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THE MECHANISM OF THE TRANSITION FROM DEFLAGRATION TO  
DETONATION IN HIGH EXPLOSIVES (U)

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THE MECHANISM OF THE TRANSITION FROM  
DEFLAGRATION TO DETONATION IN HIGH EXPLOSIVES\*

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ABSTRACT: Experimental results of the study of spontaneous transition from deflagration to detonation at the Naval Ordnance Laboratory indicate that the approach to the problem can be in two stages; the first is the formation of a shock from pressure waves engendered by a confined deflagration, and the second the shock-initiation of detonation. Since a preliminary analytical treatment of the first stage, reported previously, led to promising results, a more extensive IBM-704 program has now been undertaken. Two numerical codes have been tested, a previously developed one based on the so-called "q-method" and a special one written for this program which avoids amplitude fluctuations inherent in the "q-method" and thus gives a more realistic representation of a shock wave. Representations of spontaneous shock formation obtained by the two numerical codes and by the analytical treatment are discussed and compared. The numerical methods yield the temperature as a function of time and location during growth of the shock and thus allow a study of simple chemical kinetic models. Introduction of chemical kinetics into the program gives a basis for elucidation of the second stage of the transition problem, namely shock-initiation of detonation.

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\*Submitted to the faculty of the Graduate School of Arts and Sciences of The Catholic University of America in partial fulfillment of the requirements for the degree of Doctor of Philosophy. This dissertation was approved by Dr. Virginia Griffing, Professor Chemistry, as director and Dr. Karl F. Herzfeld and Rev. John S. Dooling as readers.

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Mechanisms of the transition from deflagration to detonation in explosives and propellants are presently of practical interest; particularly because of the potential hazard posed by large, solid propellant rocket grains. Until recently, little was known about the processes involved. Experimental studies of the transition were published in NavOrds 5748, 6104 and 6759. This report is concerned with theoretical interpretation of the transition process. While the hydrodynamic model is one dimensional, considerable insight into the mechanism of the deflagration-detonation transition is provided. It is part of a continuing investigation in this field.

This research was supported by NOL Project FR-59, Transition from Deflagration to Detonation.

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ALBERT LIGHTBODY  
By direction

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THE MECHANISM OF THE TRANSITION FROM  
DEFLAGRATION TO DETONATION IN HIGH EXPLOSIVES

INTRODUCTION

While the phenomena of deflagration (slow, pressure-dependent burning) and detonation are reasonably well understood, spontaneous transition from one regime to the other is still in early exploratory stages, and it is one of the major unsolved problems in explosives technology. Gross experimental features of the phenomenon have emerged only recently (1,2,3,4,5). It appears that the onset of detonation in condensed explosives is preceded by a relatively long (up to 80  $\mu$ sec) interval of rapid burning which propagates at a fraction (1/10 to 1/5) of the steady state detonation velocity. There is also evidence that the actual transition from rapid burning (sometimes termed "low order detonation") to steady state detonation takes place rapidly (within several microseconds) at a plane some distance ahead of the burning front.

The evidence thus far is consistent with the hypothesis that the onset of detonation is due to a shock wave which arises spontaneously as a result of deflagration, and which initiates detonation in unburnt explosive. The hypothesis was subjected to quantitative scrutiny at this Laboratory; in addition to experiments mentioned above, a preliminary theoretical treatment was carried out (6) by means of the following model:\*

A thermally initiated (slow) laminar flame progresses into a homogeneous solid explosive charge. Pressure of the hot products, because of rigid confinement, increases steeply and, in consequence, sends compression waves into unburnt explosive.

\* Reference 6 gives the conceptual and analytical basis of the computational work described below, and it will be frequently referred to in the subsequent pages.

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On this basis it was shown that compression waves thus formed coalesce into a shock wave within 10-15 cm from the region of thermal initiation. Since, experimentally, the typical pre-detonation distance is in the same range (6-14 cm), it appears reasonable to suppose that the theoretically computed shock is the direct cause of detonation.

While the analytical methods thus give promising results, it is very desirable to extend the treatment in two ways: first, by repeating the computation using different equation of state parameters and different shock-generating pressure pulses; and second, by calculating the energy (or temperature) as a function of time and distance. The latter computation can then, in principle, be used to study the chemical kinetics of the explosive reaction during build-up and thus elucidate the transition phenomenon. Such an extension clearly calls for machine computation. This report gives an introduction to the computational program which is now in progress.

The report consists of four parts. The first part describes the scope of the program treated so far and the equation of state used. The second and third parts describe two different numerical codes for the IBM-704 computer and compare the results from these codes with the previously obtained analytic results. The fourth part describes the shock formation, chemical kinetics and shock initiation.

#### SCOPE OF THE PROBLEM

##### A. Hydrodynamics

As has been stated above, the approach to the problem of transition to detonation at the Naval Ordnance Laboratory has been via two stages. The first one is formation of a shock from pressure waves engendered by a confined deflagration. The second one is shock-initiation of detonation.

The shock formation problem is programmed in the following way: The difference equations for conservation of mass, momentum and energy are written down as applied to a one-dimensional flow problem. The explosive charge, which obeys an equation of state described below, is divided into  $N$  zones ( $0 < N < 500$ ). At time  $t = 0$  the pressure throughout the

charge is fixed at a low but finite value ( $P(t=0) = 0.08$  kbar). At subsequent times, the near boundary is subjected to prescribed pressures increasing with time. The result is that compression waves of increasing amplitudes travel forward from the near boundary.

For a realistic description of the transition process the pressure at the near boundary must simulate the backing pressure rise in a confined deflagration. In such a case the theoretical relationship between  $P$  and  $t$ , derived in Ref. 6, is given by

$$t = K \int_{P_0}^P \frac{dP}{P(A-P)^2}, \quad (1)$$

where  $K$  and  $A$  are constants; at low pressures this is sufficiently well approximated by the exponential  $P = P_0 e^{kt}$  where  $P_0$  (i.e. pressure at  $t=0$ ) and  $k$  are experimental parameters. The exponential form, which was used previously in the analytical treatment, is used also in the machine computations. However, an indefinitely long exponential pressure increase would be unrealistic, because it would lead to unreasonably high pressures as well as to extremely high values of  $dP/dt$ . In reality, such a situation does not occur; rather, the pressure will increase until the confinement is broken and then decrease. As a crude simulation of such behavior the pressure in the computation is allowed to increase exponentially until about 10 microseconds after the estimated bursting pressure of the steel casing has been attained; thereafter, the pressure is assumed constant. The last stipulation may be at least partly justified if one assumes that the actual pressure decrease is relatively slow; it appears rather more realistic than the other extreme, namely a discontinuous pressure drop to zero, which would cause too rapid a rear-rarefaction to set in. Thus the assumed near boundary condition is

$$\begin{aligned} P &= P_0 e^{kt} & t &\leq t(P_{\max}) \\ P &= P_{\max} & t &\geq t(P_{\max}) \end{aligned}$$

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Hydrodynamically, the problem of coalescence of compression waves into a shock can be divided in two parts. In the first part the compression is isentropic and the flow is simple. The compression energy is  $E_s$  and the temperature attained,  $T_s$ , is given by  $T_s - T_0 = \frac{E_s}{C_v}$ , where  $C_v$  is heat capacity of the explosive and  $T_0$  the ambient temperature. This part was treated analytically in Ref. 6 by the method of characteristics. The method, in fact is valid only for such simple flow (i.e. no shocks); it does not give a basis for further calculation; in particular, it cannot show where and when the shock becomes strong enough to initiate detonation.

The second part of the problem starts with the overlap of simple waves. The flow then ceases to be simple and there is an increase of entropy across the compression wave. As  $P$  and  $dP/dt$  at the near boundary increase, shock compression conditions, described by the Rankine-Hugoniot relations, may ultimately be reached at the front of the disturbance. Since the most important part of the problem is expected to be the region of shock formation, i.e. region intermediate between simple flow and shock conditions, a parameter  $\zeta$ , which measures the extent of shock nature is hereby defined such that

$$0 < \zeta = \frac{E - E_I}{E_H - E_I} < 1 \quad (2)$$

Here  $E$ ,  $E_I$  and  $E_H$  are actual (computed), isentropic and shock (Hugoniot) compression energies respectively, corresponding to a given pressure. (Since simple flow is isentropic ( $\Delta S=0$ ), an alternative parameter,  $0 < \zeta' = \frac{\Delta S}{S_H} < 1$ , could be defined to measure the extent of shock nature). The two parts of the hydrodynamic problem are thus characterized by  $\zeta = 0$  and  $\zeta > 0$  respectively; the upper limit of the parameter,  $\zeta = 1$ , corresponds to a full grown shock.

While in a condensed medium the difference, for a given pressure, in energy (and consequently in temperature) between the two modes of compression characterized by the extreme values of  $\zeta$  will not be large, the difference in chemical reaction rates should be quite considerable and may mean a difference between failure and initiation of detonation.

Hence it is convenient that, in addition to pressure, another parameter specifying the energy be known.  $\zeta$  has been chosen because it gives a direct indication of deviation from simple flow conditions.

