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Observation of Variations in Condensed Carbon Morphology Dependent on Composition B Detonation Conditions

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transmission electron microscopy
thermochemical modeling

Abstract

Carbon particulates generated during detonation depend upon high explosive type, composition, and detonation conditions. Although the explosive composition greatly affects particulates, the focus of this work is on how detonation geometries that induce much higher temperatures and pressures in the high explosive lead to differing particulate morphologies. In this study, two geometries were used: Detonations were initiated in Composition B cylinders

at one end in conventional detonations and initiated at both ends to produce colliding detonations. Each of these detonations was observed on the sub- μ s timescale using fast radiography capturing images of the front moving through the cylinder, and colliding detonation fronts in real-time. These imaging experiments were complemented with time-resolved small-angle x-ray scattering (SAXS) experiments that were able to observe and determine the varying condensed carbon morphologies at different locations and times in each detonation. The detonations could be timed in such a way that the spatial and temporal dependence of the carbon morphology could be superimposed onto radiography images collected at the same point in time. The complementary approach is able to show that the carbon condensates are much larger when formed in the elevated temperature and pressure conditions near the location of colliding detonation fronts. Thermochemical modeling suggests that these larger particulates form either in the diamond phase or on the liquidus line of the carbon phase diagram. The increase in size observed by SAXS may correlate well with the increased residence time deeply in the diamond phase. These particulates can be described as nano-sized phases with some surface texture or otherwise near-surface intra-particle heterogeneity.

1 Introduction

Detonation nanodiamond is widely produced from RDX/TNT mixtures [1--3]. Exact mechanisms of nanodiamond formation during these detonations are uncertain; some posit a liquid to solid transition [3, 4] while others claim diamond forms under a process more similar to chemical vapor deposition [5]. Other high explosives such as HNS, TATB, and BTF produce widely varying nanostructured particulates with differing morphologies and graphitic vs. diamond content [6]. Of interest is what extent the explosive composition vs. the pressures and temperatures attained affect resultant condensed carbon particulates.

Kinetics of nanoparticle formation during detonation are also under investigation with reported formation timescales varying from microseconds [7] to hundreds of nanoseconds [8-10]. This discrepancy thus far persists and has not yet been fully resolved.

In this paper, we explore varying detonation conditions in the well-known explosive Composition B, that may lead to differing resultant condensed carbon particulates, and report

real-time ultrafast x-ray measurements during the detonation. We first explore a typical scenario: a dense, cylindrical charge is initiated at one end; the detonation propagates in one direction along the cylinder. In the second scenario, detonation is initiated at both ends of the cylinder simultaneously. Detonation fronts propagate and collide near the center of the charge, creating elevated temperatures and pressures compared to the conventional one-dimensional detonation. Furthermore, the temperatures and pressures attained in the colliding case should vary dramatically and depend upon location within the charge. We combine single-bunch radiographic scatter beam imaging [11] with single bunch time-resolved small-angle x-ray scattering (SAXS), to observe variations in both mass density and nanoscale particulate morphology during detonations. We recover condensed detonation products and use morphology observed with transmission electron microscopy (TEM) to inform the SAXS model. We also compare and contrast time-resolved SAXS data to particulates expected based on thermochemical modeling.

2 Experimental

Composition B pellets were pressed in the High Explosives Application Facility at Lawrence Livermore National Laboratory. Composition B, or Comp B, consists of about 60% 1,3,5-Trinitro-1,3,5-triazinane, also known as hexogen or RDX, and about 40% 2,4,6 trinitrotoluene, or TNT. For each sample, 2 g of the mixture was uniaxially pressed to 241 MPa (35 kpsi), for two 3-minute dwells, at 60 C, into a cylinder with a diameter of about 9.5 mm (.375 inches) resulting in cylinders about 16.6 mm tall and $1.70 \pm 0.01 \text{ g cm}^{-3}$ density.

