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Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Application of introduced nano-diamonds for the study of carbon condensation during detonation of condensed explosives

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ARTICLE INFO

Available online 3 January 2009

Keywords:

Small-angle X-ray scattering

High explosive (HE)

Products of detonation

Nano-diamonds

ABSTRACT

This paper describes experimental studies of the formation of nano-diamonds during the detonation of TNT/RDX 50/50 mixture with small-angle X-ray scattering (SAXS) method at a synchrotron radiation beam on VEPP-3 accelerator. A new experimental method with the introduction of nano-diamonds into the explosive has been applied. Inclusion of the diamonds obtained after detonation into the TNT and RDX explosives allows modelling of the case of instant creation of nano-diamonds during detonation.

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1. Introduction

Although quite large number of publications discusses the synthesis of nano-diamonds during an explosion, the question of its formation and even more common problem of carbon condensation at detonation of an explosive with negative oxygen balance is being under discussion till the present time. An answer to this question is important for both the understanding of physics of the phenomenon and the estimation of energy which is generated during exothermal coagulation of carbon clusters.

The first direct experimental studies of growth of an exploded nano-diamonds became possible due to the application of synchrotron radiation (SR), namely the possibility of the detection of photons after small-angle diffraction with a frequency up to 4 MHz. The results obtained demonstrated that the intensity of small-angle X-ray scattering (SAXS) signal started to grow from zero at the detonation front and its growth continued during several microseconds [1,2].

A hypothesis suggested based on this result that a creation of carbon particles (including nano-diamonds) takes place beyond the chemical reaction zone (in this case its size is around 0.7 mm). However, this result can be used as well as confirmation of instantaneous creation of exploded nano-diamonds. One has to suppose that the low value of SAXS intensity at the detonation front is due to the small *contrast* of nano-diamonds. In SAXS method the intensity of scattered signal at the first approximation is proportional to the squared difference of densities of the

scattering particle and the medium (detonation products—DP). At DP density in the beginning of about 2 g/cm³ the signal can be too small to be detected and only during DP decay, i.e. when the density is reduced, it can be registered. This can also explain a tendency for the increase of the zone of the signal maximum delay with the diameter of the explosive [2], as DP decay will take more time in such a case.

In the present work the study of the starting point of nano-diamonds formation is performed during the detonation of the mixture of TNT/RDX 50/50 with SAXS method at the SR beam of VEPP-3 accelerator. A new experimental arrangement is applied with nano-diamonds introduced beforehand into the charge of high explosive (HE). The known initial presence of nano-diamonds in pressed HE allows to determine the dynamics of exploded nano-diamond growth with better precision.

2. Experimental technique

Experimental work has been performed in Budker INP (at VEPP-3, wiggler with 2 T field, average X-ray energy of 20 keV, bunch frequency of 4–8 MHz and bunch duration of about 1 ns). Electronic circuit of the detector allows the recording of 32 frames with the position distribution of scattered photons performed every 500, 250 or 125 ns. In the last case the storage ring accelerates two bunches rotating at the opposite points at the orbit. As the average current of the accelerator have to stay the same, this arrangement leads to the decrease of the signal to noise ratio (the dispersion of experimental points is larger), but allows improvement of time resolution.

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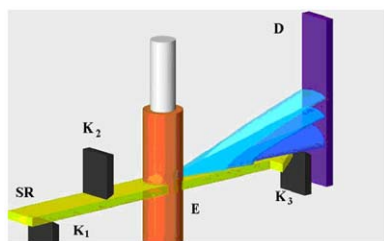


Fig. 1. Layout of the SAXS experiment using synchrotron radiation.

