

CONF-791084--1

LA-UR-79-2880

TITLE: INTRODUCTION TO DETONATION PHENOMENA

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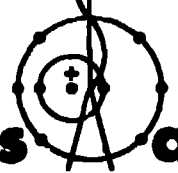
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SUBMITTED TO: U. S. Army Research Office
Working Group Meeting on Detonation
Phenomena, Menlo Park, CA,
October 23-24, 1979

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Introduction to Detonation Phenomena

W. C. Davis

LASL

My assignment is to provide an introduction to detonation phenomena for scientists who work in other fields, and to provide a broad perspective of our field as an introduction to the more specialized papers. I will try to take care of the first part, and hope it somehow serves for the second part.

Scientific research has understanding as its goal, and understanding proceeds from observation to hypothesis to deduction to comparison. Let's begin with observation and hypothesis.

Figure 1 shows a block of explosive detonating. The wave spreads almost like a Huyghens construction. The velocity of the wave is primarily determined by the composition and density of the explosive, and little affected by the time it has run, the edges, or the wave curvature. The material in front of the detonation wave is absolutely unaffected, and in particular another detonation wave in it propagates independently of the first one until they intersect. Thus the wave front is the moving surface that separates explosive material in motion from stationary material. The pressures are very high in solid or liquid explosives, perhaps a few hundred thousand atmospheres, or several tens of GigaPascals. At these high pressures, and the temperatures achieved in the compression, chemical reactions proceed very rapidly. These observations suggest that confinement of the material that allows the reaction to proceed so fast is provided by the inertia of the explosive itself. The distinguishing feature of detonation is self-inertial confinement of the chemical reaction. Detonation science is the study of this special interaction of chemistry and mechanics.

Figure 2 is a diagram showing the conventional idea of a detonation wave. The detonation front is a shock wave, supersonic relative to the material ahead of it, so no signal precedes it. Compression heats the explosive, and rapid reaction ensues. The pressure falls rapidly in the reaction region, as heat is released. Finally chemical reaction is complete, and the gases expand as an inert flow to match the boundary conditions.

After a detonation has run a long distance in explosive the reaction zone is very thin relative to the inert following flow, and it is a good approximation to neglect the reaction zone in any calculation. The reaction zone appears, in a measurement compared with such an idealized calculation, as a small rise in pressure or velocity at the front.

In summary, there is a zone of chemical reaction that is almost unaffected by the boundary conditions, followed by an inert flow that matches between the state at the end of the reaction zone and the boundaries. The state at the end of the reaction zone is the initial state for the following expansion, and must be known to define the inert flow problem. The chemical reaction zone also introduces a length, characteristic of the explosive, that provides a scale for all detonation phenomena. Lengths in the various useful explosives range from a few μm to a few mm.

The solid lines in Fig. 3 show the measured velocities of pieces of aluminum foil embedded in explosive, as the detonation wave passes over the foil. The dashed lines are calculations of the foil motions if there were no reaction zone. The four different lines show the effects of different back boundary conditions, namely different lengths of explosive. The reaction zone shows up as the departure of the solid lines from the dashed lines, as they rise near the front. We see that the reaction zone is there, at least qualitatively like we thought it would be, and that it is only slightly affected by the conditions behind.

The transient change of the reaction zone seen in Fig. 3 is a necessary consequence of a finite reaction zone length. Study of the transients gives us more information about the reaction.

Certainly the largest transient arises in getting the detonation started. Some features of the shock initiation process are shown in Fig. 4. The diagram in the upper left is intended to represent a slab of explosive, large in lateral dimensions, being driven by a piston, shown crosshatched. Beside it, on the right, is a t - x diagram showing what happens. The lowest line in the plot, showing x increasing slowly with t , gives the position of the interface between piston and explosive. The steeper line from the origin is the trajectory of the shock wave in the explosive. Material between the shock and the interface is compressed, and heated by the compression. After an induction time, the material that has been hot longest (next to the piston) begins to react in the little dark area, and quickly forms a detonation. The detonation propagates at high velocity in the shock-compressed explosive, on a trajectory indicated by the steepest line in the diagram, and overtakes the shock wave. Then after a transient phase of interaction, detonation propagates at detonation velocity in the unshocked and undisturbed explosive, along the line going up and to the right. If the explosive is a single crystal of PETN and the piston velocity is about $1.2 \text{ mm}/\mu\text{s}$, the shock will move at about $5 \text{ mm}/\mu\text{s}$ and the shocked region will be at a pressure of about 11 GPa and a temperature of 1070°K . The induction time will be a little more than $0.5 \mu\text{s}$, and overtake will occur at $1 \mu\text{s}$. The high temperature in the shocked explosive leads to an induction time that seems consistent with data about the rate from lower-temperature laboratory measurements.

All of this seems simple and obvious. However, there is a catch. If the explosive is not a single crystal of PETN, but is made by pressing PETN powder

until its density is within 1% of the crystal density, things are different. Doing this same experiment shows that a piston velocity of only 0.4 mm/ μ s, giving a shock velocity of only about 3 mm/ μ s, a pressure of 2.5 GPa, and a temperature of only 475⁰K, initiates the material with 1 μ s to overtake. Now nothing is simple and obvious. The same material can be heated in an oven to 475⁰K and decomposes only slowly. The explanation seems to be that energy is concentrated at local regions called hot spots, and that the temperature is not uniform in the material. (Careful experiments have shown that the description given above of initiation in the single crystal is also somewhat too simple to fit all the observations.) Reaction takes place quickly at the hot spots, and the shock accelerates as it runs. The instant of overtake is blurred a little, and the whole process is more complicated. Several papers at this meeting are devoted to the details.

The important point is that inhomogeneous heating and reaction are important processes in explosives. Figure 5 shows some of the ideas that have been suggested for hot spots. Energy concentration may come from jets of material generated at little vee-shaped intersections of crystallites; from impact of material thrown across a void; from viscous heating in material near the surface of a collapsing void; from shock collision around a high-impedance inclusion; from friction between two crystallites; or from internal slippage in a crystallite. I expect discussion of these processes and others at this meeting. One useful thing we could do is to define the several processes and give them names so we could be sure what process is being discussed.

Other transient effects besides initiation and the small changes in the reaction zone shown in Fig. 3 can also be found. Figure 6 shows some measurements of detonation velocity vs length of run for two explosives. The transient is not observed in PBX-9404, presumably because it is too fast, and is observable but small in Composition B-3.

The length of the reaction zone also implies that there must be edge effects in explosives. One of the simplest ones to interpret occurs in long cylindrical sticks of explosives, because they can be run long enough for all transients to die out, leaving a steady flow. Figure 7 is a diagram of detonation in such a stick. The shock front is curved, so the streamlines are deflected outward as they pass through it. Thus there is a radial component of velocity, and radial kinetic energy, so some energy is not available for the forward motion of the detonation.

Figure 8 is a plot of detonation velocity vs reciprocal radius of the stick, showing how the detonation slows down in small sticks. It also shows a large qualitative effect: the detonation will not propagate at all if the stick is too small. Failure radius for the explosives shown varies from about 0.2 mm to about 20 mm, a range of about 100 times, and the slowing at failure varies from about 1% to about 15%. We expect that both the reaction zone length and the way the rate depends on the local state influence these values.

The reaction rate and state dependence can be influenced by adding a catalyst, or by changing the hot spots. Figure 9 shows results of both of these changes. The liquid explosive nitromethane is used as the standard. Its diameter effect curve appears to be a straight line, and the detonation velocity has decreased by only $\frac{1}{2}\%$ at failure. Adding a catalyst or sensitizer, DETA (diethylenetriamine, really 2,2'-diamino diethylamine), in a very small amount, 0.03%, changes the failure diameter by a factor of two, but leaves both the detonation velocity and the amount of decrease unchanged. Adding silica particles and a little gelling agent to hold them in place also decreases the failure diameter by a factor of two, but now the velocity decrease is about 13% at failure. The slope of the curves at large diameter, shown in the inset, indicates that adding DETA shortens the reaction zone, but adding silica particles

increases it appreciably. We think that the homogeneous liquid explosives fail when a large rarefaction wave starts at some edge point and propagates in, putting out the detonation. We call this process catastrophic failure. Perhaps the silica particles diffuse the catastrophic wave, and prevent that kind of failure, so propagation continues until the energy loss to the edge is great enough to make it fail. An alternate explanation is that the hot spots produced when the wave interacts with the particles changes the effective state dependence of the reaction and thus changes the failure regime. This work is being done by Ray Engelke.

There are other detonation effects we could discuss, but the ones presented so far make up a reasonable background, except for one feature. It has been tacitly assumed that the detonation front and the reaction zone are locally smooth and uniform. If this were so, and we photograph the light emitted by a detonation front in a transparent explosive in a long cylindrical stick, the light should be nearly uniform over the front. We might expect it to be just slightly brighter in the center and a little dimmer at the edges, but without much change. Figure 10 is a photograph of the light emitted by a detonation in a transparent liquid, nitromethane/acetone 80/20 volume % mixture, in a brass tube 19 mm inside diameter. It is clear that there is a pronounced transverse structure, and that the wave is not locally smooth. Sequential photographs show that the edges move and the spots transform. Similar structures have been photographed in various liquid and solid explosives, and probably they exist in most explosives, although perhaps not in all. In gases, the structures have been studied in considerable detail. They are easier to work with in gases because the reaction zone can be lengthened by reducing the initial gas pressure. Some soot film results are shown in Fig. 11, from Strehlow. While the regular patterns are the most studied because they are the most detailed, the irregular ones are more common.

