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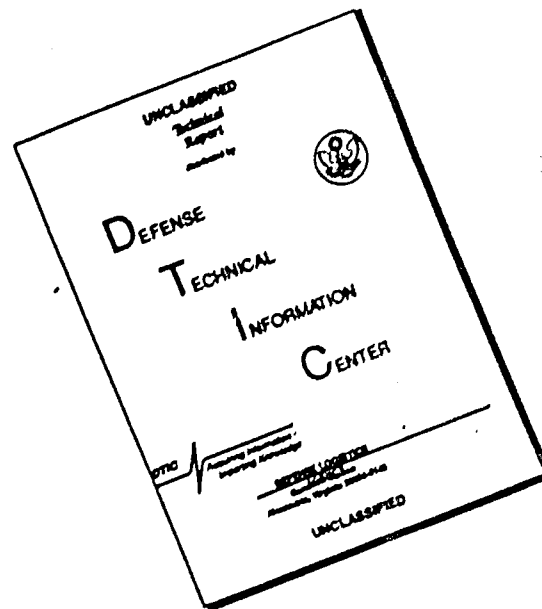
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PICATINNY ARSENAL TECHNICAL REPORT 3167

TIME TO EXPLOSION FOR AN
EXPLOSIVE SUBJECTED EXTERNALLY
TO ELEVATED TEMPERATURES

FRED P. STEIN

MARCH 1965

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PICATINNY ARSENAL
DOVER, NEW JERSEY

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TIME TO EXPLOSION FOR AN EXPLOSIVE SUBJECTED
EXTERNALLY TO ELEVATED TEMPERATURES

by

FRED P. STEIN

March 1965

Feltman Research Laboratories
Picatinny Arsenal
Dover, N. J.

Technical Report 3167

Approved:

H. J. MATSUGUMA
Chief, Explosives
Laboratory

PREFACE

This report is written primarily for the record of the Explosives Research Section prior to the author's separation from the Army. It is a companion report to one entitled "Steady-State Conditions In An Explosive Which Is Subjected Externally to Elevated Temperatures." This report contains a longer-than-usual abstract, which summarizes the author's findings and feelings on the time-to-explosion problem, followed by a somewhat detailed description of the same matter. The author recommends the abstract to all readers, and the body of the report to those inspired by the abstract and to anyone who might take up this subject. For anyone beginning such a study or continuing this one, this report could save months of time.

The report concerns lead azide throughout; however, the calculations and observations are general and may be applied to other explosives.

A large share of the detailed computational work which forms the background for this report was performed by Peter McIntyre, Co-operative Student, University of Detroit. Much credit is due him for his diligence and perseverance.

Fred P. Stein

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ABSTRACT

The most important knowledge required in the calculation of time to explosion for an explosive subjected to an elevated temperature is the kinetics of thermal decomposition of the explosive. This study of models used in computing times to explosion showed that activation energy, frequency factor, type of kinetic expression, and their interrelationships transcend in importance all other physical and chemical properties of the explosive.

Two mathematical models which describe heat conduction in an explosive accompanied by simultaneous internal generation of heat from thermal decomposition of the explosive were studied. The boundary and initial conditions for one model were such that an infinite slab of explosive of finite thickness, initially at a constant temperature, was suddenly exposed to a constant temperature on its lower surface while its upper surface was perfectly insulated. Zinn and Mader's correlation of the solutions to this problem (Ref 8) was used in the study of this model. The other model was that of a semi-infinite solid initially at a constant temperature which suddenly has its surface exposed to an elevated constant temperature. G. B. Cook's correlation of the solutions to this model (Ref 2) was studied. Both models used zero-order kinetics to describe predetonation decomposition.

The effect of each physical and chemical property considered in the correlations to have a bearing on the computed time to explosion at a given temperature was studied. The results, in capsule form, are:

1. Heat of explosion and frequency factor. These two properties always appear in the same position in the correlations so that each has exactly the same effect. As might be expected intuitively, an increase in the heat-of-explosion value (or frequency factor) results in a decrease in the computed time to explosion. In the Cook correlation, the correspondence is exactly one to one. That is to say, doubling the heat-of-explosion value (or frequency factor) halves the time to explosion. In the Zinn and Mader correlation the result is essentially the same, although not precisely so.

2. Density and thermal conductivity. These two properties do not appear in the Cook correlation; however, in the Zinn and Mader correlation they always occupy reciprocal positions in relation to one another, so that, for example, doubling density is the same as halving thermal conductivity. At high environmental temperatures, such as $360°C$ for lead azide or $260°C$ for RDX, these variables have almost no effect on the computed time to explosion even for order-of-magnitude changes. However, at temperatures near the computed minimum temperature of explosion, the effects become large. Here, an increase in the density value (which corresponds to a decrease in thermal conductivity) increases the computed time to explosion.

3. Activation energy and frequency factor. It is realized that, in the zero-order kinetic equations which are used in the differential equation, changes in the activation energy value will tend to be compensated by simultaneous changes in the frequency factor value if (and only if) the kinetic parameters are derived from adequate thermal decomposition data. Since values for activation energy and frequency factor are so often chosen from independent sources, both properties were studied independently, as well as together. It was found that computed times to explosion are extremely sensitive to activation energy. A 2% increase in the activation energy value results in a doubling of the computed time to explosion. It was learned that, if one increases the activation energy value for lead azide by 10% and then takes into consideration the compensating value of the frequency factor, the computed time to explosion is increased by a factor of two in spite of the compensating effect.

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4. Initial temperature of explosive. This effect is not strong. For example, a 20 C increase in the initial temperature would reduce the computed time to explosion by about 10%.

Comparisons were made between computed times to explosion for lead azide and those observed experimentally in an apparatus which was described, hopefully, by the mathematical models. The times to explosion computed from both the Zinn and Mader and the Cook correlations were unsatisfactory. Both gave times which were too short. Although the Zinn and Mader results were slightly better, the difference was not of great significance. When plotted in the familiar form of logarithm of time-to-explosion versus reciprocal temperature, the slopes of the computed lines were much less than those of the experimental data.

In view of the importance of the kinetic parameters, the values for activation energy and frequency factor required to force the computed line through the lead azide data for detonations in 0.4-micron air pressures were determined. An activation energy of 69,750 cal per gram mole and a frequency of $10^{21.74}$ sec^{-1} were found to permit a perfect fit of the aforementioned experimental data. These values are not thought to be very significant; however, it is of interest to compare them with 36,300 and $10^{12.5}$, respectively, which were determined from the decay portion of the thermal decomposition data (Ref 4).

In an effort to account for a difference of approximately a factor of twenty in the experimentally observed time to explosion for detonations of lead azide in high vacuum and in a helium atmosphere, computations were made with different values of the thermal conductivity of the explosive (actually, the two-phase explosive/gas system). These changes in thermal conductivity resulted in a trend in the computed times to explosion which was correct. That is to say, the thermal conductivity which represented the lead azide/helium system resulted in the shortest time to explosion, as was observed experimentally. However, close observation was required to see the trend because it was not nearly strong enough to describe the observed data in spite of the fact that thermal conductivity values were changed as much as four orders of magnitude in an effort to describe the best transfer in vacuum and in helium atmosphere.

A comparison was made for lead azide at 210 C and 246°C among

1. The rate of reaction given by the kinetic parameters used in the computations of time to explosion.

2. The initial rate of reaction which was observed experimentally, and

3. The rate of reaction given by the kinetic parameters which permitted a force fit of the data. The rates of reaction used in the time-to-explosion computations, when calculated at 210 C and 246°C, were 35-50 times greater than the estimated initial rates of reaction obtained from thermal decomposition data. On the other hand, however, the estimated initial experimental rates for the same temperatures were 230-475 times as great as the rates which permitted a force fit of explosion data.

The heat-up characteristics of lead azide, if it were inert, were determined for the same mathematical model and parameters as were used to compute times to explosion. The lead azide was presumed to have no internal heat generation at any temperature. At 350 C, where the observed time to explosion in a nitrogen atmosphere was 24 seconds, the "if inert" lead azide was all at essentially 350 C at that time; furthermore, no part of the "if inert" lead azide was below 330 C from the 5-second time to the 24-second time. At 420 C (where the experimental time to explosion is 0.0707 second), only about 20% of the "if inert" lead azide had been heated at all above its initial temperature of 30 C. Only the surface was at 420 C, and the

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temperature sloped off rapidly to $30°C$. These findings would indicate that in the experiment either considerable heating occurred by a radiative mechanism that is not accounted for in the calculations by the thermal conductivity term, or that whatever of importance that occurred happened very near to the surface of the explosive which contacted the elevated temperature.

INTRODUCTION

When an explosive is subjected to a temperature high enough to cause it to explode, it is logical to ask how much time will elapse between the application of the heat source and the detonation of the explosive. Calculations of this time to explosion can be made by considering heat conduction in the explosive in combination with the simultaneous, internal heat generation resulting from thermal decomposition of the explosive. Further, the environment of the explosive must be considered; the surroundings can markedly affect the time to explosion.

The differential equation which describes heat conduction and simultaneous, internal heat generation in an elemental volume of explosive is given in Equation 1. (Please notice the list of symbols in the latter part of the report.)

$$- \lambda \left(\frac{m}{X} \frac{\partial T}{\partial X} + \frac{\partial^2 T}{\partial X^2} \right) + \rho c \ \frac{\partial T}{\partial t} = \rho Q \ \text{(rate of reaction)} \tag{1}$$

"m" is a factor which depends on the geometry under consideration. That is $m = 0$ for an infinite slab with planar isotherms, $m = 1$ for an infinitely long cylinder with cylindrical isotherms, and $m' = 2$ for a sphere with spherical isotherms. Of principal interest here is the case of $m = 0$, which is depicted in Figure 1 below.
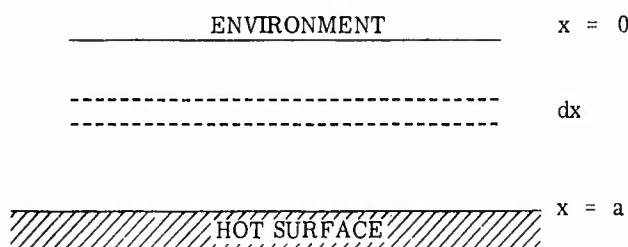
Fig. 1. Infinite slab of explosive on hot surface

Equation 1 rewritten for this case becomes

$$- \lambda \ \frac{\partial^2 T}{\partial X^2} + \rho c \ \frac{\partial T}{\partial t} = \rho Q \ \text{(rate of reaction)} \tag{2}$$

The problem is not completely defined until boundary and initial conditions are given and, in this case, until an expression is given for the rate of reaction. In the cases studied, the rate of reaction was always written as a zero-order, Arrhenius-type equation,

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$$\text{Rate of reaction} = Ze^{-E/RT} \tag{3}$$

When a rate expression which contains reciprocal temperature in an exponent, such as Equation 3, is introduced into Equation 2, the differential equation becomes strongly nonlinear with the result that no analytical solution is obtainable. Consequently, it is necessary to resort to machine solutions to Equations 2 and 3.

Each set of initial and boundary conditions specified results in a new problem. Two different sets of conditions are considered in this report.

The initial and boundary conditions for the case depicted in Figure 1 are

$$T \ (0, \ x) \ = \ T_i \qquad\qquad a > x > -a \tag{4}$$

$$T \ (t, \ \pm a) \ = \ T_{HP} \qquad\qquad t > 0 \ . \tag{5}$$

These conditions lead to symmetrical heating of the explosive, for which one can visualize two models. One can visualize a slab of twice the thickness shown in Figure 1 with hot surfaces suddenly applied to both faces, or one can visualize the slab as shown in Figure 1 with the upper face perfectly insulated from its environment, the perfect insulation being an outgrowth of the symmetry of the problem. Such a condition might be approximated if the "environment" shown in Figure 1 were a vacuum. Zinn and Mader (Ref 8) reported a usable correlation of solutions for this set of boundary and initial conditions; that is to say, the article did not contain solutions to Equations (2 through 5), but rather a correlation for time to explosion, which was derived from many solutions of the differential equation. Their correlation is used extensively and studied in detail in this report. It is

$$t_e \ = \ \frac{\rho c a^2}{\lambda} \ F \left(\frac{E}{T_m} - \frac{E}{T_{HP}} \right) \tag{6}$$

where $F\left(\dfrac{E}{T_m} - \dfrac{E}{T_{HP}}\right)$ is a function developed by Zinn and Mader to describe the correlation in compact fashion, and T_m is the minimum temperature of explosion to be calculated from

$$T_m \ = \ \frac{E}{2.303 \ R \ \log \dfrac{\rho Q Z a^2 E}{\lambda R T_m^2 \delta}} \tag{7}$$

This equation is considered in detail in a previous report by the author (Ref 7).

The explosive can also be considered as a semi-infinite solid. In Figure 1, this consideration is equivalent to moving the $x = 0$ plane infinitely far above the hot surface. The initial and boundary conditions are

$$T \ (0, \ x) \ = \ T_i \qquad\qquad x > 0 \tag{8}$$

$$T \ (t, \ 0) \ = \ T_{HP} \qquad\qquad t > 0 \tag{9}$$

$$T \ (t, \ \infty) \ = \ T_i \qquad\qquad t > 0 \tag{10}$$

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This approach is accompanied by the argument that anything of importance happening prior to the explosion occurs near the surface of the hot plate, and it occurs so rapidly that the explosion does not know or care about its surface which communicates with the environment. G. B. Cook (Ref 2) reported a usable correlation of solutions to this problem, Eq. 2, 3, 8, 9, and 10. He gives only sparse details of the correlating procedure, but he certainly had to solve the differential equation a number of times, define the explosion condition, and then correlate the resulting times to explosion. Cook's correlation is

$$t_e = \frac{CE\ 10^{14}}{RQZ}\ \left[- \left| C \right|\ \log 10^2\ \frac{RT_i}{E}\ +\ D \right] \tag{11}$$

$$\log \left| C \right| = \frac{0.4305\ E}{RT_{HP}}\ -\ 14.43 \tag{12}$$

$$\log\ D = \frac{0.4168\ E}{RT_{HP}}\ -\ 14.33 \tag{13}$$

This correlation is considered in this report but not in as much detail as the Zinn and Mader correlation, primarily because Cook's correlation is lacking in some important properties. For example, thermal conductivity does not appear at all in his correlation.

