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TITLE

INITIATION AND DETONATION MEASUREMENTS ON LIQUID NITRIC OXIDE

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INITIATION AND DETONATION MEASUREMENTS ON LIQUID NITRIC OXIDE

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Measurements are reported on detonations of homogeneous liquid nitric oxide initially at $T = 119$ K, $\rho_o = 1.28$ g/cm^3. Plane-shock initiation in mirror-covered wedges demonstrated detonation in preshocked material (superdetonation) that overtook the smooth input shock. Its transverse irregularities continued in the unsupported detonation. The input shock ran ca. 3 mm at $U_I = 3.8 \pm 0.2$ km/s. Behind it the fluid had $u_p = 0.89 \pm 0.02$ km/s, $p = 4.3 \pm 0.3$ GPa, $\rho = 1.67 \pm 0.03$ g/cm^3, and the superdetonation had $D^*_{\mathrm{lab}} \approx 6.9$ km/s. In 356-mm long graphite tubes with 25.4-mm inner diameter, measured detonation velocity is $D = 5552 \pm 3$ m/s. Failure diameter in graphite is below 8 mm. Free-surface velocities of 1- and 2-mm aluminum plates terminating the detonation axis were near 1.5 km/s, approximately as predicted for CJ flow at $D_{\mathrm{CJ}} = 5564$ m/s, $u_p = 1.34$ km/s impinging on aluminum. Electronic streak photography at $d = 20.6$ mm in brass registered a smoothly convex detonation front with apparent brightness temperature of 1735 ± 40 K as it reached an end window asynchronously.

INTRODUCTION

Liquid nitric oxide was studied as a prototype condensed-phase explosive in a multidisciplinary research program pursued during the 1980s at Los Alamos National Laboratory. Theoretical atomic/molecular treatments of this energetic, two-element substance and the products of its decomposition at condensed-phase density are simpler and correspondingly more mature than if carbon and/or hydrogen were included. Experiments with nitric oxide were favored by uniformly pure NO gas furnished as needed from the Laboratory's distillation facility operated for enrichment[1] of the stable isotopes of both O and N.

The present report assembles the dynamic measurements we completed on cryogenically liquefied nitric oxide functioning as a detonating, homogeneous high explosive. Precisely delayed initiation of detonation by a suitably weak input shock is demonstrated by a combination of orthodox wedge, impedance mirror, and gas gun techniques. The main results from long cylindrical charges are detonation velocity at selected diameters and on-axis acceleration of thin aluminum plates at the charge end. Finally, a single measurement by image-intensified, direct photography through a full-diameter window on a short cylinder is presented. This quantitatively demonstrates the convex shape and optical brightness of a detonation front in liquid nitric oxide. These measurements extend or supersede those we have published earlier.[2,3,4] The broad scope of the Laboratory's program included this work and related studies of nitric oxide, its constituent elements, and other oxidation states of nitrogen under static and transient conditions of high pressure and/or energy. Representative accounts are given in References 5-10.

The customary chemical formula of nitric oxide, NO, represents the diatomic molecular form of its gas phase under ambient conditions. Cryogenic condensation to solid or liquid is accompanied by reversible dimerization to molecular N_2O_2. The substance melts at 109.5 K and boils at 121.4 K. In all of its states of aggregation, its energy of formation significantly exceeds that of the constituent elements, O_2 and N_2, and of any of the other nitrogen oxides. We apply the name *liquid nitric oxide* to the homogeneous, neat explosive confined at temper atures near 113 K, and use one of the above chemical formulas when the molecular form or the gaseous state is pertinent.

