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SRI Technical Report No. 20
Supplement to SRI Technical Report No. 8

THERMAL STABILITY OF A REACTIVE SPHERICAL SHELL

Special Report

by

W. A. ROSSER, JR.
Y. RAJAPAKSE

JUNE 1967

DEPARTMENT OF THE ARMY
EDGEWOOD ARSENAL
Research Laboratories
Physical Research Laboratory
Edgewood Arsenal, Maryland 21010

Contract DA-18-035-AMC-122(A)

STANFORD RESEARCH INSTITUTE
Menlo Park, California

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Task 1B522301A08101
SRI Project PAU-4900

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FOREWORD

The work described in this report was authorized under Task 1B522301A08101, Dissemination Investigations of Liquid and Solid Agents (U). The work was started in July 1966 and completed in January 1967.

DIGEST

The conditions required for the spontaneous ignition of a volatile fuel drop suddenly exposed to a hot oxidizing environment have been investigated theoretically using a simplified model of the physical situation. Analysis of the model, a reactive spherical shell with unequal boundary temperatures, defined the conditions consistent with thermal stability of the shell. Two types of reaction rate were used, one with a simple Arrhenius form, and a second explicitly involving radial distance as well as a temperature dependence in Arrhenius form.

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1 INTRODUCTION

Many physical and chemical processes are involved in the ignition of a volatile fuel droplet suddenly exposed to a hot oxidizing environment. A complete mathematical description of these processes and their interactions can be formulated but is so complex that even computer solutions are impractical. Consequently, simplification is desirable. A simplifying approach to the problem is suggested by an experimental study of droplet ignition.[7] That study revealed that ignition only occurs if the initial droplet diameter, d_i, exceeds a critical diameter, d_{cr}, dependent on the nature of the fuel and on the oxidizer temperature. In the case of hexadecane droplets, d_{cr} decreases with increasing oxidizer temperature but is insensitive to oxidizer concentration for concentrations greater than the minimum required for flammability. These observations, in particular the existence of a critical initial diameter, suggest a similarity between drop ignition and smmpler steady-state problems involving attainment of a critical state in which heat released by chemical reaction cannot be dissipated to the environment.

Analyses of the thermal stability of reactive materials in the form of slabs,[2] cylinders, and spheres[3] involved a criticality parameter δ related to the problem parameters by Eq. 1:

$$\delta = \frac{E}{\lambda R T_0^2} l^2 \dot{q}_0 \qquad (1)$$

where

E = an overall activation energy

R = universal gas constant

T_0 = boundary temperature

$\dot{q}_0$ = rate of heat release per unit volume at T_0 (in Arrhenius form)

l = a characteristic dimension of the sample

λ = sample thermal conductivity.

If δ exceeds a certain critical value, δ_{cr}, dependent on sample geometry, the material will ignite or explode in a time that is related to $\dot{q}_0$. For a slab with boundaries at different temperatures, $\dot{q}_0$ refers to the reaction rate at the hot boundary and δ_{cr} depends on the temperature difference between the two boundaries.[4]

By analogy to the cited cases the critical state for droplet ignition will involve at the least: (1) the surface temperature of the droplet, (2) the ambient temperature, (3) the size of the droplet, and (4) the kinetics of the reaction between evaporated fuel and gaseous oxidizer. Because the rate of reaction will be affected by the concentration gradients associated with the movement of evaporated fuel away from the drop, the evaporation rate will also be involved if only indirectly. A characteristic dimension analogous to the term l in Eq. 1 is not well defined for a process involving diffusion from a finite source into an infinite medium. An artificial outer boundary must be imposed in order to prescribe definite boundary conditions.

This theoretical study is concerned with two simplified models of droplet ignition. Both models involve a steady-state approach (in which time does not appear as a variable) to the determination of the thermal stability of a reactive spherical shell with an outer boundary temperature greater than that of the inner boundary. The first model, the simpler case, involves a reaction rate in Arrhenius form that varies explicitly with temperature but not with position except as temperature varies with position. The second model, a more realistic and more complicated one, includes a reaction rate that varies explicitly with both position and temperature. The form of the dependence on position was chosen to simulate the variation of fuel concentration with distance from an evaporating drop. In both cases, the intention was to determine δ_{cr} as a function of the various parameters which enter the problem.