Both of these correlations apply to explosives in general, although it is to be pointed out that no provision is made for latent heat of a phase change (say melting). However, this report deals almost entirely with lead azide because that was the explosive under study. Lead azide explodes before melting.

Given these correlations, one must still have numerical values for the properties of the explosive required in the correlation before a numerical value can be obtained for the time to explosion at a particular temperature. Getting values for the properties which can be used with confidence is a difficult task. A list of such values (Ref 6), currently thought to be the best available for lead azide, has been prepared for use in the Explosives Research Section.

Much of this report is directed toward answering the question: "Which of the properties required by the correlations are really of importance in calculating time to explosion ?" Or, to state the question more quantitatively, "How much does each parameter affect calculated time to explosion ?" Answers to this question point out areas where additional research is required and also, on the other hand, areas where currently available theories and data seem adequate.

EFFECT OF HEAT OF EXPLOSION, Q, AND FREQUENCY FACTOR, Z

It can easily be observed in both Zinn and Mader's and in Cook's (Ref 2) results that Q and Z appear only once, and that, furthermore, they always appear side by side, so that the relationship $N\ Q = NZ$ holds. In other words, doubling Q is the same as doubling Z.

Intuition tells one that the greater the heat of reaction, Q, the shorter should be the delay time for a given hot-plate temperature because more heat is given off per unit of explosive decomposed, and hence the temperature rises more rapidly.

Inspection of Cook's results reveals that Q has a one-to-one effect on explosion time. Doubling Q will half t_e, the explosion time.

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The effect is not quite so straightforward in Zinn and Mader's results; however, the effect is not too complicated, since Q (and Z) appear only once. Figure 2 shows the effect of varying Q, and Figure 3 shows the effect of varying Z. It can be observed from these figures that the effect is essentially one-to-one.

It can be further observed, from Figure 2, that a large increase in Q will result in a relatively small decrease in the minimum temperature of explosion but will result in a relatively large decrease in the explosion time. To emphasize: An increase in Q from 399 cal/g to 800 cal/g will decrease the minimum temperature of explosion only from $201°C$ to $192°C$ but, at a hot-plate temperature of $340°C$, the explosion time is decreased from 4.8 sec to 2.4 sec, a 50% reduction.

It is recognized that, if kinetic data are derived from thermal decomposition experiments, the frequency factor, Z, is dependent upon activation energy and that changes in Z cannot be considered independently of activation energy. This interdependence is discussed more fully in a subsequent section entitled "Effect of Simultaneous Changes in Activation Energy and Frequency Factor." The analysis shown in Figure 3 concerning independent changes in Z was made because more often than not kinetic parameters are not taken from consistent thermal decomposition data; often Z's are selected by taking a "typical value" for explosives, like 10^{13} sec^{-1}, which is not based on any particular thermal decomposition experiment.

Computations were made for RDX as well as for lead azide. The analysis for heat of explosion of RDX uncovered the following interesting item:

Zinn and Mader (Ref 8) point out the fine compilation of physical properties of explosives to be found in Picatinny Arsenal Technical Report (PATR) 1740. They proceed to use each value for RDX in that report until they come to Q. They then say that many explosives exhibit a heat of reaction of 500 cal/g. Using PATR 1740 values plus 500 cal/g for Q, they calculate explosion times as shown in Figure 4. Excellent agreement is observed. However, PATR 1740 gives a value of 1280 cal/g for RDX. (This value has been independently rechecked and is believed to be accurate.) The line calculated using this value of Q is also shown in Figure 4. Agreement is not nearly so good, as all predicted explosion times are too short. It is interesting to speculate as to why not all PATR 1740 numbers were used.

EFFECT OF DENSITY, ρ, AND THERMAL CONDUCTIVITY, λ

There is no intuitively obvious effect of density or thermal conductivity on time to explosion.

Inspection of Cook's results (Ref 2) reveals no appearance of either ρ or λ, from which one should assume there is no effect of either variable on the time to explosion. It is possible that these terms are hidden in the constant terms 10^{14}, 10^2, 0.4305, etc., because Cook states in this article (or in some other article of his) that he assumes $\lambda = 7 \times 10^{-4}$ cal/cm sec°K for all explosives. Such an explanation, however, involves some sort of paradox, because he also assumes that c = 0.35 cal/g°K for all explosives and yet c appears to the first power in the explosion time expression. It is possible that c was inserted as itself and not in a constant in order to emphasize the dimensional correctness of the equation.

Both ρ and λ appear twice in the Zinn and Mader (Ref 8) results; therefore, one would expect the effects of these variables to be complicated and not immediately obvious. One can also observe that in both appearances the variables take the form ρ/λ; hence, it should be noted that $\ln \rho = \frac{\lambda}{\bar{n}}$. In other words, doubling the density has the same effect as halving the thermal conductivity.

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Figures 5 and 6 show the computed effect of ρ and λ. At high temperatures, such as 400°C, the effects of ρ and λ are very slight and for all practical purposes non-existent. At the low-temperature end of Figures 5 and 6, near the minimum temperature of explosion, the effects of ρ and λ are quite pronounced. It is clear that, at the low-temperature end, an increase in ρ causes an increase in t_e and conversely, that an increase in λ causes a decrease in t_e.

This latter statement backs up an experimental result obtained by James E. Abel (Ref 1) of the Explosives Research Section while exploding lead azide on a hot plate in different atmospheres, and it also confirms the qualitative explanation used to back up the experiment. At a given hot-plate temperature, explosion times were shortest in a helium atmosphere, somewhat longer in a nitrogen atmosphere, still longer in air at 4 microns pressure, and longest in air at 0.4 micron pressure. These differences in experimentally observed explosion times at a given temperature were definitely significantly different from one another. From helium atmosphere to air at 0.4 micron pressure, the times differed by a factor of roughly twenty. The qualitative explanation thus backed up by the calculations is that, to get a shorter explosion time, one must pump heat into the relatively cold (when it first hits the hot plate) lead azide as rapidly as possible. This feat is best accomplished when the surrounding atmosphere has the highest thermal conductivity; this would be He, among the atmospheres used. The effects of helium are three: (1) the surrounding atmosphere is warmer due to heat conducted away from the hot plate, (2) the heat-transfer coefficient between the lead azide and the surroundings is higher, and (3) the effective thermal conductivity of the two-phase explosive (lead azide and the gas in the interstices) is higher. All three effects favor faster heating of the lead azide.

Since the variables Q, Z, a (radius of cylinder), and ρ appear in the same position in the minimum-temperature-of-explosion equation, Equation 7, they all have the effect of lowering the minimum temperature of explosion as they are increased. The thermal conductivity, being in a reciprocal position with respect to the above-named variables, has the effect of raising the minimum temperature of explosion as it is increased. This observation is contrary to the results of the aforementioned experiments of James E. Abel, where the minimum temperature of explosion in a helium atmosphere was 341°C, in a nitrogen atmosphere 345°C, and in 4-micron air pressure 355°C. It is also contrary to the aforementioned qualitative explanation used to explain the shortness of explosion times in a helium atmosphere in terms of the mechanism of pumping in the most heat in the shortest time.

In accordance with the aforementioned qualitative theory, one should simply be able to say that, at some low temperature when the atmosphere was helium, enough heat could be pumped in to set off the lead azide whereas, at that same low temperature, not enough heat could be pumped into the lead azide to explode it if the atmosphere were low-pressure air. Hence, helium permits explosion at a lower temperature. This qualitative explanation is in accordance with the experimental data of James E. Abel but at variance with the calculations being investigated here.

On the other hand, the calculations being investigated here support on this point the erroneous theory advanced, before the experiments were run, to explain that the shortest explosion times should be accomplished under vacuum. When subsequent experiments showed the shortest explosion times to be under a helium atmosphere and the longest to be under vacuum conditions, this theory had to be discarded. This "discarded, wrong" theory said that when the surrounding atmosphere had a high thermal conductivity, as does helium, the lead azide would be cooled most; hence, the explosive could stand higher temperatures under the better cooling conditions.

If the experimental curves actually crossed one another, it could be reasonably argued that both theories—the cooling theory for low temperatures near the minimum temperature of explosion, and the pumping-in-heat theory at the higher temperatures—were correct.

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The points of inflection at the low-temperature end of the curve for $\lambda = 1.55$ x 10^{-3} (Fig. 6) result from the points of inflection in Zinn and Mader's correlating function F, at values of the abscissa near zero. Furthermore, at the low-temperature end, the temperatures are nearing those of the minimum temperature of explosion; hence, the time values must shoot up very rapidly in this region. All curves in Figures 5 and 6 would exhibit the inflection points shown in Figure 6 if they were extended far enough toward their minimum temperature of explosion.

It is not to be assumed from the previous section or from this section that a small change in the minimum temperature of explosion always means a corresponding large change in explosion time. Witness the temperature region in Figure 5 near $400°$C, where each density line means a different minimum temperature of explosion, yet each line barely represents a different explosion time.

EFFECT OF SIMULTANEOUS CHANGES IN ACTIVATION ENERGY AND FREQUENCY FACTOR

It has been the custom in explosives technology to take an activation energy from one source, any source, and combine it with an assumed frequency factor or one commonly used for other materials of the same class as the material in question. Accordingly, when the RDX study was made, the effects of activation energy and frequency factor were considered separately although it was recognized that they were interrelated.

The effect of activation energy is not obvious in either Zinn and Mader's (Ref 8) or Cook's (Ref 2) results because E appears in many places.

Figure 7 shows the results of the calculations for a one-inch slab of RDX with various activation energies. It is immediately apparent that an increase in the activation energy increases the explosion time at any given hot-plate temperature. Furthermore, the minimum temperature of explosion is also increased with a larger E. Quantitative inspection of Figure 7 shows that a 2% increase in E will at least double the explosion time. In view of the accuracies of "experimental" activation energies, this observation is so severely incriminating for the calculation method being investigated here that these computations were double checked for accuracy. There are no computational errors.

The computations performed with Cook's results are also shown in Figure 7. For values of E less than 47,500 cal/g mole, explosion times calculated from Zinn and Mader are greater than those calculated from Cook. As the value of E increases, the explosion times as calculated from Zinn and Mader become closer to those of Cook. For values of E greater than 57,000 cal/g mole, explosion times calculated from Zinn and Mader are shorter than those calculated from Cook. Hence, at a value of E somewhere between 47,500 and 57,000 cal/g mole, the explosion times of both authors will be identical.

In any event, for both correlations, the dominating term in the solutions is the activation energy, E.

If one considers zero-order kinetics, as is often done, the decomposition equation is

$$\frac{d\alpha}{dt} = Ze^{-E/RT} \tag{14}$$

From this equation it can be seen that, if both E and Z increase (or decrease), the individual effects will tend to counteract one another.

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The study was carried out by making simultaneous changes of E and Z which are exactly consistent. The results are shown in Figure 8. The reference line for the plot of the logarithm or rate versus $1/T$ was taken to be the one described by $E = 36,300$ cal/g and $Z = 10^{12 \cdot 0}$ sec^{-1}, this line was then rotated about the temperature point of 224°C, which is about the midpoint of the range of Jach's experimental data from which E and Z were obtained. The line was rotated so as to change its slope, and hence E, by 2% and 10%. The corresponding intercepts at $1/T = 0$, the frequency factors, were then determined. Thus, on this basis of consistent sets of E and Z, the analysis proceded. Near the reference line in the center, Figure 8 shows, for example, one line that represents an increase of 2% in E with no change in Z. (This is the same analysis as was done for RDX.) It can be seen that this change in E alone results in almost doubling the calculated time to explosion at a given temperature. Now, also shown on Figure 8 is a line which represents the same 2% increase in E, but also includes an exactly compensating change in Z to $10^{12 \cdot 32}$ sec^{-1}. It can be seen that the resultant change in calculated time to explosion is compensated for; in fact, it is overcorrected, for now there is about a 10% decrease in the calculated time to explosion at a given temperature. Similar arguments obtain for all of the other changes shown in the key of Figure 8.

One may not miss the "correct" activation energy by 10%, for example which does not appear from previous experience to be difficult), and then hope to have all of the problems resolved by a compensating change in frequency factor because, as Figure 8 shows, such explosion times will be different by about a factor of two. Furthermore, the greater the interval between the highest temperature at which decomposition data is obtained and the temperature at which the explosion time is calculated, the more the calculated explosion times will be in error for a given error in E and its compensating change in Z. In other words, the further one has to extrapolate the kinetic data, the more important it is to have exactly the correct kinetic expression.

EFFECT OF INITIAL TEMPERATURE OF THE EXPLOSIVE, T.

The effect of initial temperature became of interest in connection with James Abel's (Ref 1) experimental work because the lead azide in the storage hopper in his apparatus was heated somewhat above room temperature before it was dropped on the hot plate. One would expect explosion times to be shortened if the explosive hits the hot plate at a higher temperature because it has a start on the heating process. Furthermore, in the case of lead azide the possibility of a partial decomposition with the subsequent formation of self-catalyzing lead atoms would lead to shorter explosion times. When it was learned that the lead azide in the hopper was at temperatures of $50-70^\circ$C, this decomposition and self-catalysis effect was ruled out on the basis that no significant reaction would occur at such low temperatures.

Various initial temperatures were investigated for RDX using Cook's (Ref 2) correlation. The results are shown in Figure 9. Cook's correlation was chosen over Zinn and Mader's (Ref 8) for this calculation because the computations are less difficult. Zinn and Mader do discuss different initial temperatures, but for a spherical geometry only.

Figure 9 reveals that a 20 C increase in initial temperature decreases the explosion time by about 10%.

COMPARISON OF CALCULATIONS WITH EXPERIMENTAL DATA

One of the primary goals of this study was to be able to make successful predictions of explosion time as a function of temperature. To evaluate the effectiveness of the calculations, the results were compared to the available experimental data, those of James Abel (Ref 1) from his "vacuum-ignition-timer" studies.

