

Investigation of the Reaction Zone in Heterogeneous Explosives Substances Using an Electrical Conductivity Method

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A technique for measuring the electrical conductivity profile behind a detonation wave front with a resolution of about 0.1 mm was used to analyze the reaction zone in heterogeneous explosives. TNT–RDX mixtures, RDX with additives of water, NaCl, and a saturated aqueous solution of NaCl, and pure RDX of low density were studied. It was shown that the particle size of the explosive can have a significant effect on the structure of the reaction zone. The most narrow conducting zone (0.22 mm) was observed in fine RDX of density 1.2 g/cm³.

Key words: detonation, explosives, chemical reaction, electrical conductivity, meso-processes.

INTRODUCTION

Almost all widely used explosives are heterogeneous. In commercial composite high explosives (HEs), TNT/RDX compositions, plastic-bonded HEs, and others, heterogeneities play an important role in detonation initiation. In individual HEs, the presence of heterogeneity is also obvious (although it is often considered a secondary factor).

Recently, research on the effect of heterogeneity on detonation has gained in importance. A detonation wave in a heterogeneous system is a source of powerful mixing of substances and their interaction. It is only due to such mixing that mixtures of a fuel and oxidizer can detonate. In individual HE, including pressed ones, the detonation front also causes severe mechanical activation. The substance is divided into hotspots (sites of energy concentration) and relatively cold microvolumes. The temperature difference is responsible for differences in the density, and, hence, flow velocity on the scale of heterogeneities during the development of magnetohydrodynamic instabilities and mixing. Such phenomena have attracted more and more attention. At the same time, there is no consensus on the insignificance of mesoscopic processes, at least, in steady-state detonation of individual HEs.

The uncertainty in the understanding of the main detonation processes is related to difficulties in experimental studies of the narrow chemical-reaction zone behind the detonation front. We developed a procedure for measuring the electrical conductivity profile behind detonation fronts [1, 2]. For dense individual HEs of balanced compositions (PETN, RDX, and HMX), good agreement was found between the measured zones of increased electrical conductivity with the reaction zones determined by the profiles of mechanical parameters (pressure and mass velocity). In the present study, this procedure is used to examine the role of particle size and interaction of the components of heterogeneous HEs.

EXPERIMENTAL PROCEDURE

The procedure used in the experiments was described in detail in [1, 2]. An explosive was placed in a copper coaxial cell of outer diameter $b = 8$ mm (Fig. 1). The diameter of the inner electrode was $c = 2$ mm. The outer electrode was compound, and its parts 1 and 2 were tightly fixed by a screw joint. A toroidal coil which served as an electrical conductivity sensor was placed in the cavity of the outer electrode. The narrow slot connecting the cavity with the measuring volume of the cell was filled with a dielectric (fluoroplastic, 0.3–0.6 mm or Plexiglas, 0.2–1 mm). The charge was initiated through an axial channel of diameter 2.4 mm in a Plexiglas plug.

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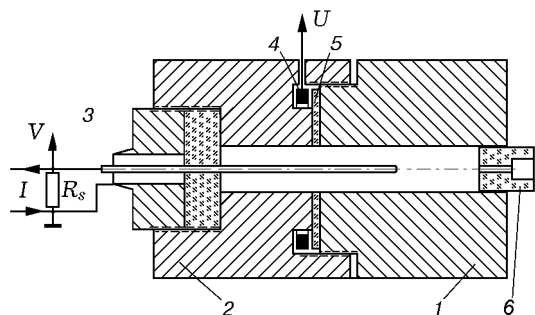


Fig. 1. Measuring circuit and cell geometry: constituents of the outer electrode (1 and 2), inner electrode (3), toroidal coil (4), dielectric (5), and Plexiglas plug (6).