INITIATION OF DETONATION BY SHOCK

BACKGROUND

Earlier investigations[2,11,12] of occurrence of detonation in dense nitric oxide demonstrated its initiation in liquid or mixed phases by shocks from donor explosives or an equivalent projectile. The results were convincing that, in its susceptibility to detonation by accidental shock, condensed nitric oxide under technologically typical conditions more closely resembles nitroglycerine than it does the less sensitive liquid high explosives. However, the previously applied methods did not examine the detailed shock processes leading to detonation or subject the explosive only to plane or divergent flows. Also, each investigation was done with nitric oxide that contained different, small percentages of NO_2, N_2O, and N_2 accumulated from gradual, spontaneous decomposition of NO kept as a compressed gas

In order to gain more precise information on initiation of detonation in liquid nitric oxide, we have aimed to control the shock geometry and the NO purity. Our preliminary experiments used plane-wave explosive systems as donors with uniform metal intermediate members to transmit and attenuate shocks at selected strength. Measurements included transit times through disk-shaped samples at a succession of depths and electrooptically intensified end-on streak photography across a diameter of a window-covered disk. An unsuccessful attempt was made to perform laser-Doppler velocimetry at the impact surface, viewing through the liquid and window above it. The investigation was culminated by two gas-gun shots that yielded clear streak-camera records of delayed detonation overtaking the steady input shock in wedges of homogeneous liquid explosive. The foregoing developments were reported in detail,[4] with emphasis on their qualitative features. Here we extend the earlier account of the wedge experiments by quantitative treatment of the wave velocities and the shocked state produced prior to the detonation

WAVE SEQUENCE

Figure 1 displays a record of initiation of detonation in a wedge of liquid nitric oxide. A very similar record from a second such shot and a schematic diagram of the gun target are available elsewhere.[4] The shot configurations had a conventional wedge shaped volume of explosive bounded downstream by a mirror coated PMMA window surface inclined to the wave front. This interface was illuminated by an explosively shocked xenon lamp and photographed through the streak-camera slit. The initiating shock was introduced at the base plane of the liquid wedge by transmission from a smooth copper plate whose opposite face was struck by a matching copper impactor delivered from the gun muzzle.

Dynamic events at the mirrored interface alter its reflection at each position viewed by the slit, beginning when the first disturbance arrives. Advancing waves in the flow are recorded as continuous tracks with positive slope. Early in the record of Fig. 1, where the explosive is thinnest, the input shock makes a smooth, straight track as it reaches the inclined mirror. Farther along, detonation makes a second track. The input-shock track terminates at a depth of 3 mm, and thereafter the single track is formed by detonation proceeding in original explosive. Before this transition point the detonation track appears in the sector to the left and identifies the beginning of detonation as a separate, fast wave in the compressed, moving material behind the input shock. In this domain the PMMA substrate of the mirror likewise is shocked, and the reflecting surface is reoriented to a decreased wedge angle. The separation between the waves and between their resulting tracks each diminishes linearly to zero when the input shock is overtaken supersonically from the rear. This event is pointed out in Fig. 1.

The sequence of waves found here duplicates, at least qualitatively, the initiation mechanism witnessed repeatedly in liquid nitromethane and recognized in other homogeneous explosive media, including single crystals and doubly-shocked gases. The PMMA substrate supporting the mirrored wedge boundary has enabled our technique to record the three principal wave fronts and the transverse structure begun in the earliest detonation motion.

CONTROL OF CONDITIONS

The straight tracks of the frontmost waves recorded in Fig. 1 determine a precise depth of overtake by their intersection and indicate sensibly constant velocities of the input shock front and later of the detonation propagating through original explosive liquid. For this depth and these velocities to be interpreted, the strength and orientation of the input shock must be adequately uniform over the wedge section whose termination on the slant face is photographed. On the indication from Fig. 1 that these criteria are met, we proceed to address the relevant material conditions.

