impact. The TNT:1-amino-4-bromonaphthalene cocrystal displays significantly reduced impact sensitivity relative to pure TNT, indicating for the first time that a new TNT cocrystal was less sensitive to impact than pure TNT component. This difference in impact sensitivities may arise from the considerably different intermolecular interactions seen in the cocrystal structure. The results highlight that cocrystallization is an effective way to alter the impact sensitivity, detonation performance, decomposition temperature, melting point, oxygen balance, crystal packing, and the density of explosives.

Key words: Trinitrotoluene, sensitivity, cocrystal, insensitive energetic material

1. Introduction

Safety (insensitivity) is one of the two most significant properties and performances of energetic materials (the other is energy). Insensitive explosives have attracted significant interest in the past three decades, owing to their potential application in numerous energetic materials. Discovery of explosives possessing low impact sensitivity and high explosive performance is a key goal in the research of energetic materials that has not yet been solved.¹⁻⁴ Generally, safety is experimentally estimated by sensitivity, which is the degree of an energetic material's response to an external stimulation: a lower sensitivity represents a higher safety. Efforts have been made to diminish sensitivity through synthesis of new molecules; however, this is usually a complex, time-consuming process with extensive validation and requirements to meet increasing challenging performance targets.⁵ Cocrystallization is an established technique for improving the solubility, bioavailability, physical, and chemical stability of drugs without altering their chemical structure, and it is extensively used for the pharmaceutical industry.⁶⁻⁹ Therefore, cocrystallization can supply a new way to tune the performance and sensitivity of an energetic material without requiring complex preparation or synthesis of novel compounds.¹⁰⁻¹³ Cocrystals are multicomponent crystalline materials formed of two or more neutral cofomers, bonded by intermolecular forces such as hydrogen bonding, π stacking, and/or van der Waals forces.¹⁴⁻¹⁶

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TNT was developed in 1863, and today it is still among the most frequently used energetic materials. It has been used for a long time by armed forces for training aims on their training grounds and has a highly negative ecological effect. This requires expensive and time-consuming enhancement and detoxification operations. The improvement of new, environmentally friendly energetic materials that still meet the performance and insensitivity necessities of the armed forces and industry and that is safe for handling, transportation, and storage is a significant step in the direction of developing ecologically friendly, nontoxic, and highly energetic materials.¹⁷

To improve the properties of vital energetic materials, a good method is to cocrystallize TNT with a more insensitive energetic material or nonenergetic substances to form a new crystal structure. In recent years, researchers have applied cocrystallization to the field of energetic materials as an effective means of altering chemical and physical properties of energetic materials.¹⁸

Cocrystals of HMX (1,3,5,7-tetranitro-1,3,5,7-tetrazocane) with a wide diversity of coformers have been reported, which also afford a remarkable decrease in sensitivity compared with pure HMX.¹² Cocrystals of HNIW with nonenergetic coformers have been reported, showing that it is possible to alter the thermal behavior, sensitivity, and detonation properties of the cocrystal when compared with the pure energetic materials.¹⁹

In this paper, nonenergetic 1-amino-4-bromonaphthalene was selected as a candidate to form a cocrystal (TNT:ABN) with TNT. Crystal structure characterization, intermolecular interaction, thermal behavior, detonation performance, and impact sensitivity of the new cocrystal explosive are discussed.

2. Results and discussion

2.1. Microscope images of TNT:ABN

The macromorphology of the crystals is a reflection of its microscopic structure, and the crystal morphologies are indicated in Figure 1. It can be observed that the cocrystals are dark red crystals, with well-defined morphology, regular structure, uniform size, and smooth surface. This differs from the crystal shape of TNT, which exists as white plates. The dissimilarity in morphology shows that cocrystal molecules have a different crystal structure.