Current I was supplied to the cell by a capacitor through a ballast resistor; a shunt R_s was connected in parallel to the cell. The supply voltage $V(t)$ and the sensor signal $U(t)$ were measured. The electrical conductivity in the plane of the slot is proportional to the sensor signal:

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

Here $x = Dt$ is the distance from the slot traveled by the wave front by the time t , D is the detonation velocity, and M is the mutual inductance. Signal correction makes it possible to eliminate the deviation of the dependence $U(t)$ due to the inductance of the sensor and to obtain the electrical conductivity profile behind the front under the assumption of a steady-state detonation wave.

The dimensional resolution is estimated at a quarter of the insulator thickness, and in the thinnest gap, it reached 0.05 mm. The measured electrical conductivity is limited by the leakage in the conducting medium over the slot due to the small but finite inductance of the sensor cavity L_c . In the experiments considered below, the leakage was significant in the case of pure TNT. The HE detonation velocity was determined from the known cell dimensions and the characteristic points in oscillograms, and the charge density $D(\rho)$ was determined from the detonation velocity by the dependences ρ taken from [3, 4] (because of losses in charging, the charge density could be several percent lower than the calculated loading density). For a 60/40 TNT/RDX composition (below, the mass ratio of the components is omitted), it was assumed that the dependence $D^2(\rho)$ is a linear (in TNT content) interpolation between the corresponding functions for TNT and RDX. For mixtures of RDX with water, NaCl, and saturated NaCl solution, the density from values of D was not calculated.

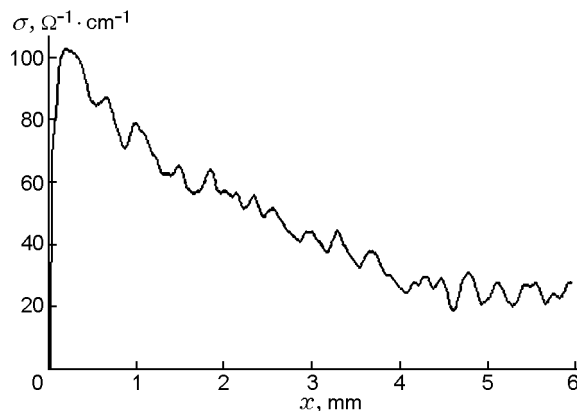


Fig. 2. Electrical conductivity profile for pressed TNT.

EXPERIMENTAL RESULTS

TNT and TNT/RDX

TNT is of interest as a classical HE with an excess of carbon, whereas the TNT-RDX composition is a sort of standard of a two-component HE. Previously, [5, 6] we have studied electrical conductivity distributions in cast and pressed TNT and TNT/RDX; the obvious effect of the particle size of RDX was observed in TNT/RDX. These data, however, were obtained in integrated measurements with a low spatial resolution (1–2 mm).

It is known that, in pure TNT, the release of free carbon leads to an anomalously high electrical conductivity [7, 8]. There is a controversy about [5, 9] the existence of a pronounced peak of electrical conductivity in TNT. Therefore, it would be of considerable interest to measure the electrical conductivity distribution in TNT with acceptable resolution.

The cell was filled with the charge in portions 5 mm high; the measuring slot was in the middle of one of the portions; the cell resolution was 0.25 mm. Figure 2 gives the electrical conductivity profile obtained for TNT of density 1.5 g/cm³. An obvious peak in the region corresponding to the reaction zone (≈ 1 mm) was not revealed. However, estimates showed that, in this case, the above-mentioned leakage could significantly distort the initial region of the signal. As shown in [1, 2], the relative error due to leakage is of order $L_c b \sigma / \tau$, where τ is the characteristic time of the signal (for example, the duration of the electrical conductivity peak). For an inductance $L_c = 0.9$ nH, an electrical conductivity $\sigma = 100 \Omega^{-1} \cdot \text{cm}^{-1}$, and an expected peak duration $\tau \approx 100$ nsec, the relative error is about 0.7, i.e., it has the order of magnitude of the measured quantity. The

roll-off of the front flattens the signal, so that in a region ≈ 1 mm wide, the result can be underestimated, and, downstream, it can be overestimated. Therefore, the question of the existence of a peak in TNT that correlates with the chemical-reaction zone remains open. Nevertheless, these measurements support data [8, 9] on the existence of high (compared to the estimate of $\approx 25 \Omega^{-1} \cdot \text{cm}^{-1}$ [5]) electrical conductivity in a region adjacent to the wave front.