#### B. Equation of State

A generalized Tait equation of state has been chosen to represent the solid explosive

$$(P + B) V - (P_0 + B) V_0 = (\gamma - 1) (E - E_0) \quad (3)$$

with appropriate values of the constants B, and  $\gamma$ , the equation gives a remarkably realistic representation of the compression of solid explosives over a wide range of pressures\*. Combined with the isentropic condition,

$$dE = -PdV, \quad (4)$$

the equation reduces to the form used in Ref. 6 (which does not include the energy):

$$P = \frac{B}{\gamma} \left( \left( \frac{V_0}{V} \right)^\gamma - 1 \right) \quad (5)$$

Explicit equations relating the various properties for isentropic compression and for shock compression on the basis of Eqn. 5 are collected in Table I.

The arbitrary parameters chosen in Ref. 6 were  $B = 105$  kbar,  $\gamma = 3$ . The choice deserves a comment.

If Eqn. (3) is to be fitted to a set of data in a certain range of pressures, the constants B and  $\gamma$  can, in general, be assigned any convenient values. If, however, the lower limit of the range is  $P = 0$ , by virtue of the relation

$$c = V \sqrt{- \left( \frac{\partial P}{\partial V} \right)_s} = \sqrt{V(\gamma P + B)}, \quad (6)$$

\*The author is indebted to Dr. S.J. Jacobs for having pointed out the promising possibilities of this extremely simple equation.

TABLE I  
 SUMMARY OF ISENTROPIC AND HUGONIOT FORMS OF THE EQUATION OF STATE  
 USED IN THIS REPORT

The equation of state in its ordinary form is

$$(P + B) V - (P_0 + B) V_0 = (\gamma - 1) (E - E_0).$$

Isentropic

$$V = V_0 \left( \frac{\gamma P_0 + B}{\gamma P + B} \right)^{\frac{1}{\gamma}}$$

Hugoniot

$$V = V_0 - \frac{2V_0(P - P_0)}{(\gamma+1)P + 2B + (\gamma-1)P_0}$$

$$E - E_0 = \frac{V_0}{\gamma-1} \left[ (P+B) \left( \frac{\gamma P_0 + B}{\gamma P + B} \right)^{\frac{1}{\gamma}} - (P_0+B) \right] \quad E - E_0 = \frac{V_0 (P^2 - P_0^2)}{(\gamma+1)P + (\gamma-1)P_0 + 2B}$$

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the value of B is fixed by

$$B = \frac{c_0^2}{V_0} \quad (6')$$

This is certainly the case in the shock formation problem, where in the early stages of shock growth, the pressure is quite low. The value  $B = 105$  kbar used in Ref. 6 corresponds to an initial sonic velocity  $c_0 = 2.56$  mm/ $\mu$ sec, which is an average of the range of 2.25 - 2.85 mm/ $\mu$ sec found by Majowicz (7) for a series of explosives. Thus the value of this parameter is realistic.

There is no doubt that the value of  $\gamma = 3$ , used in Ref. 6 (and by some earlier workers), is too low, because it gives an unrealistically high compressibility. The reason why the value has been used at all is twofold. First, it is a carry-over from calculations of high pressure gases, such as detonation products, in which the Eqn. 3 with  $B = 0$  and  $\gamma \approx 3$  gives reasonable results. Second, and perhaps more important, the choice of  $\gamma = 3$  lends convenient tractability to hydrodynamic equations. In particular, it allows the boundary path in the shock formation problem to be evaluated in closed form (see Ref. 8); this would be impossible for any value  $\gamma > 3$  (and probably for most non-integral values).

Figure 1 shows a comparison of the computed P - V relation for two different sets of parameters B and  $\gamma$  as well as experimental data of Majowicz and Jacobs (9). The high compressibility of a hypothetical material for which  $\gamma = 3$  is evident. The value of  $\gamma = 4.5$ , on the other hand (combined with  $B = 100$  kbar) is very realistic, and it is the current choice for the machine computations. However, since the analytical treatment exists (Ref. 6) in which the first set of values was used ( $\gamma = 3$ ,  $B = 105$ ), the preliminary computations discussed below, were run with this set of parameters for the sake of comparison.

The sonic velocity, corresponding to  $B = 100$  kbar, is 2.5 mm/ $\mu$ sec, a most reasonable value.

#### NUMERICAL SOLUTION OF HYDRODYNAMIC PROBLEMS (GENERAL)

The general hydrodynamic problem is a solution of the equations of motion, state and energy release subject to

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appropriate boundary conditions. The equations of motion for a one dimensional case are

$$\rho_0 \frac{\partial u}{\partial t} = - \frac{\partial P}{\partial x} \quad (\text{conservation of momentum}) \quad (7)$$

$$\frac{\partial E}{\partial t} - \frac{\partial Q}{\partial t} + P \frac{\partial V}{\partial t} = 0 \quad (\text{conservation of energy}) \quad (8)$$

$$\text{and } \rho_0 \frac{\partial V}{\partial t} = \frac{\partial u}{\partial x} \quad (\text{conservation of mass}), \quad (9)$$

where  $t$  = time

$\rho_0$  = initial density

$u$  = particle velocity

$P$  = pressure

$E$  = specific internal energy

$Q$  = heat added per unit mass (from chemical reaction)

$V$  = specific volume

$X$  = distance

$x$  = Lagrange coordinate defined by the

relation

$$\frac{\partial X(x,t)}{\partial x} = \rho_0 V(x,t).$$

The equation of state is

$$P = \Phi(E,V). \quad (10)$$

The equation of chemical energy release is

$$\frac{\partial Q}{\partial t} = R(Q,E,V). \quad (11)$$

One way to obtain a solution is by numerical techniques. This consists of dividing the Lagrange space coordinate ( $x$ ) into a number of equal zones and approximating the differentials in equations 7, 8, 9 and 11 by finite difference ratios. In the difference equations, the dependent variables are usually specified at the interfaces between the zones or at the centers of the zones.

The boundary conditions must specify the values of the dependent variables for all values of the Lagrangian space coordinate ( $x$ ) at time zero and for the end points ( $x = 0$  and  $x = x_{\max}$ ) at all times. Once the boundary conditions are specified the difference equations can then be solved to obtain the values of the variables at the interior points.

Most differencing schemes have the limitation that they cannot handle discontinuities. The equations of motion (Eqns. 7, 8 and 9) admit discontinuous solutions; in fact, the discontinuities are the most interesting parts of the solutions. Two methods of overcoming this difficulty will be discussed in the next section.

The time increment ( $\Delta t$ ) used cannot be chosen arbitrarily. A stability analysis (Ref. 10) of the problem will yield a maximum value of  $\Delta t$  with which reasonable results can be obtained. Stability analysis is an analysis of the history of an arbitrarily introduced error. Usually a critical value of  $\Delta t$  will be determined such that if  $\Delta t$  were to be made larger than this critical value, the error will increase, if  $\Delta t$  were to be made smaller than this critical value, the error will decrease and if  $\Delta t$  is made equal to this critical value the error will remain constant.

#### TWO SPECIFIC METHODS OF OBTAINING NUMERICAL SOLUTIONS TO HYDRODYNAMIC PROBLEMS

Two methods of handling discontinuities will be discussed in this section. They are the Richtmyer-von Neumann "q" method (Ref. 11) and the Lax method (Ref. 12). In both methods discontinuities are approximated by steep but finite slopes.

The "q" method eliminates discontinuities by the inclusion of an artificial dissipative term. Physically it can be considered as a one-dimensional viscosity. This dissipative term "q" is defined by the equation

$$q = - \frac{(K \Delta x)^2}{V} \cdot \frac{\partial u}{\partial x} \cdot \left| \frac{\partial u}{\partial x} \right|, \quad (12)$$

where  $K \Delta x$  = arbitrary constant.\*  $K \Delta x$  is approximately 1/3 the distance over which the shock is spread.

The term "q" is added to the pressure (P) and Eqns. (7) and (8) became respectively,

$$\rho_0 \frac{\partial u}{\partial t} = - \frac{\partial}{\partial x} (P + q) \text{ and} \quad (13)$$

$$\frac{\partial E}{\partial t} - \frac{\partial Q}{\partial t} + (P + q) \frac{\partial V}{\partial t} = 0. \quad (14)$$

Equation (9), which does not contain pressure, is unchanged.

The resultant set of equations (Eqns. (9), (10), (11), (13) and (14) do not have discontinuous solutions. The solutions of the modified equations and the original equations are very nearly the same except in regions where the solution of the original equations would have a discontinuity. Equations (9), (10), (11), (13) and (14) approximate the discontinuity by a smooth but steep curve.

The Hugoniot relation across a shock,

$$E_f - E_1 = \frac{1}{2} (P_f + P_1) (V_1 - V_f), \quad (15)$$

is not affected by the inclusion of q.

Thus the "q" method is successful in that it eliminates discontinuities and gives a good approximation to the true solution in every aspect except details of the shock.

A computer (IBM 704) program which utilizes the "q" method and a second order differencing scheme was constructed at the Naval Ordnance Laboratory by W. Walker. Some results from this code will be discussed in the next section.