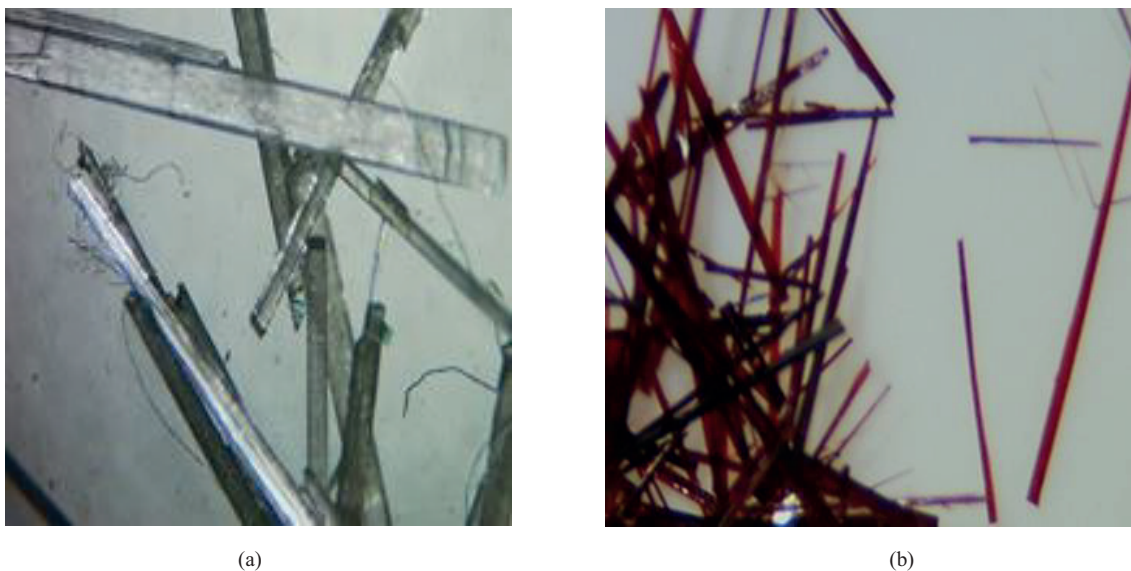


Figure 1. Microscope images of TNT (a), TNT:ABN (b).

2.2. Differential scanning calorimetry (DSC) study of TNT:ABN

DSC is useful in studying thermal behavior of materials. The DSC thermogram given in Figure 2 contains three endotherm and two exotherm peaks. The TNT:ABN cocrystal melted at 125.8 °C, which is a higher melting point compared to both TNT (82.1 °C)²⁰ and 1-amino-4-bromonaphthalene (102.13 °C). The melting point is a result of the lattice energy, namely different total intermolecular interaction in a crystal cell.²¹ The melting point increases, showing that the lattice energy is increasing. The increase in melting point shows that some intermolecular interactions occur, while the TNT:ABN cocrystal formed $\pi - \pi$ stacking interaction, hydrogen bonding, which led the lattice energy to be much higher than both components. In addition, increasing the melting point can be advantageous while creating munitions that resist deformation before detonation.¹⁸ Cocrystallization makes it possible to alter the melting point of energetic materials as needed.

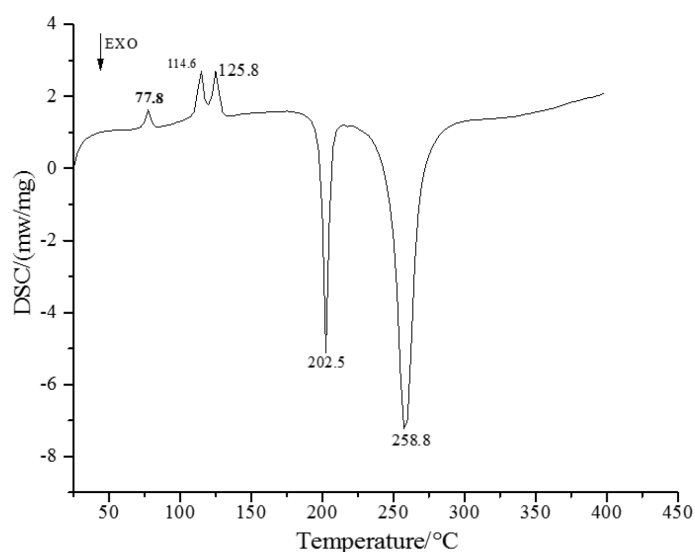


Figure 2. DSC curve for TNT:ABN.

Cocrystallization can produce important changes in another critical physical property for energetic materials: the decomposition temperature. Exothermic peaks occurred at 202.5 °C and 258.8 °C. This may be explained by the supposition that the structure of the cocrystal is destroyed with increasing temperature and is converted to liquid TNT and 1-amino-4-bromonaphthalene, leading to heterogeneous decomposition. The cocrystal's heat of decomposition temperature was lower than that of pure TNT (298 °C)¹⁸; the decomposition temperature of the TNT:ABN cocrystal changed with respect to TNT.

2.3. Structural characterisation of TNT:ABN

The powder X-ray diffraction (PXRD) patterns of the cocrystal, TNT, and the coformer are indicated in Figure 3. The characteristic diffraction peaks of TNT and 1-amino-4-bromonaphthalene do not appear in the PXRD pattern. It is clear that the diffraction patterns of the cocrystal are different from those of TNT and 1-amino-4-bromonaphthalene, indicating the formation of a new material.

The crystal structure of the TNT:ABN cocrystal was determined by single-crystal X-ray diffraction data and is indicated in Figure 4. The crystal data have been deposited at the Cambridge Crystallographic Data Centre under the CCDC number 1822174 (CSD reference code CEZCES) and the crystal data results are indicated in Table 1.