Figure 12 lists four ideas obtained from our review of detonation phenomena, and now we want to decide how we may use them for deduction. The first two items, inertial confinement and appreciable reaction zone length, tell us that if we are to make sensible deductions about detonations we must include details about the interaction of chemistry and mechanics. The second two items, hot spots and transverse waves, tell us that some technique for averaging their effects must be included in a description of most real explosives.

The equations that describe detonation are shown in Fig. 13. The first equation is the expression of the conservation of mass. The second is the conservation of momentum, or $F = ma$. The third is the conservation of energy. The next equation is the description of the material properties; it gives the specific internal energy as a function of pressure, volume, and internal variables, particularly composition. We use a vector notation for the internal variables, indicated in the next equation. Notice that u and the operator grad in the first two equations are ordinary vectors in real space, and that the composition vector is in composition space. The time derivative, represented by the dot, in the third equation means that time derivatives of the composition variables will appear. These quantities, represented by the last equation, are the chemical rates.

Application of these equations presents the difficulty that we don't know what to use for the material properties, the equation of state and the rate. Another difficulty is that although the equations, when solved correctly, presumably describe the details of the transverse wave structure, and if the local structure of the explosive were used as input, the interactions of the hot spots, we can't imagine working with that much detail.

One approach is to ask simpler questions. For example, suppose we forget about transverse waves and hot spots and assume that there could be a plane,

steady detonation. For minimum assumptions about the equation of state, and any sort of rates, what forms can the reaction zone take? This problem has been studied rather completely. With restriction to one irreversible rate the familiar ZND solutions appear. In these, the pressure falls through the reaction zone to the CJ point if the flow is supported by a piston with a velocity less than CJ particle velocity, and the flow is exactly sonic at that point. If the piston velocity is greater than that value, the whole flow is subsonic. These two solutions appear at the right in Fig. 14. If the rate is allowed to have two reversible reactions, nine new possibilities are added to the ZND solutions. Other complications of the rate or equation of state lead to other solutions. I don't have time to discuss these here; an interested person might look at Detonation by Fickett and Davis. The important point is, it seems to me, that there are lots of kinds of solutions, and we must compare them with experiment to find out which are of physical interest as approximations to real detonations.

The solutions above, the plane steady ones, have only one variable. Two variables, either plane nonsteady or steady with two space variables, make the problem much harder. Figure 15 is a diagram of a steady, cylindrically-symmetric flow of a detonation propagating in a cylindrical stick in the limit where it has become steady. John Bdzil has treated this problem. The important boundary conditions are the deflection of the flow at the edges, and the detonation jump conditions. The experimental data for calibration and comparison are the shape of the detonation front and the detonation velocity for different cylinder sizes, and the failure diameter. Figure 16 shows how the calibrated theory fits the experimental shape measurements for two sizes. The effective reaction zone thickness and the strength of the state dependence of the rate are found by this fit.

Calibration of an equation of state and a reaction rate to force agreement between a theoretical treatment and a limited set of experiments has been the almost universal procedure for testing detonation theory. If the calibration is impossible, clearly the theory must be wrong, but if it is possible, it doesn't prove that the theory is right.

One kind of experiment that gets away from this bind is shown in Fig. 17. We expect that the detonation velocity is a smooth function of the initial energy and the initial density of the explosive if the final composition can be held fixed. The upper line in the back in the figure is the detonation velocity of nitromethane for a range of temperature. Changing the temperature changes the density quite a lot, and the energy a little. The sloping line leading up is the detonation velocity for mixtures of nitromethane and acenina, and the energy changes a lot and the density a little. Acenina is an equimolar mixture of nitric acid, water, and acetonitrile, and has the same atomic composition as nitromethane; therefore the final products, if the reaction goes to completion, are always the same. What we are after is a fit to the detonation velocity surface in the neighborhood of the intersection of the two lines.

The theory for the application of the results is shown in Fig. 18. The expression for D at the top means that from experiment we know the detonation velocity as a function of initial density and energy near some point. The sketch of the p - v plane shows the theoretical result for a plane, steady, laminar detonation wave with any of a class of simple rate laws. The straight line, the Rayleigh line, and the curved line, the Hugoniot curve, are tangent at the state point that occurs at the end of the reaction zone. As a consequence, one can show that the next two equations hold. The left-hand sides are obtained from the experimental data. The right-hand sides contain two variables, which are thermodynamic derivatives evaluated at the tangent point,

and the two equations can be solved to get their values. Finally the pressure at that tangent point can be obtained from the last equations. The pressure obtained from these detonation velocity measurements interpreted using this theory can be compared with the pressure obtained from more direct measurements. The best values I have for the pressure are 12.2 ± 0.6 GPa from the detonation velocity measurements and 14.2 ± 0.4 GPa from the direct measurements. I think the most likely reason for the disagreement is that the effect of the transverse waves has not been properly accounted for in the theory.

Figure 19 shows similar data for solid and liquid TNT. The solid can be pressed to different densities below its crystal density, and the specific energy remains almost constant. The line at the back shows a small segment of the fit to measurements for a large density range. Melting the explosive adds energy to it, and heating it above the melting point adds more energy and changes the density. The theory can be applied as for nitromethane, and the disagreement is similar. However, the surface that fits these sets of measurements is not nearly plane, even though the energy change is less than 5%. Probably the data indicate the detonation in the liquid and the solid are different, perhaps because the hot spots in the solid and the transverse waves in the liquid have different effects in the reaction zone.

This concludes the introduction to detonation phenomena for scientists in other fields. I have tried to show that detonation is confined by the inertia of the material itself, that the time scale of the chemical reactions is long enough that it must be taken into account, and that hot spots and transverse waves are important in the process. Figure 20 is a diagram of detonation physics, intended to summarize the introduction, and to help place the papers presented at the meeting in perspective. At left and right at the top of the diagram are boxes representing input of the material properties, the equation

of state and the chemical rates. Down the center fluid mechanics is used to find the solutions to interesting problems for explosives with various boundary and initial conditions, leading to predictions that can be compared with experiment. In the diagram I have tried to emphasize the effects of transverse waves and hot spots that must be averaged some way to predict the measurable effect of the explosive. I think that Nunziato may talk about aspects of that problem. We will also hear two papers about the details of hot spots. Usually input from the EOS and r boxes is not available, and fluid mechanics for laminar flow is used. The results from experiments are used to get equations of state and rates that make the predictions agree with the experiments. We will hear several papers describing experiments, forms for the rates, and calibrations. The exciting new chemistry and quantum mechanics should lead us to ways to fill in the r box, and we will hear some papers about efforts in that direction. I expect the future to bring a lot of work in detonation chemistry.

At this meeting there are apparently no papers scheduled on the subject of equation of state, but the new work in statistical mechanics, and the new capabilities for static high pressure experiments will make it one of the fields of the future, too.

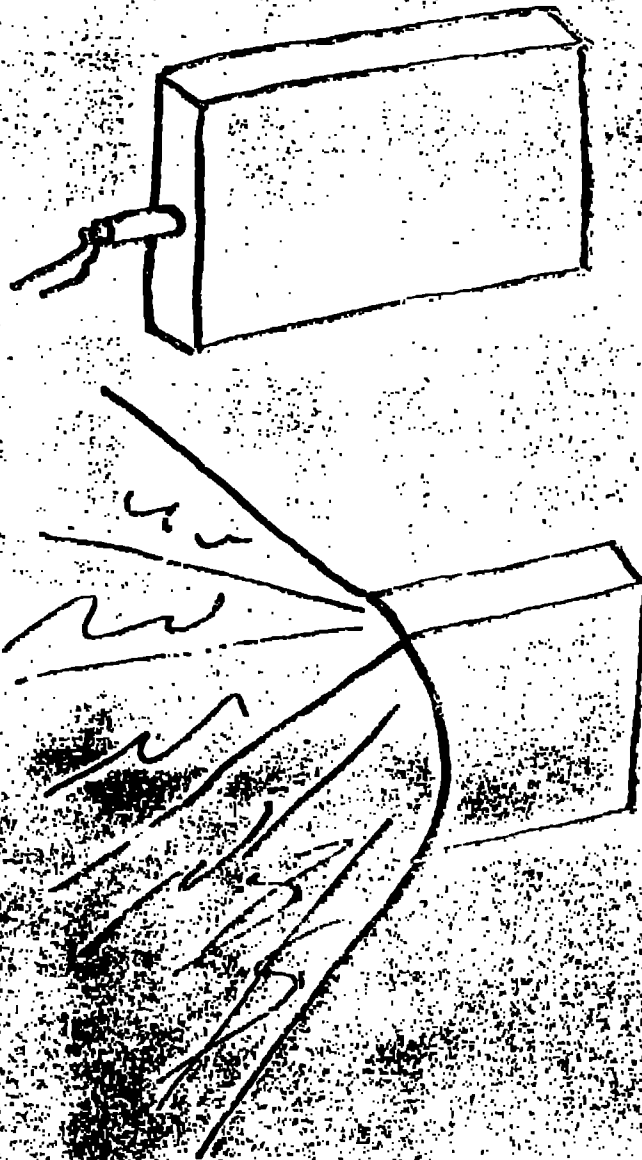
Another subject that seems to me to be given too little attention at this meeting is that some kind of averaging is needed to take account of the inhomogeneous reaction zone. Hot spots are certainly present in most of the solid materials we use in shock wave physics, but usually they can be neglected because they cause only small effects. In detonation they cannot be neglected because they are the main effect; reaction begins in their neighborhood, and large composition changes take place there. Until we can take account of the inhomogeneities, we cannot use real equations of state and real chemical rates.