\* Considering the arbitrary constant as the product of K and  $\Delta x$  is superfluous at this stage of the discussion. However, when the differential equations are replaced by finite difference equations the  $\Delta x$  mentioned above and the  $\Delta x$  used as the increment of the independent variable are identical. K is a dimensionless constant usually near unity.

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Another method for handling the problem of discontinuities was devised by Lax (Ref. 12). While the "q" method involves a quasi-physical concept and a modification of the equations of motion, the Lax method does neither. Rather, it handles discontinuities by the nature of its unusual differencing scheme. The Lax scheme requires that all of the differential equations be in perfect differential form, i.e.

$$A \frac{\partial Y}{\partial t} = \frac{\partial Z}{\partial x} \quad , \quad (16)$$

where A is a constant. This differential equation is then differenced in the following way

$$\begin{aligned} \frac{A}{\Delta t} \left( Y_x^{t+\Delta t} - \frac{1}{2} (Y_{x+\Delta x}^t + Y_{x-\Delta x}^t) \right) \\ = \frac{1}{2\Delta x} (Z_{x+\Delta x}^t - Z_{x-\Delta x}^t) \end{aligned} \quad (17)$$

In so-called normal regions where  $\frac{\partial^2 Y}{\partial x^2}$  is small,

$$\frac{1}{2} (Y_{x+\Delta x}^t + Y_{x-\Delta x}^t) \approx Y_x^t \quad . \quad (18)$$

In this case the Lax difference scheme approaches an ordinary forward difference scheme. Therefore, in normal regions, the solution obtained by the Lax scheme approaches the analytic solution.

In regions where  $\frac{\partial^2 Y}{\partial x^2}$  is high (at shocks) Eqn. (18) is not valid and the Lax scheme comes into effect. It causes any discontinuities (or other extreme changes) to be replaced by a steep but smooth change.

As mentioned earlier, the equations of motion must be in perfect differential form if the Lax scheme is to be used. The equations of conservation of mass (Eqn. (9)) and momentum (Eqn. (7)) and the equation of chemical energy release (Eqn. (11)) are already in perfect differential form. A fourth, independent, perfect differential equation must be constructed.

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This can be done by multiplying Eqn. (7) by  $u$ , Eqn. (8) by  $\rho_0$  and Eqn. (9) by  $-P$  and adding the results

$$\begin{aligned} \rho_0 \left( u \frac{\partial u}{\partial t} + \frac{\partial E}{\partial t} - \frac{\partial Q}{\partial t} + P \frac{\partial V}{\partial t} - P \frac{\partial V}{\partial t} \right) \\ = -u \frac{\partial P}{\partial x} - P \frac{\partial u}{\partial x} \end{aligned} \quad (19)$$

Simplifying;

$$\rho_0 \frac{\partial}{\partial t} \left( \frac{1}{2} u^2 + E - Q \right) = - \frac{\partial}{\partial x} (Pu) \quad (20)$$

One of the results of this task is a computer (IBM 704) program to solve hydrodynamic problems by the Lax method. Appendix I gives a description of this program.

Comparison of Analytic, "q" and Lax Methods

The general hydrodynamic problem solved numerically by the "q" and Lax methods as described above, will now be compared to the previously obtained analytic solution (6). The same equation of state and boundary conditions were used in all three calculations. Equation (3) was used as the equation of state. It was assumed that no reaction took place so  $Q(x,t)$  was set equal to zero.

The following boundary conditions were used in all three calculations

$$\begin{aligned} P(0,t) &= P(0,0) e^{kt} && \text{for } t \leq 60 \mu\text{sec;} \\ k &= .1 \mu\text{sec}^{-1} \\ P(0,t) &= P(0,60) && \text{for } t \geq 60 \mu\text{sec;} \\ P(x_{\text{max}},t) &= P(0,0) \\ P(x,0) &= P(0,0) \\ u(x,0) &= 0 \\ P(0,0) &= .08 \text{ kbars} \end{aligned}$$

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$V(x,0) = .6245$  cc/gm and  $E(x,0) = 2 \times 10^4$  ergs/gm were the values calculated for an adiabatic compression from .001 to .08 kbars. These boundary conditions approximate the boundary conditions realized in the experimental work.

Figure 2 compares the P - X plots at 60  $\mu$ sec obtained from the analytic, "q" and Lax methods. Except for fluctuations in the plateau, the "q" method agrees more closely with the analytic method than does the Lax method.

Figure 3 compares P - X plots at 100  $\mu$ sec obtained from the "q" and Lax methods. At this time a real solution would have a discontinuity extending slightly below the plateau. The "q" method gives a somewhat closer approximation to this discontinuity than does the Lax method. However, the "q" method gives severe fluctuations in the plateau, while the Lax method gives none.

Because of the exponential dependence of reaction rates on temperature (i.e. energy), the spurious fluctuations inherent in the "q" method render the "q" method almost useless for reaction rate studies. Therefore all further numerical work discussed in this report is based on the Lax scheme.

#### SHOCK FORMATION AND INITIATION

##### A. Shock Formation

The analytic solution (Ref. 6) to this problem showed that a shock had started to form at about 12 cm from the boundary at 90  $\mu$ sec. The analytic method cannot give the rate of shock growth.

The rate of growth of the shock is illustrated in Fig. 4 which gives energy-distance (or temperature-distance) profiles at several different times as computed numerically by the Lax method using the previously defined boundary conditions and equation of state. The generating pressure pulse was allowed to increase exponentially for 60  $\mu$ sec so that the maximum pressure reached was  $P_{\max} = 32.27$  kbars; thereafter the boundary pressure remained at 32.27 kbars. In Fig. 4, the upper horizontal line gives the energy that would result from a shock compression to 32.27 kbars; the lower line gives the energy that would result from an isentropic compression to

32.27 kbars. The actually computed energy has increased perceptibly above the limiting isentropic value at 72.1  $\mu$ sec, at which time the compression front is about 5.3 cm from the boundary. However, the transition from the isentropic compression to the shock compression is continuous; there is no sharp point of shock formation.

The growth of the shock is also shown in Fig. 5, in which the parameter  $\zeta$  (evaluated at the compression front) is plotted against time. The figure also gives the location of the compression front as a function of time, so that the extent of the shock nature ( $\zeta$ ) in the front can be read both as a function of time and distance.

#### B. Snock Initiation

Figures 4 and 5 show that, assuming a chemically inert medium, the shock wave is half developed ( $\zeta = .5$ ) when the compression wave has travelled about 16 cm into the charge. The next step was to see whether the temperatures generated were sufficient to start a detonation in an actual (i.e. chemically reactive) explosive, and where the detonation would start. The latter point is of particular interest because, experimentally in the NOL-DDT test (Ref. 6), the detonation starts about 15 cm. into the charge.

The computed point of initiation, in general, could be located anywhere between the boundary and the wave front. As seen in Fig. 4, the layer of explosive near the compression front is at a temperature higher than that of the boundary, but its residence time at that temperature is shorter. The point at which the chemical reaction rate becomes sufficiently high to generate a detonation wave will evidently depend on the specific parameters used in the computation.

In order to see if the theoretical model agrees with the experiments, a simple first order kinetic model was used, i.e.

$$\frac{\partial F(x,t)}{\partial t} = (1 - F(x,t)) A e^{-\frac{E_a}{RT(x,t)}} \quad (21)$$

where,

$F(x,t)$  = mass fraction of burnt explosive,

$A$  = preexponential factor,

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$E_a$  = activation energy,

$R$  = gas constant,

and  $T(x,t)$  = temperature.

$T(x,t)$  is defined by the equation

$$T(x,t) = T_0 + \frac{E(x,t)}{C_v} \quad (22)$$

Since  $Q(x,t)$  is the energy liberated by the chemical reaction at a point  $(x,t)$  and  $H$  is the heat of explosion of the explosive, then

$$F(x,t) = \frac{Q(x,t)}{\Delta H} \quad (23)$$

Therefore, equation (21) can be rewritten as

$$\frac{\partial Q(x,t)}{\partial t} = \Delta H \left( 1 - \frac{Q(x,t)}{\Delta H} \right) A e^{-\frac{E_a}{RT(x,t)}} \quad (24)$$

This is the explicit form of equation (11) that was used in the following calculations. The constants in equations (22) and (24) are

$$C_v = 1.254 \times 10^7 \text{ ergs/gm}^\circ\text{C}$$

$$\Delta H = 5.016 \times 10^{10} \text{ ergs/gm}$$

$$A = 10^{14} \text{ sec}^{-1}$$

$$E_a = 35,000 \text{ cal/mole.}$$

A computer run was made using the previously discussed boundary conditions and equation (24) to compute the reaction rate. The maximum pressure was 32.27 kbars. The computed temperatures were too low to cause any appreciable reaction. The run gave results almost identical to the run represented in Fig. 4.

Figure 6 illustrates the most important result of this run. It is a plot of  $F(x,t)$  vs  $x$  at several different times. It shows that after the shock is partly developed, the greater reaction rate in the interior (due to the greater temperature

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increase from the partly developed shock) causes the reaction to proceed farther than it does at the boundary.