The density of the initial nitric oxide liquid is governed by its temperature. Data from References 13–15 are represented between $T = 110$ and 130 K by

$$\rho_{\mathrm{liq}}(\mathrm{g/cm^3}) = 1.274 + .00469(120.0 - T) \quad . \qquad (1)$$

Under radiative heat load from gun components in both axial directions, the copper structure of the gas gun target attained an approximately uniform temperature after cooldown. A thermocouple in a blind well next to the explosive-filled space stably registered 120.0 ± 0.3 K over times exceeding 10^3 s before firing each shot. The condition in the nitric oxide liquid was also measured by the vapor pressure recorded during filling of the visible wedge volume. This indicated a somewhat lower temperature, 117.6 K. The average of these temperatures, 118.8 K, is taken to determine $\rho = 1.28 \pm 0.005\ \mathrm{g/cm^3}$. Gravitational stratification is relied upon to keep nonuniformity within the horizontal wedge section less than the absolute uncertainty of the liquid density.

A test of our control of conditions that might influence the initiation of detonation was made by a duplicate experiment. The same target and impactor materials were used, and each face of the pure copper elements in the shock-forming path was diamond-tool machined to 6 x 10^{-5} mm smoothness. The impact velocity from the gun was repeated to within 0.2%. The two shots used deliberately different batches of nitric oxide. Figure 1 is from the shot with the purest liquefied NO gas we could manage; absence of detectable infrared absorption through 100-mm path at 85-kPa gas pressure determined the sum of mass fractions of N_2O and NO_2 to be below 10^{-4}. However, the liquefied state appeared bluish and not as colorless as if the nitric oxide were

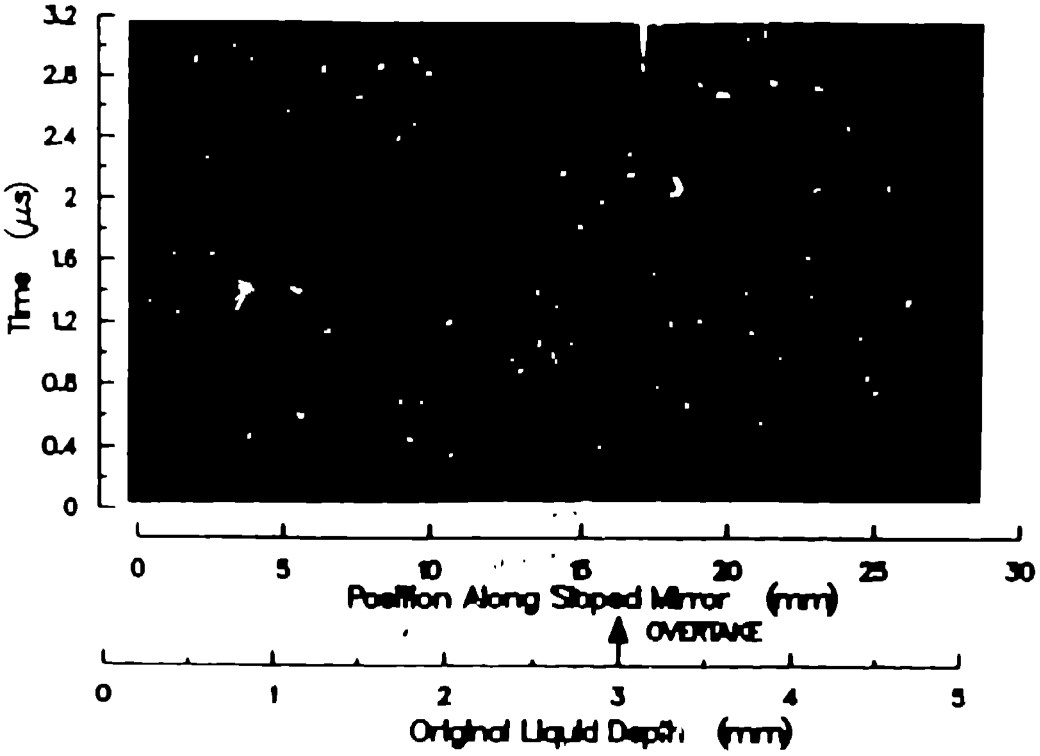

FIGURE 1. ANNOTATED STREAK PHOTOGRAPH OF DELAYED INITIATION OF DETONATION IN WEDGE OF LIQUID NITRIC OXIDE. TIME AXIS ORIGIN IS ARBITRARY, SLIT AXIS PROJECTIONS AS SHOWN