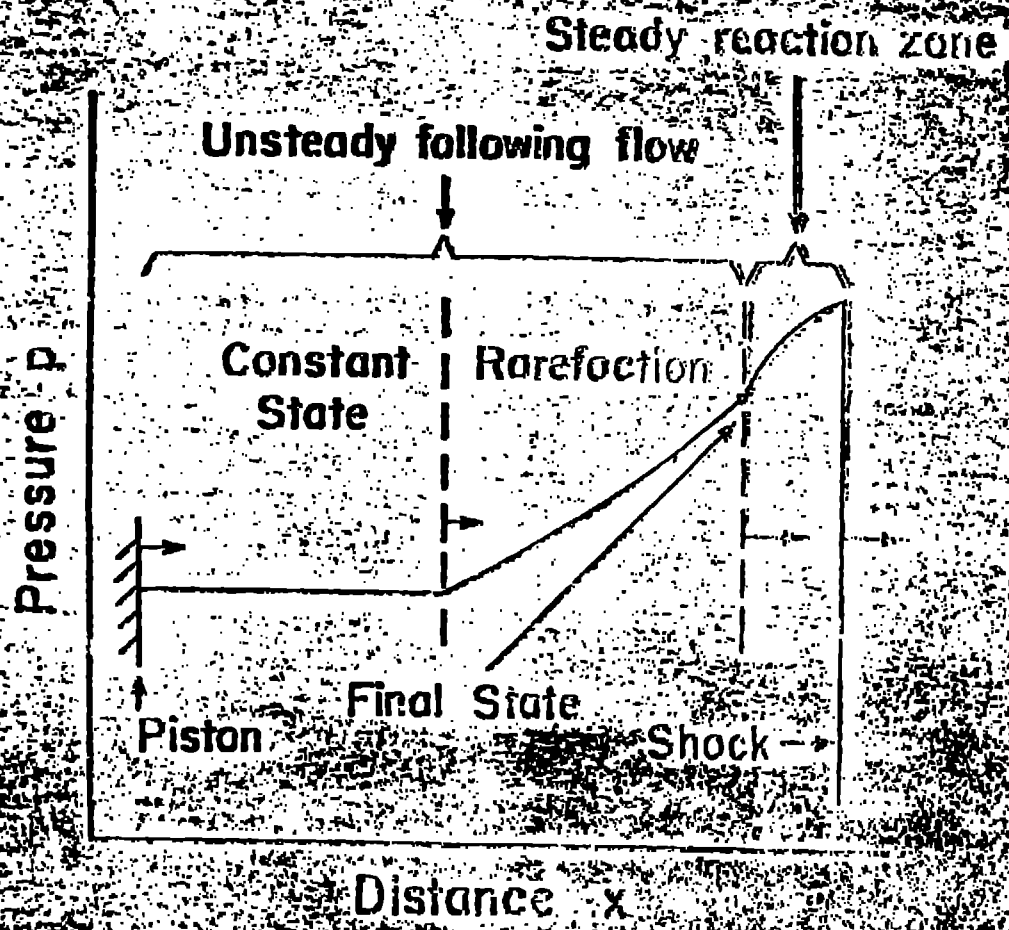
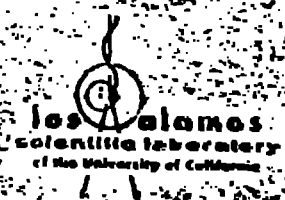
So far, I have been talking about scientific understanding. Another topic is practical application. Does our research have value for engineering, or

are we just enjoying ourselves by satisfying our curiosity? Put another way, can we get anyone to pay for the research? It is always hard to guess what new things might come from research, but we can extrapolate a little. A safe explosive system is one that has all its explosive parts near failure size, so that when they are initiated intentionally they operate well, but accidental initiation will almost always fail. In such a system, the reaction zone is important, and we have to understand it to make one. We are already a long way in that direction with the insensitive explosives now in use. Further, the failure diameter is controlled by the inhomogeneities in the reaction zone, and they have to be understood if the manufacture is to be satisfactorily controlled. Any systems designed for safety will need knowledge of transients, edge effects, and failure.

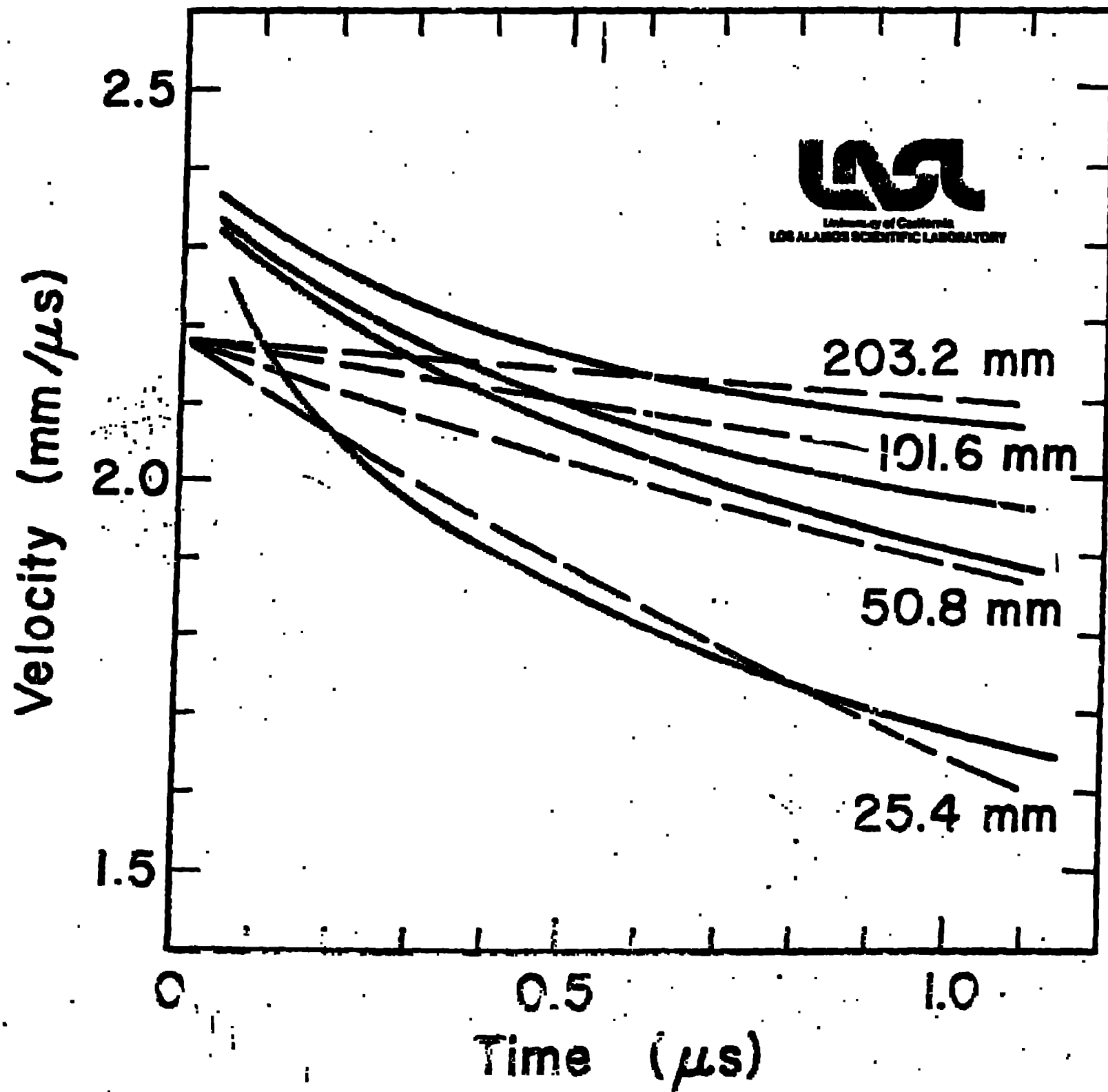
The next step beyond good engineering of systems using explosive is designing the explosive itself. If we could fill out the boxes of Fig. 20 in detail, we wouldn't even have to make samples of the explosive molecules for testing, or experiment to find the best particle size distribution. Certainly we can't go that far very soon, but every step toward explosive design is important.

I'm sure the discussion sessions and the papers to be presented will provide a new view of our subject. Just in case there is any newcomer to the business who feels diffident about speaking up in disagreement with the majority when so many of us have been working so long with explosives, let me quote from E. Bright Wilson, Jr. "No one can be so obstructive of progress as the 'expert' who has worked all his life on a single subject."