These pellets were then detonated using exploding foil-based detonators that use 70 mg of hexanitrohexaazaisowurtzitane (HNIW or CL-20) to accelerate a small aluminum flyer into the base of each high explosive cylinder. For conventional detonations, one detonator was used to initiate detonation from one end of the cylinder. For the colliding case,

a detonator was affixed to each end of the cylinder and these were simultaneously detonated to produce detonation fronts traveling towards and colliding at the center of the charge.

Time-resolved small-angle x-ray scattering (TR-SAXS) and scatter beam imaging (SBI) were performed at the Advanced Photon Source, Argonne National Laboratory, at the Dynamic Compression Sector 35-ID-B [9,10,12,13], and at Sector 32-ID-B [14-16] in the LLNL detonation tank [10]. Detonations were fired in the vacuum vessel at <200 mTorr, with KaptonTM (polyimide) windows sealing the vacuum and providing x-ray transmission. Within the vacuum vessel, two LexanTM (polycarbonate) panels each ranging from 2 mm in the conventional to 2.5 mm thick in the colliding case were placed between the sample and Kapton vacuum windows as shrapnel shields.

The detonators were fired synchronously with the storage ring to achieve the desired timing. Experiments presented herein were performed in the 24-bunch mode, where ~80 ps FWHM x-ray pulses are generated at 153.4 ns intervals. Detonation times, t_{det} , were determined relative to the time the detonation front passes the center of the cylindrical charge. SAXS data were collected in both the normal and colliding case at this central position.

The timings for conventional shots were calibrated using a spring-loaded sample holder with a piezo timing pin (Dynasen) positioned at the center, top of the sample. A pin was also placed in the x-ray beam between experimental detonations, calibrating the detonation front arrival times relative the x-ray pulses arriving at the sample [8].

Both the sectors 35 and 32 at the Advanced Photon Source (APS), at Argonne National Laboratory, possess short-period undulators conducive to single-bunch TR-SAXS [17] and radiography. At sector 35, the U17.2 undulator, with a closed gap and Kirkpatrick-Baez-optics with a platinum coating to focus and reject higher harmonics, provides a beam profile with a peak energy of about 23.5 keV. At sector 32, the U18 undulator provides

similar spectral output. A Be refractive lensing system was used to both focus the beam and to partially reject higher harmonics in this case at sector 32.

Images and SAXS patterns were recorded using an array of four identical area detectors (PI-MAX4 1024i ICCD, Princeton Instruments) focused on the output of a scintillator and image intensifier [10, 16, 18, 19]. The two-dimensional camera intensity images were reduced to one-dimensional SAXS traces using the Nika package [20] for the four cameras independently. Silver behenate and glassy carbon data acquired intermittently at the sample position were used for calibration of the q -range [21] and absolute intensity [22], respectively. Small-angle x-ray scattering data were modeled by the least-squares fitting routine (lmfit [23]) that included the undulator profile. Centimeter-scale radiographic scatter beam images were collected by placing 2 mm of glassy carbon upstream from the sample in the focused x-ray beam and acquiring point-source projection images in the detection scheme commonly used for SAXS [11]. The scattered x-rays from glassy carbon expose the entire sample to a non-uniform x-ray flux. After flat and dark field corrections to each image, the data provides a time-resolved, 2D map of the X-ray transmission during detonation. Common timings were used for both imaging and SAXS; additional interleaved shots provided additional data for the SAXS measurements at early (sub ~ 500 ns) timescales.

Recovery of detonation products for subsequent analysis, performed separately from TR-SAXS measurements, maintained soot purity. The recovery shots were performed in deionized ice capture vessels; explosive samples were placed in the center of a bore with a larger diameter than the explosive and detonated within this ice capture layer [24, 25]. Ice both quenches the products and provides a convenient medium to capture and retain detonation products at purity limited only by the water, and detonator and sample housing material. After detonation, soots were recovered from the ice surface layer of the inner, nominally spherical, cavity left by the detonation.

For transmission electron microscopy (TEM), suspensions of carbonaceous products were further diluted in DI water, and a few microliters of these suspensions were drop-cast on lacey carbon TEM grids (Ted Pella). The sample droplets were left on the TEM grids for up to five minutes prior to wicking away the residual liquid with a microfiber cloth and drying with nitrogen gas. The samples thus prepared were kept in a TEM grid box under ambient conditions until analysis in the TEM.