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A comparison between predictions and some experimental data is shown in Figure 10. The data for detonations of the lead azide in nitrogen at atmospheric pressure are represented by a line. Many experimental points scatter about this line. For the purposes of this discussion, the data, as represented by this line, are "truth." Also shown are the values computed from the Zinn and Mader (Ref 8) correlation and those computed from Cook (Ref 2). It is difficult to choose which correlation better describes the data. The Cook correlation has the advantage of not curving down away from the data at the lower temperatures. In neither case is the slope of the line great enough.

Since the kinetic parameters produce by far the greatest changes in the computed explosion times, it is of interest and natural to ask what values for the kinetic parameters are needed to fit the experimental data. Figure 10 shows, among other things, the line which may be forced through the data with the Cook correlation if the kinetic parameters are allowed to take on any desired values. It coincides with the line representing the data for most of the range. The values of $E = 73,500$ cal/g mole and $Z = 10^{24.24}$ sec^{-1} were obtained by trial and error. The "best" line that can be forced through the data with the Zinn and Mader correlation is also shown in Figure 10. It is not as good as the forced fit with the Cook correlation at the low-temperature end of the range where it curves down away from the data. In order to obtain the kinetic parameters of 77,600 cal/g mole and $10^{25.3}$ sec^{-1}, which were used in the fit, the Zinn and Mader correlating function, F, was forced into a straight line. The function is essentially a straight line over most of its range, and this straight portion was merely extended in both directions.

The Zinn and Mader model, in the sense used here with the top surface of the slab considered to be perfectly insulated, would be expected to best describe the experimental data taken under conditions of high vacuum, which, of course, presents good insulation. Accordingly, the analysis shown in Figure 11 was undertaken. The dashed lines represent the data taken by James Abel for detonations of lead azide in atmospheres of HE (1 atm pressure), N_2 (1 atm pressure), air (4 microns pressure), and air (0.4 micron pressure). Again, the line calculated directly from Cook's correlation is shown. The line from the Zinn and Mader model is also shown; however, it differs from the one shown previously in Figure 10 in that a thermal conductivity of 2.51×10^{-6} cal/sec cm^3K, which is supposed to correspond to the fine lead azide powder in high vacuum, was used. The low thermal conductivities make for low minimum temperatures of explosion and, as a result, the calculated line is quite straight in the range covered by these experiments and does not curve down away from the data at the low-temperature end. The Zinn and Mader correlation gives results which are nearer to the experimental data than those of Cook; however, neither is satisfactory, and both have the same incorrect slope, which is not nearly great enough.

The line in Figure 11, which is coincident with the line representing the data for 0.4-micron air, is the one which was forced through the data using the Zinn and Mader correlation with adjustable kinetic parameters. The parameters producing this perfect fit are $E = 69,750$ cal g mole and $Z = 10^{21.3}$ sec^{-1}.

Figure 12 is an extension of the analysis begun in Figure 11. Again the four data lines are shown along with the forced-fit line through the data for 0.4-micron air. For the remainder of the analysis, the parameters of $E = 69,750$ and $Z = 10^{21.74}$ were retained and the thermal conductivity was varied from $2.51 \times 10^{-}$ to 2.08×10^{-7} to 0.945×10^{-4} to 4.11×10^{-4} cal/sec cm C in order to try to account for the variations in experimental data observed among vacuum, nitrogen, and helium atmospheres. These results are shown in Figure 12. It is to be observed that the trend is correct, but that it is not nearly strong enough. That is to say, the change in the values for thermal conductivity from high vacuum to helium atmosphere produces calculated explosion times which go from longer to shorter as do the experimental data, but the difference in these computed times is not great in view of the four-orders-of-magnitude change in the thermal conductivities used. Also, at the

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low-temperature end of the range, the lines representing the higher thermal conductivities, which produce the higher minimum temperatures of explosion, spread quite a bit, as do all of the computations from the Zinn and Mader correlation. In fact, the lines for the two largest thermal conductivities would cross one another at $355°$C when the one heads toward infinity.

COMPARISON OF "FORCED-FIT" KINETICS AND EXPERIMENTALLY OBSERVED KINETICS

Since kinetic parameters have the strongest effect on the calculated times to explosion, it is logical to ask how the parameters which force a fit through the data compare with experimentally obtained thermal decompositions. Figure 13 shows such a comparison. Consider first only the lines which are concerned with $210.6°$C. Near the bottom of the graph is the line representing Jach's experimental data for $210.6°$C up to a decomposition of 40%. Jach has determined $36,300$ cal/g to be the activation energy and $10^{12.5}$ sec^{-1} to be the frequency factor for the decay portions of his sigmoid-shaped curves. If these parameters are employed in a zero-order rate equation, as is done in both the Cook (Ref 2) and the Zinn and Mader (Ref 8) correlations, one of the steepest lines on Figure 13 is obtained. If the order is taken to be first, then the curve is obtained which has the same initial slope as the zero order but at all later points exhibits a lesser rate. Both of these rates are much greater than the observed initial rates. In fact, an inspection of Figure 13 as it is drawn shows that the zero-order rate is approximately 50 times greater than the experimentally observed initial rate of decomposition. If one uses the forced fit kinetics of $E = 69,750$ cal/g mole and $Z = 10^{21.74}$ sec^{-1} in a zero-order rate expression, the dashed line which nearly lies on the abscissa is obtained. At first glance, one is tempted to say that this rate is much nearer to the experimentally observed initial rate than is the aforementioned rate. However, again from the figure, the observed initial rate seems to be about 475 times greater than the zero-order rate obtained from forced-fit kinetic parameters. In any event, the rate which has been used is far too great, and the forced-fit rate seems to be much less than the observed rate.

The other lines on Figure 13, which are for $246.0°$C, reveal much the same story. Jach's data line shows the observed decomposition for $246.0°$C. It can be seen that the slope of the zero-order rate kinetic expression is far too great to describe the initial rate. This zero-order rate, however, would be a rather good description of the rate when about 35% of the material had decomposed. Of course, beyond that amount of material the zero-order rate is again always too great because it does not recede to zero when all of the material is consumed. The first-order expression is also shown. It is not much better than the zero-order one in the important early stages of decomposition. Here, the zero-order rate, from the graph, is about 35 times as great as the observed initial rate. The result of the forced-fit parameters used in the zero-order form is shown by the dashed line which is nearly on the abscissa. From the graph it appears to be about $1/230$th of the observed initial rate. Again a rate which is much too great was used and apparently the forced-fit rate which does work is, for the zero-order form, much less than the observed initial rate.

Figure 13 shows that, for both temperatures, a rate which is quite in excess of the observed initial rate has been used in calculations of times to explosion. From this observation, one should be able to conclude that the predicted delay times should be shorter than the measured times. This is indeed the case, as can be seen in Figure 12. However, such is not the complete story, for the comparisons were made at 210.6 and $246.0°$C ($246.0°$C is the maximum temperature at which Jach obtained data) and the explosion data were obtained for hot-plate temperatures of about $340-440°$C. The equation for thermal decomposition must thus be extrapolated approximately 200 C from $246°$C to about $440°$C. In this interval, it would be possible for the relationship to reverse so that the "observed" (if they could be observed in practice) initial rates would become greater than the zero-order rates. Such an analysis is shown in Figure 14.

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Figure 14 shows rate of reaction as a function of temperature. One line gives the rate as determined by the zero-order expression which uses Jach's decay-period values for E and Z. The other line gives the same information for the forced fit E of 69,750 cal/g mole and Z of $10^{21.74}$ sec^{-1}. Certainly, unless two rate expressions have exactly the same activation energy, there will be some temperature at which the rate curves will cross. At temperatures above this crossover temperature, one reaction will be faster. For the two curves shown in Figure 14, this temperature is about 480°C. Now, at lower temperatures as shown on Figure 13, the forced-fit kinetics give much slower rates of reaction; and, futhermore, the observation recorded in the previous paragraph is confirmed. That is, calculated explosion times are shorter than observed ones, but now this statement must be qualified by adding the phrase, "at temperatures below 480°C." Measurements were not obtained for temperatures higher than 440°C. Since the forced-fit kinetics give the correct explosion time at all temperatures, it can be concluded from Figure 14 that, since the 480°C point is common to both lines, Jach's decay-period kinetics would give the correct time to explosion at 480°C. Inspection of Figure 12 reveals that the extrapolated Zinn and Mader curve seems to be headed for an intersection with the extrapolated 0.4-micron-air data line in the neighborhood of 480°C. Of course, at temperatures greater than 480°C, the computed explosion times would be longer than the observed ones.

HEAT-UP OF LEAD AZIDE IF INERT

Calculations were made of the heat-up characteristics of lead azide if inert (internal heat generation considered to be zero) in an infinite slab 1.56 mm thick. The solution to this problem, where the infinite slab is initially at a uniform temperature and where the surfaces are suddenly exposed to an elevated temperature and maintained at that temperature, was taken from Ingersoll et al (Ref 3). Of course, this model is exactly the one used by Zinn and Mader except that here heat generated by slow thermal decomposition is considered to be zero. Such an assumption leads to a simpler set of equations for which a fairly simple series solution exists. The temperature/distance profiles for several times after the exposure are shown for hot-plate temperatures of 350°, 385°, and 420°C in Figures 15, 16, and 17, respectively. In all of these models, the slab thickness was taken to be 1.56 mm. It is interesting to notice the temperature profile for the time which corresponds to the experimentally observed explosion time. At 350°C on the hot plate, this time is 24 seconds; Figure 15 shows that, even if the material were inert, the entire slab would have been essentially at the hot-plate temperature at the explosion time. Further, from 5 to 24 seconds no part of the sample would be cooler than 330°C. On the other hand, at 420°C, where the time to explosion is 0.0707 second, it can be seen in Figure 17 that essentially none of the "if inert" lead azide has reached the hot-plate temperature and only roughly 20% of the slab has increased in temperature at all above the initial temperature of 30°C. In all of these calculations, the thermal conductivity of lead azide was taken to be 0.945 x 10^{-4} cal/sec cm°C, which is currently believed to be the best value available for the azide powder in a nitrogen atmosphere; the bulk density was taken to be 0.54 g/cm^3, which was observed in the photographs; and the specific heat was taken to be 0.116 cal/g°C.

Figure 18 shows the results of a calculation like those described in the previous paragraph except that the slab thickness was taken to be 1.018 mm.

It is currently thought that a thermal conductivity of 2.51 x 10^{-} cal/sec cm°C is descriptive of the lead azide powder in a high vacuum medium. Accordingly, this value was used to compute the heat-up characteristics, with all other parameters remaining the same. The results are shown in Figure 19. Since such long times are involved (in particular, since a period of 100 seconds would be required to increase the temperature of roughly 20% of the material significantly above its initial temperature), compared to the measured explosion time of only one second for this temperature of 420°C in a vacuum of 0.4 micron one would be tempted to

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say that the heat transfer is much greater than can be accounted for by the thermal conductivity value, and that apparently heat transfer occurs by radiation from particle to particle. The only other possible explanation is that all of the significant action occurs essentially on the surface of the explosive which is in contact with the . hot surface.

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SYMBOLS

ρ = density of explosive, g/cm^3

Q = heat of reaction of explosive, cal/g

Z = frequency factor in Arrhenius equation, sec^{-1}

E = activation energy, cal/g mole

R = universal gas constant, cal/g mole$^\circ$ K

T = temperature in explosive at any time or position, $^\circ$ K

λ = thermal conductivity of explosive, cal/cm sec° K

C = specific heat of explosive, cal/g° K

t = time, sec

T_i = initial temperature of explosive, $^\circ$ K

T_{HP} = hot-plate temperature, $^\circ$ K

T_m = minimum temperature of explosion at hot plate, $^\circ$ K

a = half thickness of slab, or radius of cylinder or sphere, cm

δ = geometric factor

F = function defined by Zinn and Mader (Ref 8)

t_e = explosion time, sec

x = distance, cm

c = constant

D = constant

n = constant

α = fraction decomposed

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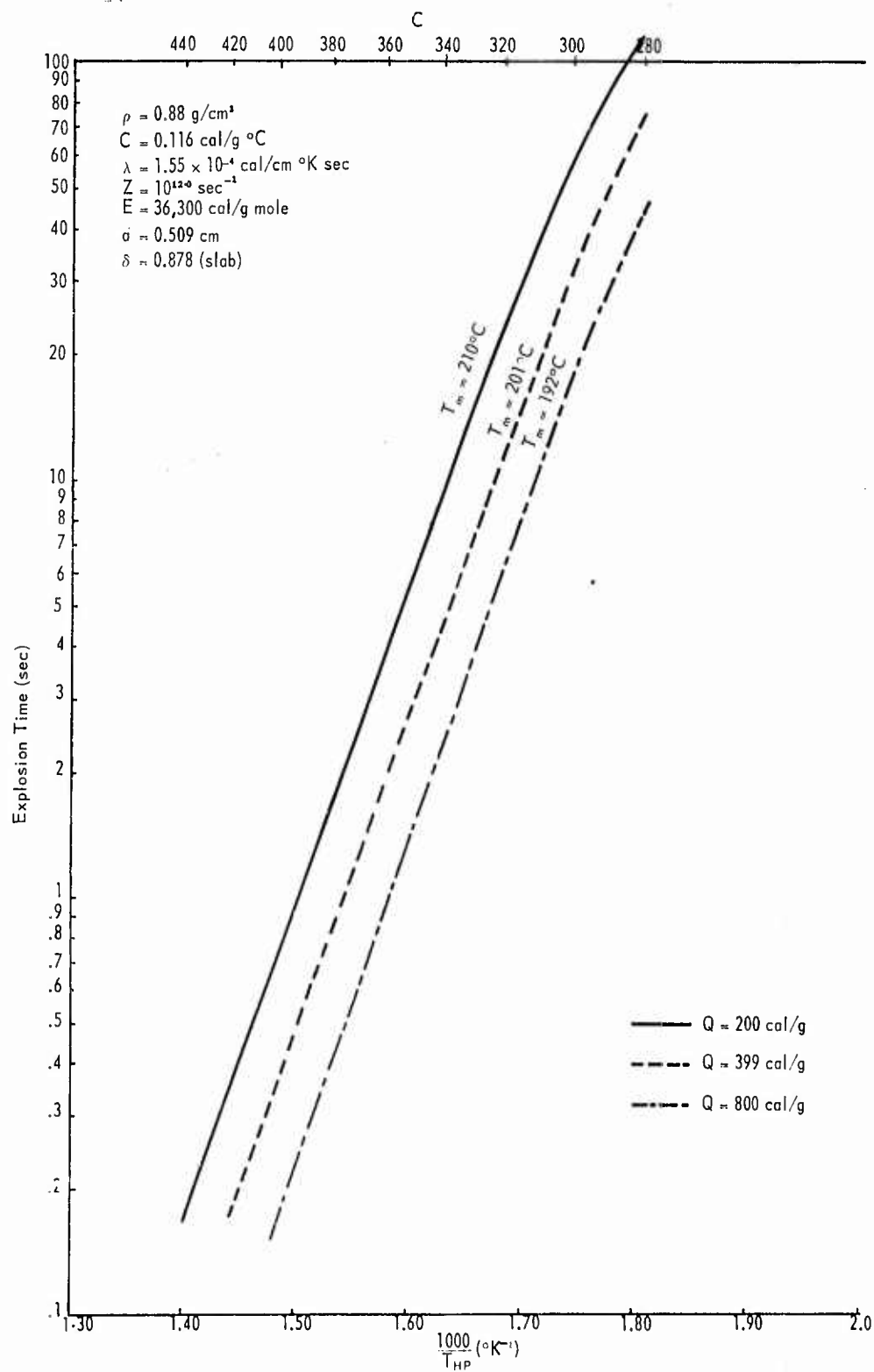
8. Zinn, J. and Mader, C.L., J. Appl. Phys. 31, 323, 1960