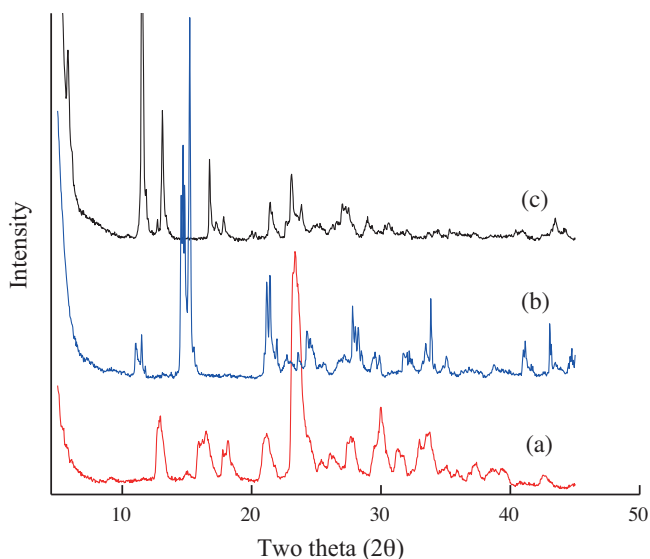


Figure 3. PXRD patterns for TNT (a), 1-amino-4-bromonaphthalene (b), TNT:ABN (c).

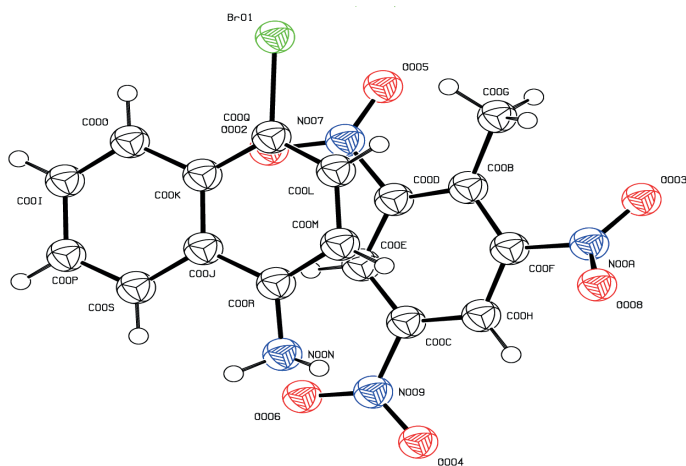


Figure 4. ORTEP drawing of TNT:ABN cocrystal.

TNT is an electron-poor π system, whereas 1-amino-4-bromonaphthalene is a relatively electron-rich aromatic compound. When choosing electron-rich aromatic substances, the COSMOtherm software package was used to predict the likelihood of cocrystal formation by calculating the excess enthalpy of formation of the coformer.²² The calculations were generated by the Turbomole package using the BP86 density functional with a TZVP34 basis set (BPTZVP-COSMO level of theory). TNT and the cocrystal formers of electrostatic potential surfaces was calculated to understand intermolecular interactions. Calculated electrostatic potential surfaces are indicated in Figure 5a and 5b.

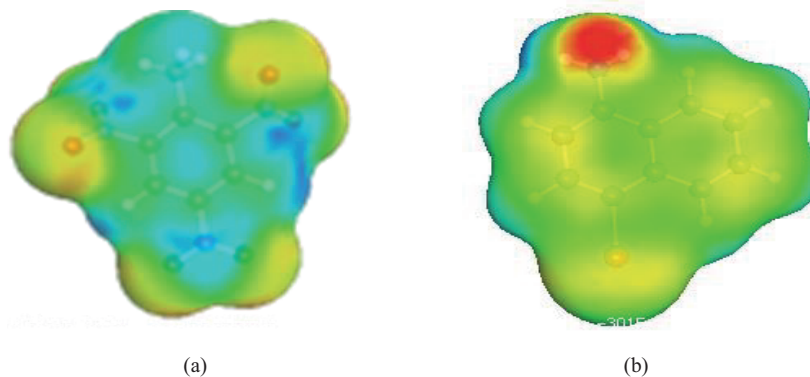


Figure 5. Electrostatic potential surfaces of TNT (a), the 1-amino-4-bromonaphthalene (b).

The TNT:ABN cocrystal possesses hydrogen bonds, halogen bonds, and $\pi - \pi$ stacking interactions and are indicated in Figures 6a–6c. Weak hydrogen bond interactions formed between amino nitrogen and nitro oxygen (N(H)...O) groups and halogen bond interactions formed between bromine and nitro oxygen.

Aromatic–aromatic or $\pi - \pi$ interactions are significant noncovalent intermolecular forces akin to hydrogen bonding. Utilization of $\pi - \pi$ stacking is of essential significance for the further development of supramolecular

Table 1. Single-crystal X-ray refinement factor for TNT:ABN.