TEM analysis was performed on a JEOL 2100F field-emission microscope operating at either 120 kV or 200 kV. Images and diffraction patterns were acquired with an Orius SC1000 CCD at 1x binning, and electron energy loss spectra (EELS) were collected with a GIF Tridiem spectrometer (Gatan, Inc.) using dispersions of 0.1-0.3 eV. Substantial graphitization of nanodiamond was not observed during typical TEM imaging.

Thermochemical calculations were performed for comparison to the experimental time-resolved data. Sizes of the carbon condensates were input as empirical parameters to correct for surface energy effects [26, 27]. The size was deduced from the SAXS experiments at early (sub-microsecond) times. Carbon phase diagrams, calculated for these sizes [28], take into account the surface energy contributions. Additionally, thermochemical calculations of detonation and expansion were performed for each expected detonation product. (These calculations included condensed carbon diamond, graphite, and liquid; N₂, CO₂, and CO as major products, as well as minor gaseous products NO, NO₂, N₂O, O₂, etc.) With these assumptions, the most probable (pressure, temperature) expansion paths for Comp B, at different locations within particularly the colliding detonation, were calculated and appear subsequently in this text.

The kinetics of particulate growth, measured by SAXS, requires that the data be fit to a model that can account for the unique morphology of detonation nanodiamond, namely a diamond or diamond-like core surrounded by heterogeneous surface layers that both

contribute to the scattered intensity. [24] Briefly, the model assumes a spherically symmetric particulate, whose scattering is calculated by the equation: <ffr1><ffr2>

$$\langle \text{ffr1} \rangle I(q) = \left[4\pi \int_0^R \rho(r) \frac{\sin qr}{qr} r^2 dr \right]^2 \langle \text{ZS} \rangle (1)$$

$$\langle \text{ffr2} \rangle q = 4\pi \frac{\sin \theta/2}{\lambda} \langle \text{ZS} \rangle (2)$$

where q is the magnitude of the scattering vector, θ is the angle that the scattered x-rays were measured, $\rho(r)$, is the radial scattering length density profile and R is the radius of the phase. [29] Generally, Equation 1 can be used to model the scattering from any spherically symmetric particulate as long as the function, $\rho(r)$, is known or parameterized; the scattering length density is proportional to the electron density. [29] In the case of detonation nanodiamond, $\rho(r)$, contains a radial segment that is associated with diamond, as well as some surface texture modeled by three segments. [24] Carbon onions, produced by, and observed in the soots of hydrogen-free high pressure and temperature explosives [30], can also be modeled with Equation 1 by parameterizing $\rho(r)$ in such a way that it is a Heaviside function that mimics the graphitic layers. In this study, the nanodiamond formulation is used to model the SAXS from the conventional detonation, as well as the colliding detonation. As temperature-pressure conditions are different in the central position, these data are also additionally described by the carbon onion model and both results are presented. Details of these formulations can be found in the supporting information.

3 Results

Copious detonation nanodiamond appears in TEM imaging of recovered soots from both the conventional and the colliding detonation, and determination of the most prevalent morphology in the products facilitates the SAXS modeling. Collected recovered detonation products from colliding detonations contain an abundance of detonation nanodiamond (Figure 1) and some graphitic fibers, all similar in size and morphology to what has been observed during conventional Composition B detonations [24]. Electron diffraction confirms a mixture of graphite and diamond, with extensive regions of high diamond content. The limited volume that encompasses the elevated temperature and pressure where detonation

fronts collide makes recovery of soot originating from this exact volume extremely challenging. Even so, the soot recovered from a colliding Composition B shot, in one instance, exhibited stacked graphitic sheets (supporting information) that have not appeared in conventional Composition B detonations. However, the TEM data indicate that most of the detonation products in general, and the average morphologies of particulates considering the entire charge, are not much different from those produced by a conventional detonation. The observed diamond core surrounded by heterogeneous surface layers thus forms the basis for the model used to fit the SAXS data.