A subsequent run was made with one important change. The near boundary pressure was,

$$P(0,t) = .08 e^{kt} \text{ kbars for } t \leq 67.5 \text{ } \mu\text{sec}$$

$$k = .1 \text{ } \mu\text{sec}^{-1}$$

$$P(0,t) = .08 e^{6.75} \text{ kbars} = 68.32 \text{ kbars for } t \geq 67.5 \text{ } \mu\text{sec}$$

The higher pressure caused the temperature (i.e. energy) to reach higher values than in the previous run. The reaction rates from these higher temperatures were great enough to cause the reaction to go to completion.\* The reaction first went to completion\* 17.7 cm in from the boundary. Figure 7 illustrates the course of the reaction. It is a plot of  $P(x,t)$  vs  $x$  at several different times.

Figure 8 is a plot of  $T(x,t)$  vs  $x$  at two different times from two different computer runs. In one run the material was assumed to be non reactive; in the other run the material was assumed to be reactive. The times chosen were slightly before and slightly after the reaction went to completion in the reactive run. The interior temperature is higher than the boundary temperature; this coupled with the exponential dependence of reaction rate on temperature caused the reaction to go to completion in the interior before it went to completion at the boundary.

Figure 9 is a plot of pressure vs  $x$  at several different times for the 68.32 kbar maximum pressure, reactive explosive calculation. It shows the development of the detonation wave.

\* The first order reaction assumed here would never actually go to completion, but in a numerical computation the reaction goes to completion. The error is completely negligible.

#### DISCUSSION

Measurements and rough calculations (Ref. 6) indicate that the maximum boundary pressure attained in the NOL-DDT test is about 32 kbars. Calculations based on this maximum pressure and homogeneous first order kinetics show no appreciable reaction. This is not surprising since it is almost certain that initiation by weak stimuli (e.g. weak shocks) requires some mechanism of stress concentration (e.g. occluded grit or gas bubbles). The important result from the 32 kbar calculation was the observation that the reaction inside the charge surpassed the reaction at the boundary.

A later calculation was made based on a maximum boundary pressure of 68 kbars. This was done to compensate for the absence of stress concentrating mechanisms in the model used. This pressure was adequate to cause initiation of the explosive. The initiation started in the interior at a location comparable to the experimental results.

It was concluded from these calculations that the shock formation-shock initiation model for the transition from deflagration to detonation is essentially correct.

#### ACKNOWLEDGEMENT

The author wishes to acknowledge the advice and suggestions of Dr. A. Macek, who suggested this problem and supervised the work.

APPENDIX I

As a part of this task a computer (IBM 704) program was constructed to solve hydrodynamic problems on the basis of the Lax scheme. The program consists of a main routine in which the equations of motion are integrated and certain other unchanging operations are performed. Calculations involving the equation of state reaction rates, boundary conditions and stability are carried out in subroutines. Thus if any of these things must be changed, only the appropriate subroutine need be reprogrammed.

The program runs according to the flow diagram in Figure 10. The following are notes to Figure 10:

- (1) The stability analysis of this system shows that

$$\Delta t < \rho_0 \Delta x \frac{V}{C} .$$

$t$  is computed at every interface and the smallest value is used.

- (2) At this step the equations of conservation of mass and momentum are integrated. The equation of conservation of mass (Eqn. 9), when differenced according to the Lax scheme, becomes

$$\frac{\rho_0}{\Delta t} \left( v_x^{t+\Delta t} - \frac{1}{2} (v_{x+\Delta x}^t + v_{x-\Delta x}^t) \right) = \frac{1}{2\Delta x} (U_{x+\Delta x}^t - U_{x-\Delta x}^t).$$

Since the values of all of the variables are known at  $t$ , this equation can be used to evaluate  $v_x^{t+\Delta t}$ . Likewise, the equation of conservation of momentum (Eqn. 7<sup>x</sup>) when differenced according to the Lax scheme becomes,

$$\frac{\rho_0}{\Delta t} \left( U_x^{t+\Delta t} - \frac{1}{2} (U_{x+\Delta x}^t + U_{x-\Delta x}^t) \right) = \frac{1}{2\Delta x} (P_{x+\Delta x}^t - P_{x-\Delta x}^t).$$

Since the values of all of the variables are known at  $t$ , this equation can be used to evaluate  $U_x^{t+\Delta t}$ .

Since  $0 < x < x_{\max}$ , the above two equations cannot be used to evaluate the variables at  $t+\Delta t$  where  $x = 0$  or  $x_{\max}$  because this would demand values of the variables at  $x$  outside the range  $0 < x < x_{\max}$ .

(3) At this step the equations of chemical energy release and conservation of energy are integrated. The equation of chemical energy release, when differenced according to the Lax scheme, becomes

$$\frac{1}{\Delta t} \left( Q_x^{t+\Delta t} - \frac{1}{2} (Q_{x+\Delta x}^t + Q_{x-\Delta x}^t) \right) = R_x^t .$$

Since the right hand side of this equation can be evaluated directly, it will not be differenced or averaged.  $R_x^t$  are evaluated in the reaction rate subroutine. Therefore, this equation can be used to evaluate  $Q_x^{t+\Delta t}$ .

(4) The values of the variables at the boundaries (i.e. at  $x = 0$  and  $x = x_{\max}$ ) are computed in the boundary value subroutine. The values of the pressure  $P$  are specified by the equations,

$$\begin{aligned} P(0,t) &= .08 e^{0.1t} \text{ kbar for } t \leq t_{co} \\ P(0,t) &= .08 e^{0.1t_{co}} \text{ kbar for } t \geq t_{co} \\ P(x_{\max},t) &= .08 \text{ kbar for all values of } t \end{aligned}$$

The values of the other variables at the boundaries are computed from  $P(0,t)$ ,  $P(x_{\max},t)$  and equations 7, 9, 11 and 19. The Lax differencing scheme cannot be used at the boundaries because it would require values of the variables outside the range of  $0 < x < x_{\max}$ . Therefore a different differencing scheme is used. For a partial differential equation of the following general type,

$$A \left( \frac{\partial Y}{\partial t} \right) = \left( \frac{\partial Z}{\partial x} \right),$$

the following differencing scheme is used;

$$\begin{aligned} \frac{A}{2\Delta t} \left( Y_x^{t+\Delta t} - Y_x^t + Y_{x+\Delta t}^{t+\Delta t} - Y_{x+\Delta t}^t \right) \\ = \frac{1}{2\Delta x} \left( Z_{x+\Delta x}^t - Z_x^t + Z_{x+\Delta x}^{t+\Delta t} - Z_x^{t+\Delta t} \right) . \end{aligned}$$

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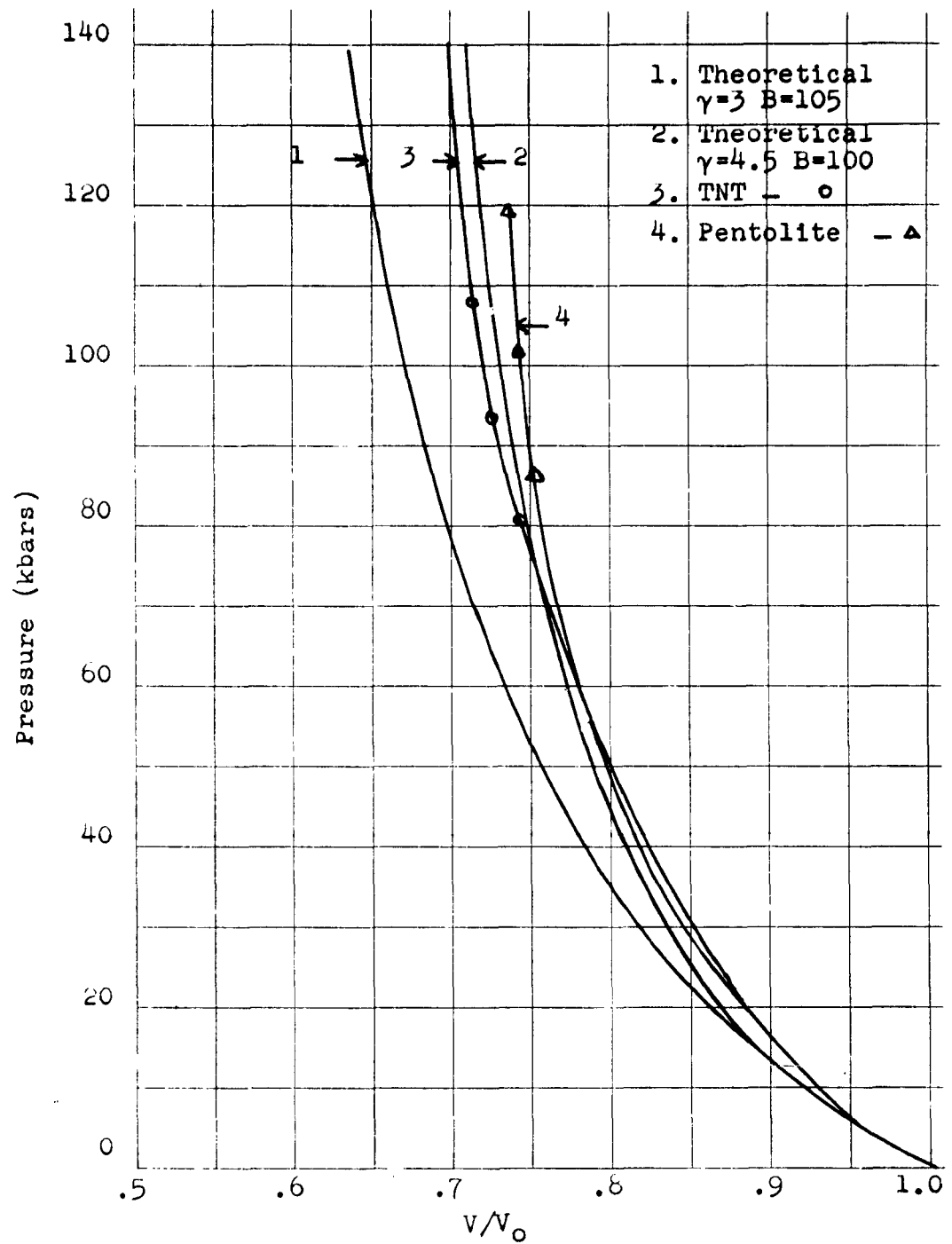