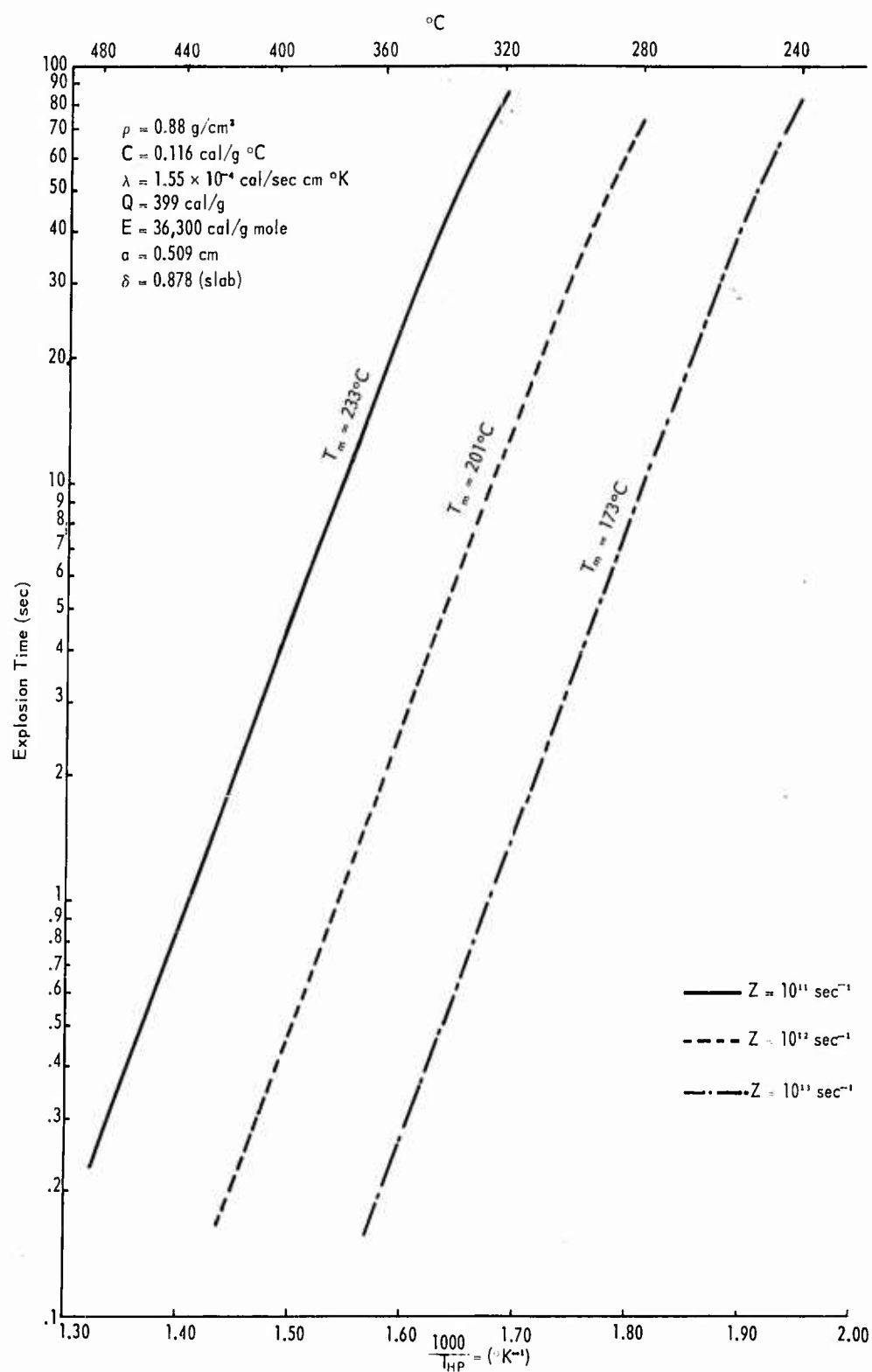Fig 2 — Effect of heat of explosion on explosion time of lead azide

°C

ρ = 0.88 g/cm³
C = 0.116 cal/g °C
λ = 1.55 × 10⁻⁴ cal/sec cm °K
Q = 399 cal/g
E = 36,300 cal/g mole
a = 0.509 cm
δ = 0.878 (slab)

T_m = 233°C

T_m = 201°C

T_m = 173°C

Z = 10¹¹ sec⁻¹

Z = 10¹² sec⁻¹

Z = 10¹³ sec⁻¹

Explosion Time (sec)

$\frac{1000}{T_{HP}}$ = (°K⁻¹)

Fig 3   Effect of frequency factor on explosion time of lead azide

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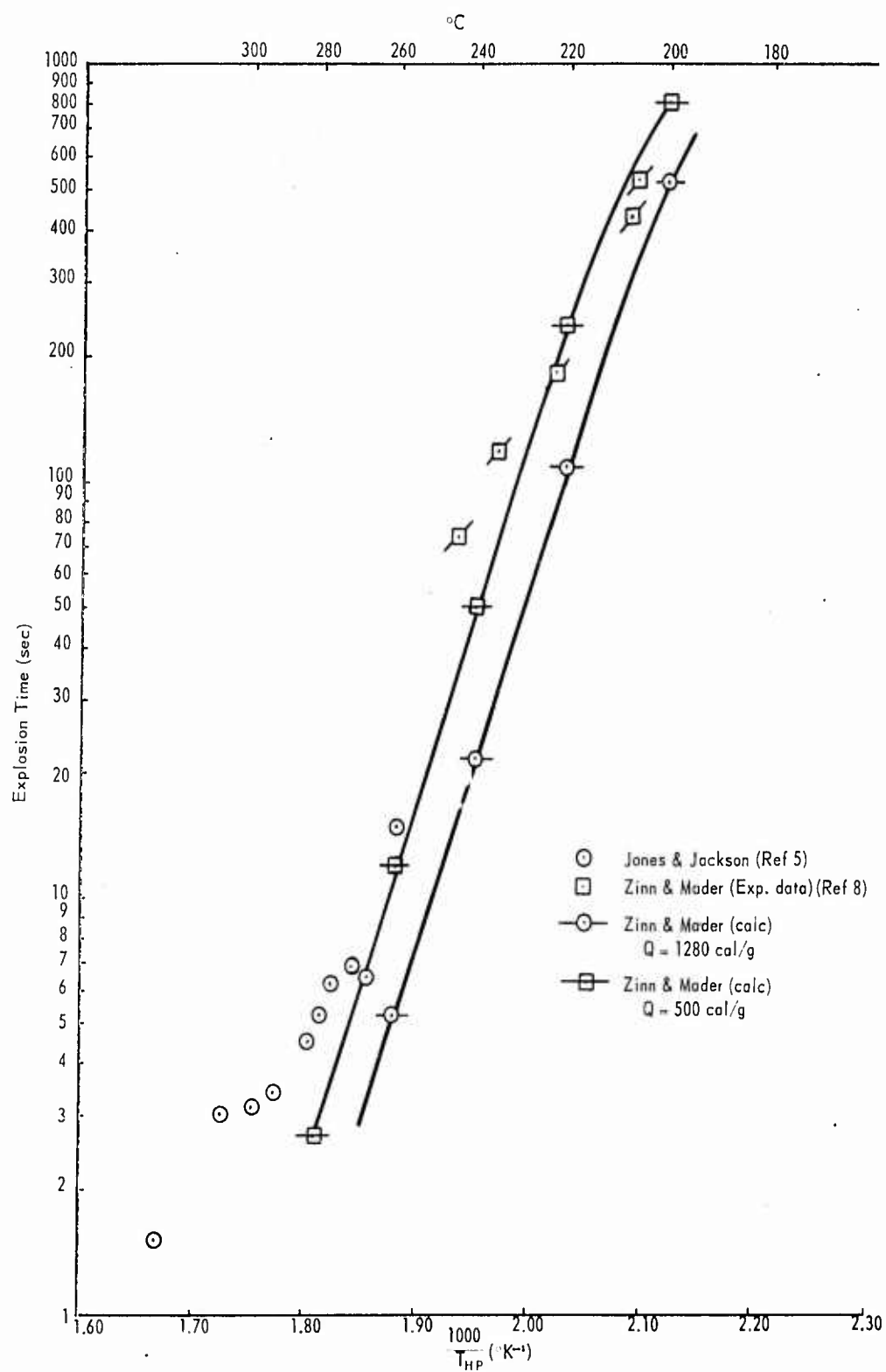
Fig 4   Comparison of calculations with experimental data of Zinn and Mader for RDX in one-inch cylinders
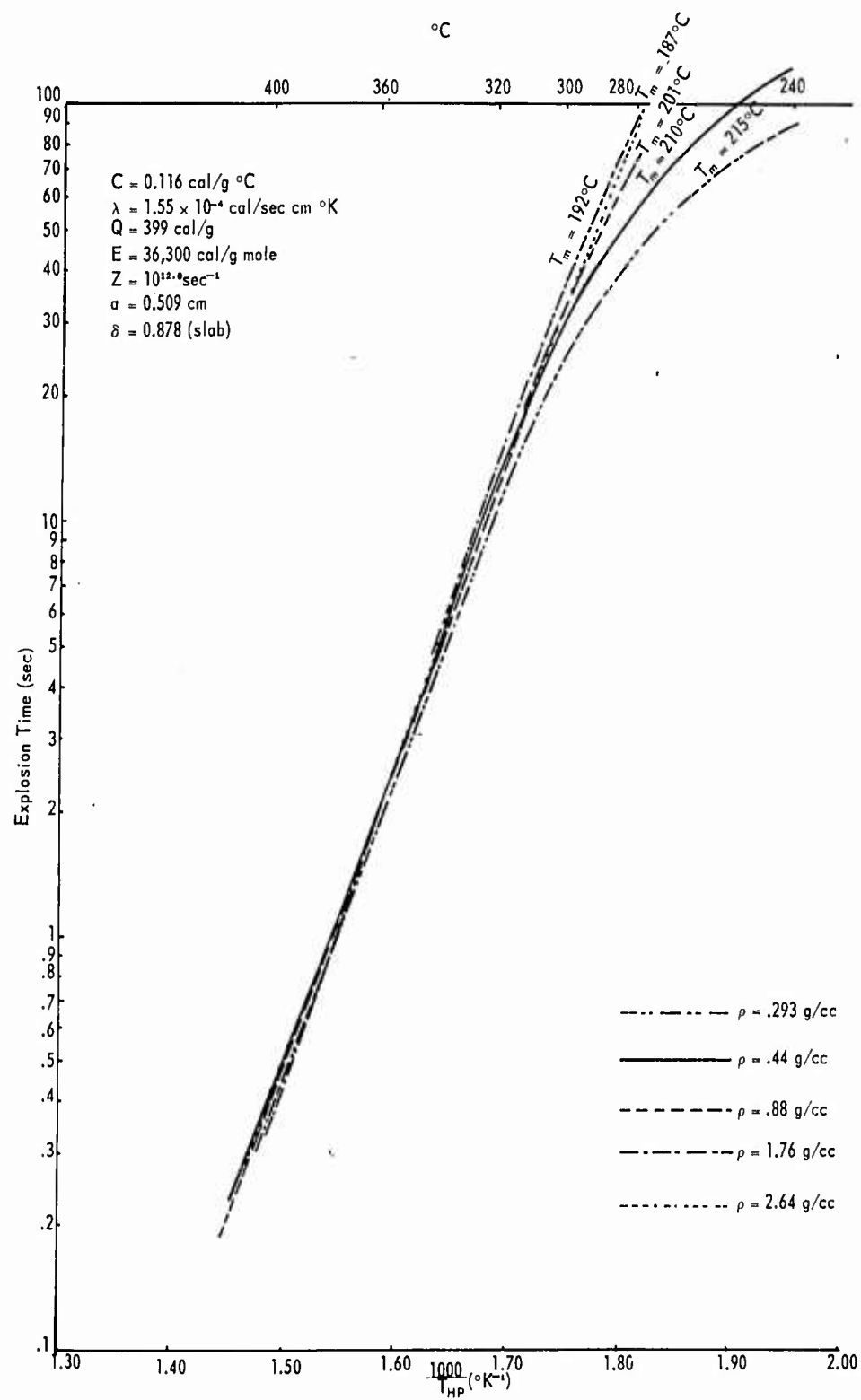
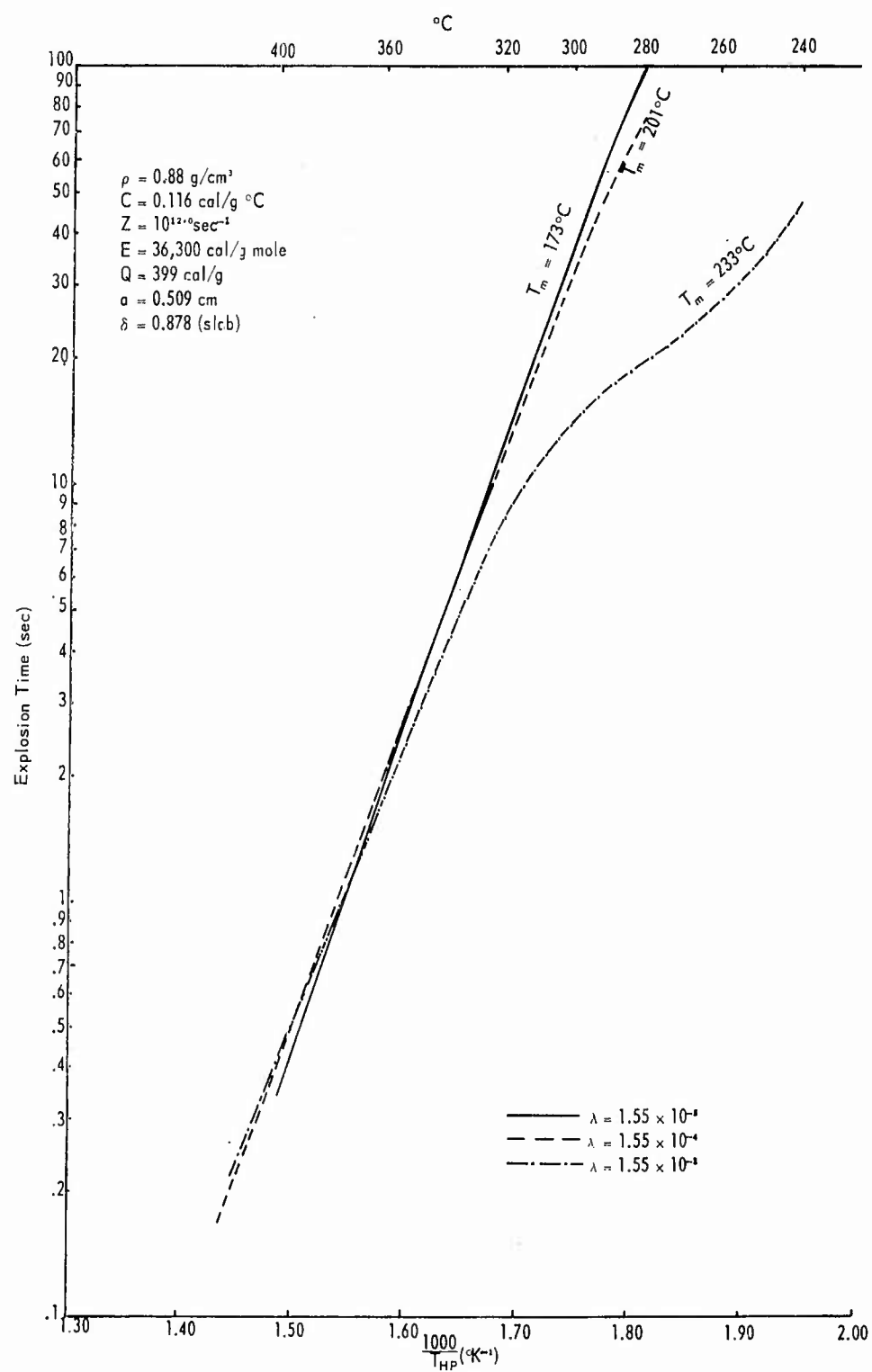Fig 5   Effect of density on explosion time of lead azide