Dilution of TNT with RDX reduces the electrical conductivity of the mixture and, thus, the leakage. A 60/40 TNT/RDX mixture was studied. The TNT used in this study had a particle size of $\approx 10 \mu\text{m}$ and RDX was of two types: standard ($200 \mu\text{m}$) and fine ($\approx 10 \mu\text{m}$). Figure 3 gives the measured electrical conductivity profiles in TNT/RDX with fine and standard RDX of the same identical charge density of 1.54 g/cm^3 , and the profile in pure (standard) RDX of the same density. The experiments were performed with a resolution of ≈ 0.125 mm.

It is evident that the contribution of RDX to the electrical conductivity can be marked only at the very beginning of the peaks obtained for TNT/RDX. Hence, the conductivity of these compositions is determined primarily by TNT. The electrical conductivity is an order of magnitude lower than that in pure RDX, and the role of leakage is insignificant. Thus, the addition of RDX made it possible to determine the electrical conductivity peak of TNT. The peak is followed by an extended electrical conductivity tail characteristic of TNT containing mixtures, which is also related to the release of free carbon [5].

The width of the electrical conductivity peak was determined from the inflection of the profile (Fig. 3 shows the procedure of its determination for the profile in RDX as an example): for the mixture of the HE with a small particle size (profile 1), the average value was 0.66 mm, which corresponds to a peak duration of 90 nsec, and for mixtures with standard RDX (profile 2), 0.42 mm and 59 nsec, respectively. It should be taken into account that our procedure determines the time of passage of the detonation wave by the measuring device, whereas the literature gives Lagrangian reaction times, which are 1.4–1.5 times larger. In view of this, our results agree with known data on the reaction zone of TNT/RDX mixtures of close compositions (0.44–0.68 mm and 80–130 nsec, see [10–12]). The results of [13] (0.13 mm) and the causes of their deviation from those obtained later are discussed critically in [10, 12].

The role of RDX particle size can be explained as follows. If the RDX particles are larger than the TNT particles, the peak is much more pronounced and the

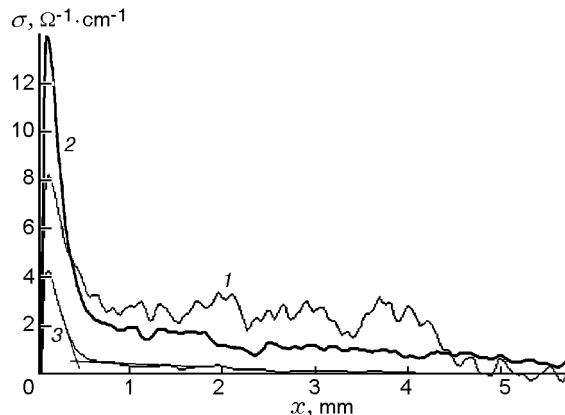


Fig. 3. Electrical conductivity distributions in 60/40 TNT/RDX and RDX (charge density of 1.54 g/cm^3): 1) TNT/RDX, fine RDX; 2) TNT/RDX, standard RDX; 3) pure RDX.

electrical conductivity decay outside the reaction zone is more rapid. In such a system, the TNT particles initially form thin layers around the larger RDX particles, and this geometry is primarily kept during pressing. Behind the detonation front, this structure is first well conducting (along TNT layers), but, fairly soon, it collapses because of mesoscopic flows [14]. If the particle sizes are approximately identical, part of the TNT is surrounded by RDX particles and is eliminated from the conductivity at the peak, resulting in a decrease in the peak amplitude. Later, however, the mixing of the components restores the contact between these volumes; as a result, the decay of the electrical conductivity is decelerated and the tail of the profile becomes more pronounced. Thus, besides the reaction time, the profile shape also reflects mesoprocesses in heterogeneous substances. At the same time, the difference in peak width is within the spread of the data obtained by traditional methods, i.e., the peak duration is less sensitive to the structure of the charge than the details of its shape.