The combined TR-SAXS and scatter beam radiographic imaging results provide a means to observe the development of nanoscale particulate morphology relative to the detonation front(s) traversing the explosive. The top two rows of Figure 2 present time-resolved radiographs of the conventional and colliding detonation fronts progressing through the explosive. The conventional detonation appears in the top panes and colliding is shown in the 2nd row, with 2-bunch (0.3068 μ s) interframe timing. The 4th through 8th frames are highlighted and the corresponding TR-SAXS is depicted in the third, bottom row. These SAXS traces in each of these panes were acquired at different positions within the explosive, at equivalent timings. Small angle scattering traces were acquired at positions indicated in the leftmost radiographs; namely blue and red for the center position of conventional and colliding, respectively; black was acquired at the edge of the explosive at the central height during colliding experiments, and finally, green was acquired at the quarter-height position during colliding experiments. Blue, red, black and green traces in the SAXS panes correspond to these measurement positions.

The first SAXS frame corresponds to the time of about 0.045 μ s before the front arrives at the center measurement position. The SAXS curves from center positions (red and blue) exhibit only the characteristic surface scattering from static Comp B, as expected, while

the detonation front has already passed the quarter-height position and carbon condensate scattering is evident in the green trace.

In the next SAXS plot, acquired about 0.26 μ s, the scattering from the mid points changes dramatically, and clear differences appear between the two additional positions as well. At low q , the colliding (red) has a higher intensity than the conventional (blue), while at high q , the colliding is less intense than the conventional case. Some scattering also appears at the side position at this time.

As the time after detonation increases, some slight changes in shape occur in the region where the intensity decays in a “knee-like” fashion, which follows the Guinier approximation for any phase:

$$I(q) = NV^2\Delta\rho^2 \exp\left(\frac{-q^2R_g^2}{3}\right) \langle ZS \rangle \quad (3)$$

where N is the total number density of the phase, V is the mean volume, $\Delta\rho$ is the x-ray contrast, which is proportional to the difference in electron density between the particulate and its immediate surroundings and R_g is the radius of gyration, which is the root-mean-square distance from the center of electron mass of the phase. [29] Referring to Equation 3, the q -dependent shape of the intensity is attributed solely to the particle size and shape via the radius of gyration, R_g , while the scaling can be attributed to one of three factors: N , V , and $\Delta\rho$. An apparent shifting of the Guinier knees in Figure 2 indicates that the size of the particulates depends on the detonation conditions. In the normal detonation, the Guinier knee is located at $q \sim 0.12 \text{ \AA}^{-1}$ at 0.26 μ s and shifts towards a steady-state position of $q \sim 0.1 \text{ \AA}^{-1}$ at 1.18 μ s. To a first approximation, the same knee-like feature shifts to $q \sim 0.05 \text{ \AA}^{-1}$ in the SAXS data collected at the colliding point in the middle of the pellet. From detonation times beyond 0.57 μ s, little observable shift occurs in these Guinier knees. On the other hand, the Guinier knee obtained from the side position of the colliding detonation continuously shifts from $q \sim 0.15 \text{ \AA}^{-1}$ at 0.26 μ s to $q \sim 0.07 \text{ \AA}^{-1}$ at 1.18 μ s. Conversely, the Guinier knee of the SAXS data at the quarter-point does not change at all. The intensity decays at q -values greater than the Guinier knees deviate slightly from Porod scattering, which is commonly observed in small angle scattering of detonation nanodiamond [31--33].

Scattering intensities vary and evolve differently depending upon measurement position. At early times through $0.26\mu\text{s}$ the intensity of the SAXS data collected from the mid-point of the colliding detonation resembles intensity collected from the conventional detonation; these are less intense than the quarter-point. For a given number density, N , and contrast, $\Delta\rho$, the scaling should be higher since the particulates are clearly larger at the mid-point of the colliding detonation. At times after $0.26\mu\text{s}$, the scaling of the SAXS data collected at the mid-point of the colliding fronts increases dramatically. Therefore, between $0.26\mu\text{s}$ and $1.18\mu\text{s}$, either or both the total number of particulates and/or the x-ray contrast is increasing. Also, note the relative intensities between conventional and colliding vary at high- q . Greater than about 0.2 \AA^{-1} , the colliding case begins less intense than the conventional at $0.26\mu\text{s}$, and increases to more intense as seen in the $0.87\mu\text{s}$ and $1.18\mu\text{s}$ traces.