ultrapure.[16] Because it was impractical to eliminate the triatomic constituents, we tested their possible influence upon initiation of detonation by increasing their concentrations to a total mass fraction near 1×10^{-3}, measured by infrared analysis. This alteration of composition was not enough to perceptibly influence the density, detonation velocity, or initial-shock Hugoniot of the liquid explosive. The increase in run to detonation that it produced, from 3.0 mm to 3.4 mm, is small enough that the apparent densitization is of uncertain significance. The important indication is that impurities that accumulate in NO by its slow, spontaneous disproportionation do not profoundly alter the initiation of detonation of its liquid form.

PRIMARY WAVE VELOCITIES

In Fig. 1 the arrival tracks for the input shock and the final detonation have positive slopes; their angular departures from the horizontal are respectively denoted ϕ_I and ϕ_D. The corresponding apparent velocities at which the respective waves in the wedge progressed along the photographed band on the mirrored face are $S_I = (W \cot\phi_I)/M_a$ and $S_D = (W \cot\phi_D)/M_a$, where W is the writing rate of the camera and M_a is the apparent magnification at which distances along the sloped object are projected through the camera slit and onto the film. These and other raw results of our two wedge experiments are assembled in Table 1.

To evaluate the velocities of these waves within the wedge of explosive, we consider their ideal and real orientations. Coordinates are adopted with the x, y plane in the impact face of the wedge and the positive z axis directed normally into the explosive. The axis of the photographic object band on the slant face lies in the x, z plane by definition, and has direction cosines $(\cos\psi, 0, \sin\psi)$, where ψ is the wedge angle. Planar waves, identified by subscript w, are oriented in this coordinate system by their unit normal vectors (a_w, b_w, c_w). Waves that are ideally parallel to the wedge base have $c_w = 1$, $a_w = b_w = 0$, and their wave speed is simply $U_w = S_w \sin\psi$. For tilted waves,

$$U_w = S_w(a_w \cos\psi + c_w \sin\psi) \quad . \tag{2}$$

Waves crossing $z = z_o$ planes with a small delay gradient along the x– and/or y–directions possess small tilt components a_w and/or b_w that are proportional to U_w itself. For detonation following a slower input shock by a uniform time, this determines

$$a_D = a_I(U_D/U_I) \quad . \tag{3}$$

The analogous acceptor/donor relation between b_D and b_I is presumed, but is inconsequential.

Table 2 traces the application of these wave orientation relationships to the speeds of Table 1. The detonation speeds, D, in the "ideal" columns for the two shots differ from each other, and both are larger than the velocity near 5.55 km/s expected for unsupported detonation. The input shock speeds assuming ideal orientation are likewise unequal, whereas the gun

TABLE 1. LIQUID NITRIC OXIDE WEDGE PARAMETERS

Gas gun Shot No.	660	661
Photograph	Fig. 1	Ref. 4
Projectile Speed (km/s)	4.008	4.010
Wedge Angle, ψ (rad)	0.172	0.173
S_D (km/s)	35.9	33.8
S_I (km/s)	24.0	22.8

TABLE 2. WAVE VELOCITIES IN LIQUID NITRIC OXIDE*

	Shot 660		Shot 661	
	Ideal	Tilted	Ideal	Tilted
Detonation:				
c_D	(1.0)	.99985	(1.0)	.99996
a_D	(0.0)	.0179	(0.0)	-.0084
D (km/s)	6.15	(5.55)	5.82	(5.55)
Input Shock				
a_I (= a_D)	(0.0)	.0170	(0.0)	.0084
U_I (km/s)	4.10	3.70	3.92	3.74
a_I (= $a_D U_I/D$)		.0117		.0057
U_I (km/s)		3.84		3.80

* Assumed values in parentheses; symbols defined in text.

projectile speed was duplicated. However in each shot we find $(U_I/D) = 0.67 \pm .01$.