HE with Inert Additives

It is natural to expect that the processes occurring on a scale of the order of the HE particle size will manifest themselves markedly in electrical conductivity measurements in HE–inert additive systems. We studied standard RDX with the following additives: 1) 10% NaCl with a particle size of $10 \mu\text{m}$; 2) water up to pore filling; 3) saturated aqueous solution of NaCl up to pore filling. In cases 2 and 3, the partial density of RDX was close to the bulk density ($\approx 1.15 \text{ g/cm}^3$), and in case 1, it was $\approx 10\%$ smaller. The amount of NaCl per unit volume of the mixture in cases 1 and 3 was almost

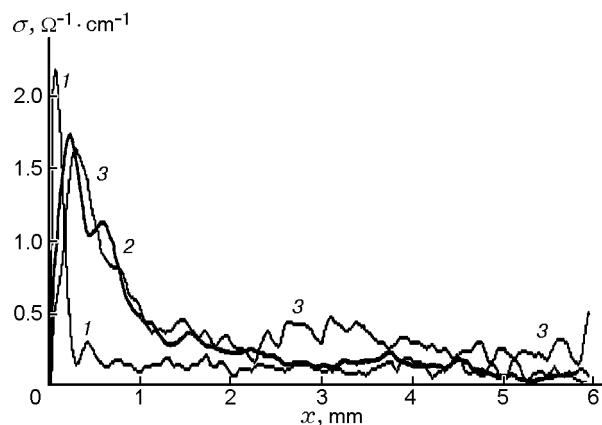


Fig. 4. Electrical conductivity distributions in RDX with additives: 1) 10% NaCl ($D = 5955$ m/sec); 2) water ($D = 7388$ m/sec); 3) saturated NaCl solution in water ($D = 7186$ m/sec); the resolution is 0.15 mm.

identical. The measured distributions of electrical conductivity are given in Fig. 4. In all cases, except for dry NaCl, the additive increases the peak width (on the average, 0.8 mm with the addition of water and 1.1 mm for the NaCl solution, whereas for dry NaCl, 0.32 mm). In measurements using an electromagnetic method [15], the width of the chemical-reaction zone in RDX filled with water was 2.17 mm, which is appreciably larger than the electrical conductivity peak width in our work. Such a difference in measurement data obtained by different methods is not something unusual; in this case, the width of the chemical-reaction zone [15] also exceeds the reaction-zone width (1.29 mm) in dry RDX of density 1.1 g/cm³ measured by an electromagnetic method [10]. The addition of water, NaCl, and especially NaCl solution increases the electrical conductivity in the tail; with the addition of the salt, the decay in the equilibrium electrical conductivity with time is very slow. The amplitude of the peak is almost the same in all cases.

Low Density RDX — Particle Size Effect

Attempts to determine the effect of particle size on the reaction time at the detonation wave front have been repeatedly undertaken. The obtained results were contradictory. Electromagnetic measurements [10] have not shown a marked effect. In experiments with higher resolution [16, 17], the particle size effect was observed although it did not reduce only to a change in the reaction time. Our experiments with high-density RDX and HMX [2] have not found a marked effect of the particle size on the electrical conductivity profile.

