Carbon Condensation During Detonation of High Explosives of Various Diameters

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Abstract. This paper presents experimental data on monitoring of condensed carbon nanoparticles during the detonation of TNT/RDX charges 20, 30 and 40 mm in diameter. The dynamics of nanoparticles sizes were derived from processing of measured distributions of small-angle X-ray scattering (SAXS). SAXS is investigated via registering the diffraction signal from a sample in the small area. This method is widely used for analysis of the structure of disperse systems. The work was carried out at the accelerator complex VEPP-4M (BINP SB RAS). Nanoparticles of 4 nm size was detected behind the detonation front, after that the average size of particles increases up to 9 nm during 3–4 microsecond for all charges.

Introduction

Studies of carbon condensation in detonation of oxygen-deficient HEs were initiated at Lavrentyev Institute of Hydrodynamics in 1983 in the context of works on synthesis of detonation nanodiamonds. Now there are over 1000 papers[1, 2] on this theme in the world. Most studies investigated the saved detonation products (soot) collected after an explosion.

In the early 2000s, studying of triaminotrinitrobenzene (TATB) in the nuclear centers of Russia and USA renewed interest of carbon condensation in detonation of high explosives (HEs) with negative oxygen balance. The results[3, 4] have shown that the assumption of the additional allocation of energy behind a chemical reaction zone better describes the experimental data on acceleration of the metal plates. The results of experiments can be explained by the additional energy released due to the exothermic coagulation of carbon clusters. Because of that the problem of carbon condensation at detonation of oxygen-deficient high explosives is still open to question. It is important both for understanding the physics aspect of the phenomenon and for evaluation of the amount of energy released at coagulation of carbon clusters.

At the same time there were the first experiments on the dynamic registration of small-angle X-ray scattering (SAXS) distributions, which showed a long signal growth in the detonation of TNT/RDX charges.[5]

In recent years, small-angle X-ray scattering (SAXS) of synchrotron radiation (SR) has been widely used to study carbon condensation during an explosion.[6, 7, 8, 9] SAXS patterns depend on nanoscale density fluctuations in the detonation process. During the detonation of oxygen-deficient high explosives (HEs), the occurrence of these fluc-

tuations is related to the formation of condensed carbon in different phases. For the time being, sizes of condensed carbon nanoparticles at detonation of high explosives can be experimentally registered only through diffraction methods using synchrotron radiation. The ability to use short high-periodical SR flashes of high-energy accelerators allowed us to reveal the time evolution of the signal at detonation of HEs.

A new experimental station on accelerator complex VEPP-4M was put into operation in 2015. This new station gives much higher SR intensity than the similar station 'Extreme States of Matter' on the VEPP-3 accelerator complex. It allowed us to X-ray explosive charges of larger diameter (up to 40 mm) and increase the mass of the charges studied by ten times up to 200 grams.[10]

Similar experiments was carried out at the Advanced Photon Source (Argonne National Laboratory, USA).[8, 11, 9, 12] These studies investigated small charges of about 10 mm in diameter, but dynamics of carbon condensation in big charges still remain an actual problem.

Earlier, experiments at the VEPP-3 accelerator complex showed a long time of carbon condensation behind the chemical reaction zone during the detonation of HE charges up to 20 mm in diameter. For TNT/RDX charges this time was about 1 μs.[7, 13]

Here we present the experimental data on particle size dynamics during TNT/RDX detonation of different diameters (20, 30 and 40 mm). The study of TNT/RDX is important because this explosive is the main one used in the commercial production of ultrafine diamonds.

SAXS measurement experiments

Cylindrical cast charges of TNT/RDX of 20, 30 and 40 mm in diameter and a length of 55 mm (weighing 29, 65 and 120 g respectively) was used. HE charges were initiated by an electric detonator through a plane wave generator.

The SAXS measurements were carried out on new station located at accelerator complex VEPP-4M. The experimental set-up is described in ref. [10, 14, 15].

The high-energy accelerator was operated in a 'standard' two-bunch mode with a current of

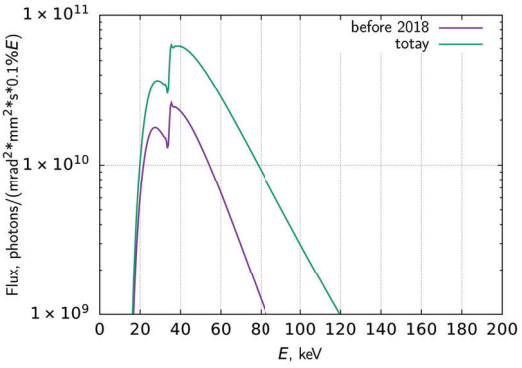

Fig. 1. Real radiation spectrum at the station.