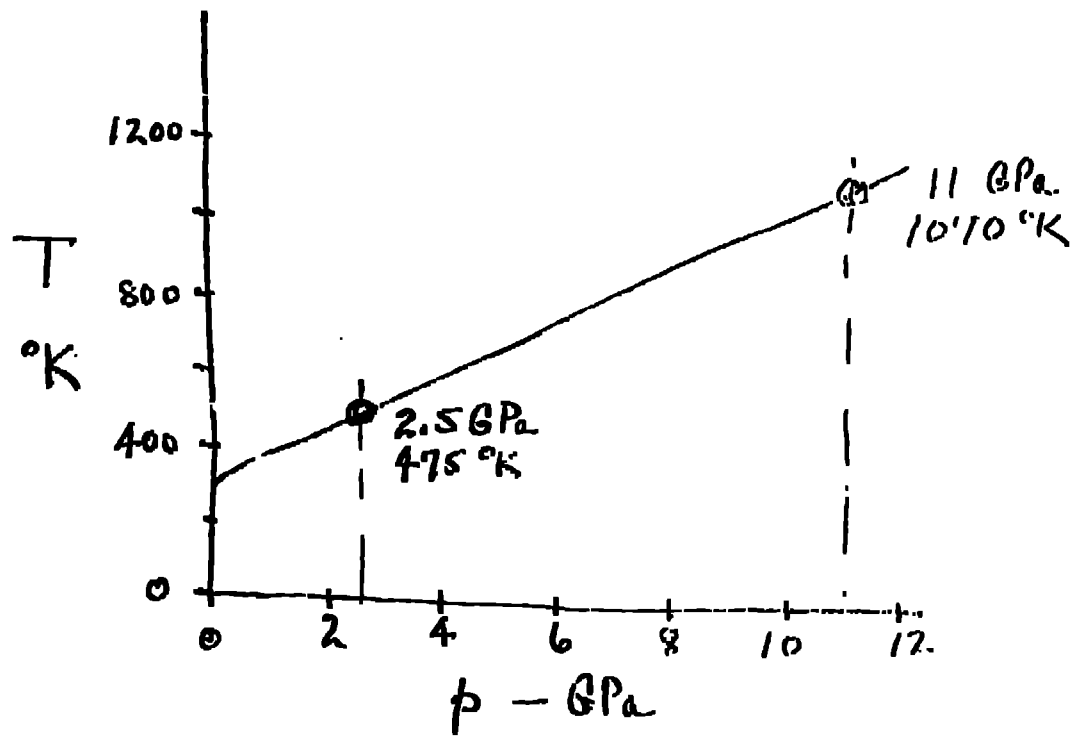
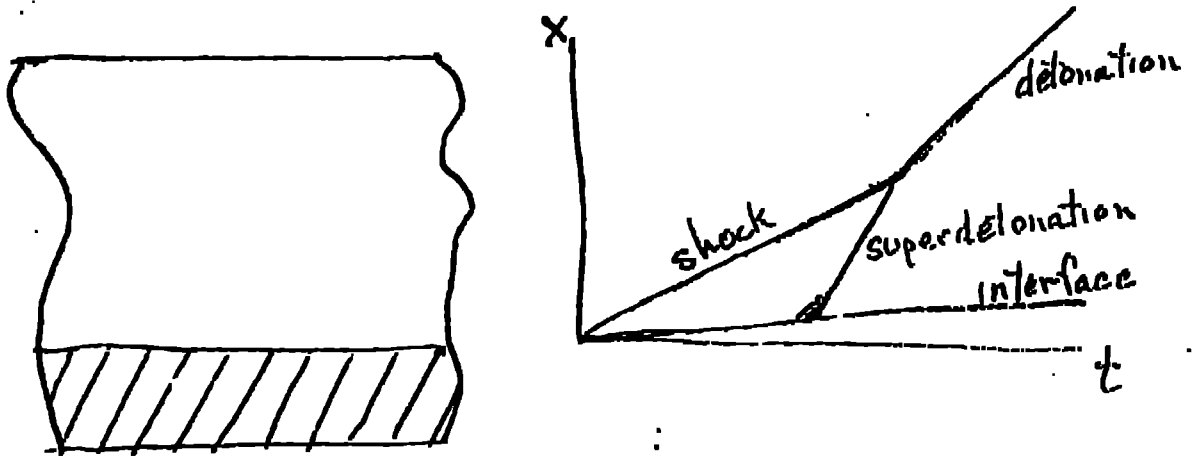


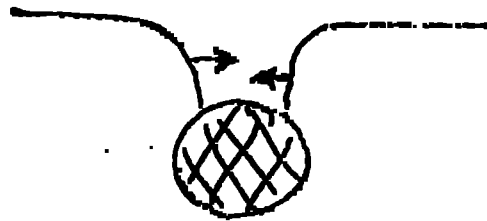
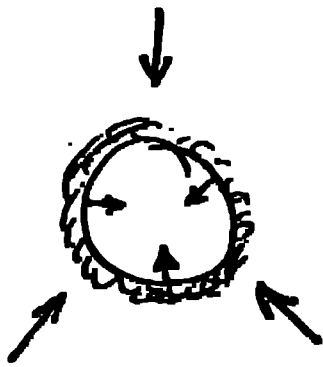
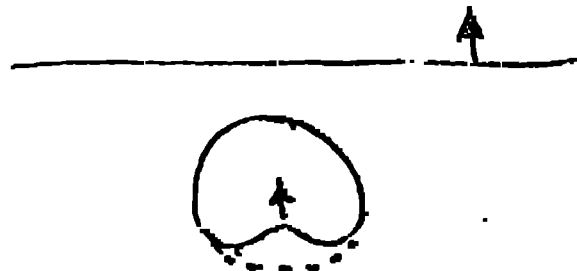
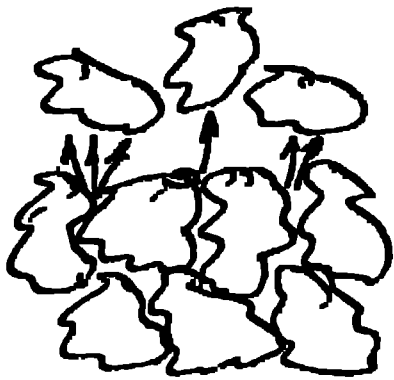


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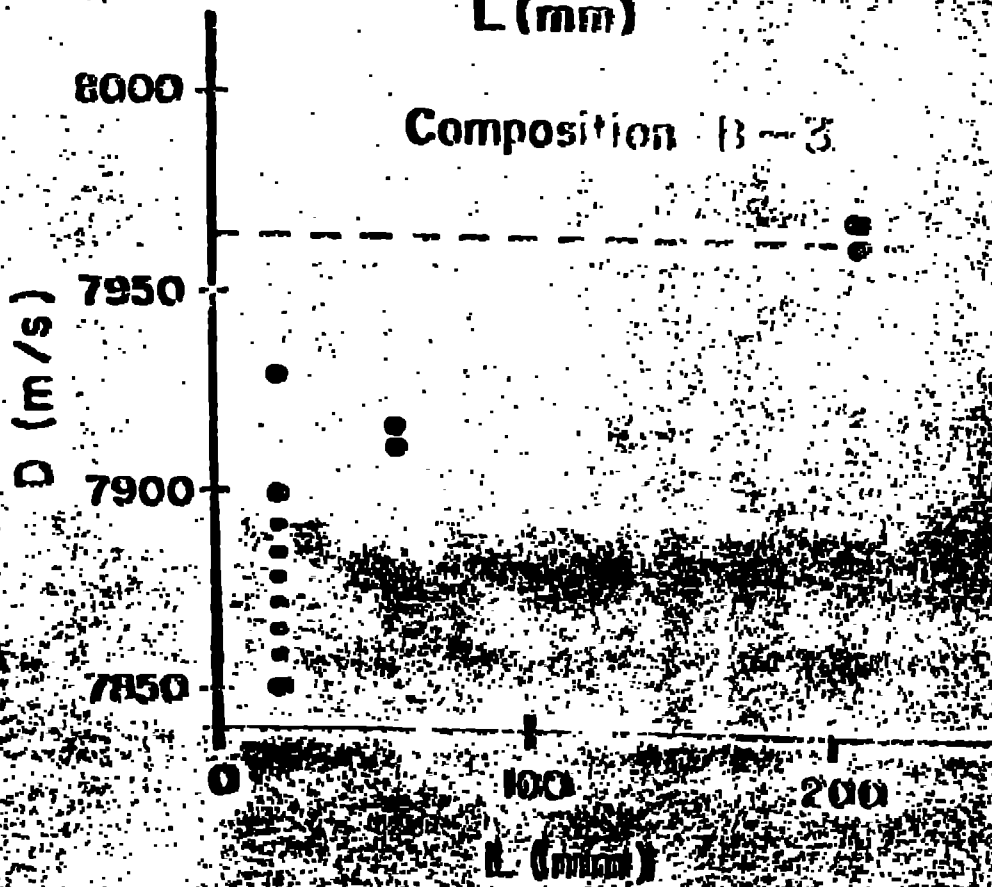
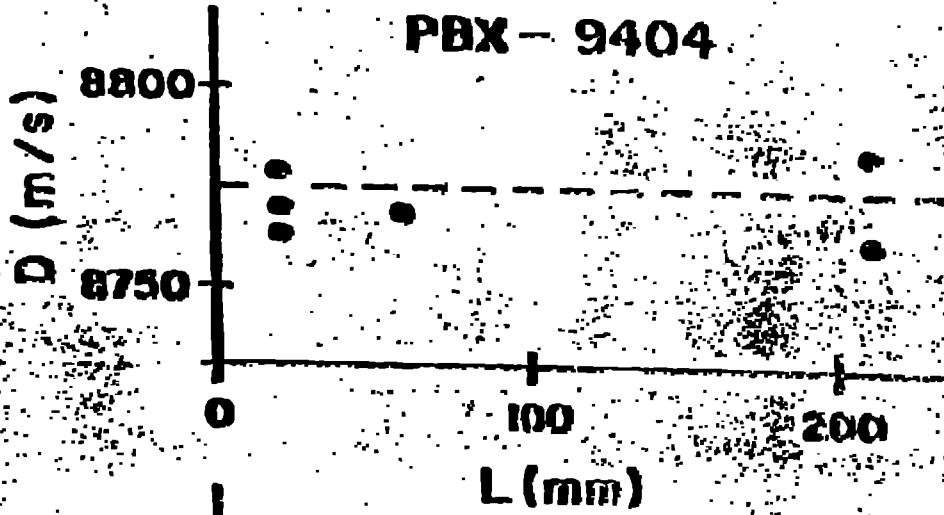