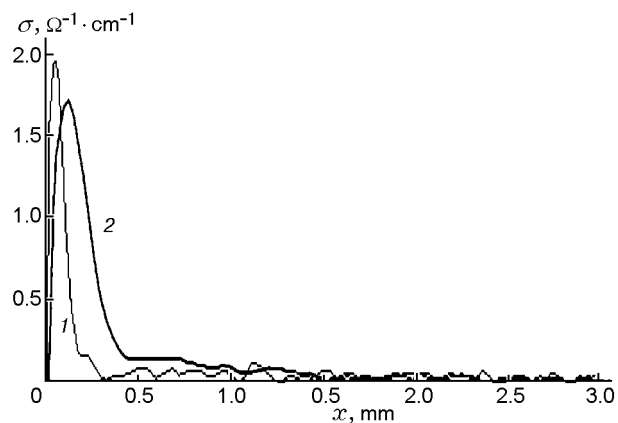


Fig. 5. Effect of particle size for low-density RDX: 1) particle size $10\ \mu\text{m}$ (resolution 0.05 mm); 2) particle size $200\ \mu\text{m}$ (resolution 0.1 mm).

It would be natural to expect a reduction in the reaction zone for HEs with a small particle size, due an increase in the density of hot spots which, according to most calculated models, are primary reaction centers. However, as noted in [10], a number of circumstances can interfere with a straight-line dependence of the reaction-zone width on particle size. Among such factors are the crushing of the particles to a size weakly dependent on the initial one in a shock wave or under pressing. The role of pressing can be minimized by using HEs of nearly bulk density, in which the initial structure is practically preserved. We note that, in experiments [16, 17], the role of particle size manifested itself at low densities. Therefore, we attempted to compare the electrical conductivity profiles in RDX of density 1.2 g/cm³ with an initial particle size of 200 and $10\ \mu\text{m}$. A HE charge with a particle size of $200\ \mu\text{m}$ was prepared almost without pressing (easy compaction by a small load). In the case of particles of size $10\ \mu\text{m}$, a hand-operated press was used. In the rest, the procedures of preparing the charges were identical.

Figure 5 shows the obtained electrical conductivity profiles. In the HE with a small particle size, the peak length is approximately half shorter at comparable amplitudes. The average width of the conducting zone for standard RDX is 0.4 mm, and for fine RDX, it is 0.22 mm. Thus, the particle size effect is obvious. We note that the peak in fine RDX was so narrow that, to measure it correctly, we had to increase the slot width to 0.2 mm (resolution 0.05 mm); at a slot width of 0.6 mm, the conducting zone is entirely covered by insulator, resulting in an appreciable surge of the cell voltage due to an increase in the cell resistance.

DISCUSSION

Electrical diagnostics allows investigation of meso-processes on a scale of the order of the HE particle size, to which other methods are insensitive. For example, traditional methods for measuring mechanical parameters do not detect any marked effect of particle size in pressed TNT/RDX, whereas the electrical conductivity method shows an obvious dependence on particle size. Previous studies [5] have shown an increase in conductivity in coarse-grained cast TNT/RDX containing up to 50% RDX, but peaks were not identified in these experiments because of their low resolution. The peak durations agree with known data on the reaction time in TNT/RDX obtained in electromagnetic and optical measurements. The marked effect of the RDX particle size on the electrical conductivity profiles suggests that, at least, part of the spread of the literature data on the size and duration of the reaction zones in TNT/RDX can be related to different particle size distribution of the substance.

The question of the presence of an electrical conductivity peak in dense TNT remain open. Such a peak with an amplitude of $200\text{--}250\ \Omega^{-1}\cdot\text{cm}^{-1}$ and about 1 mm wide, followed by a zone of lower electrical conductivity ($30\ \Omega^{-1}\cdot\text{cm}^{-1}$) was reported in [9]. It should be noted that, with sufficient electromagnetic resolution, the experimental procedure of [9] was characterized by gas-dynamic nonstationary, which could be reflected in the measured profile. Our experiments described above showed results closer to [9] compared to integrated measurements [5], but a peak was not recorded. Recording a peak in pure TNT would be of considerable interest. This would allow the correlation between increased electrical conductivity and reaction zone to be extended to the case of a HE with large excess of carbon. The authors are planning to return to this problem in the future.