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Fig 6   Effect of thermal conductivity on explosion time of lead azide
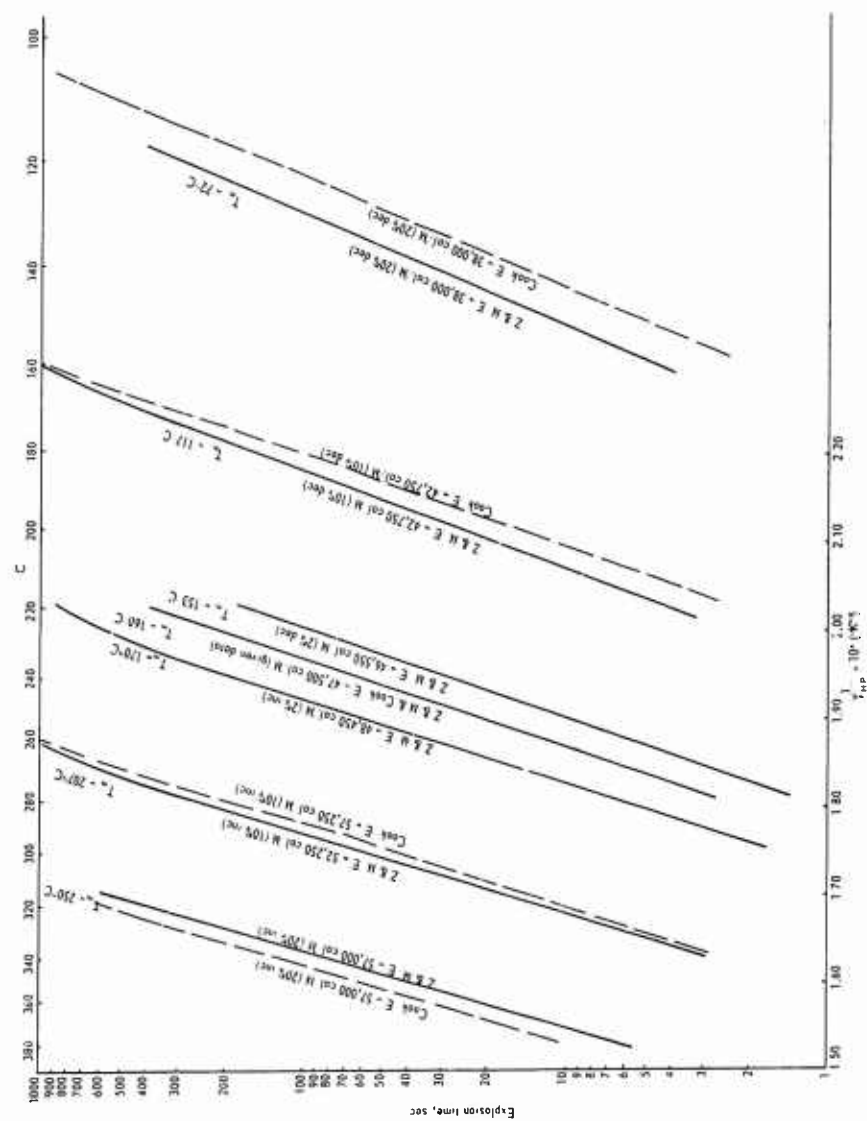
Fig 7   Effect of activation energy on explosion time of RDX
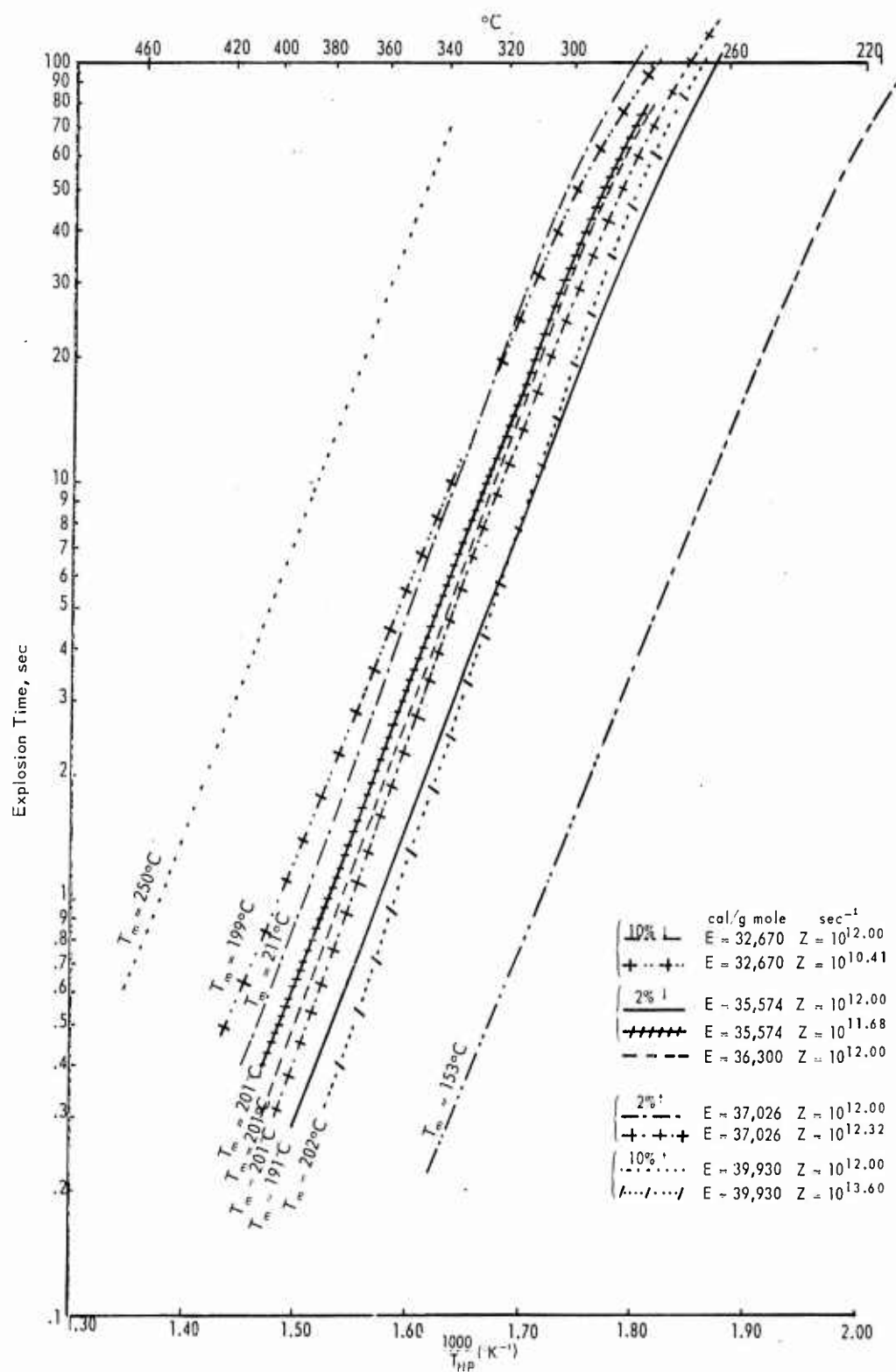
Fig 8   Effect of consistent activation energy – frequency factor changes on explosion times of lead azide
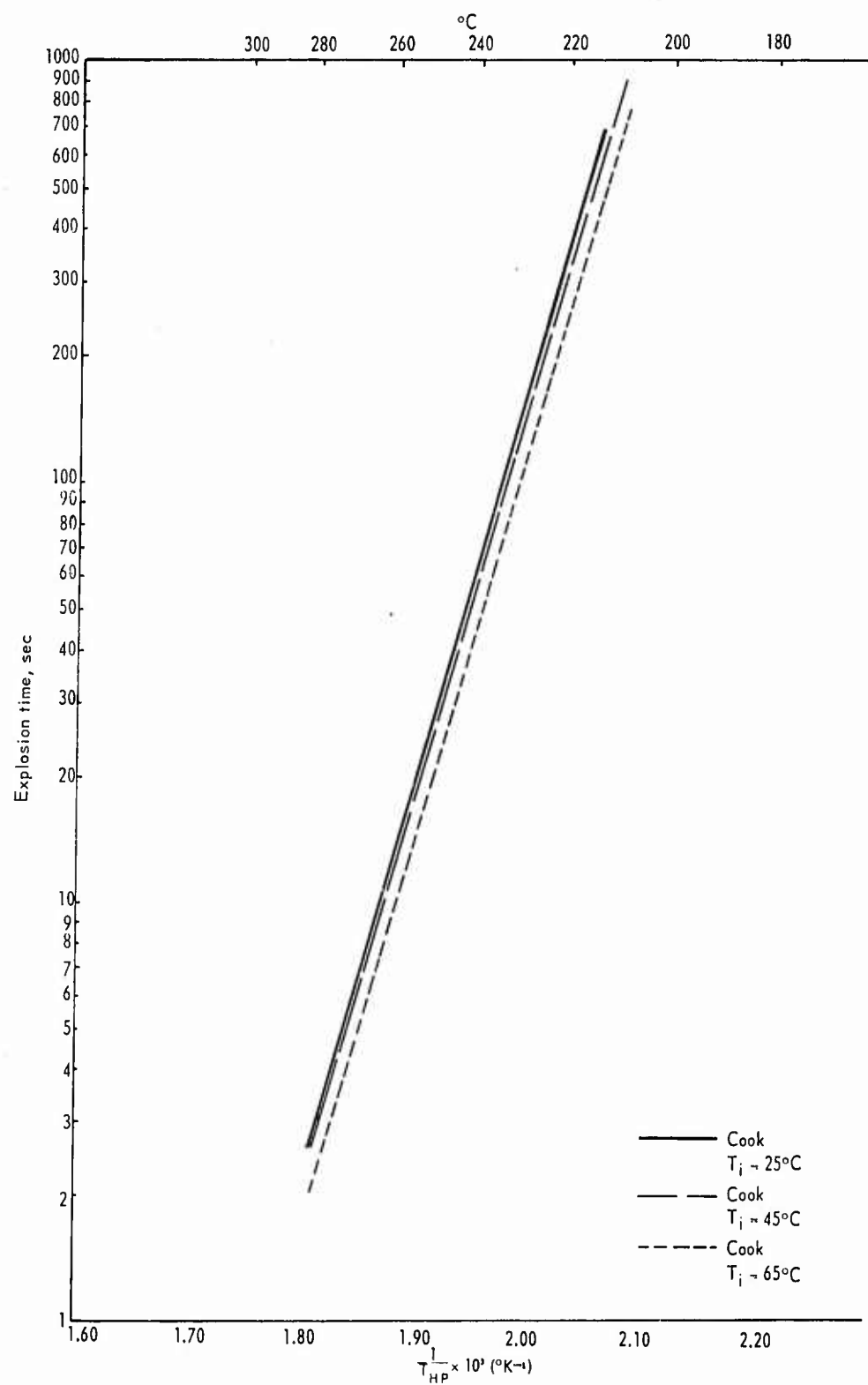
Fig 9   Effect of initial temperature on explosion time of RDX

To about 345°C

460 440 420 400 380 360 340

$T_m = 327°C$

$T_m = 263°C$

Explosion time (sec)

Abel's exp. data for explosions under
N_2. Curve represents many pts.

