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Electrical conductivity distributions in detonating low-density explosives – Grain size effect

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ABSTRACT

Electrical conductivity profiles were measured in three detonating high explosives with initial densities slightly greater than loose-packed ones. Behind the detonation fronts, the conductivity peaks were found presumably correlating with the reaction zones. A distinct grain size effect was demonstrated: the conductivity peaks were clearly shorter in fine-grained materials. This suggests that the chemical reaction rate is faster for smaller grains. This supports the concept of a "hot spots" reaction mechanism prevailing at comparatively low pressures.

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1. Introduction

Determining the structure of a chemical reaction zone is perhaps the most difficult problem in detonation physics. Currently, the reaction course is traced using a marker sensitive to the evolution of the mechanical state of the material. The time resolution of embedded gauges (e.g., the electro-magnetic ones [1]) is limited by inertia, and the flow around the gauge is evidently disturbed. The interface between the explosive and inert window is also used as marker (laser interferometer methods for example [2]). In this case, the reflection at the interface is the major disturbing factor. Some window materials provide good impedance matches with explosives in general. However, ideal matching throughout the reaction zone seems to be questionable because of sharp changes in the parameters during the reaction. The measurements should therefore be accompanied by modeling. Since the model is usually adjusted to reproduce a variety of practically important features like shock initiation, acceleration of metal layers, etc., the kinetics in the thin reaction zone may be described in a rather rough way. This is probably one of the reasons why the data concerning the reaction zone structure and the characteristic reaction time are often contradictory. Alternative approaches are therefore of interest. In this paper we report the results of a high-resolution electrical conductivity method, which is virtually non-perturbative and does not require model fitting. A limitation of this method is that the conductivity value, at least at present, cannot be directly related to the dynamical state of the explosive. Therefore the conductivity data should be used preferably as a comparative material. In this work, we examine the influence of grain size on the conductivity distribution in the detonation wave front.

Most authors agree that the reaction behind the shock front starts in multiple local ignition sites called hot spots, and then spreads to engulf the rest of the volume. However, an *a priori* way to estimate the density of hot spots does not exist. In practice it is adjusted independently of the other parameters, or is not explicitly introduced to the models at all.

One may expect that explosives with small grain sizes should have higher hot spot density and consequently a faster reaction rate. Indeed, to the certain extent, the shock-to-detonation transition is easier in fine-grained materials. At some grain size the shock sensitivity reaches a maximum, because too small hot spots cannot be self-sustaining. Thus both the increase of sensitivity and its subsequent decrease are easily explained within the hot spots framework. However, no marked influence of the grain size on the reaction time has yet been found for the stationary detonation wave. The potential effect of initial grain size may be eliminated when the explosive is compacted to a high density (which is the case for most studies). Evidently the majority of pores are closed upon compaction, and fresh surfaces appear instead of old ones.

Search for the grain size effect on reaction rate seems therefore to be more promising for low-density explosives, in which the initial structure of the powdered charge is not destroyed. In an earlier work we presented some preliminary results supporting this suggestion [3]. The study of the detonation wave structure in low-density explosives of different grain sizes using the electrical conductivity method is reported below.

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2. Experimental methods

There is a long history of electrical conductivity measurements in detonation waves [4,5]. However, the specific difficulties limiting the resolution of the measuring cells in an aggressive medium were resolved much later. The key idea was to use the electric field perpendicular to the conductivity gradient [6] to minimize parasite current spreading and distortions caused by space charge (which were first discussed in [5]). In later works the method was improved and applied to the detonation of high-density explosives [7–9] and to metastable intermolecular composites [10]. The measurement procedure is discussed in length in [8,9]. We provide only a brief summary here.