II DISCUSSION

A. Model I: Reaction Rate, a Function of Temperature

For a steady-state temperature distribution to exist in the situation represented by Fig. 1, the temperature within the shell must satisfy the differential equation

$$\lambda \nabla^2 T = -\dot{q} \tag{2}$$

where λ is the thermal conductivity of the shell material and $\dot{q}$ is the rate of heat release per unit volume within the shell. The prescribed boundary conditions are $T = T_a$ at $r = a$, and $T = T_b$ at $r = b$. If the rate of heat release $\dot{q}$ is assumed to have the Arrhenius form $\dot{q} = Ae^{-E/RT}$, a steady-state temperature distribution is not possible for all values of the parameters a, b, T_a, T_b, A, E, and λ. The task is to determine the range of values that do permit a steady-state temperature distribution.

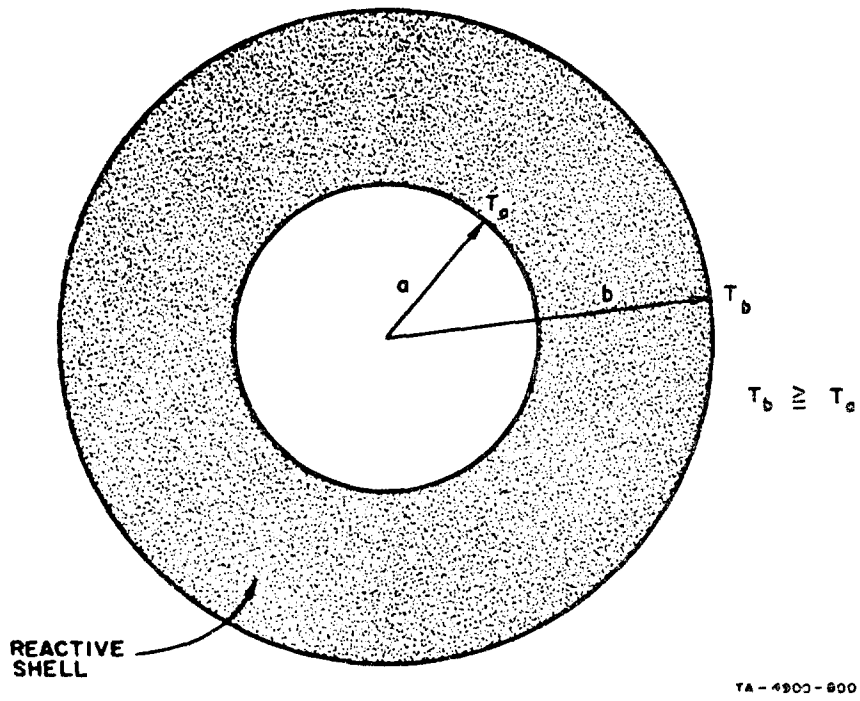

FIG. 1 REACTIVE SPHERICAL SHELL

Some mathematical difficulties associated with integration of Eq. 2. are a consequence of the Arrhenius form of the reaction rate $\dot{q}$. Although simplification is not necessary, the approximation

$$-\frac{E}{RT} \cong -\frac{E}{RT_b}\left(1-\frac{(T-T_b)}{T_b}\right) \tag{3}$$

usually involves only a small error,[5] simplifies the problem, and permits comparison with other studies.[2-5] In terms of the dimensionless variables

$$\theta = \frac{E}{RT_b^2}(T-T_b) \tag{4}$$

$$y = 1-\frac{r}{b} = 1-z \tag{5}$$

Eq. 2 takes the form

$$\frac{d^2\theta}{dy^2} - \frac{2}{1-y}\frac{d\theta}{dy} = -\delta e^{\theta} \tag{6}$$

with the boundary conditions

$$\theta = 0 \quad \text{at} \quad y = 0 \tag{7}$$

$$\theta = \theta_0 \leq 0 \quad \text{at} \quad y = y_0 = 1-z_0 \tag{8}$$

where $z_0 = a/b$, $0_0 = E(T_a - T_b)/RT_b^2$, and

$$\delta = \frac{Eb^2}{\lambda RT_b^2} Ae^{-E/RT_b} \tag{9}$$

Solutions of the system of Eqs. (6)-(8) are only possible for values of δ less than a critical value δ_{cr} that depends on the boundary conditions. Because analytic solution of Eq. 6 is not possible, specific values of δ_{cr} must be obtained by numerical methods. These values of δ_{cr} define the possible ranges of variation of the parameters a, b, T_a, T_b, A, E, and λ.