Best forced fit of Zinn & Mader corr.
Q = 399 cal./g
ρ = 0.54 g/cm³
C = 0.116 cal/g °C
λ = 1.55 × 10⁻⁴ cal/cm °K sec
a = 0.78 mm
δ = 0.878

Forced { E = 77,600 cal /g mole
Parameters { Z = 10²⁵·³ sec⁻¹

Forced fit of
G. B. Cook's corr.
Forced { E = 73,500 cal/g mole
{ Z = 10²⁴·²⁴⁴ sec⁻¹
C = 0.116, Q = 399, T_i = 30°C

Calc. directly from
G.B. Cook
E = 36,300 cal g mole Z = 10¹²·⁰ sec⁻¹
C = 0.116 cal. g K   Q = 399 cal g
T_i = 30°C

Calc. directly from Zinn & Mader
Parameters as above except:
{ E = 36,300 cal./g mole
{ Z = 10¹²·⁰ sec⁻¹

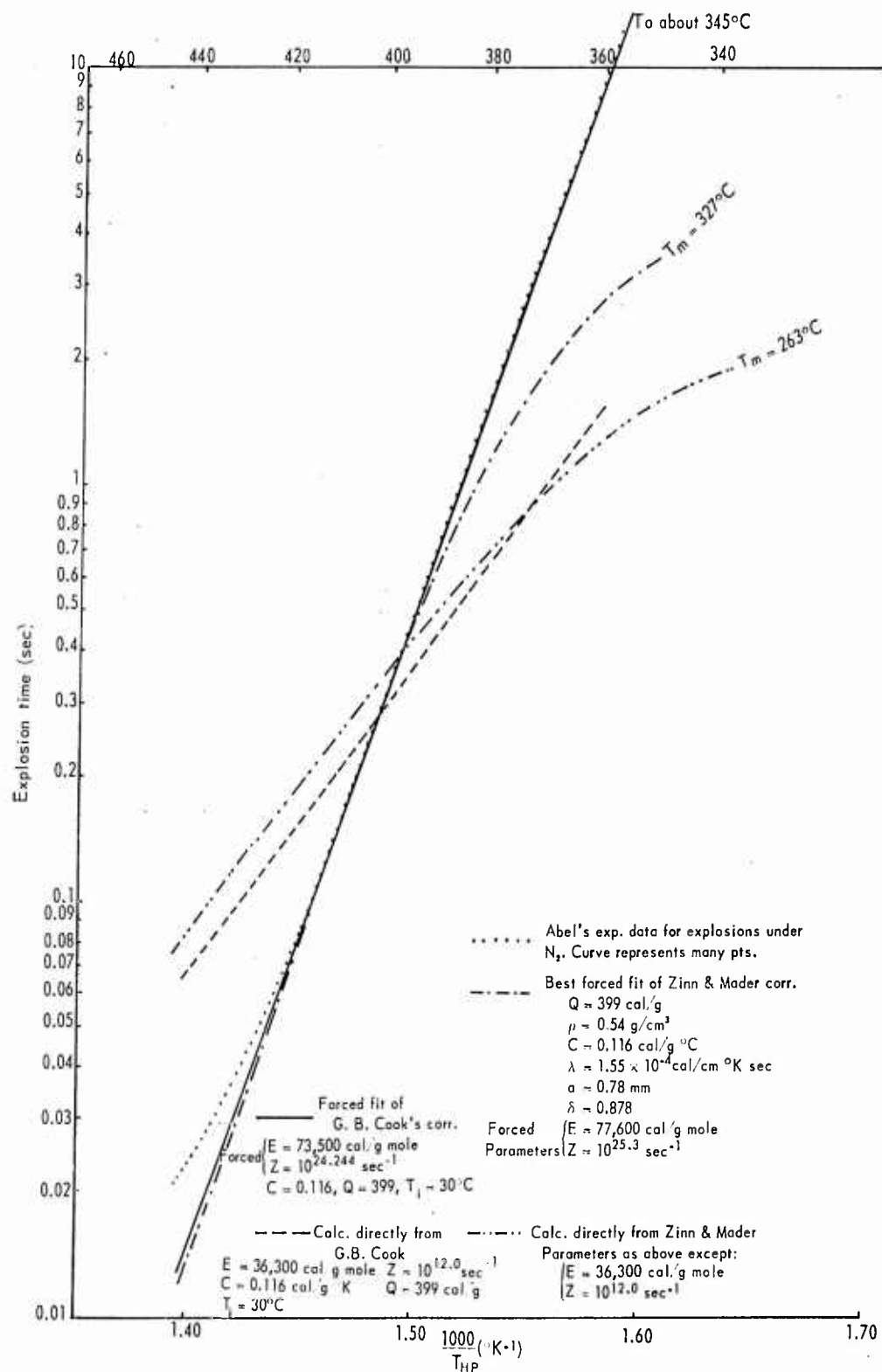1.40   1.50 $\frac{1000}{T_{HP}}$ (°K⁻¹)   1.60   1.70

Fig 10   Comparison of calculated curves with the experimental curve for
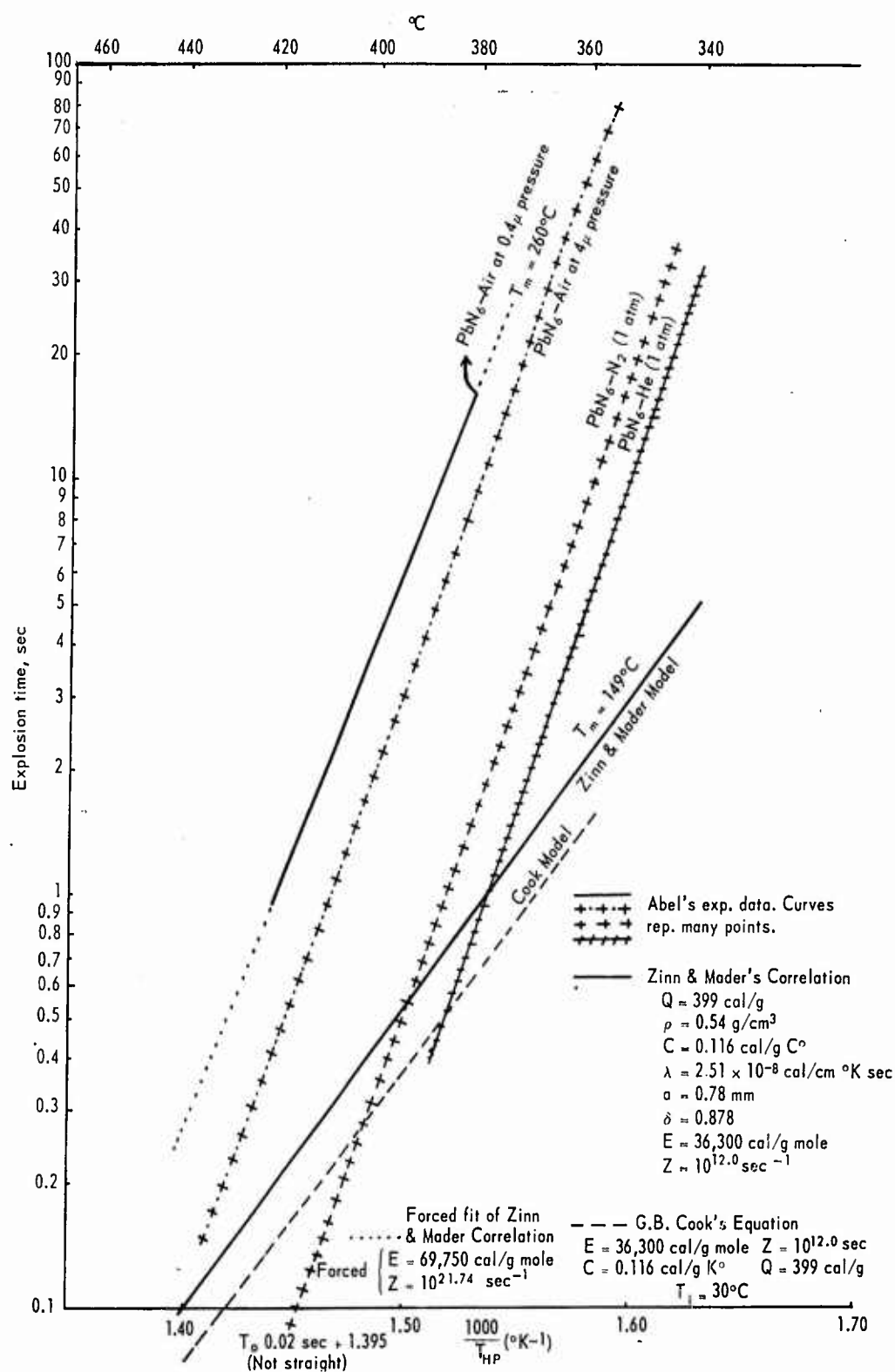explosions of lead azide under nitrogen