Empirical formula	C ₁₇ H ₁₃ BrN ₄ O ₆
Formula weight	449.22
Temperature/K	120.01 (11)
Crystal system	monoclinic
Space group	P2 ₁ /c
a/Å	7.9237 (2)
b/Å	7.12180 (10)
c/Å	30.5659 (5)
α/°	90
β/°	94.775 (2)
γ/°	90
Volume/Å ³	1718.88 (6)
Z	3
ρ _{calc} g/cm ³	1.302
μ/mm ⁻¹	1.829
F(000)	678
Crystal size/mm ³	0.789 × 0.242 × 0.12
Radiation	MoKα (λ = 0.71073)
2θ range for data collection/°	5.61 to 65.53
Index ranges	-11 ≤ h ≤ 11, -10 ≤ k ≤ 10, -46 ≤ l ≤ 46
Reflections collected	85344
Independent reflections	6153 [R _{int} = 0.0398, R _{sigma} = 0.0194]
Data/restraints/parameters	6153/0/255
Goodness-of-fit on F ²	1.187
Final R indexes [I ≥ 2σ (I)]	R ₁ = 0.0716, wR ₂ = 0.1624
Final R indexes [all data]	R ₁ = 0.0773, wR ₂ = 0.1651
Largest diff. peak/hole / e Å ⁻³	1.88/-0.90
CSD reference code	CEZCES

chemistry and crystal structures prediction.^{23,24} The intermolecular distance between parallel aromatic groups was calculated and is displayed in Table 2.

Table 2. The lengths and angles of intermolecular bonds.

Hydrogen bonding							
Cocrystal	D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/deg
TNT-ABN	N	H	O	0.871	2.618	3.172	122.52
TNT-ABN	N	H	O	0.871	3.175	3.827	133.54
Halogen bonding							
Cocrystal	D	Br	A	d(D-Br)/Å	d(Br-A)/Å	d(D-A)/Å	D-Br-A/deg
TNT-ABN	C	Br	O	1.925	3.863	3.403	139.77

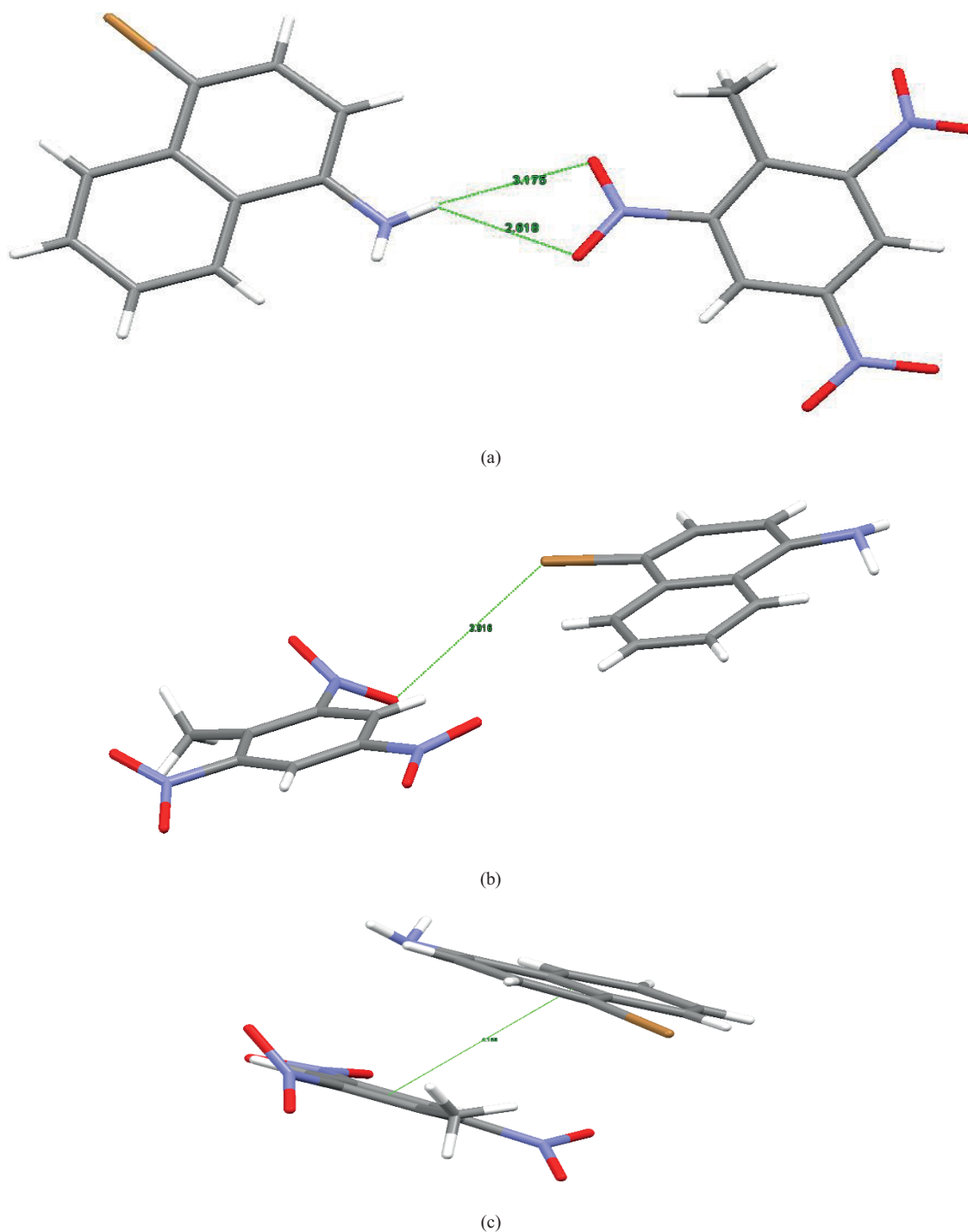


Figure 6. Hydrogen bonding (amino-nitro) (a), halogen bonding (Br-nitro) (b), $\pi - \pi$ stacking (c).