Thermochemical modeling provides temporal pressure-temperature evolution for both the conventional and colliding Composition B detonations. The colliding detonation results in much higher temperature and pressures near the mid-point where the opposing fronts collide. Figure 3 a indicates in this region, approximately $0.25\mu\text{s}$ after fronts hit center position, a fraction of carbon condensed at pressures and temperatures conducive to the liquid phase. Particulates that form in the presence of liquid carbon extend over a limited region at the mid-point and encompass a small fraction of the overall pellet volume.

The evolution of the temperature and pressure, obtained by thermochemical modeling, superimposed on the carbon phase diagram, appears in Figure 3b. The conventional detonation C-J is deeply within pressure and temperature conditions for diamond as previously demonstrated. [24,25] In the colliding case, the pressure-temperature evolution is highly position-dependent because the x-ray scattering measurement samples all points along a diameter through the part with a beam that is $\sim 100\mu\text{m}$ wide and $50\mu\text{m}$ tall, while the angle between curved detonation fronts locally continuously changes with position from planar at

the central position to angled at the cylinder edge. The two lines enclose a manifold of pressure-temperature states seen along the measurement diameter through the part. The central position exceeds 60GPa and 5000K as fronts collide; the top-right of the pressure-temperature line for the central position at 0.04 μ s after the detonation fronts collide. As time from detonation progresses, pressures and temperatures drop out of the diamond portion of the phase diagram at \sim 0.6 μ s. Based on the extreme pressures and temperatures generated, one might expect particulates to remain deeply in the diamond part of the phase diagram longer than the conventional detonation, and one might expect particulates with significantly differing morphology in the colliding case.

4 Discussion

These results show that detonation carbon condensate morphology varies with parameters beyond simply the composition of the explosive. Particularly, detonations from nominally identical composition B charges produce particles that depend on conventional vs. colliding detonation conditions as well as position within the charge undergoing the colliding detonation.

Colliding detonations produce larger particles in a narrow band in the center of the charge where temperatures and pressures are much higher than the conventional case. Conversely, the SAXS from just a few mm below this central point, at the quarter height position, resembles that obtained from a conventional detonation. These observations agree well with thermochemical calculations that indicate a small region with elevated temperature and pressure conditions approaching the liquid phase portion of the carbon phase diagram (Figure 3). Therefore, the quarter-point vs. center point SAXS data (Figure 2) and thermochemical calculations (Figure 3) together indicate that the particulates formed under different conditions at and near the colliding point constitute a very small fraction of the overall recovered detonation products. Thus, detonation nanodiamond very similar to that

obtained from a conventional detonation dominates particulates observed by TEM imaging (Figure 1). Particulates originating at that central position are not easily isolated.

While the Guinier approximation is sufficient to conclude that larger particulates form at the colliding point, it says nothing about the size distribution and particulate morphology. Additional assumptions and appropriately refined models are necessary to extract this additional information. Many models exist for small angle scattering from detonation nanodiamond and each has its advantages and limitations. In most cases, detonation nanodiamond is considered to be aggregated spherical particles with a surface that is not necessarily smooth nor well-defined, as the high- q scattering does not follow a perfect Porod decay. [12,24,32,34--36] Here, the model accounts for the departure from Porod scattering by including surface texture, as it mimics the morphology observed by TEM imaging and has been proven to reproduce an accurate size distribution from the SAXS data. [24] This model contains four fit parameters: scaling constant K_{DND} , background b , median radius \bar{R} , and a (log-normal) standard deviation σ . This model fits all of the SAXS data collected from the conventional detonation, as well as the quarter-point and side point of the colliding detonations. Details of the model fitting can be found in the supporting information. The result of the fitting is that the mean diameters of the whole particle, \bar{D} , and mean diamond core, \bar{D}_{core} can be extracted from the SAXS data and are shown in Figure 4a and Figure 4b, respectively. In general, ~ 4 nm diamond core sizes are consistent with expected detonation nanodiamond [37].