To determine the maximum value of δ, $\delta_{cr} = \delta_c(z_0, \theta_0)$ which permits the solution of differential Eq. (6), subject to the boundary conditions (7) and (8), the following method was adopted:

Starting with a definite value of δ, Eq. (6) was integrated subject to the boundary conditions

$$\theta = 0 \quad \text{and} \quad \frac{d\theta}{dy} = m \quad \text{at} \quad y = 0 \tag{10}$$

where m is a parameter. The solution $\theta = \theta(y, \delta, m)$ is a function of the variable y and the parameters δ and m. The value of θ at $y = y_0$ is $\theta(y_0, \delta, m)$. As m varies, this quantity has a maximum value $\bar{\theta} = \bar{\theta}(y_0, \delta)$, *i.e.*,

$$\bar{\theta} = \bar{\theta}(y_0, \delta) = \max_{-\infty < m < \infty} \theta(y_0, \delta, m) \quad . \tag{11}$$

If $\bar{\theta}$ is less than θ_0, it is evident that Eq. (6), subject to the boundary conditions (7) and (8), has no solution for the particular value of δ chosen, *i.e.*, $\delta < \delta_{cr}(\theta_0, z_0)$. If θ is greater than θ_0, there are two solutions of the system of Eqs. (6)-(8) and $\delta < \delta_{cr}$. Finally, if θ is equal to θ_0, there is only one solution to the problem and the value of δ is the critical value δ_{cr} corresponding to given values of θ_0 and y_0 (or z_0).

As noted, the numerical procedure provides two solutions to Eq. 6 if $\delta < \delta_{cr}$. Other studies[6,7] have revealed multiple solutions in cases involving spherical symmetry. In particular, a large number of solutions (not necessarily stable) are possible for some values of δ. We are concerned in this study with values of δ in the vicinity of δ_{cr} for which there are probably at most two solutions. Of these two solutions the one corresponding to the smaller value of m is probably the significant one, for in a physical problem the stable temperature distribution would be approached from below and the eventual steady-state distribution would be that corresponding to the lowest value of m.

A computer program based on the method described was carried out to determine δ_{cr} as a function of z_0 and θ_0. The results are shown in Table I. The range of values chosen for θ_0 (0 to -14) corresponds to the range of values for T_a, T_b, and E that might be encountered in an experimental study of drop ignition. Graphical interpolation may be used to obtain other

Table I

CRITICAL VALUES OF δ: MODEL I

	$\theta_0 = 0$	$\theta_0 = -2.0$	$\theta_0 = -4.0$	$\theta_0 = -6.0$	$\theta_0 = -8.0$	$\theta_0 = -10.0$	$\theta_0 = -14.0$
$z_0 = 0.05$	3.74	4.21	4.69	5.16	5.64	6.13	7.13
$z_0 = 0.125$	4.47	5.72	7.04	8.43	9.90	11.45	14.79
$z_0 = 0.25$	6.16	9.21	12.75	16.75	21.21	26.12	37.30
$z_0 = 0.5$	14.00	26.01	42.30	62.75	87.31	115.92	185.25
$z_0 = 0.75$	56.18	121.78	222.66	360.04	534.00	744.20	1273.2
$z_0 = 0.90$	351.34	820.45	1591.2	2685.5	4107.1	5855.3	10328.5

values of δ_{cr} for θ_0 from 0 to -14 and for z_0 from 0.05 to 0.90. Extrapolation of the tabulated results to values of z_0 in the range $0 \leq z_0 \leq 0.05$ is also readily accomplished. As suggested by the data in Tables I and II, for all θ_0, as $z_0 \to 0$, δ_{cr} appears to approach 3.32, the value for a solid sphere.[3] The convergence was not investigated analytically. Extrapolation of the data to values of $z_0 > 0.9$ is not feasible. However, for $1 > z_0 > 0.9$, the reactive shell of Fig. 1 may be approximated by a thin slab and the solution to the thin slab problem[4] used to estimate values of δ_{cr} for the shell. To utilize the slab solution it must be noted that δ for a slab is defined in terms of the characteristic dimension $(b - a)/2$.