The coaxial cell used is shown in Fig. 1. An explosive charge with a diameter $b = 8$ mm was placed in an axial channel of the copper cylinder (40 mm outer diameter) made of two sections threaded together. A copper electrode ($c = 2$ mm in diameter) was mounted along the charge axis in the PMMA plug, and was supported by the bolt. The conductivity gauge, a small Rogovsky coil, was inserted into a special chamber within the outer electrode. A thin slit connected the gauge chamber to the coaxial cell volume. The slit width was fixed by the dielectric layer. The mutual inductance M between the gauge and the chamber contour was 10–15 nH. The explosive was initiated through a 2.4 mm axial opening drilled in a PMMA stopper to maintain the cylindrical symmetry. The external source produced a nearly constant feeding current I (usually 30 A), which was distributed between the cell and the shunt resistor R_s.

The cell becomes conductive as the detonation wave reaches the central electrode. The cell current flows around the gauge chamber. A magnetic flux change across the coil produces an output voltage pulse U. This first pulse was utilized for real-time gauge calibration. As the wave passes by the slit, the current around the gauge decreases, as does the magnetic flux. The pulse $U(t)$ is again generated with an inverse polarity. This second pulse represents the electrical conductivity σ in the slit plane:

$$\sigma(x) = \frac{\ln(b/c)}{2\pi DM} \frac{U(t)}{V}. \tag{1}$$

Here D is the detonation velocity, V is the feeding voltage across the cell, and $x = Dt$ is the distance between the detonation front and slit at a time t. The relation (1) assumes that the wave is stationary as well as the moving conductivity distribution. Actually somewhat smoothed voltage U_{in} is observed due to the coil inductance $L \approx 1$ μH, and the corrected voltage in Eq. (1) is $U = U_{in} + (L/R)dU_{in}/dt$; here $R = 50$ Ω is the input oscilloscope impedance.

The spatial resolution was estimated [9] to be about a quarter of the slit width. Dielectric layers made of PMMA or polyvinyl-chloride 0.2–0.3 mm thick were used in the tests providing the
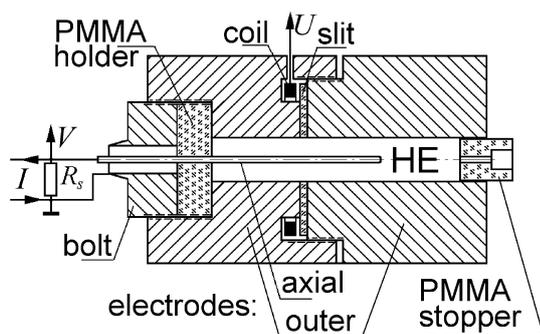
resolution of 50–75 μm (about 10 ns). Thinner slits could be prematurely shorted due to deformation of electrodes by the detonation flow. In such cases the magnetic flux in the gauge chamber is frozen and the $U(t)$ signal is zeroed.

Let us briefly discuss the gas-dynamical perturbations in this conductivity cell. Both the outer casing and the central electrode are deformed by the detonation pressure. The inclination angle of the electrode surface behind the detonation wave front was estimated to be less than 3°. This will produce a rather weak flow expansion, and is not likely to introduce significant distortions. At the tip of the central electrode the reflected shock is inevitable, as is a small change in the flow area. To minimize this problem, the tip was placed far enough from the slit (about 15 mm). This distance is equal to several dozen reaction zone widths, or about eight electrode diameters, so the flow at the measuring plane is steady enough. The leak of detonation gases through a thin slit is also small. In summary, the gas-dynamical disturbances in our system are weak because the detonation generally propagates along the boundaries, so neither reflected shocks nor sharp rarefactions occur.

Secondary explosives cyclotrimethylene-trinitramine $C_3H_6N_6O_6$, or hexogen (RDX), cyclotetramethylene-tetranitramine $C_4H_8N_8O_8$, or octogen (HMX) and pentaerythritol tetranitrate $C_5H_8N_4O_{12}$ (PETN) were tested in this work. The same three explosives were previously studied at high densities [8,9]. Quite non-uniform conductivity profiles were obtained: the distinct peak at the wave front was followed by the "tail" where the conductivity was several times lower. The durations of the peaks were found to be close to characteristic reaction times determined by conventional methods. This allowed us to assume that the conductivity peak is produced by the ionization accompanying the reaction. It can therefore be used as an indicator for the reaction in more general situations. It is notable that no influence of the grain size on the conductivity peak was found in dense explosives.