Fig 11   Comparison of calculated curves with the experimental curves for explosions of lead azide
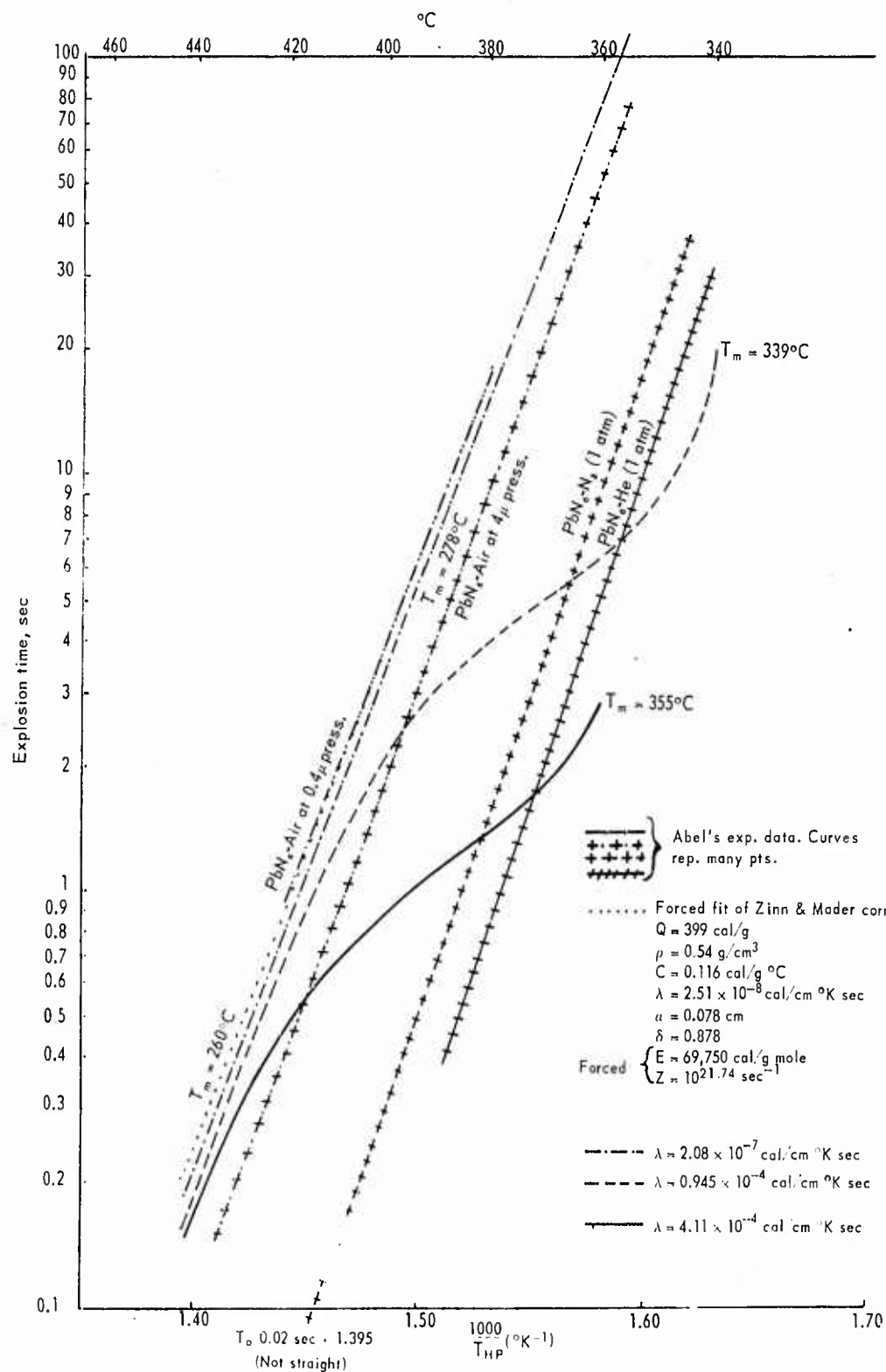
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Fig 12   Comparison of calculated curves with experimental curves for explosions of lead azide under different atmospheres
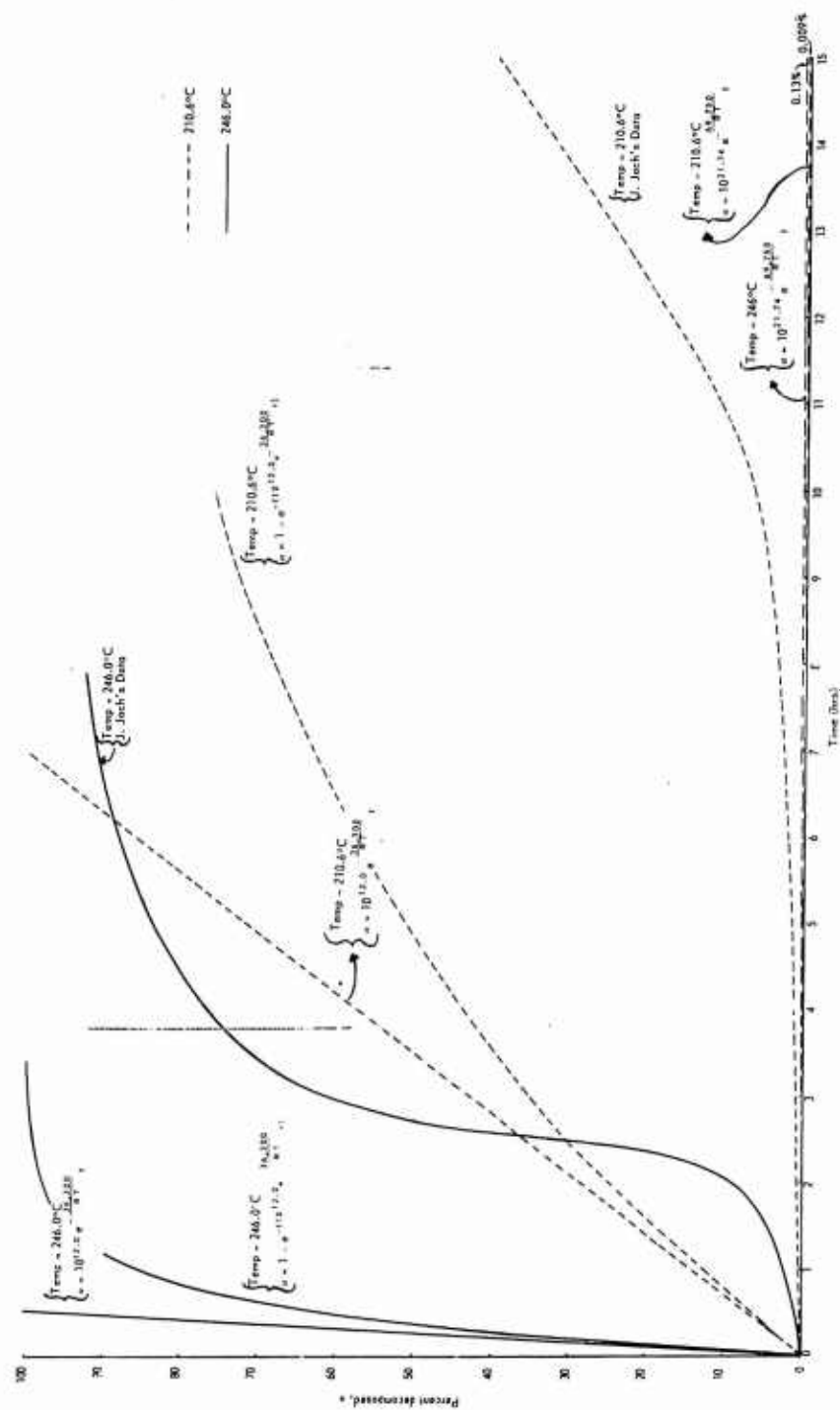
Fig 13   Comparison of experimental decomposition curves with those used for calculating explosion times of lead azide
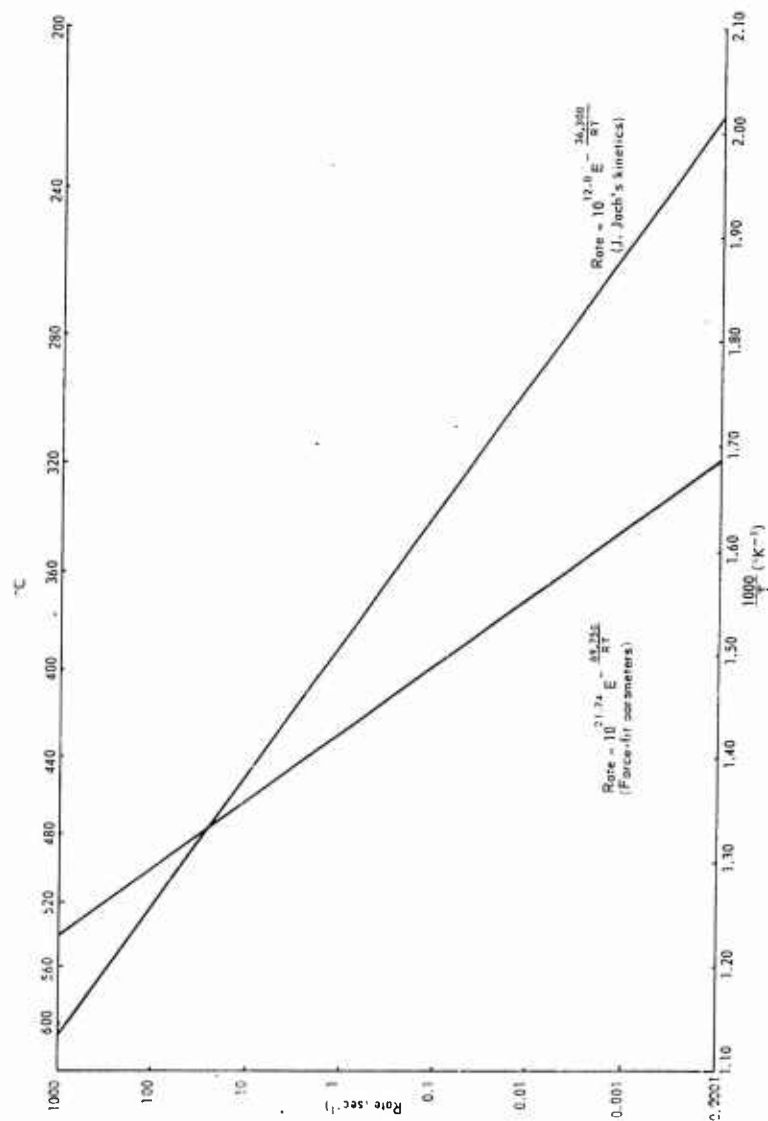
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Fig 14   Rate of reaction of lead azide as a function of temperature
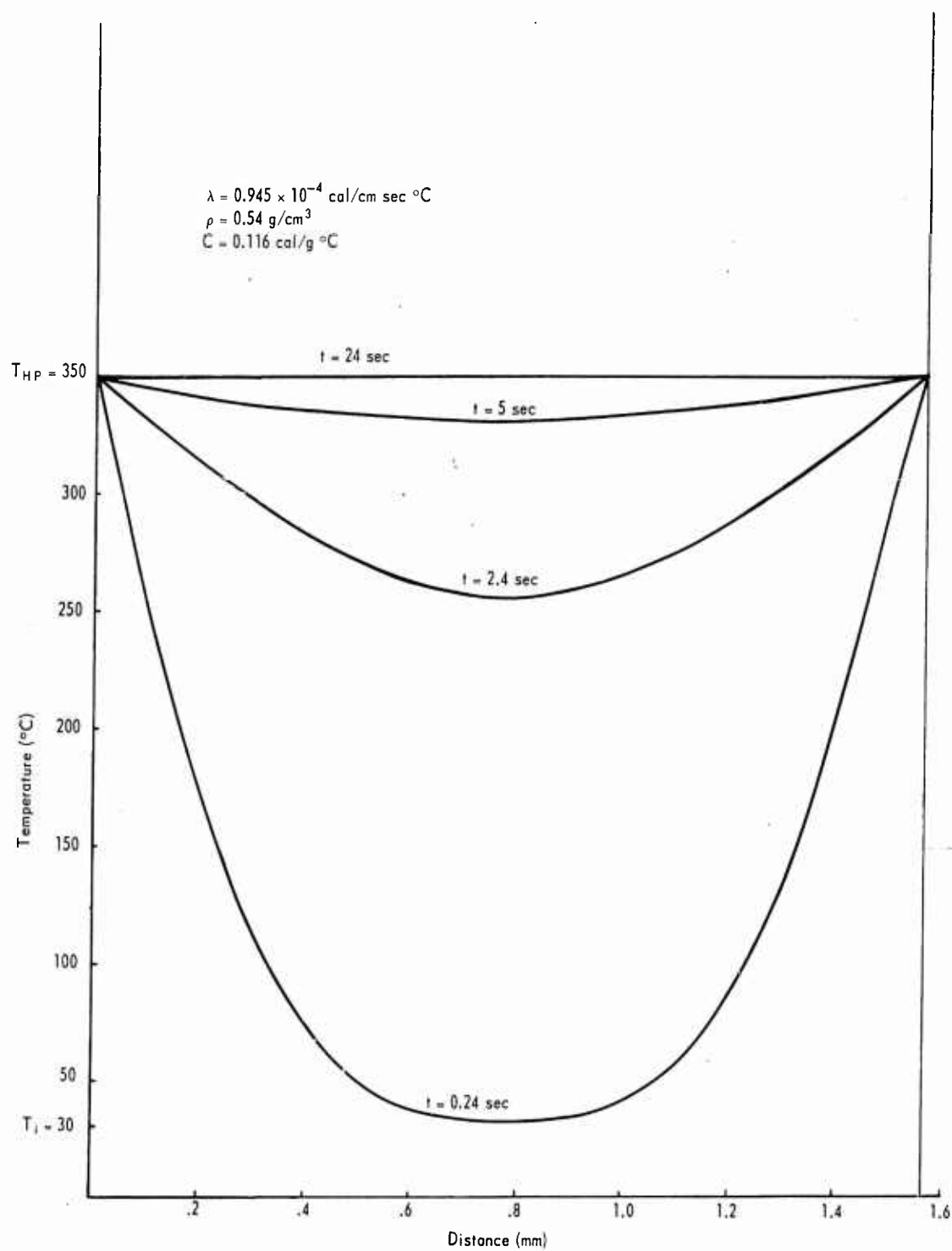
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Fig 15   Heat-up of lead azide if inert (infinite slab 1.56 mm thick). Experimentally observed explosion time at 350°C in N_2 is 24 sec
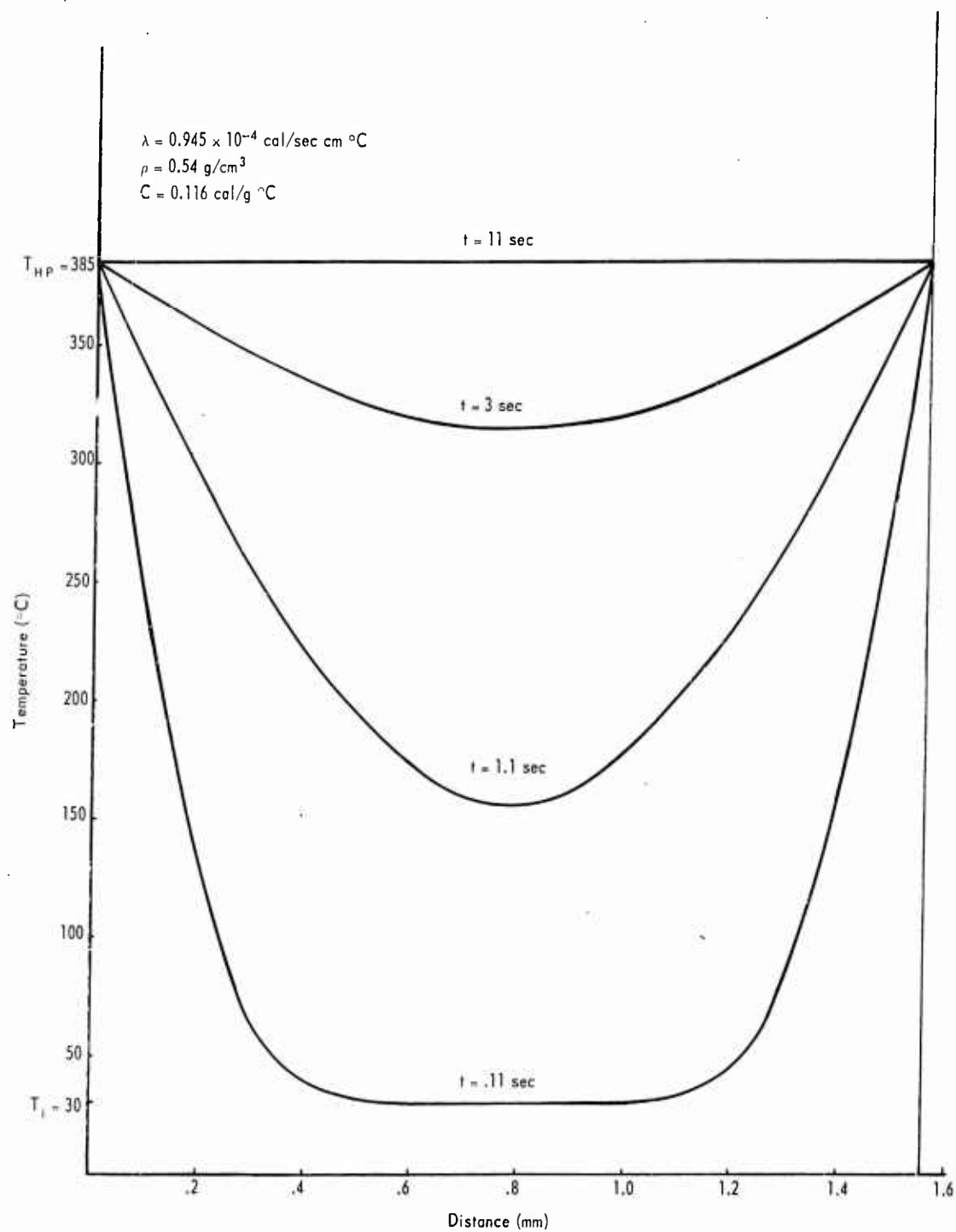
Fig 16   Heat-up of lead azide if inert (infinite slab 1.56 mm thick). Experimentally observed explosion time at 385°C in N_2 is 1.1 sec