2.4. Hirshfeld surfaces analysis

Hirshfeld surfaces are a method of visualizing intermolecular interactions by color-coding close contacts. The color intensity shows the relative strength of the interactions. The size and shape of a Hirshfeld surface also reveal the interplay between different atoms and intermolecular contacts in a crystal, enabling supramolecular

chemists and crystal engineers to gain insight into crystal packing behavior.²⁵ Hirshfeld surfaces provide a three-dimensional picture of close contacts in a crystal, and these contacts can be summarized in a 2-D fingerprint plot. Crystal Explorer 17 was used to produce the fingerprint plot and Hirshfeld surfaces of the TNT:ABN cocrystal, shown in Figures 7a and 7b, respectively. These display surfaces that have been mapped over d_{norm} -0.25 to 1.12 Å. The contact contribution ratio of the cocrystal is shown in Figure 8.

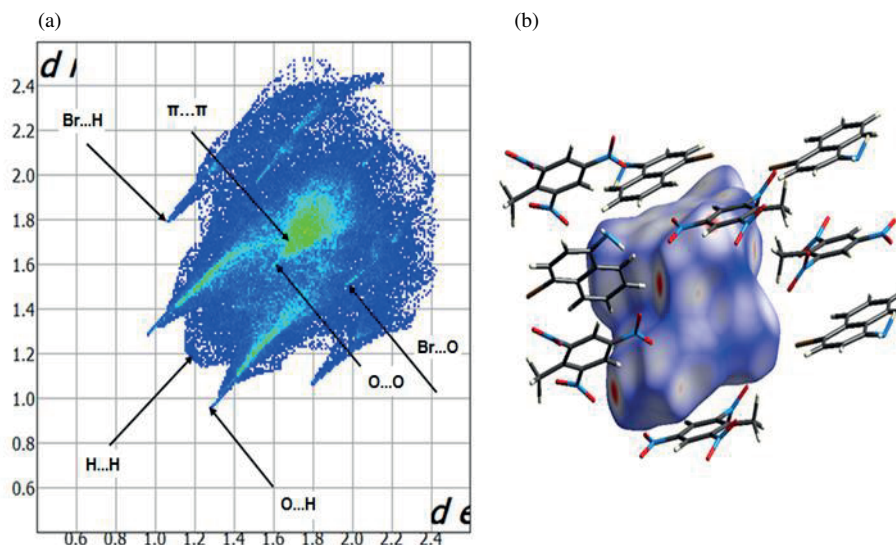


Figure 7. The two-dimensional fingerprint of TNT:ABN cocrystals (a), surface analysis of TNT:ABN (b).

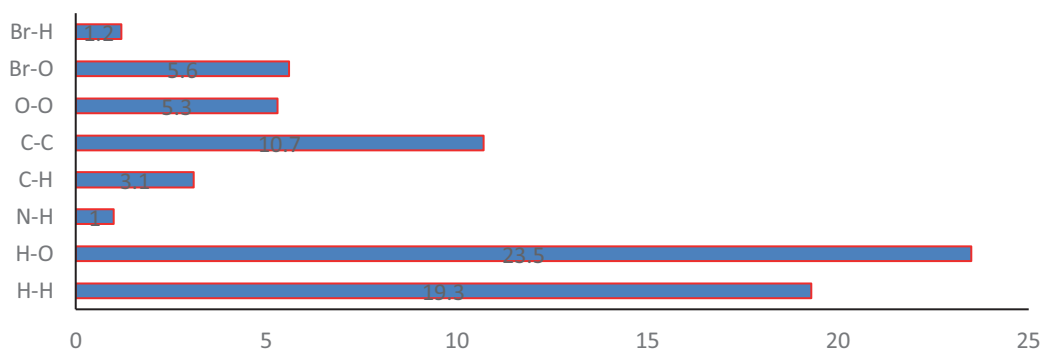


Figure 8. Summary of the various contact contributions to the TNT Hirshfeld surface area in cocrystal 1-amino-4-bromonaphthalene.