In this paper low-density explosives were tested. Electrical conductivity distributions behind the detonation front were compared for two grain sizes. The coarse-grained explosives used were typical commercial ones, with a grain size of hundreds of μm. Fine-grained materials (dozens of microns) were prepared from warm acetone solutions by adding water. Photographs of the powders used are shown in Fig. 2. The figure shows that in all cases the typical grain size in fine-grained materials was distinctly smaller than that in coarse-grained ones. However, the initial powders contained certain part of fines. The average particle sizes from the images are presented in Table 1.

The charges were slightly pressed to eliminate large voids and agglomerates that are common in loose-packed explosives (especially fine-grained ones). The degree of compaction was adjusted for each explosive to obtain material that was uniform enough while maintaining the initial structure. The fine-grained explosives were harder to compress, but we succeeded in obtaining nearly equal densities for both types of the same explosive by applying a moderate force. The explosive was loaded by 5 mm increments, and the slit was situated near the middle of one of these portions. The actual density was estimated from the detonation velocity D, which was measured using characteristic changes of $U(t)$ and $V(t)$ signals and known distances. $D(\rho)$ dependences from [11] were used.

3. Experimental results

Six tests were done for each explosive, three with the common grain size and three with the fine-grained material. Typical conductivity graphs are shown in Fig. 3. In all cases a distinct initial conductivity peak was observed followed by a "tail", i.e., a region

Fig. 1. Schematic of the experimental cell. HE indicates a high explosive. The detonation wave propagates from right to left. U and V are the recorded signals.
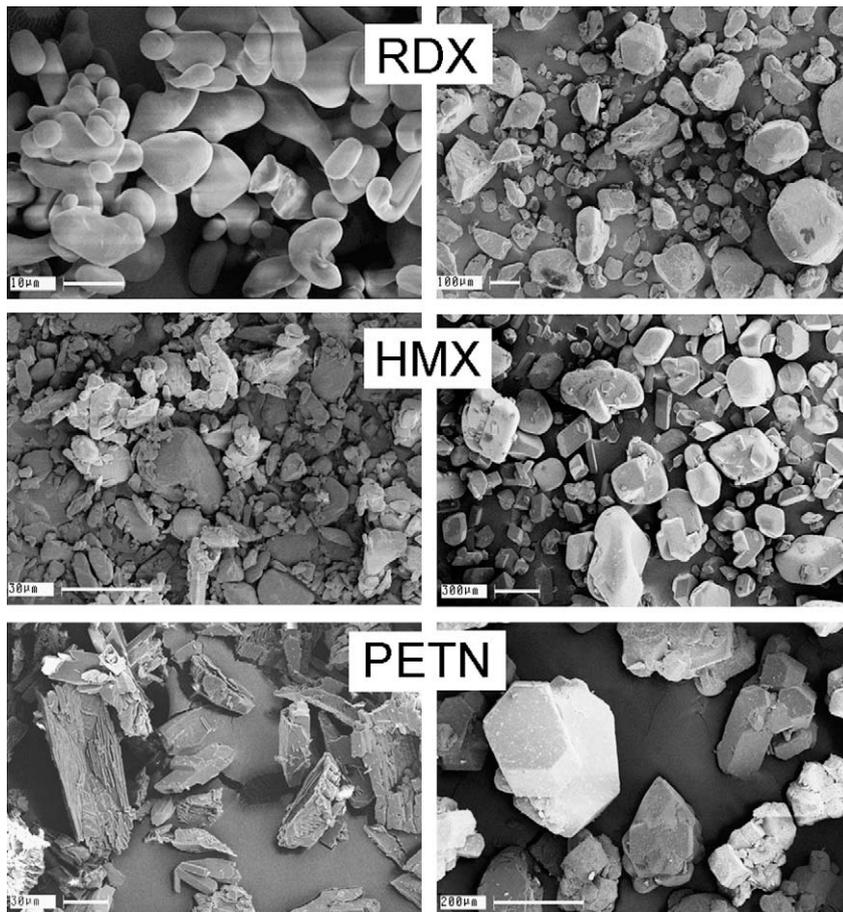
Fig. 2. SEM micrographs of the powders used. Left column: fine-grained samples. Right column: coarse-grained samples. Scale bar, from left to right for RDX: 10 and 100 μm, for HMX: 30 and 300 μm, for PETN: 30 and 200 μm.