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$\lambda = 0.945 \times 10^{-4}$ cal/cm sec °C
$\rho = 0.54$ g/cm^3
$C = 0.116$ cal/g °C

$t = 7.07$ sec

$t = 0.707$ sec

$t = 0.0707$ sec

$T_{HP} = 420$

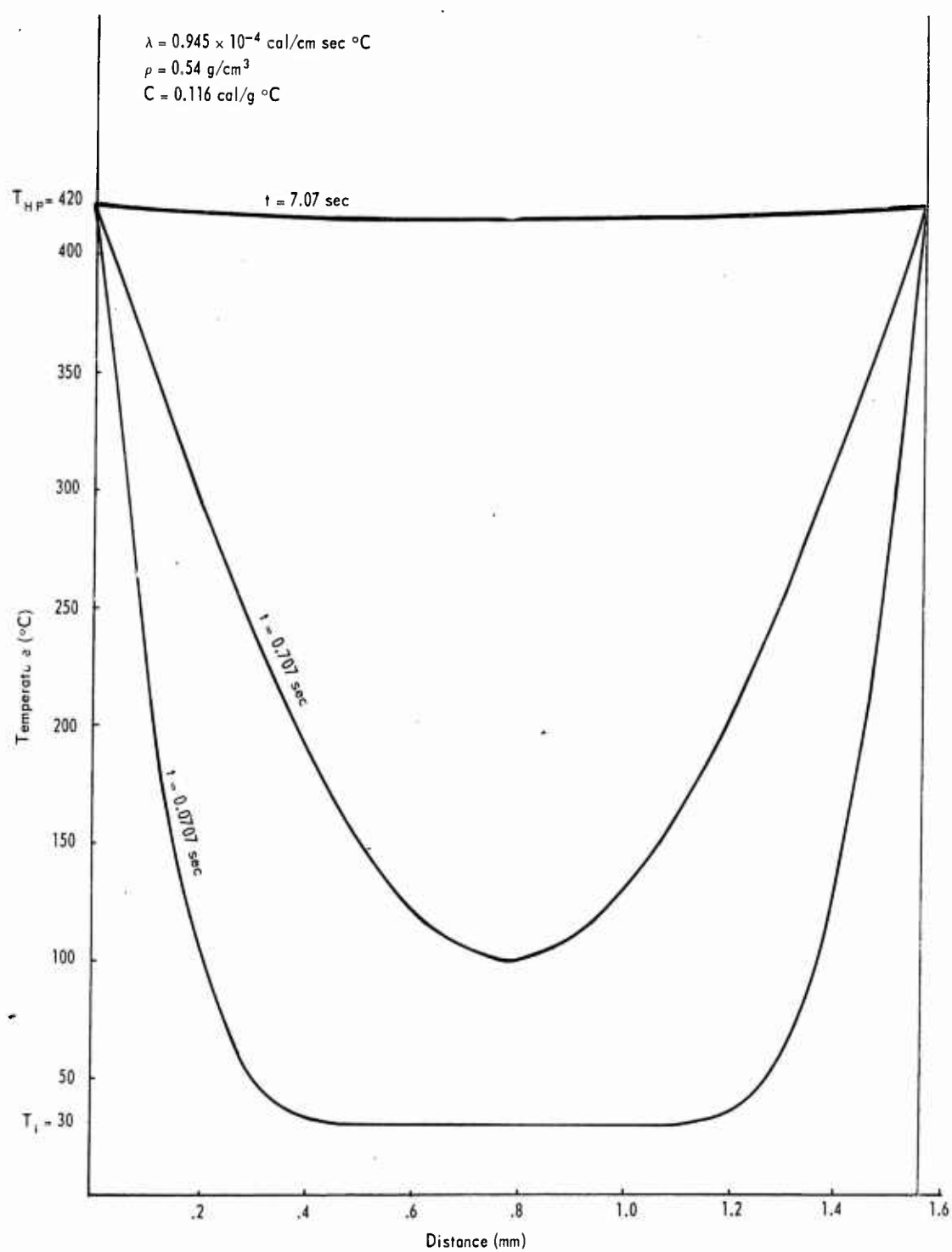$T_i = 30$

Temperature (°C)

Distance (mm)

Fig 17   Heat-up of lead azide if inert (infinite slab 1.56 mm thick). Experimentally observed explosion time at 420°C in N_2 is 0.0707 sec
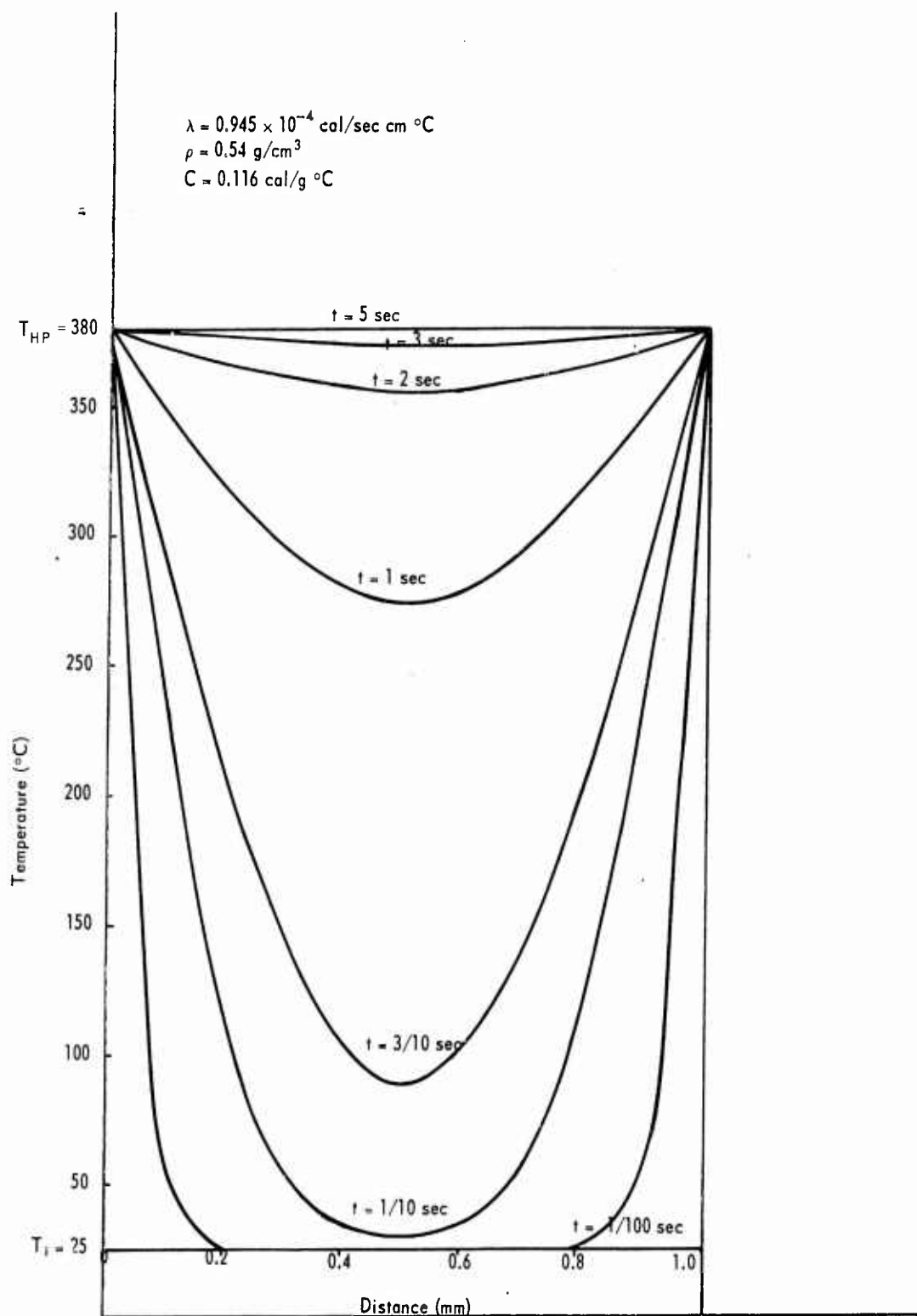
Fig 18 Heat-up of lead azide if inert (infinite slab 1.15 mm thick). Experimentally observed explosion time at 380°C in N_2 is 1.6 sec
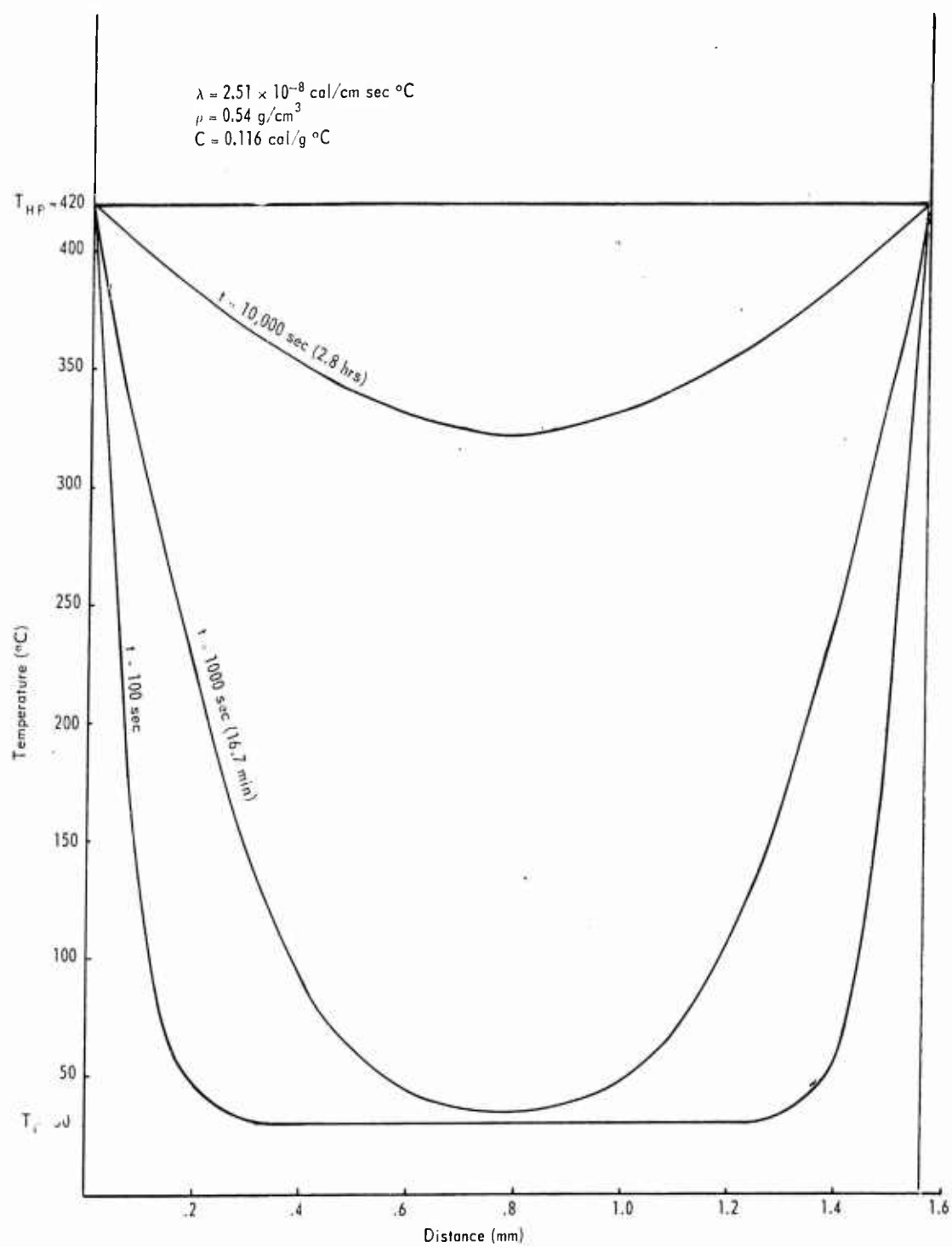
Fig 19   Heat-up of lead azide if inert (infinite slab 1.56 mm thick). Experimentally observed explosion time at 120 C in 0.1μ vacuum is 1 sec

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13. ABSTRACT

The most important knowledge required in the calculation of time to explosion for an explosive subjected to an elevated temperature is the kinetics of thermal decomposition of the explosive. This study of models used in computing times to explosion showed that activation energy, frequency factor, type of kinetic expression, and their interrelationships transcend in importance all other physical and chemical properties of the explosive.

Two mathematical models which describe heat conduction in an explosive accompanied by simultaneous internal generation of heat from thermal decomposition of the explosive were studied. The boundary and initial conditions for one model were such that an infinite slab of explosive of finite thickness, initially at a constant temperature, was suddenly exposed to a constant temperature on its lower surface while its upper surface was perfectly insulated. Zinn and Mader's correlation of the solutions to this problem (Ref 8) was used in the study of this model. The other model was that of a semi-infinite solid initially at a constant temperature which suddenly has its surface exposed to an elevated constant temperature. G. B. Cook's correlation of the solutions to this model (Ref 2) was studied. Both models used zero-order kinetics to describe predetonation decomposition.

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Time to explosion						
Lead azide						
Activation energy						
Frequency factor						
Zero-order kinetics						
Zinn and Mader correlation						
Cook's correlation						
RDX						
Temperature of explosion						

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