The Hirshfeld surfaces in Figure 7 revealed the close contacts of hydrogen bond donors and acceptors, but other close contacts were also obvious. The H...O interactions comprise 23.5% of the total Hirshfeld surface and appear as narrow spikes in the 2D fingerprint plot. The $\pi - \pi$ (C...C) interactions have a relatively high contribution (10.7%) to the total Hirshfeld surface of the cocrystal and appear as the large spike in the middle of the 2-D fingerprint plot. These highlight the close contacts between the molecules of TNT and 1-amino-4-bromonaphthalene. The H...H interactions have a more noteworthy contribution to the total Hirshfeld surfaces of the cocrystal, comprising 19.3%, shown in the middle of the scattered points in the two-dimensional fingerprint plot. Br...O interactions comprise 5.6% of the total Hirshfeld surfaces of cocrystal TNT:ABN.

2.5. Sensitivity

2.5.1. Impact sensitivity

Impact sensitivity is largely dependent on the physical and chemical properties of the material. Indeed, a major index of the sensitivity of an explosive to mechanical impact is the BAM fall-hammer impact test. The impact sensitivity, i.e. drop energy, is the height, h_{50} , from which a weight of given mass must be dropped onto the sample to produce an explosion 50% of the time.²⁶ In this study, we used a 10 kg drop weight. The h_{50} values of TNT and TNT:ABN were 25.1 cm (impact energy 25.1 J) and 100 cm (impact energy 100 J), respectively, and are indicated in Figures 9a and 9b. The outcomes of the experiments imply it is possible to reduce sensitivity by crystallizing TNT with a coformer.

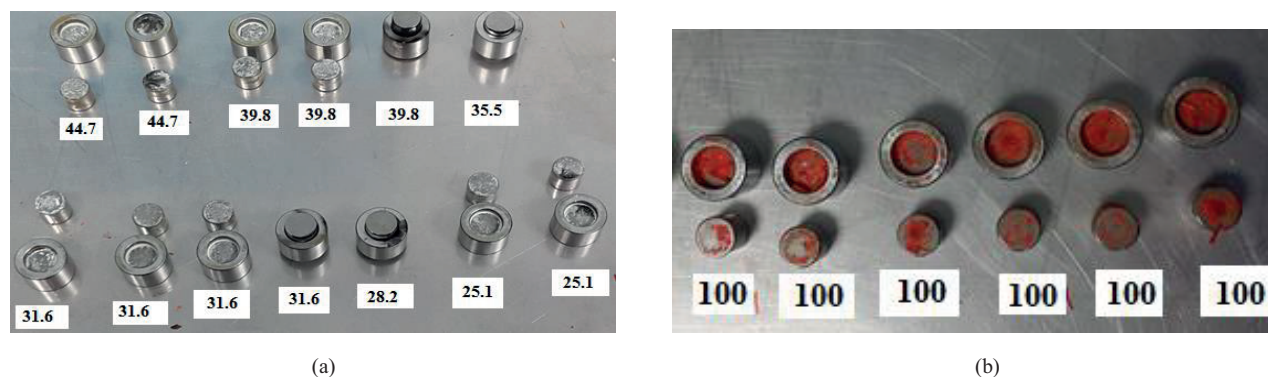


Figure 9. BAM fall hammer impact testing results of TNT (a) and TNT:ABN cocrystal (b).

In the case of TNT:ABN, not only have improved sensitivity and detonation performance been added to this material, but multiuse has as well. Through cocrystallization, a halogen (bromine) was successfully added to TNT. The production of reactive halogen species within the explosion plume provides a means of detonating biological and chemical warfare agents remotely.²⁷

2.5.2. Crystal packing effect on impact sensitivity

Though cocrystallization, explosive materials with nonenergetic materials can frequently improve the performance of explosives by reducing the sensitivity to impact and improving chemical and thermal stability. Earlier research has shown that numerous factors can affect these properties: crystal packing, density, oxygen balance within the molecular structure, intermolecular hydrogen bonding, long- and short-range interactions, voids, defects, decomposition, and molecular motion within a crystal lattice.²⁸ Crystal packing is vitally important to both the safety and detonation performance, and thus becomes of interest in energetic crystal engineering.

Four types of stacking are used to clarify the crystal packing and rationalize the stacking effect on impact sensitivity. All π stacking can be classified: face-to-face stacking, wavelike stacking, crossing stacking, and mixed stacking.²⁵

The TNT:ABN cocrystal is composed of layers along two crossed directions, whereas TNT has a herringbone packing structure, as indicated in Figures 10a and 10b. This is a different characteristic of the HBs connected within this crossing stacked crystals. This $\pi - \pi$ stacking is supported by intermolecular hydrogen bonding indicated in Figures 11a and 11b. This facilitates the aid of the crystal packing by improving intermolecular intersections.