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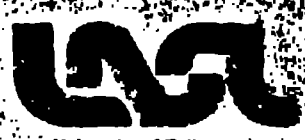
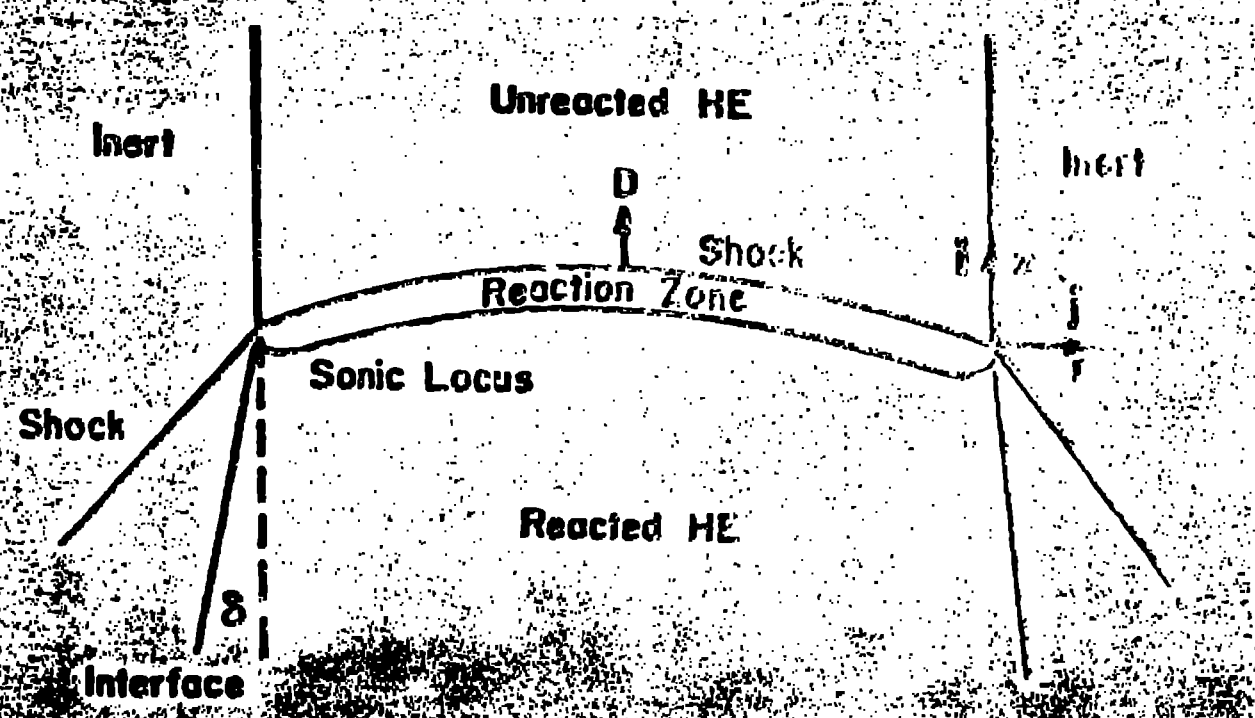




HOT SPOTS



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University of California
JPL/AMES SCIENTIFIC LABORATORY

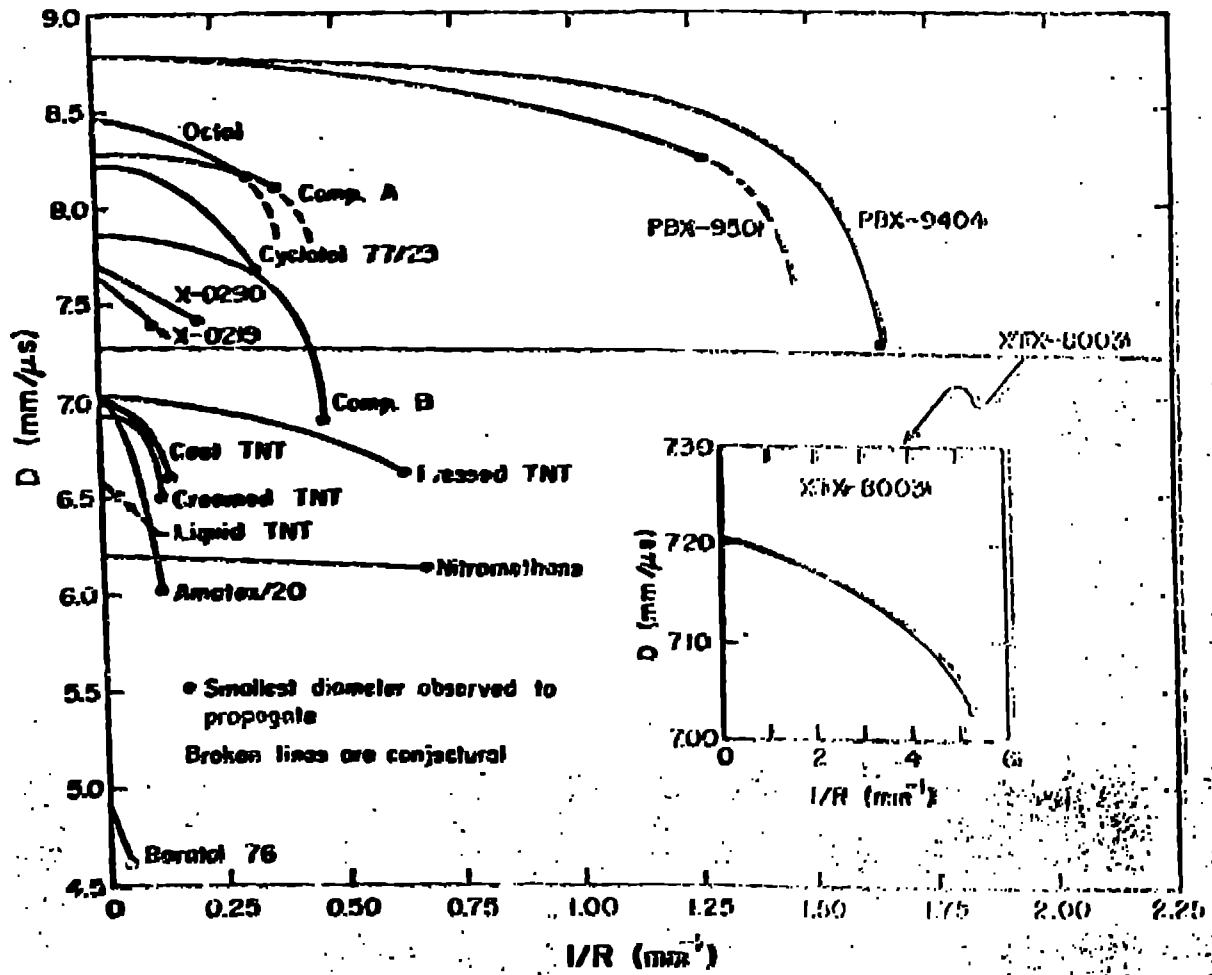
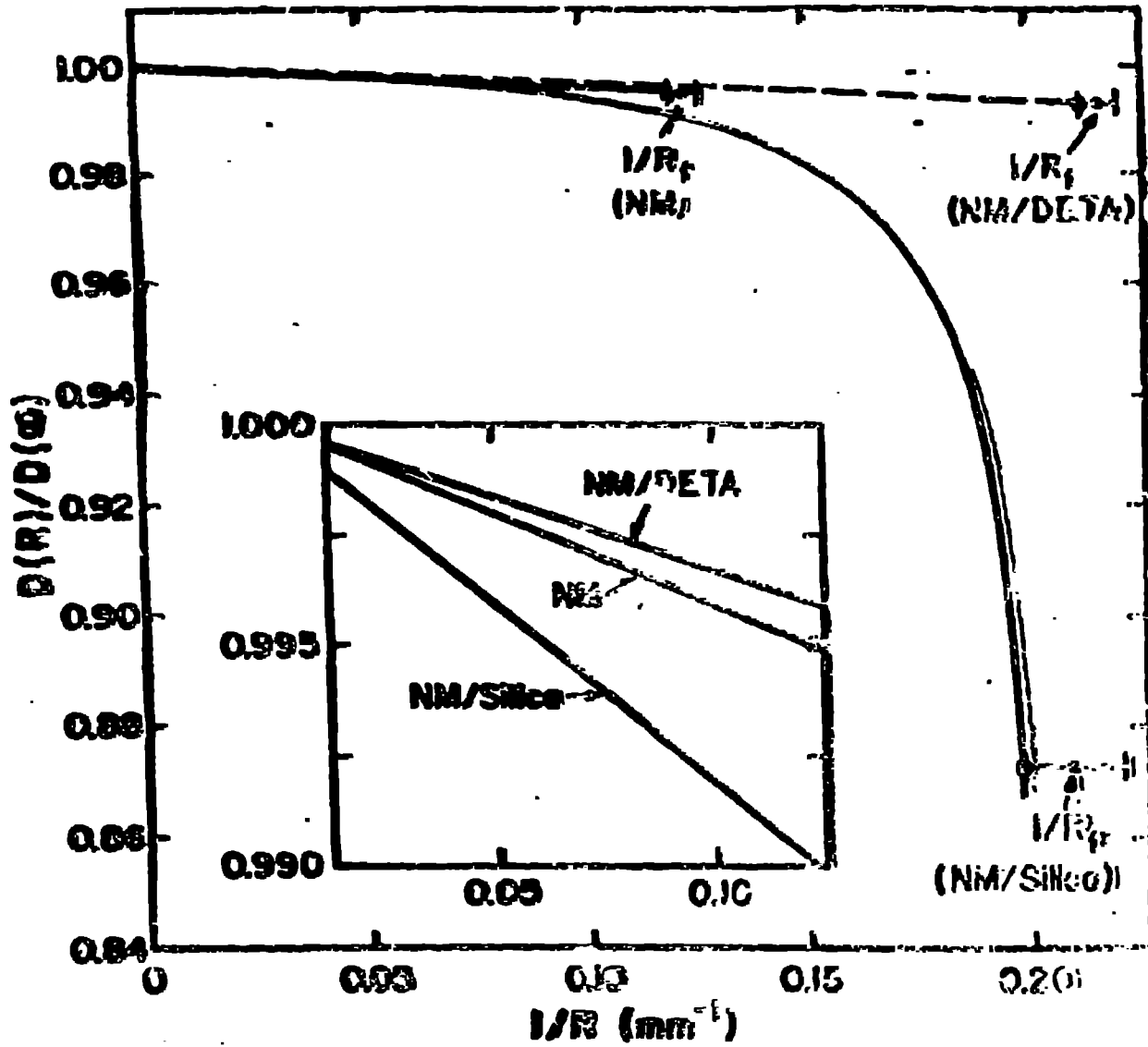