Table 1
Results of experiments.

Explosive	Type, d (μm)	Shot #	ρ (g/cc)	σ_{max} (S/cm)	Δ (mm)	τ (ns)
RDX	Coarse 160	102	1.20	1.72	0.36	56
		105	1.17	1.56	0.43	68
		143	1.15	2.1	0.28	45
	Fine 11	140	1.19	0.94	0.28	44
		141	1.18	1.77	0.16	25
		144	1.20	2.25	0.15	23
HMX	Coarse 430	146	1.28	3.3	0.43	64
		148	1.30	2.0	0.55	80
		150	1.31	3.2	0.43	63
	Fine 21	147	1.29	2.6	0.25	37
		149	1.36	3.9	0.25	35
		151	1.35	4.4	0.22	31
PETN	Coarse 260	152	1.05	0.50	0.59	102
		154	1.07	0.49	0.61	106
		156	1.08	0.78	0.39	67
	Fine 80	153	1.11	0.60	0.31	52
		155	1.09	0.52	0.43	73
		157	1.12	0.73	0.31	52

ρ is the initial density, σ_{max} is the maximum conductivity measured in the peak, Δ is the peak width, and τ is the peak duration. The mean particle diameter d indicates that half of the total powder mass consists of particles whose volumes are less or equal to $\pi d^3/6$.

of much lower and slowly decreasing conductivity. The threshold point between these regions was defined as an intersection of straight lines fitting the tail and the rear part of the peak. This procedure, though somewhat arbitrarily, did not introduce significant data spread against other sources of experimental scattering.

An explicit effect of grain size on the peak width was detected. Fig. 3 shows that the peak is much narrower for fine-grained explosive, although the tails are quite similar for both types. All experimental results are summarized in Table 1. The experimental scattering that could not be eliminated in low-density explosives
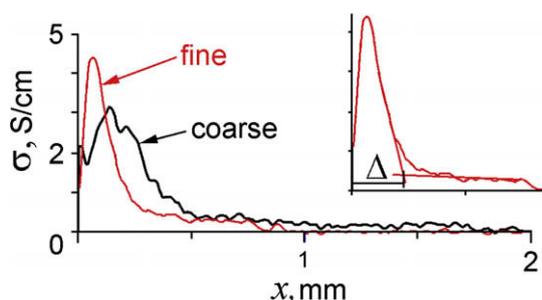
Fig. 3. Conductivity profiles in two HMX charges of different grain sizes. Conductivity σ in S/cm, S (Siemens) is $1/\Omega$. Coordinate x is measured from the detonation front. The records are terminated at ≈ 0.8 mm (shot 151, fine-grained) and at ≈ 1.8 mm (shot 150, coarse-grained) because of slit shortening. The definition of the peak width Δ is shown in the upper right.

is present. In PETN "coarse" and "fine" intervals of the peak widths overlap (shots 155 and 156), and in RDX results from shots 140 and 143 are quite similar. Nevertheless, all tests can be arranged into pairs in which the "fine" peak is shorter than the "coarse" one. For each individual explosive the confidence level of grain size effect was about 90% or more. Due to the small size of statistic samples these estimates are rather crude. In order to take all 18 tests into account, the peak widths were normalized (i.e., divided by average coarse-grained value for each explosive). Average ratio of the peak width in fine-grained explosives to that in coarse-grained ones was 0.57 for all experiments. The Student's t-test gives a value of about 5.77 and the significance level of the null hypothesis (that the "fine" peaks are accidentally shorter) is 3.7×10^{-5}. The latter number seems to be small enough to verify that the grain size does affect the peak width.