While the detonation nanodiamond scattering can be modeled based on the TEM imaging and consistent with prior studies, the morphology of the particulates formed at the colliding point cannot be easily assessed by recovered products and must be inferred from the SAXS. By observation, the colliding has a Guinier knee at lower q , more intensity at lower q , a steeper primary particle power law, and significant deviation from Porod scattering at the

high- q , as seen, for example, in the SAXS acquired at 1.81 μs . Similar qualitative SAXS features occur during DNTF detonations. [25] In these DNTF detonations SAXS, thermochemical modeling, and recovered particulates point to relatively larger nanoparticles formed through carbonaceous liquid nanodroplet formation and subsequent solidification during detonation [25]. Based on these qualitative SAXS similarities and thermochemical modeling, we posit that a portion of the particulates in the Comp B colliding detonation form from larger liquid nanodroplets that freeze in similar fashion. A qualitative difference between colliding Comp B and DNTF is that the colliding Comp B retains a very low- q ($<0.02 \text{ \AA}^{-1}$) power-law decay of about 1.8 to 1.9 prior to the Guinier knee, which is characteristic of aggregated particles [38]. Thus, qualitatively the SAXS from the colliding point also retains features resembling nanodiamond from a conventional detonation as well. The chord through the diameter likely sees an ensemble of various nanodiamond and liquid-to-nano-onion structures.

In this study, two candidate models are chosen to fit the data at these diamond and nano-onion extremes: 1) the same detonation nanodiamond model but with enhanced contrast and 2) a model that assumes a small diamond core that is surrounded by graphitic layers. Though many more models could be fit to the data, these two models were chosen because they resulted in the largest disparity in the diameter of the diamond core and because they are physically possible since that Comp B produces diamond, and graphitic layers could arise from either liquid-to-solid transitions observed previously [25], or nanodiamond graphitization under extreme conditions [39]. Details of these model fits can be found in the supporting information.

From the model fitting, clearly much larger particulates form at the colliding point (Figure 4a) and both models obtain similar overall particle diameter \bar{D} sizes. However, the actual internal structure of these particulates remains ambiguous; the nanodiamond fit (closed

circles) and nanodiamond core onion shell fit (open boxes) match data equivalently well. The maximum onion shell thickness obtained during fitting to generate Figure 4b then gives an approximate possible range of particulate core sizes, with the caveat that the particles likely have a distribution of varying morphology for a variety of temperature-pressure conditions as discussed above.

Kinetics of particulate growth appear in the model fits of \bar{D} and \bar{D}_{core} . The quarter-point case exhibits little or no discernible change in size; in the timings used, the detonation front passed the measurement position prior to $t_{det}=0$ and the SAXS is of fully formed particulates throughout the series. The shock from the opposing end is expected to arrive at the particulates at the quarter-height position between 0.87 and 1.18 μs based on the imaging, thus it does not appreciably change particulate size or morphology within the accuracy of the SAXS measurement. Note that particulate size at the quarter-point is slightly smaller than the center conventional detonation. The center point of the conventional case has a value of \bar{D} that is ~ 5.4 nm at 0.11 μs increasing to ~ 5.8 nm through the rest of the time series. The colliding case also shows relatively monotonic growth from ~ 7.4 nm at 0.11 μs to ~ 8.5 nm by 0.41 μs . Generally, primary particulate growth for these charges occurs over the first few hundred nanoseconds and ceases within about 0.3 to 0.4 μs .

We posit the side case provides insight into different mechanisms of apparent particulate growth originating at differing radial locations along the central, colliding plane. Shock release occurs rapidly at the cylinder edges, and very early time particulate formation observed just beyond the surface represents much smaller particulates whose composition cannot be quantitatively determined by SAXS alone; these particulates are about 4 nm in diameter at the first measurable time. The measured particulate sizes increase to over 6.5 nm about 1 μs later and continue to grow. In this region at early times, particulates originating from the near-surface region contribute to the SAXS while as time passes, particulates

originating nearer to the center of the charge increasingly dominate the SAXS as the detonation products expand and encroach along the measurement chord. This implies a distribution of sizes ranging from small, ~ 4 nm particles (with, if diamond, cores possibly as small as ~ 2 nm) to large, ~ 8.6 nm particles originate near the center. Measurement of these varying sizes dependent on detonation conditions opens potential possibilities for collecting varying detonation nanodiamond sizes with similar chemical/atomic composition.