In the columns headed "tilted," we reconcile these results by considering first the tilt component a_D needed to adjust each detonation wave speed to an assumed value of 5.55 km/s. The effect upon $c_D = \sqrt{1 - a_D^2 - b_D^2}$ of an unknown y-component, b_D, is ignored. Extending the adjustment to the input shock is less definite, and two naive estimates for a_I are evaluated. Taking $a_I = a_D$ gives values of U_I near 3.7 km/s from both shots. Considering the detonation as acceptor and input shock as donor according to Eq. 3 diminishes the adjustment of each U_I from its "ideal" value, and the resulting values are both near 3.8 km/s.

HUGONIOT STATE AND SUPERDETONATION

$U_I = 3.8 \pm 0.2$ km/s is adopted to determine the shocked state of nitric oxide fluid in which the superdetonations occurred. With the symmetric approximation for the shock and release of pressure and particle velocity in copper, the following properties are found from the conservation equations for the matching shock in the explosive: pressure $p_I = 4.3 \pm 0.3$ GPa; particle velocity, $u_I = 0.89 \pm 0.02$ km/s; density, $\rho_I = 1.67 \pm 0.03$ g/cm^3 = $1.305\ \rho_o$; specific internal energy, $E_I = E_o + 0.40 \pm 0.02$ kJ/g. E_o for the original liquefied nitric oxide has the value $+2.65 \pm 0.01$ kJ/g, relative to the elements O_2 and N_2 as ideal gases at zero K[8]. From the averages of the corresponding angles of the tracks registered by the superdetonations in Fig. 1 and Reference 4 before they overtook the input shocks, we find $S^* = 45.5$ km/s and $D^*_{\text{mat}} = 6.0$ km/s. This superdetonation velocity is in the frame of the compressed, moving material. In an Eulerian frame D^*_{lab} has the value $6.0 + 0.9 = 6.9$ km/s. These values of D^* are subject to large uncertainity from tilt, and are presented primarily to illustrate the relationships applicable in the compressed wedge confined by a window with matching shock impedance.

DETONATION PRESSURE AND VELOCITY MEASUREMENTS

The experimental arrangements have been described previously.[3] The tubes confining the explosive were made of graphite. They were cooled with liquid nitrogen in a bath with isopentane as the heat transfer fluid. With vigorous stirring, the temperature was maintained uniform along the length of the tube. Table 3 gives a list of temperatures. Except for the heat leak from the booster explosive warming the first thermocouple station, the temperature control is very good.

TABLE 3. TEMPERATURE UNIFORMITY ALONG GRAPHITE-WALLED CHARGES OF LIQUID NITRIC OXIDE.

Station Number	Distance from Booster (mm)	Temperature (°C)	
		Shot F-5730	Shot 8-184
1	25.4	− 153.89	− 153.75
2	76.2	− 154.11	− 154.05
3	127.0	− 154.25	− 154.10
4	177.8	− 154.24	− 154.14
5	228.6	− 154.16	− 154.05
6	279.4	− 154.07	− 154.06
7	330.2	− 153.98	− 154.01

Graphite was chosen as the confining material because it is available commercially in the form of tubes, it has a relatively low shock impedance and offers minimal confinement, and it was thought that there would be no region of subsonic flow in the graphite that might cause a perturbation where the wave in the explosive intersects the tube wall. Later calculations have shown that the uncertainties in the equations of state are large enough that there could be a small subsonic region.

The detonation pressure is inferred from the measured free-surface velocity of a 6061 dural plate driven by the explosive at the end of its run through 355.6 mm of nitric oxide in the 25.4-mm diameter tube. The velocity was measured using the method described by Davis and Craig.[17] It has been shown by Davis and Venable[18] that the measured free-surface velocity is a linear function of $(x/d)^{1/2}$, where x is the thickness of the plate, and d is the diameter of the detonation tube. The data are plotted in Fig. 2. The intercept with the axis gives the desired value of the free-surface velocity, which is then used to find the particle velocity in the dural at the instant it is contacted by the detonation wave.