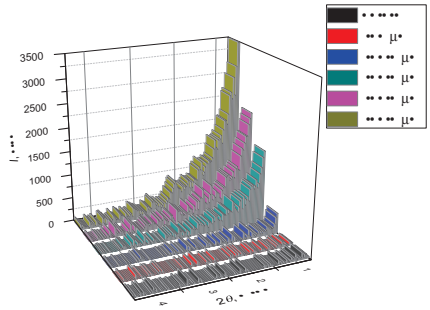

Fig. 2. The SAXS dynamics of detonation of cast TNT/RDX charges 30 mm in diameter

10 mA, yielding SR pulses of 73 ps and 611 ns spacing between pulses. A new 9-pole wiggler was used for generation SR beam on station. It create a non-monochromatic SR beam whose real spectrum shown in figure 1. Real radiation spectrum include wiggler spectrum, sample absorption and registration efficiency of the gas detector.[10, 15] The possibility of using such 'pink' SR beam was shown in ref. [10, 15].

SAXS was recorded with a special DIMEX-3 detector [16] with an angular resolution of 3×10^{-5} rad, located at 3432 mm from the explosive charges.[14, 15] The detector was filled with a mixture of Xe-CO_2 (75%-25%) at a pressure of 7 bar. The distance between detector channels is 0.1 mm. The detector is capable of recording 100 frames every 124 ns.[16]

The SAXS intensity distributions during the det-

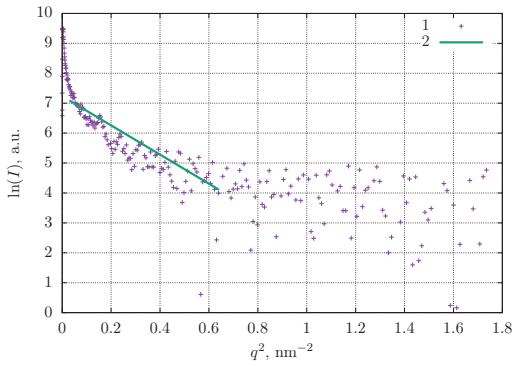

Fig. 3. 1 – SAXS for TNT detonation at 6 μs behind the front, 2 – Guinier approximation.

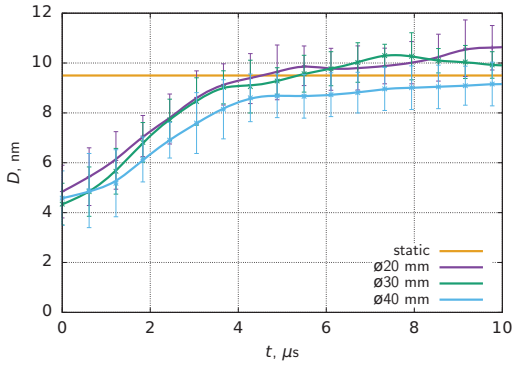

Fig. 4. Average size of carbon particles versus time in detonation of TNT/RDX charges.

onation of TNT/RDX charges are shown in figure 2. Each colored line represents the distribution of SAXS signal at a certain time. The sequence of the angular distributions gives the time dependence of the process or its dynamics. Time is measured from the moment of passage of the detonation front.

The sizes of the scattering centers were determined using the Guinier approximation:

$$I(q, R) = I_0 \exp(-q^2 R_g^2/3), \quad (1)$$

where R_g is the radius of gyration of the particle, $q = 4\pi \sin(\theta)/\lambda$ is the scattering vector, λ is the scattered radiation wavelength and 2θ is the scattering angle [17, 18]. For a spherical homogeneous particle of radius R, $R_g = \sqrt{3/5}R$.

Taking the logarithm of (1):

$$\ln(I(q, R)) = \ln(I_0) - q^2 R^2/5, \quad (2)$$

we obtain a function which decreases linearly versus q^2. One can restore the size of the spherical particle $D = 2R = 2\sqrt{-5k}$ using the slope k of this line.[17, 18]

Figure 3 represented the Guinier approximations at the 6 μs behind the detonation front in one experiment on detonation of TNT/RDX charge of 30 mm in diameter. The slope of straight line approximation is k $\approx$ -4.9, corresponding to the scattering of particles of the diameter of D $\approx$ 9.9 nm. After processing all frames we obtained dynamics of average particle sizes for one experiment. Figure 4 presents the dynamics of the average size of nanoparticles for all experiments during the detonation of TNT/RDX charges of 20, 30 and 40 mm in diameter.

The dynamics of nanoparticle sizes (figure 4) obtained under the following assumptions. The scattering centers are a sphere with uniform density (excluding the phase state). Carbon in the diamond form gives a more intensive SAXS signal. In addition to that we replacement the real radiation spectrum by a monochromatic effective energy and the size distribution of nanoparticles by average size.

Results

SAXS dynamics in detonation of cast TNT/RDX charges of 20, 30 and 40 mm in diameter were measured. Dynamics of the average size of the condensed carbon nanoparticles behind a chemical reaction zone were recovered from the SAXS distributions in the Guinier approximation. The minimal particle size registered in experiments is 4 nm. Particles of this size are registered immediately behind the detonation front. Then the average size of particles increases. The time of fast growth of scattering centers up to 9 nm is 3–4 microseconds (figure 4).

The experiments do not allow us to speak about the phase state of the nanoparticles. However, a further increase in the intensity of SR will enable us to measure the diffraction at large angles, and, consequently, to determine the phase composition of condensed carbon in the detonation products.

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