FIGURE 1 - Experimental and Theoretical Hugoniot Curves for Unreactive Explosives

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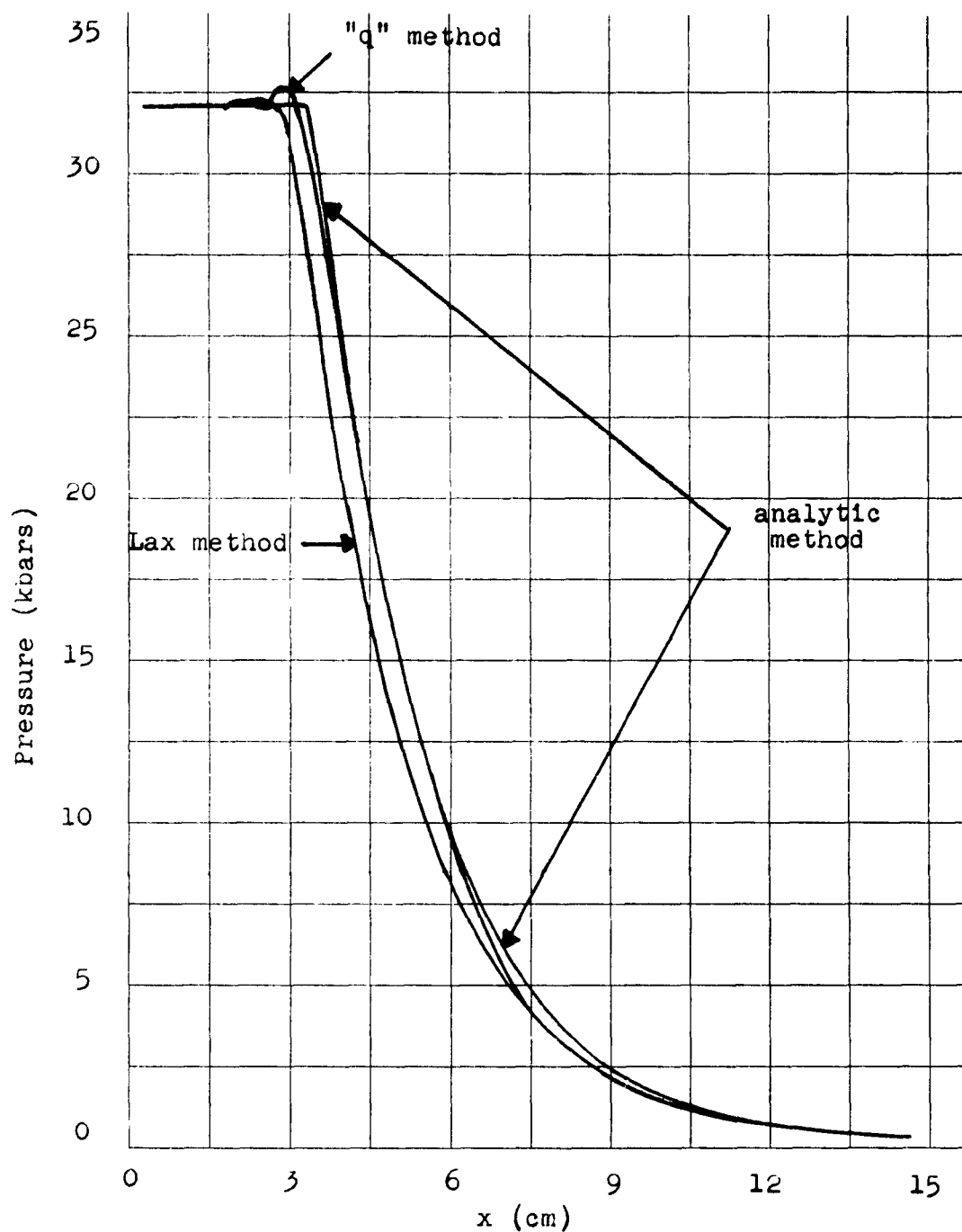


FIGURE 2  
Pressure-Distance Profiles as Computed by the  
Lax, "q" and Analytic Methods  
The time of the computation is 66.1  $\mu$ sec after  
the first application of pressure.

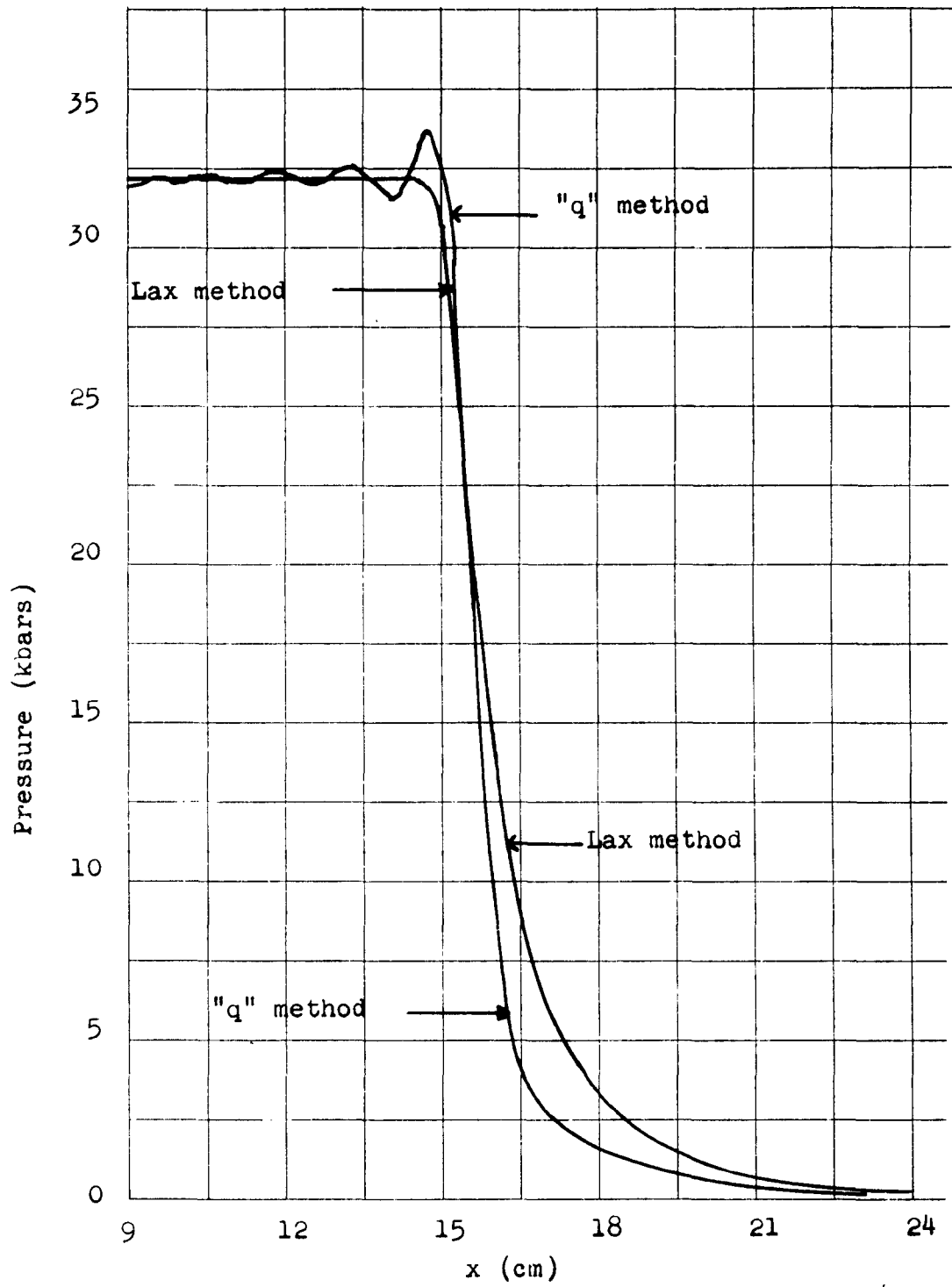


FIGURE 3  
Pressure-Distance Profiles as Computed by the  
Lax and "q" Methods

The time of the computation is 100  $\mu$ sec after  
the first application of pressure.