Fig. 2. Composite of diameter-effect curves in the D -vs- $1/R$ plane. Broken lines are conjectural.



100 WT % NI

$$D(\infty) = 6.247 + 0.032 \text{ MW} / \mu \text{S}$$

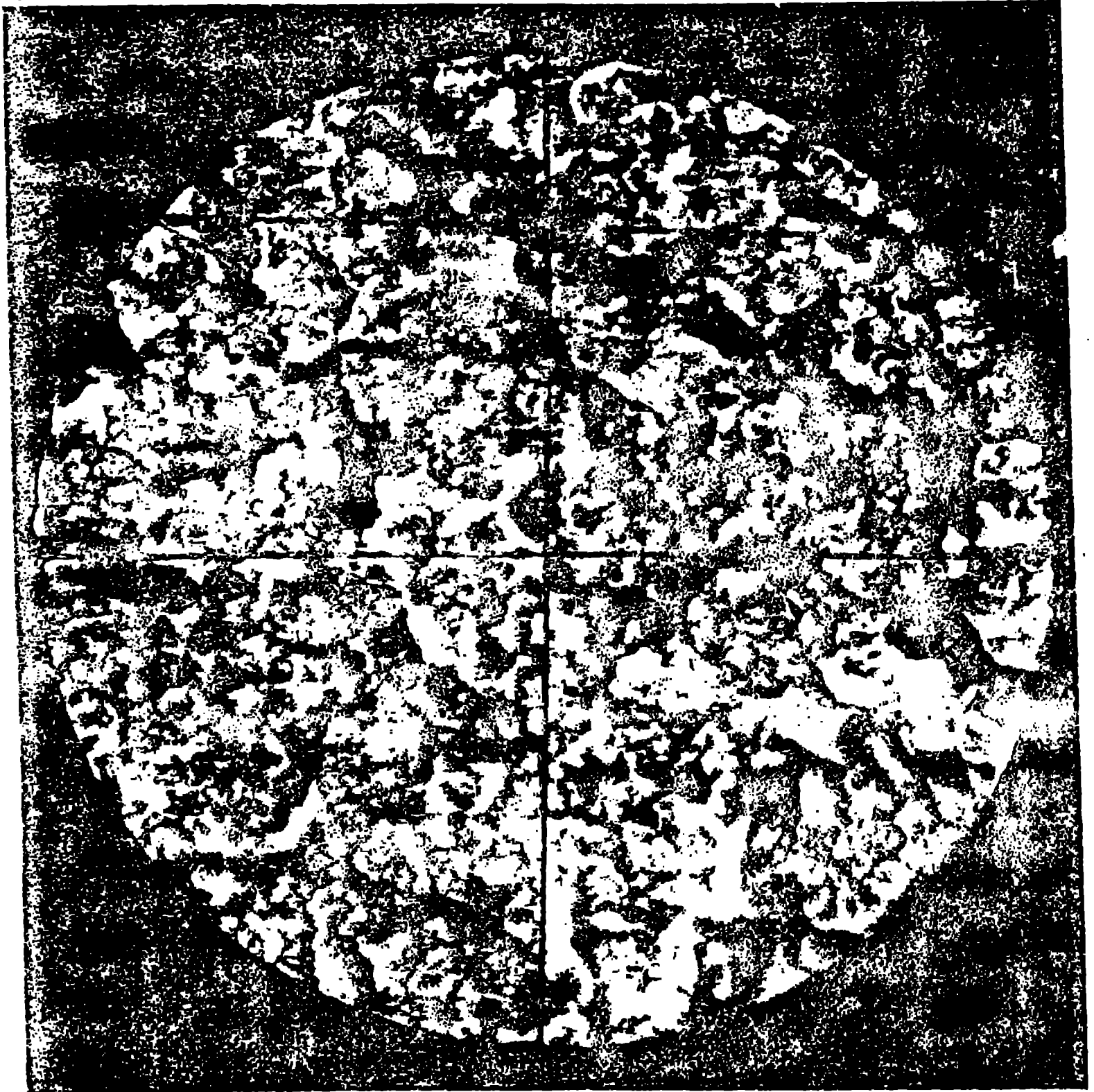
99.97/0.030 WT % NI/DETA

$$D(\infty) = 6.238 + 0.031 \text{ MW} / \mu \text{S}$$

92.75/5.00/1.25 WT % NI/SILICA/GLASS

$$D(\infty) = 6.157 + 0.032 \text{ MW} / \mu \text{S}$$







EXCELLENT: $2C_2H_4 + 3O_2 - 75\% Ar$

GOOD: $126H_2 + C_2 - 49\% Ar + 16\% N_2$

P = 100 mm

P = 150 mm

PROPAGATION DIRECTION

2 IN



POOR: $2H_2 + C_2 - 25\% Ar$

IRREGULAR: $CH_4 + 2O_2 - P = 50 mm$

INERTIAL CONFINEMENT

REACTION ZONE LENGTH

HOT SPOTS

TRANSVERSE WAVES



$$\dot{\rho} + \rho \operatorname{div} u = 0$$

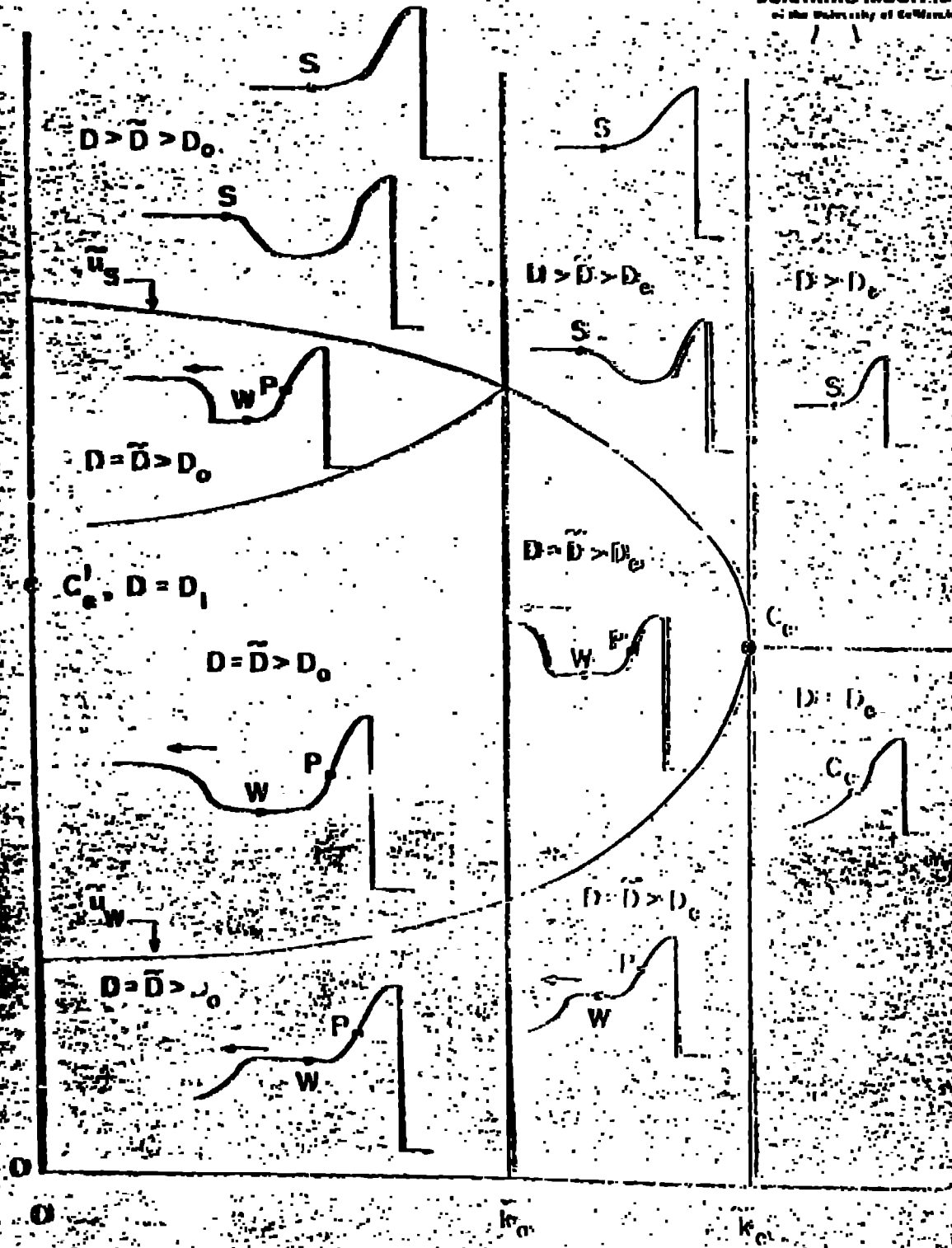
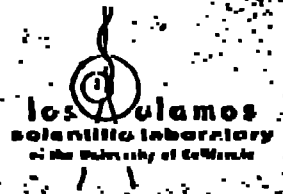
$$\dot{u} + v \operatorname{grad} p = 0$$

