JUL 29 1965

LA-AC-6993 (Full Paper)

CONF-651003-J

MASTER

ON ELECTRICAL CONDUCTIVITY IN DETONATION PRODUCTS

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ABSTRACT

The purpose of this paper is threefold: to discuss electronic techniques appropriate to high-resolution electrical conductivity measurements in the detonation products of condensed explosives; to present evidence to show that if the detonation products contain free carbon, that carbon is the main conductor of electricity; and to demonstrate the usefulness of the measurement technique to explore reaction-zone structure.

INTRODUCTION

Until recently, electrical conductivity measurements in the detonation products of high explosives have been made with electronic systems having a least resolved time of a few tenths of a microsecond.^{1,2} Improved time resolution, which increases at the rate at which information can be recorded, requires an increase in the frequency bandwidth of all components used in the measuring system. This paper reports on conductivity experiments utilizing modern traveling wave oscilloscopes along with dynamic conductivity cells to improve time resolution from a few tenths of a microsecond to a few tenths of a nanosecond — three orders of magnitude. In terms of frequency, the bandwidth has been increased from a few megacycles to over a thousand megacycles. As a consequence of this increase in resolution, electrical conductivity measurements have taken on increased significance by revealing information which was previously unavailable.

Thus, instead of obtaining a single value for the conductivity of detonation products, a conductivity-time profile with subnanosecond time resolution can be generated. These profiles, in which micron distances behind detonation fronts are being resolved, reveal that, while explosive products become conductive very quickly after the passage of the front, relatively large differences occur in the times required for different explosives to reach peak conductivity. Also, each explosive presents a different profile as well as different value for the peak conductivity.

The value of the peak conductivity is strongly correlated with the amount of solid carbon present in the detonation products. So much so that the data suggest the principal mechanism for electrical conduction in detonation products is a continuous network of solid carbon. That is to say, in detonation products containing free carbon, the phenomenon of charge conduction can be attributed to the conduction electrons available in the carbon, and the bulk conductivity is governed primarily by the fractional carbon density. On the other hand, explosives with little or no free carbon in their products have relatively low conductivities, and the

charge carriers are probably ions produced by high temperature.

Finally, it is believed that the high time resolution offered by the technique affords the best hope for resolving reaction zones in liquid explosives. However, of those studied only liquid TNT has yielded informative data.

THEORY AND ASSUMPTIONS

The basis for determining electrical conductivity in detonating high explosives is the proper use of a suitable conductivity cell. The cell employed herein to obtain profile data consisted of a long cylindrical tube with a small hemispherically shaped probe mounted on one end and with a plane-wave initiation system on the other end. The tube serves as the explosive container as well as the common return for the probe current. This arrangement is particularly good for liquid explosives and with proper probe fitting may be used with solids. Measurements are made in times that are less than the time required for a detonation wave to traverse a probe radius. For this situation the hydrodynamic disturbance is negligible and the electrical boundary conditions are well known.

In this connection, Mader³ has computed the interaction of a nitromethane detonation wave with an aluminum probe using his two-dimensional hydrodynamic code. His results are shown in Fig. 1. The change in probe radius is linear with time and in this case results in a reduction in probe length of 16% when the wave front just covers the hemisphere. The products under the probe can be thought of as being doubly shocked: the first shock causes the nitromethane to detonate, the second shock arises from the interaction of the detonation wave with the probe. The temperature rise in the products as a result of the second shock is quite small, about 200 degrees in this case. The shock velocity in the products is about half the detonation velocity, and therefore the reflected shock does not expand to an important extent compared to the electric sensing distance of about ten probe radii. For dense probe materials such as steel, changes in probe radius are even less pronounced. The effect of radius changes can be taken into account when distances behind the detonation front are required; however, for convenience in plotting, time rather than distance will be used.