The addition of dry NaCl to RDX changes significantly the electrical conductivity in the reaction zone but gives an extended tail (see Fig. 4) which is absent in pure RDX (see Fig. 5). This is due to the gradual dissolution of NaCl in detonation products (this process also includes the crushing of the additive particles and mixing of the substances).

Water-filled RDX and RDX with a saturated NaCl solution are characterized by a wide peak (≈ 1 mm). At the measured detonation velocity of 7.2 km/sec, the Chapman–Jouguet pressure in the mixture is about 15 GPa (this follows from the data of [15]). Under such conditions, the electrical conductivity of shock-compressed water reaches $1\text{--}2\ \Omega^{-1}\cdot\text{cm}^{-1}$, according to [18, 19]. The peak electrical conductivity of dry RDX of

bulk density falls in the same range. Hence, for water-filled RDX, the peak electrical conductivity should have the same values as is observed. The broadening of the reaction zone compared to pure HE is quite natural, due to the obviously superequilibrium amount of water. This is reflected in the electrical conductivity profile. It should be noted, however, that the electrical conductivity of the mixture has a more complex nature than in the case of pure HE, and the observed distribution is determined by both chemical reactions (in RDX and between the RDX detonation products and water) and the change in the degree of water dissociation [20] during gas-dynamic expansion. Both these processes depend significantly on the hydrodynamic mixing of the substances behind the detonation front. The gradual increase in the electrical conductivity (front ≈ 0.3 mm) can be related to the interaction of the substances but bending of the wave front due to the heterogeneity of the charge (which is difficult to avoid for these compositions) is possible.

The peaks for samples with additives of pure water and NaCl solution differ little up to a distance of ≈ 2.5 mm, i.e., the contribution of NaCl in this region does not play a marked role against the background of water dissociation (the molar concentration of NaCl in the saturated solution is ≈ 9 times smaller than that of water, even ignoring water in the RDX detonation products). Subsequently, with expansion of the substance, the difference between the profiles becomes appreciable: in the experiment with the NaCl solution, the electrical conductivity in the tail is higher and its decay is slower since NaCl is an easily ionizable additive (decomposes into ions in solution even under normal conditions).

The role of particle size in low-density RDX deserves special attention. In the HE with a small particle size, the conductivity peak is narrower than in standard RDX, as one might expect. It was unexpected, however, that this peak was even narrower than that in RDX of density $1.5\text{--}1.7\ \text{g}/\text{cm}^3$, which is evident from a comparison with data [1, 2]. This behavior can be explained on the basis of the concept developed by Dremin et al. (see, for example, [21]), which assumes superposition of two main processes: surface combustion originating at hotspots and a bulk reaction, the bulk mechanism dominating at pressures exceeding about half of the Chapman–Jouguet pressure in the substance of maximum density. At high densities, the bulk homogeneous mechanism dominates, so that the initial particle size of the HE is not important. This agrees with our measurements of electrical conductivity profiles [1, 2]. At low densities, the hotspot mechanism dominates, for which the particle size effect is significant. This is manifested, for example, in increased sen-

sitivity of HEs with small-size particles. For small particles, the rate of the overall reaction can exceed that at high densities, which is apparently responsible for the anomalously narrow peak of electrical conductivity. It would be of great interest to perform direct measurements of the reaction time under such conditions using traditional methods. Some anomalies in HEs consisting of small particles were observed in [16, 17]. We note that, for low-density HEs, optical measurements of mechanical parameters are complicated by difficulties in providing homogeneity of the sample and formation of interfaces. The electrical conductivity method proved more convenient since the HE sample is placed in a rigid shell and measurements are performed in the bulk of the substance.

Thus, the high-resolution electrical conductivity method proved a useful and informative means for studying detonations of heterogeneous systems.

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