The experimental set-up is shown in Fig. 1. The SR beam was collimated with the bottom (K1) and top (K2) edges, and irradiation zone was formed at the central part of the explosive charge with 1 mm height and 12–15 mm width. In front of the detector (D) the direct beam was closed with another bottom edge (K3). Scattered photons were registered by the detector. For additional monitoring a part of the direct beam irradiated the detector through an absorber (1 mm thick copper filter). The distance between the edges (K1) and (K2) was ~ 200 mm. The HE charge was at the distance of 700 mm from the edge K2 and at ~ 640 mm from the edge K3. The distance between the detector and the edge K3 was ~ 200 mm. Scattered radiation was registered by the detector DIMEX [3] with the vertical channel pitch of 0.1 mm. Thus the angular range covered by the measurements was ~ 0.0068 – 0.34° . During one flash of SR the detector recorded all channels (make one frame) and measured the distribution of SAXS as a function of the angle. As the detonation front moved along the charge axis with a constant speed 7.6 km/s the series of frames gave the time dependence of the SAXS distribution.

In the investigated HE TNT/RDX 50/50 the duration of the chemical reaction zone is ~ 0.1 s [4] and the measurements of SAXS were performed every 125 ns. VEPP-3 was operating in a two-bunch mode for this with bunches rotating in the opposite points of the orbit. To realize this regime the problems of positioning, focusing and phase parameters of the bunches were solved.

3. Measurement of the dynamics of SAXS from HEs with introduced nano-diamonds

The intensity of SAXS from mono-dispersed system of nano-particles can be described by the following formulae:

$$I(s) = n(t)(\rho - \rho_0)^2 F(s),$$

where $\mathbf{s} = \mathbf{s} - \mathbf{s}_0$ is the vector of scattering ($|\mathbf{s}| = 4\pi \sin \theta/\lambda$), 2θ the scattering angle, λ the wavelength of radiation, $n(t)$ the number of nano-particles of density ρ in a unit volume, ρ_0 the density of the medium, and $F(s)$ the form-factor of an individual particle.

In the present experiments the maximal registered scattering angle was $2\theta_{\max} = 0.014$, minimum angle was $2\theta_{\min} = 0.0006$. Integration in this range of angles leads to the following formulae for the integral intensity of SAXS:

$$I_0(\lambda, R) \sim R^6 (\rho - \rho_0)^2 N. \quad (1)$$

Total SAXS intensity can be obtained by integration through all wavelengths according to the spectrum of irradiation from VEPP-3 storage ring.

In the performed experiments the intensity in Eq. (1) is summed from scattering on atoms (molecules) and scattering on nano-particles. The analysis shows that scattering on atoms is constant and small due to strong dependence on the size in Eq. (1). The registered dependence can be obtained only from relatively large particles of ~ 2 – 4 nm size. In the conditions of the performed experiments the minimal size of detected particles is

equal to $d_{\min} \approx \lambda/\theta_{\max} \approx 2.8$ nm, where $\lambda = 0.04$ nm is the wavelength at the energy $E = 30$ keV.

Maximal size of particles is determined by the minimal registered scattering angle θ_{\min} , which is given by the accuracy of the alignment of the edge that is closing the direct SR beam (around 0.5 mm). For the present experiments $d_{\max} \approx \lambda/\theta_{\min} \approx 70$.

SAXS method does not allow the distinguishing of signals from nano-diamonds and non-diamond carbon forms. As shown in Ref. [5] the fraction of nano-diamond in a solid residue can reach 80% (for initial HE TNT/RDX 50/50).

In all experiments detecting SAXS [2] the signal started from zero in the zone of maximal compression. One of the possible reasons of such a behavior of SAXS can be the very low contrast $(\rho_D - \rho_1)^2$ of nano-diamond in this moment. At the density of HE in the chemical reaction zone of about $\rho_1 \sim 2.1$ g/m³ the SAXS signal might not be visible for the detector and only during the decay of DP, i.e. the reduction of the density, it becomes detectable. Thus it is necessary to know if this method can distinguish condensed nano-diamonds ($\rho_D \sim 3.5$ g/cm³) at the background of shock compressed HE.

In order to answer this question the experiments on SAXS measurements were performed with the detonation of TNT and RDX with the introduced 8% (weight fraction) of nano-diamonds produced by NPO "Altaj". Such an amount of nano-diamonds corresponds to its yield at the detonation of TNT/RDX 50/50 [5].