The electric current stream lines emerging from a hemispherical probe as it is being engulfed by a detonation wave constitute the electric boundary value problem. The exact solution to this problem for a homogeneous isotropic medium can be obtained by solving Laplace's equation in toroidal co-ordinates.^{4,5}

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Omitting the details of the derivation, the solution for the normalized conductance under a hemispherical probe as a function of time after a detonation front first contacts the probe (horizontal line 1 in Fig. 1) can be written

$$\frac{g(t)}{4a\sigma} = \frac{\pi(2 - Dt/a) \sqrt{Dt/a}}{2 \cos^{-1}(Dt/a - 1)} \int_0^{\infty} \frac{\sqrt{\cosh w + 1} dw}{(\cosh w + 1 - Dt/a) \cosh [\pi w/2 \cos^{-1}(Dt/a - 1)]} \quad (1)$$

where g is the conductance and σ is the conductivity of the material under the probe; a is the probe radius, D is the detonation velocity, and t is the time; w is a parameter to account for the continuous spectrum of separation constants which naturally occur in the solution of the Laplace equation. The integral can be easily evaluated for $t = 0$ and $Dt = a$. Between these two limits it is necessary to resort to numerical integration methods for a solution.

A simple approximation for the normalized conductance is²

$$g(t)/4a\sigma = \cos^{-1}(1 - Dt/a). \quad (2)$$

This expression matches the exact expression at the probe tips and, for a hemispherical section, has the same general shape but results in a value of conductance at position 2 in Fig. 1 which is low by 5.6%. As it turns out, changes in probe shape due to the interaction of the detonation wave with the probe favor results which lie between these two limits. Furthermore, beyond position 2 in Fig. 1 the results become less and less accurate as the probe shape deviates from a true hemisphere.

The experimental record consists of an oscilloscope trace of voltage vs time, the voltage being the probe voltage as it penetrates the detonation products. The initial voltage on the probe is supplied by a transmission line of impedance Z . The reason for using a transmission line as the electrical energy source is that no ordinary power supply, such as a battery or a condenser, has the necessary high-frequency response required for this application. Also, as we shall see, the cable impedance serves as a conductance multiplier.

The waveform to be recorded is carried on a separate cable of impedance R to the recording oscilloscope. Both the feed line and the receiving line are connected to the probe with very short leads to preserve high-frequency response. A useful guide to charge radius, probe length, and lead dress, in this respect, is that no dimension should exceed a quarter wavelength of the upper bandwidth frequency. Otherwise the bandwidth will be reduced. Decreased bandwidth is most harmful at the instant of contact between probe and detonation wave, when voltage gradients are greatest.

Treating the source transmission line, supplied by a voltage V , as a series impedance Z shunted by both the fixed receiving line impedance R and the variable conductance of the cell, the cell conductance as a function of the measured voltage-time waveform $f(t)$ at the oscilloscope is

$$g(t) = (1/Z + 1/R)(V/f(t) - 1), \quad (3)$$

where V is the initial voltage on the probe. The actual value of the voltage is not important since only voltage ratios are required, and these can be obtained from the shot record. In general, the waveform $f(t)$ is a monotonically decreasing

function similar to the record of a nitromethane shot shown in Fig. 2. From measurements made on the shot record the instantaneous conductance as a function of time, $g(t)$, can be computed using equation (3), and if the conductivity of the products were constant the value of the conductivity could be obtained directly from equations (1) or (2). However, since the conductivity may not be constant a different approach is required.

To this end, the detonation products are imagined to consist of a large number of thin layers stacked one on top of the other. A layer thickness will depend on the time resolution employed, and in this connection it is convenient to use microns per nanosecond as the velocity dimension. The layer thickness is then written

$$\Delta x = (D - u_p) \Delta t \quad (4)$$

For nitromethane with $D \approx 6.4$, $u_p \approx 2.0$, and a resolution time of 0.25 nanoseconds, the approximate layer thickness, Δx , is 1.1 microns.