Figure 3 is a plot in the pressure - particle velocity plane of the Hugoniot curve for dural and the reflected shock Hugoniot curve for the explosive. Their intersection should be at the point measured in the free-surface velocity experiments if the reflected shock Hugoniot curve is the correct one. Within the accuracy of the experimental result, the curve is satisfactory.

An important reason for performing these measurements was to try to answer questions raised by speculation that real detonations are weak detonations, not Chapman Jouguet detonations. Nitric oxide is the only condensed phase material for which it has been possi

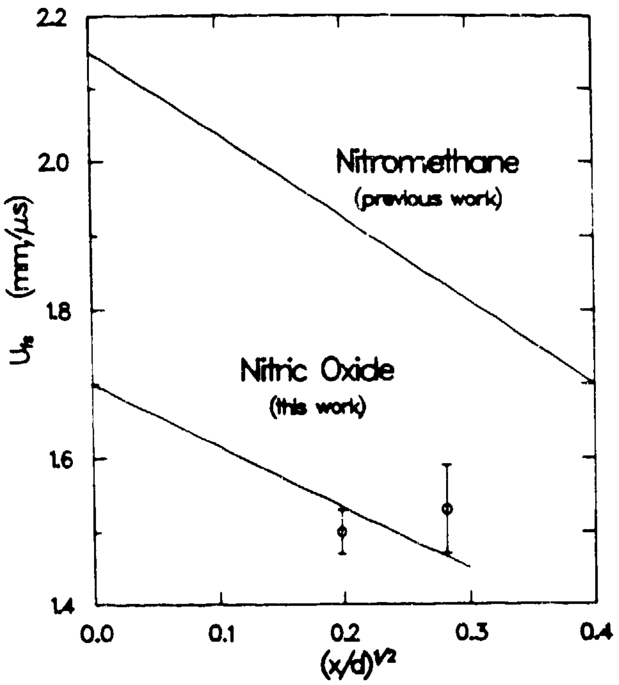

FIGURE 2. PLOT OF FREE-SURFACE VELOCITY VS PLATE THICKNESS FOR NITROMETHANE FROM REFERENCE 18 AND FOR THE MEASUREMENTS FROM SHOTS F-5730 AND 8-184 DESCRIBED HERE. THE EXTRAPOLATION TO ZERO PLATE THICKNESS IS MADE USING THE SAME FRACTIONAL DECREMENT FOR EACH.

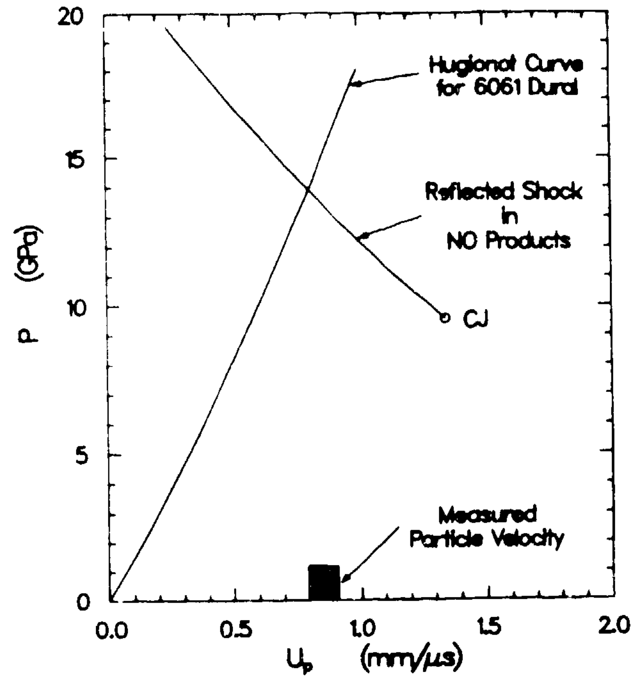

FIGURE 3. MATCHING OF NITRIC OXIDE DETONATION PRODUCTS INTO THE DURAL PLATE, PLOTTED IN THE PRESSURE VS PARTICLE VELOCITY PLANE. THE VALUE OF PARTICLE VELOCITY INFERRED FROM THE FREE-SURFACE VELOCITY MEASUREMENTS IS PLOTTED ON THE PARTICLE VELOCITY AXIS, WITH ESTIMATED ERROR SPAN. THERE IS NO DISAGREEMENT WITH THE ASSUMPTION THAT THE DETONATION IS A CJ DETONATION.