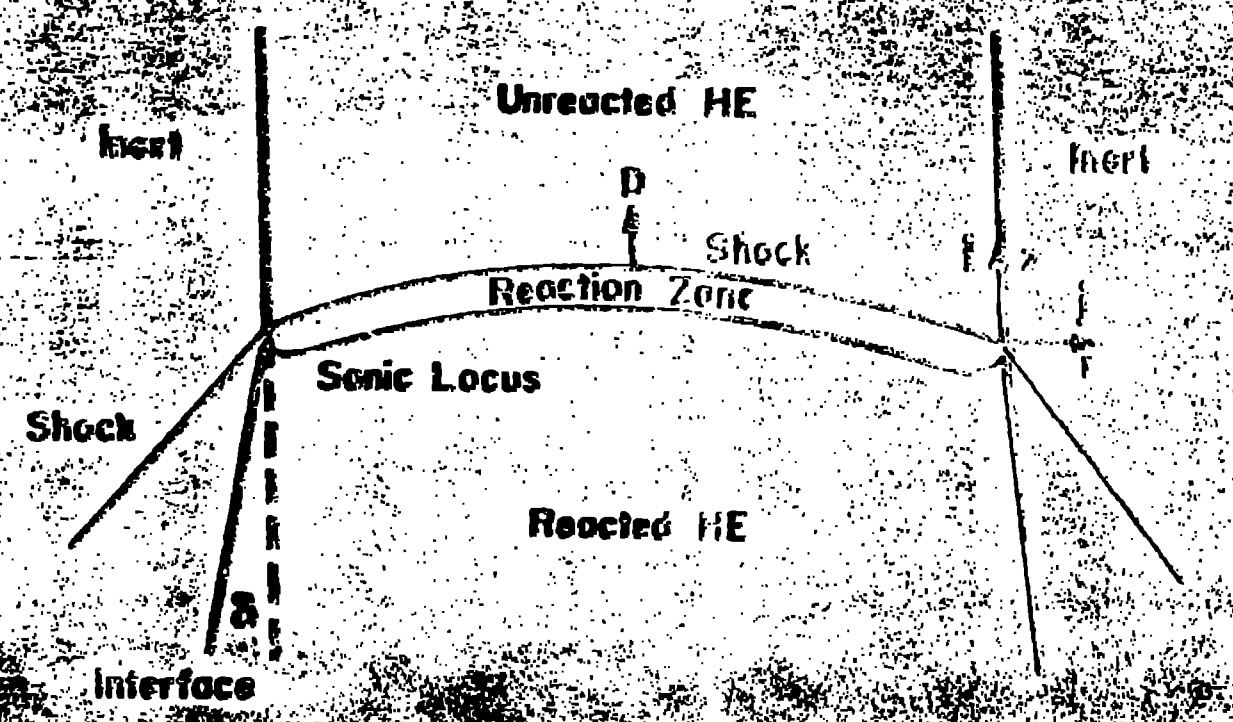
$$\dot{E} + p\dot{v} = 0$$

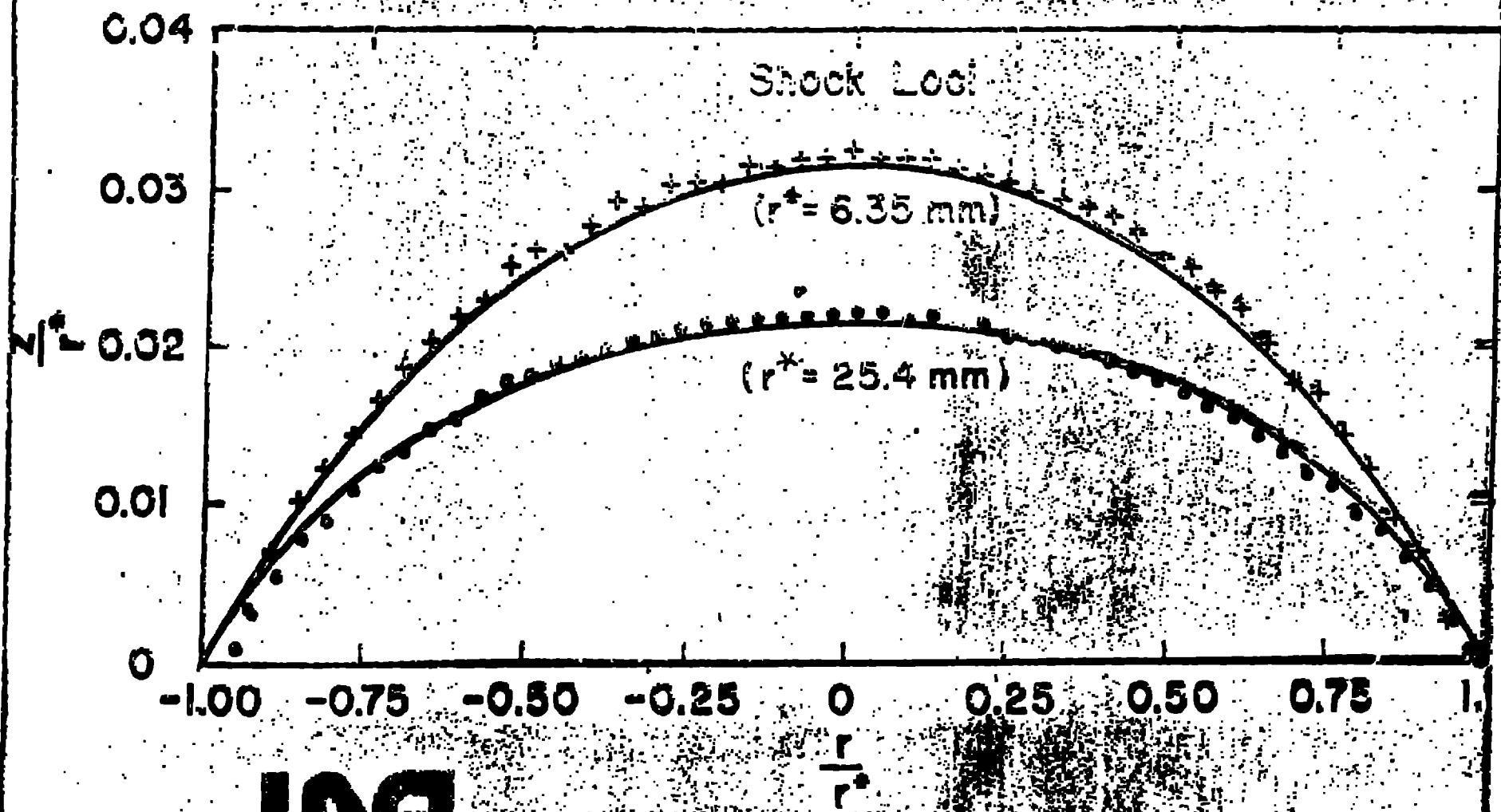
$$E = E(p, v, \lambda).$$

$$\lambda = (\lambda_1, \lambda_2, \dots, \lambda_n).$$

$$\dot{\lambda} = r.$$

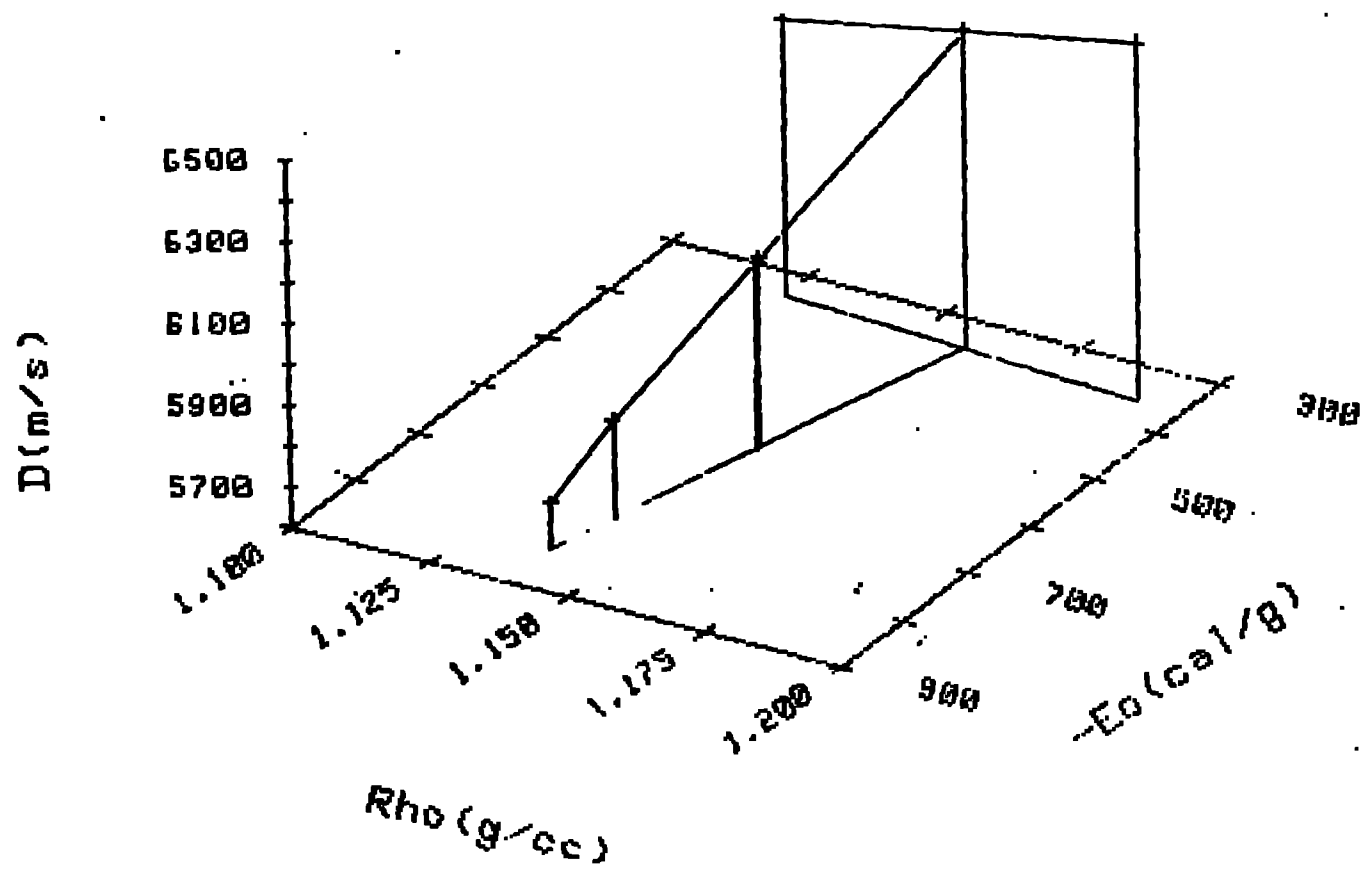




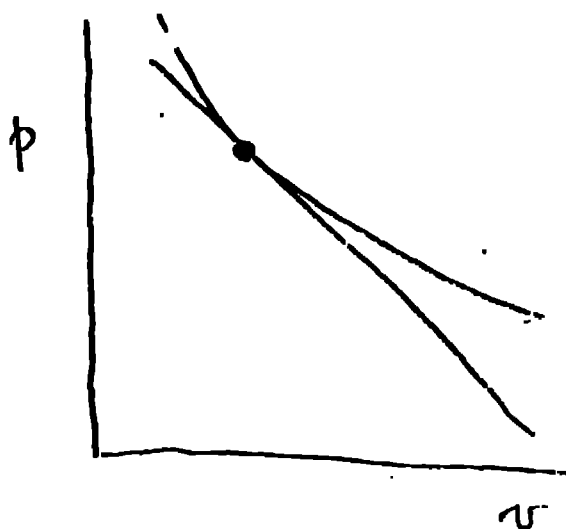