4 Conclusion

This study combines sub-microsecond, time-resolved x-ray radiography, and SAXS, as well as thermochemical modeling to gain a broader understanding of the conditions present during carbon condensation and resultant nanoparticle formation from differing detonation conditions (i.e. pressure and temperature) independent of the explosive composition. The radiography and SAXS can be collected from separate experiments at similar times from nearly identical composition B explosive pellets to resolve the relative density and size of the detonation products as a function of time. When Composition B is initiated from both ends, at the point where the detonation fronts collide, carbonaceous particulates are almost twice the size of the diamond formed during conventional detonations. These large particulates are likely formed under conditions where an appreciable fraction of the carbon is expected to be in the liquid phase, and resultant particulates are likely a mixture of detonation nanodiamond and carbon nano onions. Away from this central region, at 4.15 mm from one of the edges along the diameter of the cylinder, particulates resemble but are slightly smaller than, detonations at the central position initiated from one end. Measuring particulates as they traverse an area just to the side of the explosive charge, particulates at early times are very small (~ 4 nm). Later time measurements show larger particulates that originated from core regions of the charge but move into the measurement position with product mixing and expansion. Particulates of various sizes can be generated by a single explosive under varying

pressure and temperature conditions by colliding two detonation fronts. In this geometry, there is a clear size dependence of the carbon condensates formed at different points of the high explosive.

Symbols and Abbreviations

SBI scatter beam imaging,

SAXS small angle X-ray scattering,

Comp B composition B,

HE high explosives

t_{det} time after detonation front passes the mid-point of the Comp B pellet,

\bar{D} mean whole particle diameter obtained from the SAXS modeling,

\bar{D}_{core} mean diameter of the particle core (assumed to be diamond) obtained from the SAXS modeling

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Figure 1 TEM images showing typical detonation nanodiamond aggregates obtained from the colliding detonation (a) and the electron diffraction obtained from the entire aggregate (b).

Figure 2 Radiographic imaging (top) and SAXS data (bottom) of the Composition B detonation. The radiographic scatter beam imaging sequences, spaced 2 bunches ($0.3068 \mu\text{s}$ apart) were obtained from both colliding (top row) and conventional (2nd row) detonations. SAXS data was obtained from detonation times similar to rightmost five of the imaging times; these temporally synchronous images & SAXS are denoted with commonly colored frames. In each log-log plot of the SAXS data, there are four curves that represent the particulate scattering acquired from the middle of the HE pellet under a conventional detonation (blue), and three from the colliding detonation with middle (red), quarter (green) and off-set (black). The location of each of these is shown in the first scatter beam images on the far left.

Figure 3 Thermochemical simulations of the fraction of detonation products lying within the liquid carbon phase during a small-scale colliding detonation of Comp B at $\sim 0.26 \mu\text{s}$ (a) and the temporal evolution of the pressure and temperature of the colliding detonation (blue) and conventional detonation (green) through the carbon phase diagram for a 6 nm carbon particles. The times relative to the detonation front for the colliding detonation in (b) correspond to $0.04 \mu\text{s}$ (60 GPa) and $0.6 \mu\text{s}$ (~ 10 GPa).

Figure 4 the time evolution of the mean diameter of the whole particle (a) and mean diameter of the core (b) obtained from model fitting of the SAXS data collected from the mid-point (red), quarter-point (green), side-point (black) of the colliding detonation and the mid-point of the normal detonation (blue). The modeling results from the two candidate models applied to the mid-point colliding detonations were a detonation nanodiamond model with enhanced contrast (closed circles) and a model that assumes significant graphitization of the diamond, which results in a diamond core surrounded by onion-like layers of graphite (open squares).