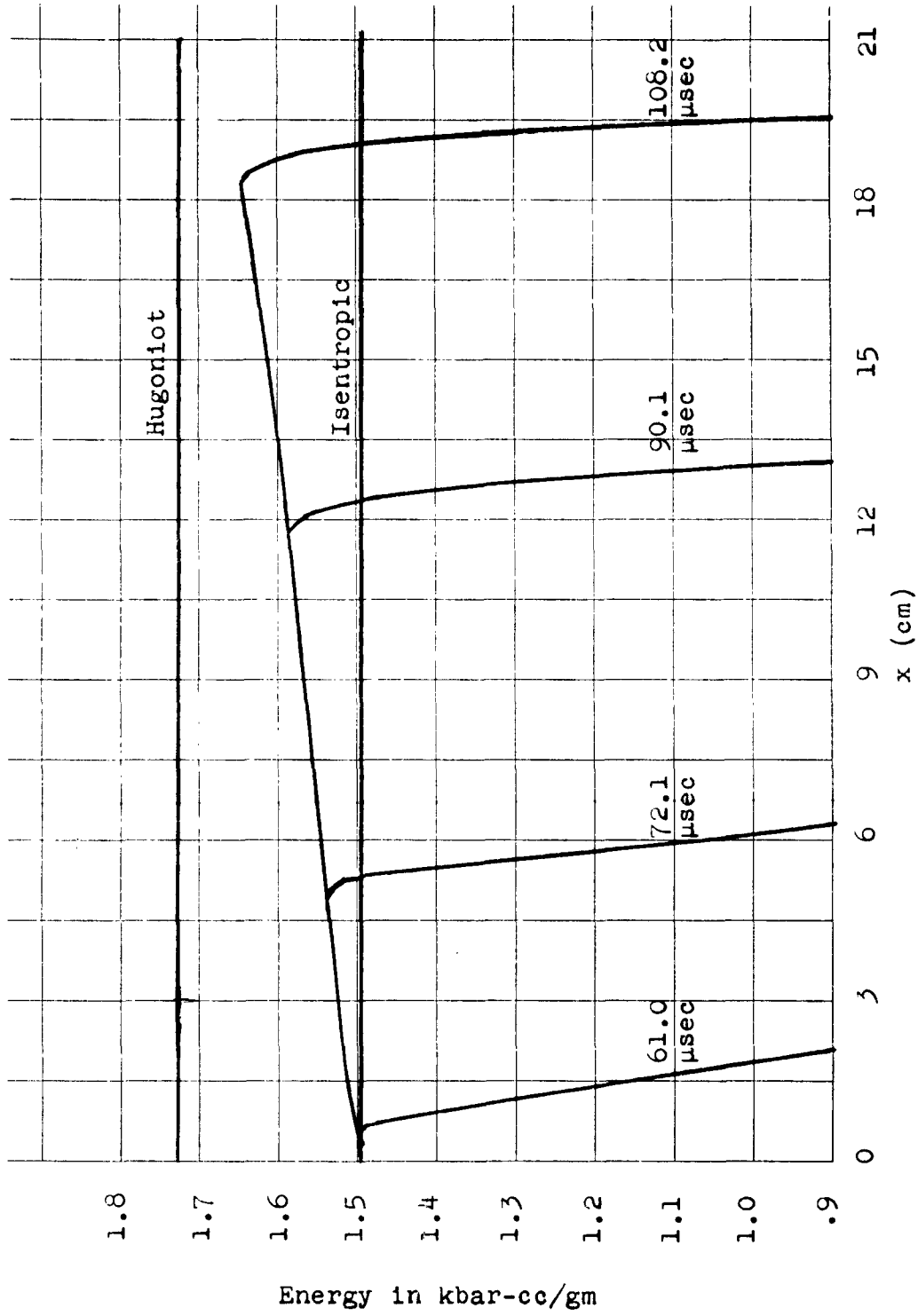
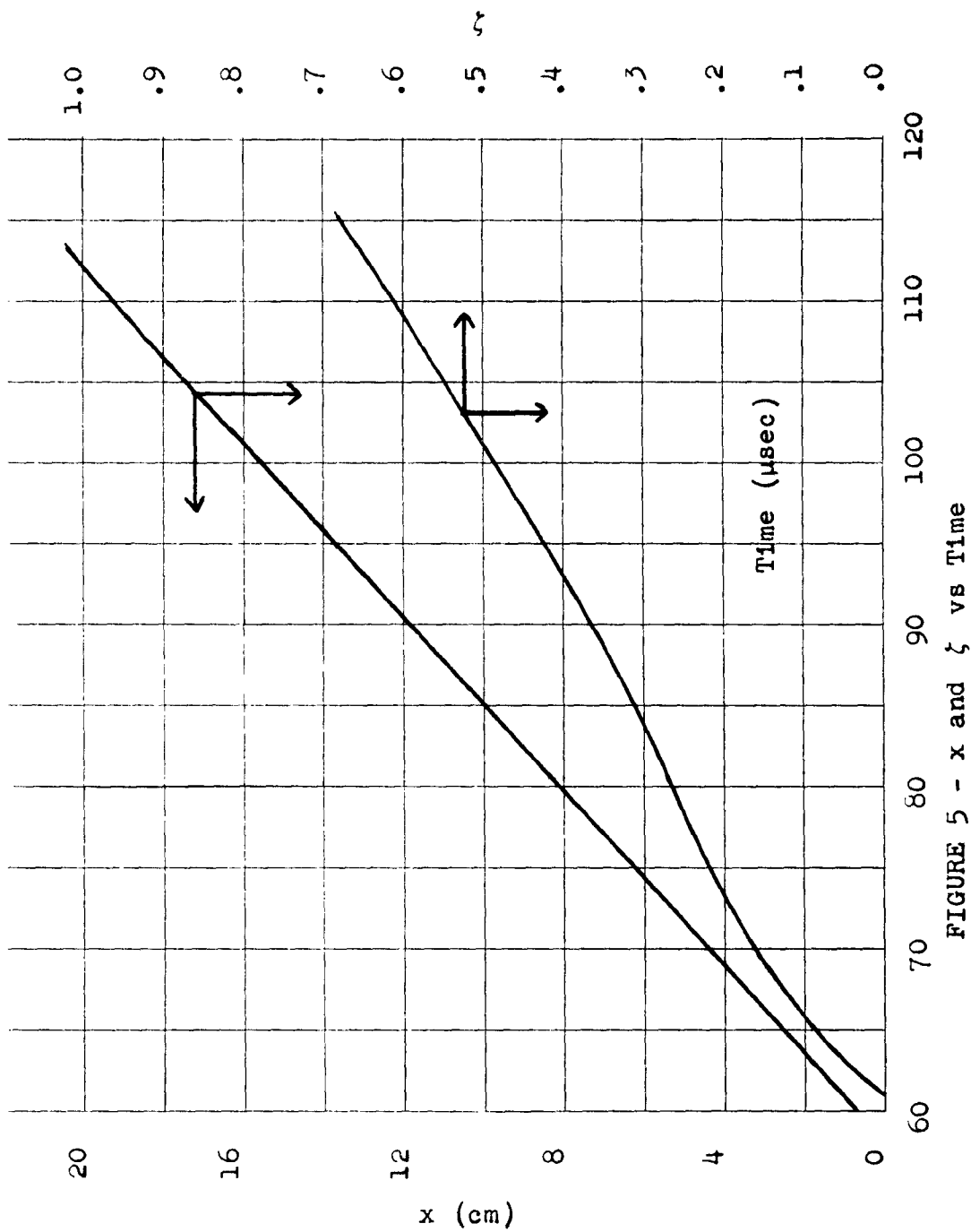


FIGURE 4 - Energy vs Distance at Several Different Times in a Non-Reactive Material