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Nitromethane/Acetona



$$D = D(\rho_0, E_0)$$



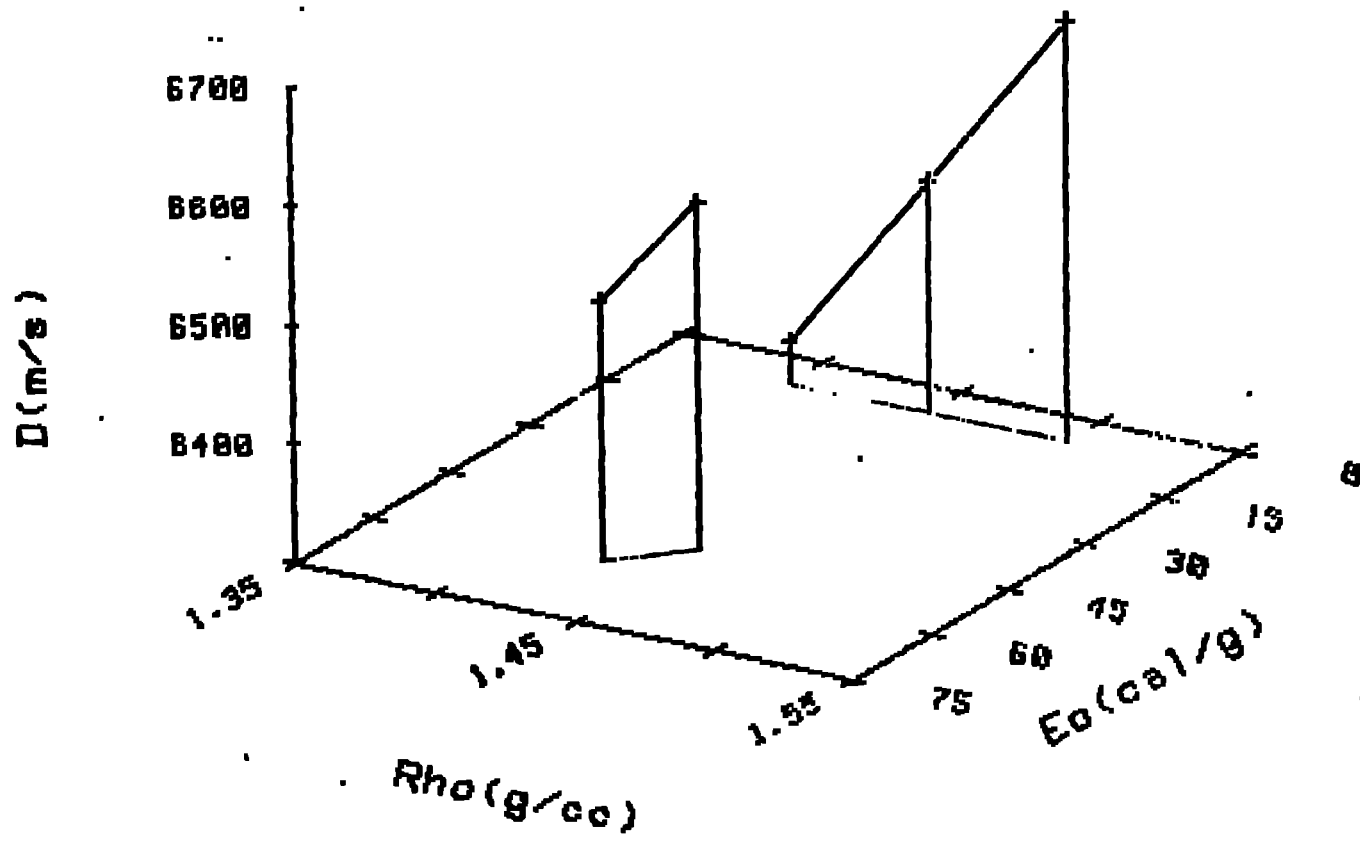
$$D \frac{\partial D}{\partial E_0} = \frac{\alpha(\nu+1)^2}{2+\alpha}$$

$$\frac{\rho_0}{D} \frac{\partial D}{\partial \rho_0} = \frac{\nu-1-\alpha}{2+\alpha}$$

$$p_j = \frac{\rho_0 D^2}{\nu+1}$$



Solid/Liquid TNT



EOS

r