As each layer of products contacts the probe, conductance which is characteristic of the conductivity of the layer is added to the electrical circuit. Consequently, the shape of the electric waveform reflects the changes in conductivity as they take place. By accounting for the conductance of each layer as it moves up the probe an estimate of the conductivity of the n th layer can be made from the residual conductance up to that layer. To rephrase this mathematically,

$$\sigma_n = [0.25g_n - \sum_{j=1}^{n-1} \sigma_j (S_{n+1-j} - S_{n-j})]/S_1, \quad (5)$$

where σ_n is the conductivity of the n th layer, g_n is the conductance of the n th layer, and the summation accounts for preceding layers which have moved up the probe. Each layer with a conductivity σ_j is assigned a new value of conductance in accordance with the requirements of equations (1) or (2) to approximate closely the electrical boundary conditions with S being the geometric factor. Equation (5) is somewhat in error because it does not take into account refraction of the electric streamlines at the boundary between layers when conductivity changes are large. In the neighborhood of large conductivity gradients (except the jump at the front of the detonation wave) changes are averaged over too great a distance, which is to say the gradient is reduced. Thus, the analysis will produce a value of conductivity for each layer which is somewhat higher than the actual value should be. Nevertheless, the qualitative profile will reasonably approximate the true profile, showing changes in conductivity without generating spurious steps. Equation (5) is

valid, however, for regions of constant conductivity which sometimes exist well behind a detonation front. Because of the difficulty in handling the geometric factor S , equation (5) was coded in Version III Fortran to accommodate equations (1) and (2). Typical results for nitromethane are shown in Fig. 3, where the lower curve is the result of using the geometric factor from equation (1) and while the upper curve results from equation (2).

In situations where it is not convenient to use hemispherical probes a flat-bottomed probe can be employed to obtain a good estimate of the detonation products' bulk conductivity.⁶ For this case the normalized conductance is

$$g/4a\sigma = 1, \quad (6)$$

where a is the radius of the wire or rod contacting the explosive. This configuration is particularly useful when comparisons of different explosives in the same geometry are desired or when a parameter such as density is the only variable. Again, good time resolution is required as equation (6) does not hold if the conducting products have covered any appreciable length of the probe.

Typical shot records for two different PETN densities are shown in Fig. 4. Divergent or tilted detonation waves affect the electric waveform causing the discontinuity to be somewhat rounded so that conductance measurements (using equation 3) in the 7 to 15 nanosecond time interval are averaged to obtain an estimate of the conductivity from equation (6).

Alternatively, conductance plots alone furnish interesting comparison data such as those shown in Fig. 5 for PETN with different initial densities. It is apparent from this plot (which should not be construed as a profile) that as the initial density of the explosive decreases the conductance decreases, implying that the conductivity also decreases.

The geometry for this series of shots was quite simple. It consisted of a small brass cylinder with a 5/16 in. diameter hole 0.2 in. deep for the explosive. The booster was an SE-1 detonator with a 40-mil-thick 9404 pellet driving a 15-mil-thick brass shim which formed the bottom of the acceptor charge. The probe was the center wire of the coaxial line from the recording oscilloscope, and the source line in this case was a 93-ohm coaxial cable. The geometry in this situation is marginal for obtaining a good value of conductivity, because the detonation wave is divergent and the ratio of charge diameter to probe diameter is also marginal: 12.4 to 1. As a general rule charge diameter to probe diameter should exceed a 25 to 1 ratio.

RESULTS

Conductivity profiles for nitromethane, Composition B, and liquid TNT have been obtained, and the results are shown in the log-log plot of Fig. 5. The onset of conductivity is seen to be quite rapid in the nitromethane and the liquid TNT relative to the Composition B. The time to the first inflection in both liquid TNT and

Composition B is thought to be connected with reaction-zone thickness, although further experimental evidence appears warranted before a final judgement can be made. In the case of nitromethane there has been no indication of an inflection, which tends to confirm the hypothesis that the reaction zone is too thin to be resolved. The conductivity peak evidenced in all three explosives has been attributed to carbon precipitation which takes place near the end of chemical reaction.

Conductivity profiles of PETN have not been generated as yet because of the difficulty in making small hemispherical holes in this material. However, since the profile is thought to be similar to nitromethane (very thin reaction zone, fast-rising, flat-topped conductivity profile) bulk conductivity measurements have been made. The best estimate of PETN conductivity is 60 mhos per meter for an initial density of 1.699 g/cc, a charge diameter of 1.625 in., a charge length of 1 in., and a flat-bottomed probe of 30 mils — see Fig. 4a for a typical shot record.