The shape of the peak is also grain size dependent. In coarse-grained explosives the noise was present with the spatial scale of about one grain size, while the profiles were smoother for fine grains (see Fig. 3). This is natural since the cell averages the non-uniformities around the axis. When the grains are smaller there are a larger number of them taking part in this averaging. Similar behavior was observed in VISAR experiments with low-density explosives [12].

Note that some experiments, especially at small grain sizes, approached the resolution limit, thus the corresponding peaks may be somewhat distorted. Nevertheless, since the same data handling procedure was used throughout the experiments, the grain size effect on the peak width is reliably demonstrated.

4. Discussion

In contrast with the most methods in which mechanical parameters are measured, the electrical conductivity method is nearly non-perturbative. It is also direct, i.e., it does not involve any kinetic model to obtain results. A disadvantage of this method is the lack of a strict connection between the conductivity and energy released in the reaction. The results are therefore of a qualitative nature and it is desirable to compare them with conventional approaches.

In dense explosives the durations of conductivity peaks agree with the "mechanical" reaction times within the dispersion of literature data [8,9]. It is reasonable to suppose that at low density the same correlation holds. The following discussion exploits this assumption.

According to previous studies, reaction rate increases with initial explosive density [13], but does not depend on grain size [2,13]. Our results show that the conductivity peak in fine-grained low-density explosives is shorter than in coarse-grained ones. Moreover, the

peak may be narrower than for the same explosive compacted to nearly the theoretical maximum density (TMD). These findings can be explained using the concept introduced by Dremin [14]. Two main processes are assumed to proceed in parallel, namely the hot spot mechanism and homogeneous decomposition. The hot spots govern the low-pressure domain while the homogeneous reaction prevails at pressures higher than about half the Chapman-Jouguet pressure at TMD. This explains why the grain size does not matter in a stationary wave at high initial densities, and is in agreement with our conductivity data [8,9]. At low pressure the relative contribution of the hot spot mechanism increases and grain size becomes important. With sufficiently small grains, the effective reaction rate can exceed that at high density, which presumably leads to narrow conductivity peaks. Such an effect should be most pronounced in loose-packed explosives.

One may observe that the decomposition at high pressures needs not be strictly homogeneous. The hot spots may be replaced by multiple local heterogeneities like triple shock configurations. For such cases Dremin's arguments can be modified: the pressure determines whether the scale of the reaction spots is determined by the initial charge structure, or this scale is dynamically generated in the course of wave development. With this generalization, Dremin's concept can be applied to our data equally well.

Although grain size effect on the peak width is certainly present, its functional dependence is not as simple as a direct ratio. There are no grounds to expect a direct proportion between the reaction time and grain size. Even if this proportion were the case, the effect will be blurred in polydisperse powders.

Comparative measurements of mechanical wave structure would be of interest. Unfortunately, at low densities, fluctuations complicate the laser interferometry at the interface [12]. The method [15] using thick foils which provide averaging of the signal demonstrated peculiarities of grain size effect in some way similar to that seen in the present work. New approaches like dynamic X-ray measurements that use a synchrotron radiation source [16] as well as protonography [17], both employing natural averaging along the ray direction, could provide important information if their resolution were improved.

The explosives studied produce moderate quantities of the solid carbon. In an opposite case, for example, in trinitrotoluene (TNT) free carbon contribution to the conductivity dominates because of its high yield. It is not yet clear whether the chemical reaction zone can contrast against a background of high quasi-equilibrium carbon-produced conductivity. We did not observe a peak for TNT [3], perhaps due to limitations taking effect at high conductivity level. In the non-stationary window version [18], a peak of ~ 250 S/cm was reported, though it may have been produced by gas-dynamic disturbances. RDX, HMX, and PETN have lower yields of free carbon, the "tail" conductivity is rather low, and distinct conductivity peaks rise above this background.

In conclusion, the high-resolution conductivity method gives useful information that is complementary to the data from conventional methods on the reaction within the detonation wave. This work presents evidence that the reaction rate is higher in fine-grained explosives at low charge densities.

Acknowledgments

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