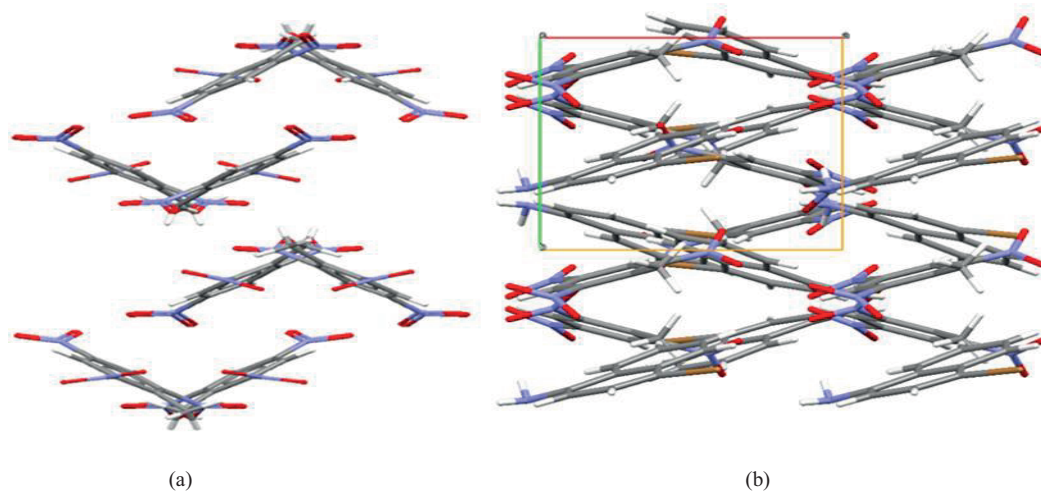


Figure 10. A comparison of the herringbone packing of TNT (CSD code ZZZMUCO8)²⁹ (a), crossing stacking present in TNT:ABN cocrystal (b).

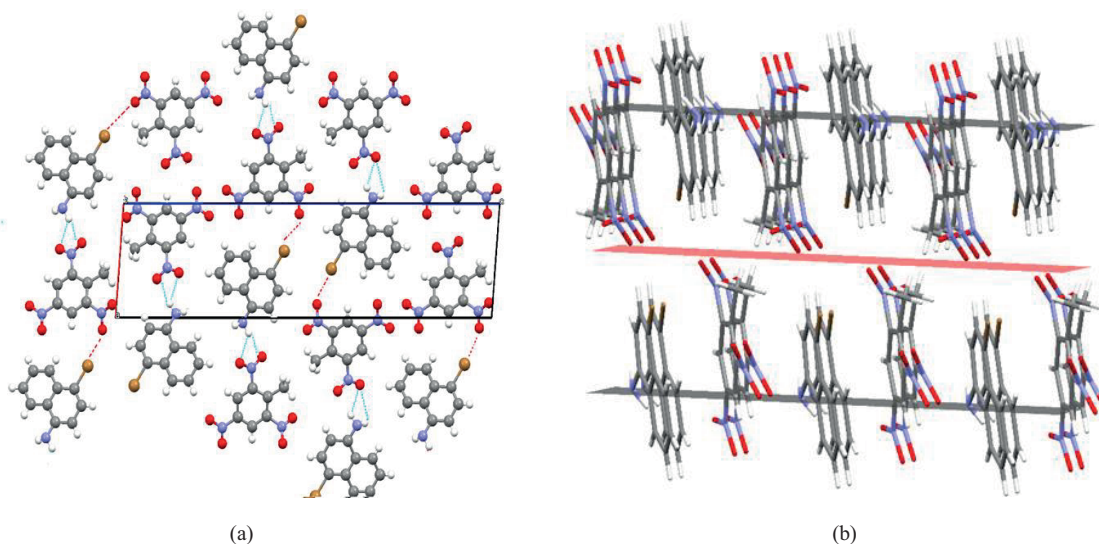


Figure 11. Intermolecular hydrogen bonding of TNT:ABN (a), Crystal structure packing of TNT:ABN with planes (b).

In the structure of the TNT:ABN cocrystal, crossing stacked crystals occur along the molecular plane and represent change sliding mechanism, suggesting that this TNT- ABN cocrystal would exhibit low impact sensitivity.

2.6. Calculation of detonation performance

Detonation parameters are essential for gaining a basic understanding of a material's energetic performance. Two important parameters, detonation pressure and detonation velocity, are used to calculate the performance of explosive compounds. In the present study, Kamlet–Jacobs empirical equations (1) and (2) were used to determine $C_a H_b O_c N_d Br$ (TNT:ABN) compounds.^{30–33}

$$D = 1.01(N^{0.5} \times M_{ave}^{0.25} \times Q^{0.25})(1 + 1.30\rho) \quad (1)$$