The maximum values of the conductivities for the above four explosives were plotted against carbon content as shown in Fig. 7. The values of the carbon content were computed by Mader using the BKW equation of state.⁷ Because of the uncertainties in the form of the equation of state used to calculate the solid carbon content, the numbers may not be accurate, but their relative values should be quite reliable. The correlation is excellent, and it is believed that in these explosives electricity is conducted in a network of solid carbon.

As further evidence of the validity of the carbon hypothesis, the bulk conductivity of PETN as a function of initial density can be estimated from the data of Fig. 5. The computed values are somewhat high compared to the best estimate for high-density PETN (probably due to marginal geometry), but since firing conditions and geometry were identical in all cases the relative conductivities should be quite satisfactory. Figure 8 is a plot of the results along with a few physical properties of PETN obtained from Mader-type calculations.

The most revealing feature in connection with Fig. 8 is that all parameters except temperature increase with increasing density. At low density the temperature is high, there is no solid carbon present (the carbon combines with oxygen to form carbon monoxide), and yet the conductivity is low relative to the high-density material. The point is that thermal ionization is not as effective as solid carbon in enhancing electrical conduction. In fact, at low density the conductivity is only a little more than an order of magnitude higher than that of detonation products from a gas mixture of acetylene and oxygen. Furthermore, while references are not available it is known that the conductivity of tetranitromethane and nitroglycerine are also quite low and that these materials also have carbon-free products.

SUMMARY

To conclude, this investigation has shown that the electrical conductivity of detonation products for different explosives ranges from 1 to 10^4 mhos per meter. A strong correlation between the value of the peak conductivity and the calculated carbon content in the detonation products was noted. That is, in explosion products containing free carbon the conductivity is primarily determined by the fractional solid carbon density in the products. This suggests that a network of solid carbon forms and is the dominating conductor. In detonation products which do not contain free carbon the electrical conductivity is low and probably conduction is primarily by ions which are present because of the high temperature of the products.

The technique described here, yielding two to three orders of magnitude improvement in time resolution over conventional conductivity methods, constitutes a new diagnostic tool for probing into detonation products. The full benefits of the technique have not been fully assessed as yet, but further experiments to elucidate reaction-zone structure are being considered.

ACKNOWLEDGEMENT

The author wishes to thank C. L. Mader for carrying out the probe interaction computations, B. G. Craig for his assistance in shot preparation, and W. C. Davis for his assistance in the preparation of this paper. This work was performed under the auspices of the United States Atomic Energy Commission.

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Fig. 1. Hemispherical probe shape changes (curved lines numbered 1, 2, 3) and shock wave position (curved dashed lines 2 and 3) for corresponding nitromethane detonation front (horizontal dashed lines 1, 2, and 3) advancing over an aluminum probe. The percentage change in radius is shown on the left for a detonation advance of $a/2$ and a .

Fig. 2. A nitromethane shot record. The timing wave is a 2000 megacycle sine wave ($t = 1/f$). The probe radius for this shot was 3.0 ± 0.3 mils; and $Z = 33$ ohms and $R = 125$ ohms.

Fig. 3. Typical conductivity profiles of nitromethane near the detonation front. The lower curve makes use of equation (1) while the upper curve makes use of equation (2).

Fig. 4. Conductance waveforms using flat probes. The timing trace is a 400 Mc/sec sine wave

Fig. 5. Electrical conductance measurements in PETN products for different initial densities. See text for shot geometry configuration.

Fig. 6. Conductivity profiles for nitromethane, Composition B, and liquid TNT. The inflection on the front of Composition B and liquid TNT curves are thought to be associated with the ends of the reaction zones in these materials and the peaks with carbon precipitation.

Fig. 7. Correlation of electrical conductivity with the solid carbon content in the detonation products of various explosives.

Fig. 8. PETN properties as a function of initial density. Illustrating the inverse relationship between temperature and conductivity.