ble to measure the equation of state of the detonation products in separate experiments that are not detonation experiments.[8] In Fig. 3 the Chapman-Jouguet state and its reflected-shock Hugoniot are the ones calculated from a semiempirical equation of state of equilibrium products from liquid nitric oxide.[8,19] This is based on the measured shock Hugoniots of oxygen and nitrogen individually and of nitric oxide products in overdriven detonation states. Hence it is independent of assumptions about the nature of the unsupported detonation of nitric oxide. The measured point plotted in Fig. 3 is not in disagreement with the assumption that the real detonation is a Chapman Jouguet detonation. The data are not adequate to put any severe limit on the range of weak (or strong) detonations that might also be in agreement with the data.

Detonation velocity measurements were made incidental to the pressure measurements. A plot of detonation velocity vs charge diameter is shown in Fig. 4. The infinite medium detonation velocity is estimated to be 5564 m/s at a temperature of 119 K, corresponding to a density of 1.280 g/cm^3

DETONATION FRONT PHOTOGRAPH

We had ambitiously intended to use measured wavefront shapes in long, detonating charges of several different diameters to determine rates of energy release in the reaction zone.[20] However, we completed only a prototype experiment demonstrating wavefront curvature. Electronically intensified cameras of two types, single-frame and streak, were used to photograph self-light from the transparent end of a detonating cylinder of N_2O_2 capped by a flat PMMA window. The cylinder interior was 40 mm long and 20 mm in diameter, and detonation was initiated promptly by a flat, axially directed shock introduced from detonating TNT through the brass end wall opposite the window.

The streak photograph from this experiment is displayed in Fig. 5, with axes to indicate the diametrical slit and advancing time, t. The midpoint of the streak record

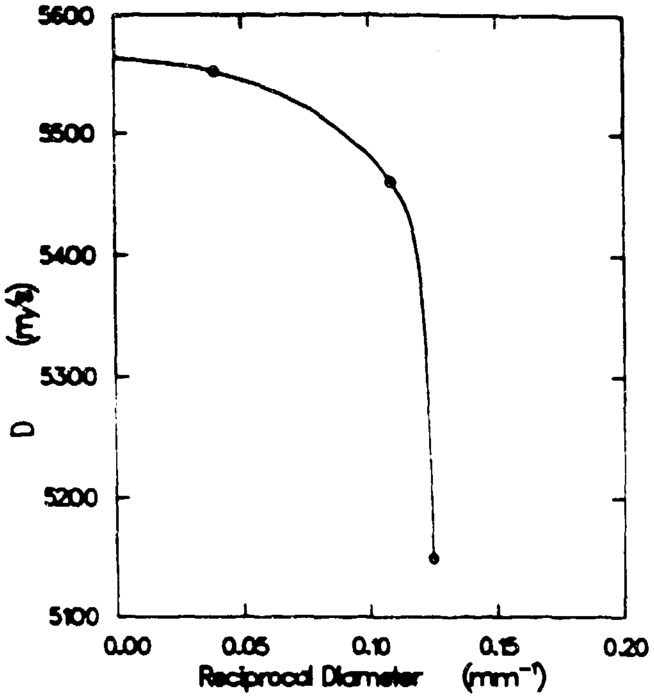

FIGURE 4. PLOT OF DETONATION VELOCITY VS RECIPROCAL DIAMETER FOR NITRIC OXIDE AT 119 K AND DENSITY 1.280 g/cm^3. THE LARGE DECREMENT IS UNUSUAL FOR A LIQUID EXPLOSIVE.