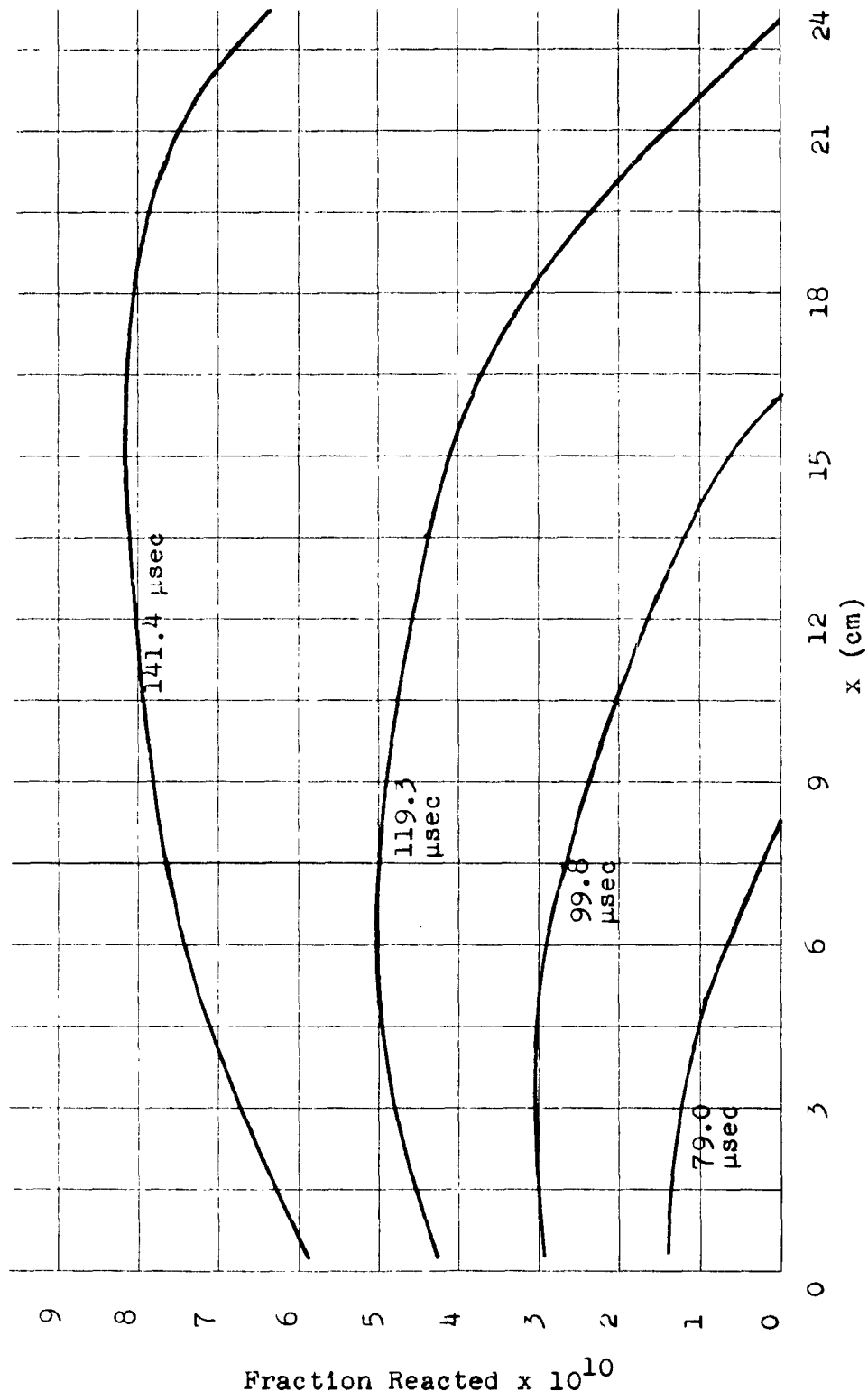


FIGURE 6 - Fraction Reacted vs Distance at Several Different Times

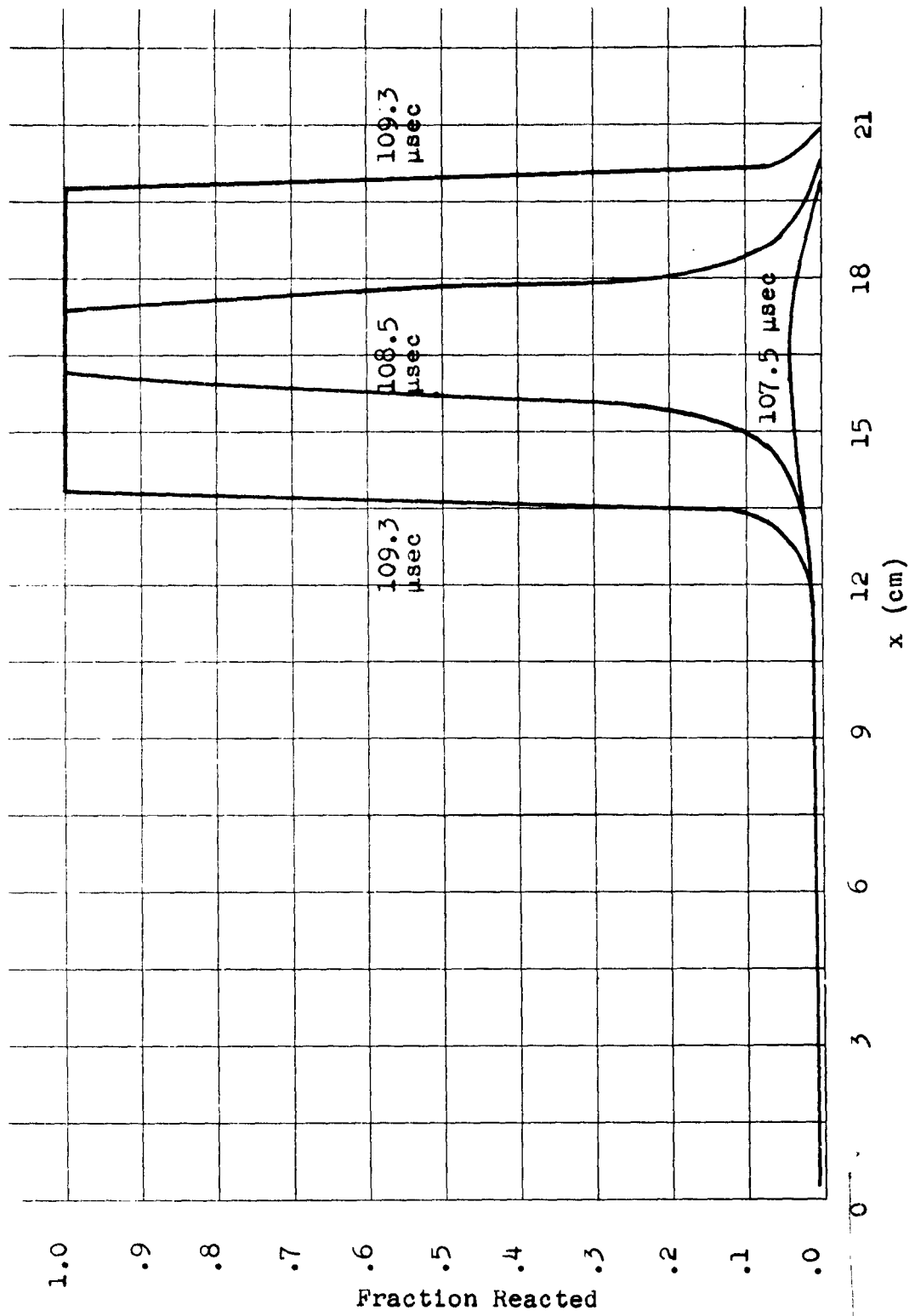


FIGURE 7 - Fraction Reacted vs Distance at Several Different Times

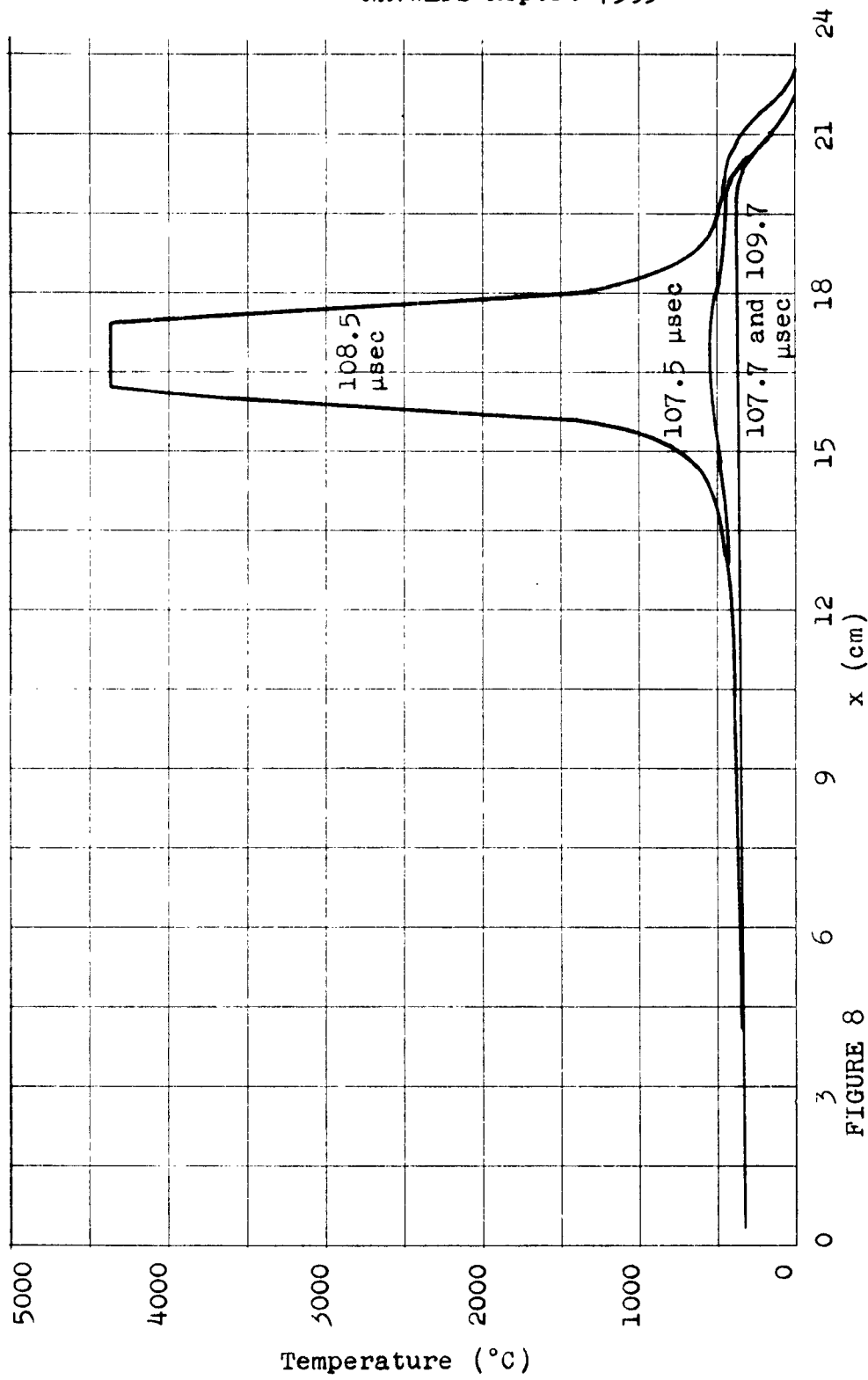


FIGURE 8  
Temperature vs Distance at Several Different Times with Reactive and Non-Reactive  
Materials