$$P = 1.558(N \times M_{ave}^{0.5} \times Q^{0.5} \rho^2) \quad (2)$$

The terms in Kamlet–Jacobs equations (1) and (2) are: D , detonation velocity (km/s); P , detonation pressure (GPa); ρ , density of a compound (g/cm^3); N , moles of gaseous detonation products per gram of explosive; M_{ave} , average molecular weight of gaseous products; Q , chemical energy of detonation (kcal/g). The parameters N , M_{ave} , and Q are calculated according to the chemical composition of each explosive as revealed in the literature.³⁴ For a $\text{C}_a\text{H}_b\text{O}_c\text{N}_d$ type explosive, if $2a + b/2 > c \geq b/2$ is fulfilled, N is calculated using $(b + 2c + 2d) / 4M$, M_{ave} is calculated as $(56d + 88c - 8b) / (b + 2c + 2d)$, and Q is calculated as $[28.9b + 94.05(c/2 - b/4) + 0.239 \Delta H^\circ f]/M$. If $b/2 > c$ is satisfied, N is calculated as $(b + d/2M)$, M_{ave} is calculated as $(2b + 28d + 32c) / (b + d)$, Q is calculated as $[(57.8c + 0.239 \Delta H^\circ f) / M]$. The coefficient (0.239) of $\Delta H^\circ f$ in the relations is a conversion factor from kJ/mol to kcal/mol. M is the molecular weight of the compound (in g/mol); $\Delta H^\circ f$ is the standard heat of formation of the compound (in kJ/mol). The standard heat of formation ($\Delta H^\circ f$) was measured using DSC under constant pressure. The calculated oxygen balance ($\Omega\%$)³⁵, measured heat of formation ($\Delta H^\circ f$), density (ρ), the chemical energy of detonation (Q), detonation velocity (D), and pressure (P) are listed in Table 3.

Table 3. Calculated detonation properties for TNT:ABN cocrystal.

Molecule	% Ω	$\Delta H^\circ f$ (kJ mol ⁻¹)	Q(kcal g ⁻¹)	(g cm ⁻³)	D(km s ⁻¹)	P(Gpa)
TNT	-73.97	50.57	1400.5	1.64	7.11	19
TNT:ABN	-122.87	530	1214.83	1.302	7.88	22.03

A novel energetic cocrystal explosive composed of TNT and 1-amino-4-bromonaphthalene with a 1:1 molar ratio has been discovered and characterized. The TNT:ABN cocrystal exhibited $\pi - \pi$ stacking interactions, hydrogen bonding interactions (amino–nitro), and halogen bonding interactions (Br–O). Crossing stacking in TNT:ABN, combined with strong intermolecular bonds, could potentially lead to a reduced impact sensitivity for this cocrystal. Properties crucial to energetic materials, including the density, melting point, decomposition temperature, crystal structure packing, and impact sensitivity have all changed as a result of co-crystallization. This shows that cocrystallization is indeed a favorable way to enhance safety of energetic materials by tuning crystal packing structure.

3. Experimental

3.1. Materials and sample preparation

TNT was synthesized from 2,4-dinitrotoluene (DNT) according to a known procedure.³⁶ 1-Amino-4-bromonaphthalene was purchased from Sigma-Aldrich (St. Louis, MO, USA). Cocrystallization was performed by dissolving a 1:1 molar ratio of TNT (1.57 mg), and 1-amino-4-bromonaphthalene (1.53 mg) in 85% anhydrous methanol (approximately 3–5 mL) at 50 °C and stirring for 60 min. The solvent was allowed to evaporate at room temperature over a period of several days. Dark red crystals of TNT:1-amino-4-bromonaphthalene (TNT:ABN) formed.

3.2. Single-crystal X-ray diffraction

The single-crystal X-ray diffraction data of the cocrystals was collected on an Agilent Technologies Super Nova diffractometer (Santa Clara, CA, USA) equipped with an Oxford Cryosystems device. The crystal was kept at $T = 120$ K during data collection. Using Olex2, the structure was solved with the ShelXT structure solution program, using the intrinsic phasing solution method. The model was refined with version 2017/1 of ShelXL using least squares minimization.^{37–39}

3.3. Powder X-ray diffraction (PXRD)

PXRD patterns were recorded using the D2 Phaser instrument using Cu-K α radiation ($\lambda = 1.54439$ Å) and an operating voltage and current 40 kV and 10 mA, respectively. The data were collected over an angle range of $2\theta = 5$ – 50° . This technique provided an initial screening of samples for cocrystal formation.

3.4. Optical microscopy

Optical micrographs of all the crystals were taken using an SK2005A polarization microscope.

3.5. Differential scanning calorimetry

DSC was performed on a NETZSCH STA 449 F1 differential scanning calorimeter (Selb, Germany). The sample (1.43 mg) was placed in aluminum pans and the thermal behavior of the samples was studied under a nitrogen (30.0 mL/min) purge at a heating rate of $10^\circ\text{C}/\text{min}$ over a range from 25 to 400°C .

3.6. Impact sensitivity properties

The impact sensitivity was determined by using a BAM fall hammer device (BFH-12). Impact testing on the TNT and TNT:ABN cocrystal was conducted at the Cavendish Laboratory at Cambridge University. A sample of 40 mm^3 was enclosed in an anvil device consisting of two coaxial steel cylinders. Anvil devices were disposed of along with the sample after test. A load of 10 kg was dropped on to a sample from heights ranging from 10 cm to 100 cm. The “one-in-six” test procedure was performed to obtain limiting impact energy for both tested sample. The method used throughout was the 30 trial Bruceton method.^{40,41}

Acknowledgments

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