coincides with the framing picture exposure discussed below, and is taken as the time origin. Somewhat later, when the detonation reached the window, it abruptly intensified the otherwise slowly varying exposure on the film. The resulting arrival track appeared earliest at the charge axis (t_{axis} = 0.34 μs) and formed a smooth, symmetric arc indicating that the detonation front is likewise curved and at the walls lagged by 0.09 μs, or 0.5 mm (t_{wall} = 0.43 μs). Transverse irregularity indicative of proximity to the failure diameter is absent, and any fine-scaled manifestations of nonlaminar detonation flow are not resolved by our electronically intensified photography.

The single-frame picture, exposed for 30 ns, showed the full circular area of the approaching detonation when its front was still about 0.4 μs, or 2 mm, from reaching the window surface. It showed a uniformly exposed disc whose apparent brightness we matched through the same photographic system by an incandescent tungsten-ribbon lamp. The equivalent black-body temperature of the tungsten source was 1706±37 K, and exposures at nearby temperatures indicated the film coefficient. Densitometry of the streak photograph showed that the brightness of the detonation increased steadily, and understandably, as the remaining depth of undisturbed, bluish-gray N_2O_2 liquid diminished. The abrupt increase of brightness when the PMMA window was reached was followed by a steep, steady decrease. The apparent brightness temperatures determined from the axial densitometric scan are plotted against time at the top of Fig. 5.

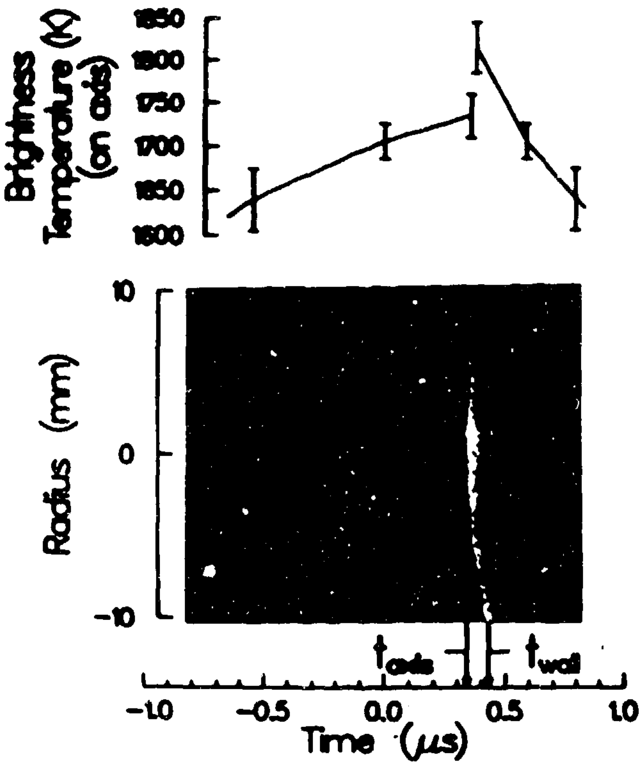

FIGURE 5. ELECTRONIC STREAK PHOTOGRAPH OF SELF-LIGHT FROM CONVEX FRONT OF LIQUID NITRIC OXIDE DETONATION REACHING FULL-DIAMETER PMMA WINDOW. BRIGHTNESS TEMPERATURES AT ZERO RADIUS PLOTTED ABOVE, TO SAME TIME AXIS.

All these brightness temperatures are far below the C-J temperature of approximately 2600 K we expect[19] from the equation of state of products from detonating N_2O_2 at liquid initial density. The simplest interpretation, sufficient but not unique, is that the reaction zone and/or unreacted layer is significantly opaque and behaves in the limit as a black body near 1735 K. The effective emissivity of the detonation product state at or near C-J conditions remains unknown, as does the complex net effect of encountering the PMMA boundary.

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