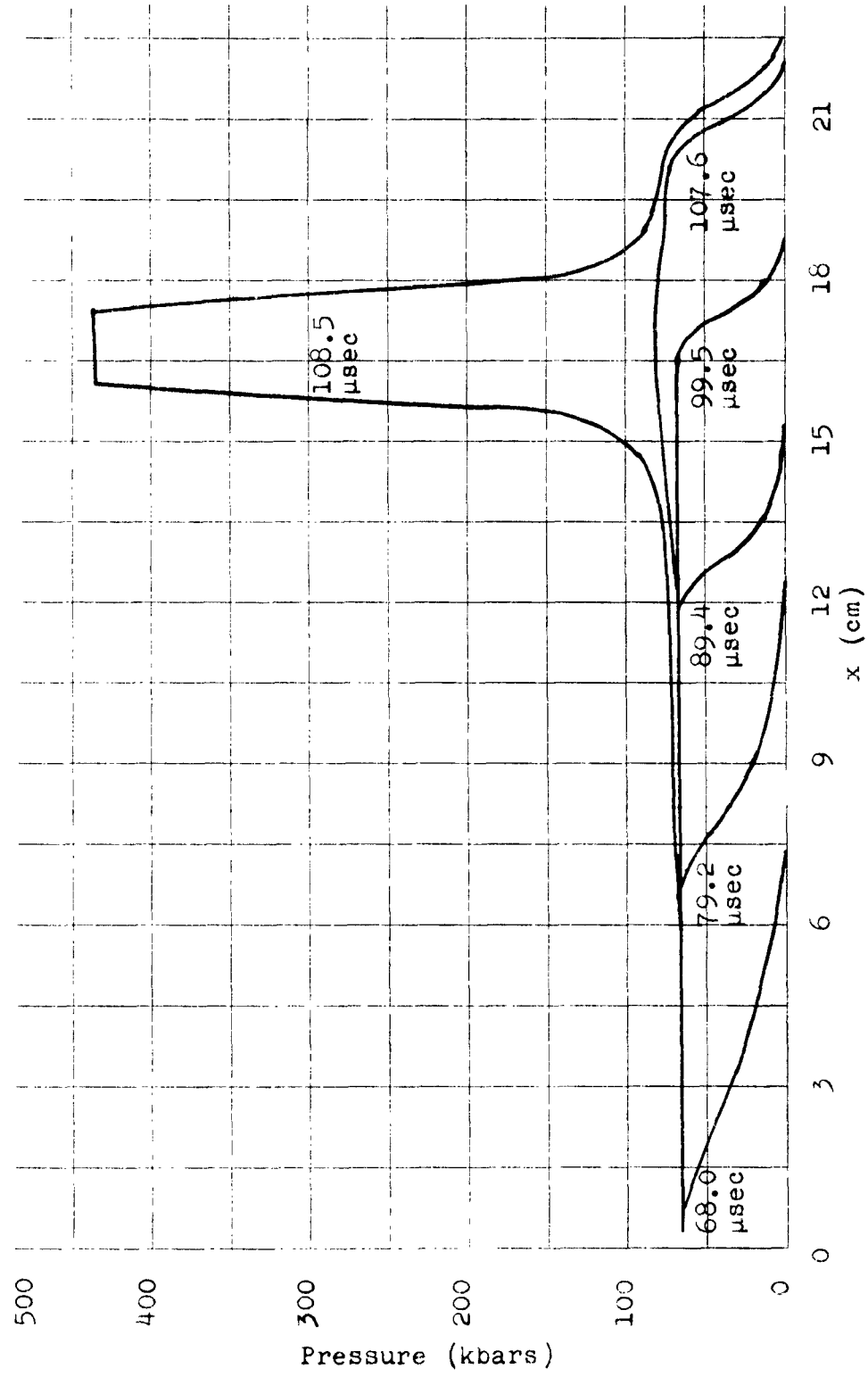


FIGURE 9 - Pressure vs Distance at Several Different Times with a Reactive Material

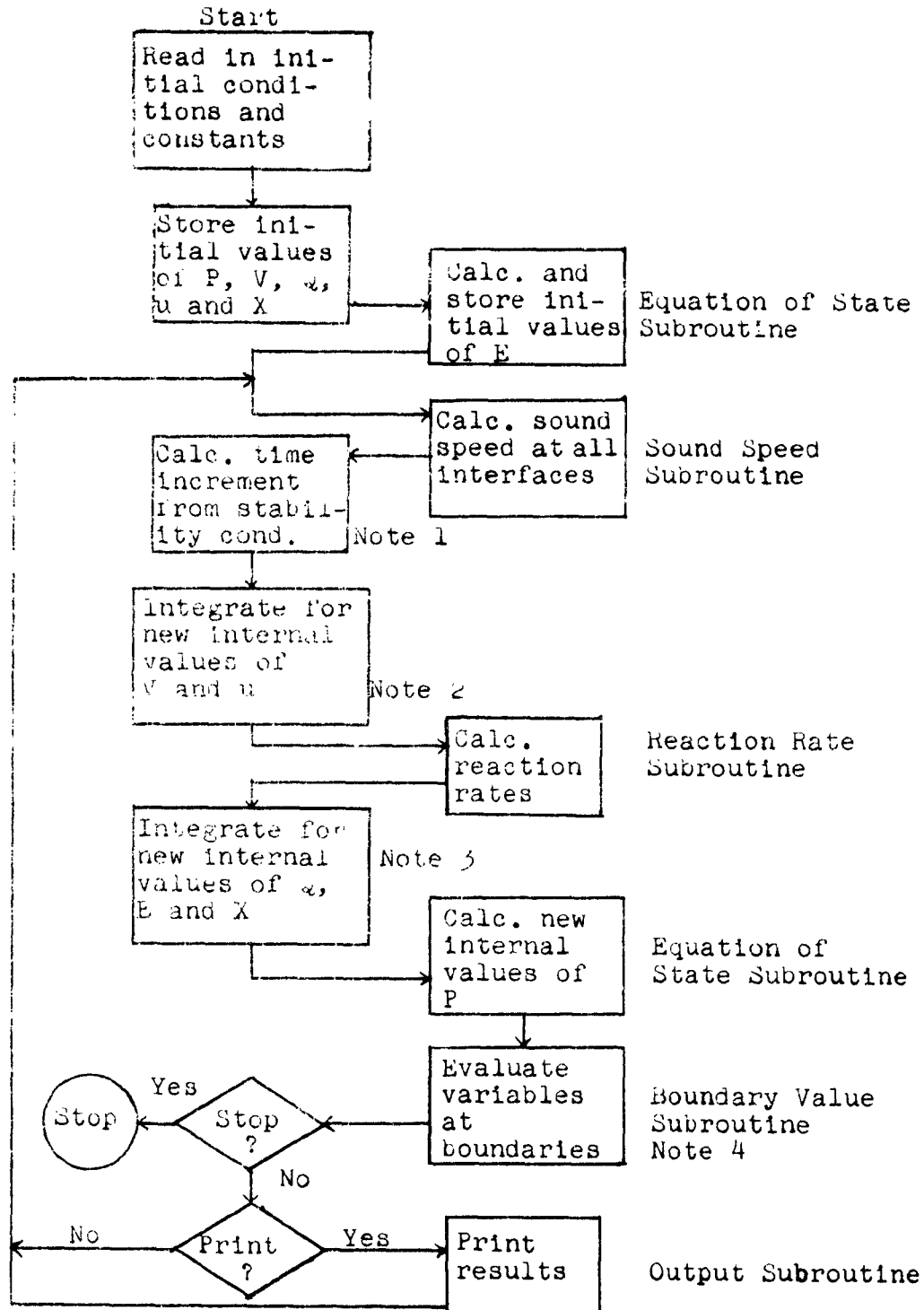


FIGURE 10. Flow Chart of Computer Program

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