



Extreme condition nanocarbon formation under air and argon atmospheres during detonation of composition B-3



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ABSTRACT

Novel nanocarbons synthesized by extreme conditions produce products with various chemical and physical properties. Herein through high explosive detonation of Composition B-3 (40% TNT, 60% RDX), pressures and temperatures not classically attainable in laboratory syntheses provide access to interesting carbon allotropes. For example, detonations of Composition B-3 are regularly used to synthesize the now commercially-available nanodiamond (3–5 nm spherical diamond particles) through quenching of the detonation by ice collars at high pressures and temperatures. Detonation conditions of Composition B-3, in this study, were modified by altering the atmosphere (air versus Ar) without quenching, consequently directing nanocarbon formation. X-ray scattering and microscopy elucidate that detonations performed in air produced spherical hollow core-shell nanocarbons, whereas detonations in Ar produced elongated nanocarbons. Although morphology bifurcates, spectroscopy reveals that both major detonation products are comprised of sp^2 hybridized carbon. As expected from air atmosphere detonations, surface functionalization is dominated by C–O bonding. The absence of oxygen in the Ar atmosphere detonations may propagate extended sheets of graphene that either stack together due to electrostatics or fold upon itself. This work demonstrates that modification of the detonation atmosphere provides an alternative route for the production of new and interesting nanocarbons.

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1. Introduction

High explosive (HE) detonations provide access to extreme conditions that explore the recognized boundaries of the carbon phase diagram (Fig. S1A) [1]. Carbon allotropes are the backbone of numerous scientific areas; however molecular dynamics (MD) simulations suggests the possibility of various undiscovered nanocarbons [2,3]. Allotropes such as graphite (hexagonal structure, sp^2 -hybridized) have high electronic transport, the ability to store electrons, and high tensile strength [4]. With an additional bond to carbon, diamond (tetrahedral, sp^3 -hybridized) is the hardest known material, an electronic insulator and a good thermal

conductor [5]. Diamond is synthesized at high pressures (HP) and high temperatures (HT). Q-carbon is an example of a recently discovered carbon allotrope consisting of sp^2/sp^3 mixture, synthesized by nanosecond laser melting of amorphous carbon at HT (4000 K) but low P (<1 GPa); Q-carbon possesses properties similar to diamond, additionally it is ferromagnetic, a property not typically observed in carbon allotropes [6]. Detonation nanodiamonds (DND), a carbon allotrope discovered during detonation of cyclo-trimethylenetrinitramine/trinitrotoluene (RDX/TNT, Composition B (Fig. S1 A, B), [7–9] have a variety of uses including: thickeners in mechanical lubricants, fluorescent biomarkers for medical applications and epitaxial substrates for Au nano-platelet growth [10,11]. Extreme condition synthesis, such as the P and T coupled to produce the above nanocarbons, can be explored to synthesize various carbon structures.

Nanocarbon formation (size, morphology, hybridization) under extreme conditions produce particles where resultant properties are dependent upon time, P , T , amount of detonation material and

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atmosphere. HE detonations are a rapid and violent form of combustion in which energy transfer is by mass flow in extremely strong compression waves [12]. The detonation process in HEs is described by the Zel'dovich-von Neumann-Döring (ZND) model. The leading edge of the detonation front is a strong shock wave called the von Neumann (VN) spike [13]. In the VN spike, the explosive molecules are compressed to *HP*, *HT* and density [14]. Following the VN spike is the Chemical Reaction Zone (CRZ), where large metastable HE molecules are converted to a dense fluid of simple molecules such as N_2 , CO_2 , H_2O and carbon with a rapid energy release that drives the leading shock wave. The Chapman-Jouguet (CJ) state marks the end of the CRZ. Typical pressure at the CJ state for Composition B-3 (Comp B-3, 40% TNT/60% RDX, Fig. S1A, B) is ~28 GPa (versus 38 GPa at the VN spike). Ahead of the CJ state the sound velocity is greater than the detonation velocity and energy released within the CRZ then drives the detonation front. Behind the CJ point the products expand along the CJ isentrope (Fig. S1C). [15–17]; the CJ state at ~28 GPa, 3500 K is the upper terminus. Carbon-based detonation products are formed in the CRZ ($VN = 38 \text{ GPa} > P > CJ = 28 \text{ GPa}$) or on the CJ isentrope ($CJ = 28 \text{ GPa} > P$).

Understanding particle formation within the CRZ and along the isentrope proves difficult due to the short reaction times and violent nature of HE detonations. MD simulations suggest that initial product formation commences with gaseous (H_2O , N_2 , CO_2) and solid carbon products produced on picoseconds timescales, where carbon coagulation is known to evolve out to microseconds [18–20]. CRZ dynamics were previously determined through measuring particle velocity wave profiles, resulting in a Comp B-3 CRZ time of ~50 ns.¹ Complete mixing of this turbulent system with the surrounding atmosphere (i.e. afterburn) does not transpire until the detonation has relaxed to lower *P* and *T* along the CJ isentrope. At the *P* and *T* reached during detonation, both graphite and diamond are expected in the post detonation soot [21]; however, the introduction of oxygen to similar syntheses are shown to modify product formation at lower *P* [22]. Herein we study how detonation atmosphere (air vs Ar) affects nanocarbon formation, specifically how does the presence of oxygen direct allotrope type, morphology, size and surface functionalization. Through the x-ray scattering, microscopy and spectroscopy of the untreated post detonation soot we are able to resolve specific atomic-level to micron-level variations resultant from the two atmospheric conditions without quenching.