The results of the measurements of SAXS are shown in Fig. 2. The initial signal level in TNT and RDX (curves B and D) corresponds to 8% of introduced nano-diamonds. At the moment when detonation front passes through (zero moment in the figure) their SAXS signal is reduced proportionally to the change of contrast. For RDX the change of signal has to be equal to a factor of $(\rho_D - \rho_1)^2/(\rho_D - \rho_0)^2 = 1.89$ where $\rho_D \sim 3.5$ g/cm³ is the density of nano-diamonds, $\rho_0 \sim 1.71$ g/cm³ is the initial density of RDX and $\rho_1 \sim 2.2$ g/cm³ is its maximal density.

For TNT the reduction of signal is equal to a factor of $(\rho_D - \rho_1)^2/(\rho_D - \rho_0)^2 = 1.6$, where $\rho_0 = 1.69$ g/cm³ and $\rho_1 = 2.08$ g/cm³ are the corresponding densities for TNT. From the plot one can see that the level of SAXS signal is one order of magnitude higher than the zero level (noise level). If at detonation of TNT/RDX 50/50 all nano-diamonds were produced within a narrow chemical reaction zone (in ~ 0.1 μ s), the jump of SAXS signal to the same level as in TNT and RDX with introduced nano-diamonds must be observed in this zone. The behavior of SAXS for

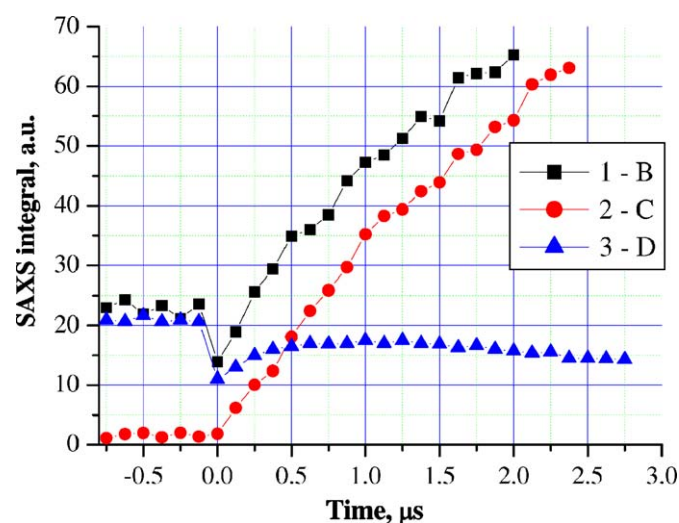


Fig. 2. Dependence of integral small-angle scattering on time at the detonation of charges of TNT (B), RDX (D) with the introduced 8% of ultra-dispersed diamonds, and also of mixture of TNT/RDX 50/50 (C).

TNT/RDX 50/50 (curve C) demonstrates that in the chemical reaction zone the signal is practically equal to zero and minimal level of curves B and D is reached after $\sim 0.75 \mu\text{s}$ that significantly exceeds the duration of the chemical reaction. The *contrast factor* in experiments B and C is practically the same as their decay dynamics of the explosion products differs a little. The SAXS signal in TNT/RDX 50/50 smoothly grows due to the increase of the number of nano-particles and their sizes (1), which does not confirm the instant creation of the detonation of nano-diamonds. Due to specially taken measures the accuracy of time alignment of the results of different experiments is not worse than 20 ns. Phase shift between curves B and C at similar level of the signal significantly exceeds this value.

Behind the detonation front the curve of SAXS of RDX with nano-diamonds passes far below the one of TNT and in spite of that the decay of DP in these two cases is approximately the same. Such a behavior of SAXS means the destruction of nano-diamonds in the decaying DP of RDX. And *combustion* happens not instantly, but behind the chemical reaction zone.

4. Conclusion

The performed experiments demonstrate that the proposed method of measurement of SAXS reliably distinguishes exploded nano-diamonds at the background of substance compressed in a detonation wave. At the detonation of TNT/RDX 50/50 carbon condensation into nano-diamonds (with sizes $> 2 \text{ nm}$) occurs behind the chemical reaction zone.

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