2. Experimental

2.1. Synthesis of nanocarbons

HE Comp B-3 (lot #HOL85G461-004, Holston) composed of 40% trinitrotoluene (TNT), 60% cyclotrimethylenetrinitramine (RDX: 6.5% octogen HMX) (Fig. S1A, B), was warm-pressed into pellets weighing ~2.7 g, (L: 20 mm; OD: 10 mm; Density: 1.7 g/cm^3). Four pellets were epoxied together with Aralhex B (Los Alamos urethane-based adhesive) to create a detonation stick weighing ~10.8 g. Detonations were initiated using an RP-1 detonator (Reynolds). Detonations were conducted in a stainless steel cylinder (Fig. S1D), (OD = 0.3 m; Wall Thickness = 0.013 m; Length = 0.61 m; Aluminum caps: $0.356 \text{ m} \times 0.356 \text{ m} \times 0.0254 \text{ m}$) under either ambient (i.e., in air) atmosphere or after a 10 min Ar purge with the Ar continuing to flow throughout the detonation.

The amount of oxygen present in the cylinder after 10 min Ar purge was 0% O_2 (Fig. S1E). Oxygen levels were measured with an ALTAIR® 5 × Gas Detector (MSA, PA). After detonation the solid residues were collected by vacuum onto an Ag filter. Detonation cylinders were thoroughly cleaned between detonations by sweeping out soot particles, as well as cleaning the walls and caps with industrial solvents. The post detonation soot collected after detonation was analyzed without acidic or oxidative treatment, the one caveat being soot studied by 1H NMR was extracted into hexanes. Impurities present in the detonation includes brass from the detonator caps, stainless steel from the chamber and vacuum grease.

2.2. X-ray scattering

Small-angle X-ray scattering (SAXS) measurements were performed at 12-ID-C (12 keV) at the Advanced Photon Source (APS) at Argonne National Laboratory. These data were collected using a custom built CCD detector composed of 4 CCD chips and features a 175 mm square active area with 1000×1000 pixel resolution. The sample to detector distance provided a *q*-range from 0.07 to 1 \AA^{-1} . The scattering vector, *q*, was calibrated with silver behenate's characteristic diffraction ring at $q = 0.1076 \text{ \AA}^{-1}$. The collected scattering images were azimuthally averaged to produce plots of scattering intensity, *I*(*q*), versus the scattering vector, *q*, where $q = 4\pi/\lambda \sin(\theta)$. The samples were placed between pressure-sensitive tape for measurements.

Wide-angle X-ray scattering (WAXS) measurements were performed at 12-ID-B and 12-ID-C. The *q*-range for 12-ID-B was $2.3\text{--}2.9 \text{ \AA}^{-1}$ (Pilatus, Switzerland). Samples were also placed between tape for measurements.

2.3. High energy diffraction

High energy diffraction was performed at APS on Beamline 11-ID-B at a wavelength of 0.2112 \AA , 58.66 KeV. The data were collected on a Perkin Elmer amorphous silicon detector (Santa Clara, CA) with a sample to detector distance that provided a *q*-range from 0.38 to 8.65 \AA^{-1} . Cerium dioxide (Acros Organics, 99.9%) was used to calibrate all collected data. The data was corrected and azimuthally averaged in Fit2D (ESRF, France) to create one-dimensional plots with scattering intensity, *I*(*q*), versus the scattering vector, *q*, where $q = 4\pi/\lambda \sin(\theta)$. Samples were sealed within polyimide tubes (OD: 0.0432 inch Cole-Parmer, IL) with epoxy.

2.4. Electron microscopy

Samples were prepared by suspending soot in water through sonication and then placed onto the transmission electron microscopy (TEM) grids (SiN windows) and dried. Analysis was performed on a FEI Tecnai G(2) F30 S-Twin 300 kV transmission electron microscope.

2.5. Photoelectron spectroscopy

Solid carbon residues were characterized for amount of carbon and oxygen by X-ray photoelectron spectroscopy (XPS). XPS was performed using a Physical Electronics VersaProbe (ULVAC-PHI, MN). Soot collected on the Ag filter was directly inserted into the XPS for analysis. First a survey scan was performed, then a high resolution scan of the carbon and oxygen peaks was collected.

2.6. Raman spectroscopy

Raman spectroscopy was performed with an Invictus diode 785 nm laser (Kaiser Optical Systems, Inc., MI). Short acquisition

¹ Dattelbaum DM, Sheffield SA, Gustavsen RL, Gibson LL, Johnson CE. A comparison of the shock initiation sensitivities, and resulting reactive flow of several 2,4,6-trinitrotoluene-based explosives. In Preparation.

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