Table II

CRITICAL VALUES OF δ: MODEL I

	$\theta_0 = 0$	$\theta_0 = -1$	$\theta_0 = -2$
$z_0 = 0.001$	3.330	3.335	3.340
$z_0 = 0.01$	3.403	--	3.500
$z_0 = 0.02$	3.485	--	3.677

B. Model I: Drop Ignition

In order to apply Model I criticality data to drop ignition it is necessary to consider the variation of a, the drop radius, and of b, the radius of the effective outer boundary, with time. The surface temperature of a drop suddenly exposed to a hot oxidizing atmosphere will quickly approach a value T_a corresponding to a balance between heat transfer inward and diffusion of evaporated fuel outward. Thereafter, the surface temperature T_a will remain nearly constant as evaporation proceeds and the radius of the drop decreases. Initially, the radius b of the effective outer boundary equals a and then increases as a decreases and evaporated fuel diffuses into the surrounding atmosphere. The outer radius b, although poorly defined, must eventually approach a maximum value corresponding to complete evaporation. As shown schematically in Fig. 2, during this time, δ will approach a maximum value as a approaches zero. If this maximum

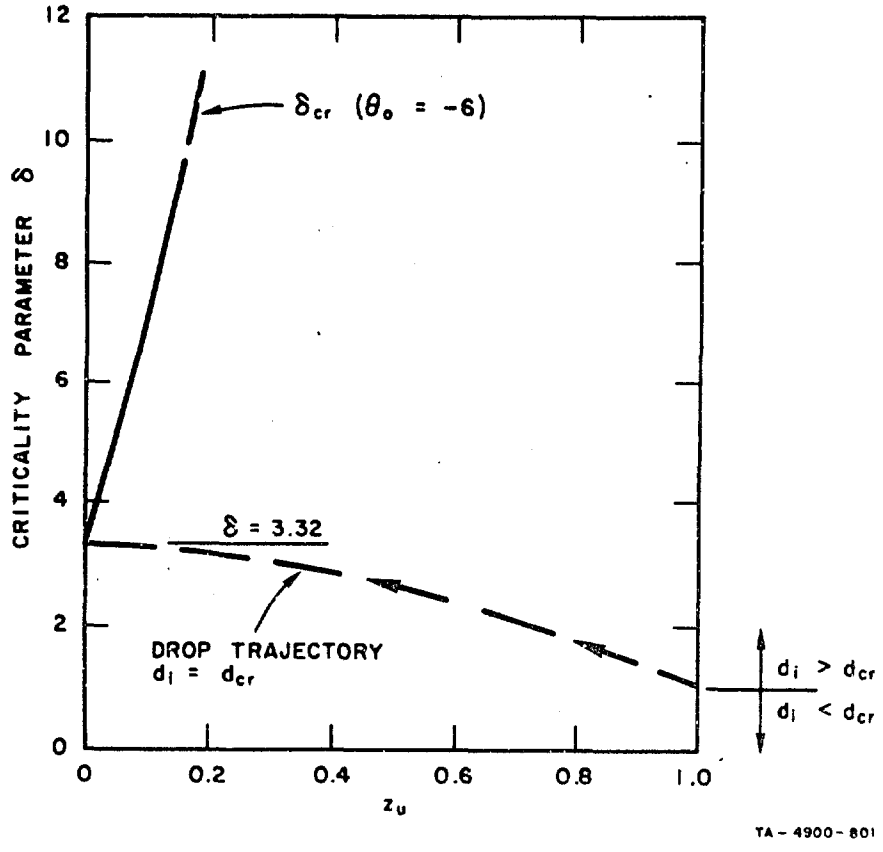

FIG. 2 DROP TRAJECTORY IN $\delta - Z_o$ PLANE: MODEL I

value exceeds 3.32, the apparent limit of δ_{cr} as $z_0 \to 0$ for all θ_0, the drop can ignite. For fixed T_b, the above qualitative description implies the existence of a d_{cr} such that a drop with an initial diameter $d_i < d_{cr}$ will not ignite.

As noted, all δ_{cr} as a function of θ_0 appear to approach the value of $\delta_0 = 3.32$ as $z_0 \to 0$. If the final value of b corresponding to complete evaporation of a drop is proportional to the initial drop diameter d_i, then the definition of δ implies that d_{cr} will decrease as T_b increases with an apparent activation energy that can be deduced from the definition of δ (Eq. 9). That definition results in the proportionality:

$$d_{cr}^2 \propto \delta_0 \frac{\lambda R T_b^2}{E} \frac{1}{A} e^{E/RT_b} \tag{12}$$

The pre-exponential term A is not strictly independent of temperature because it includes an average reactant concentration that varies with T_a. If $T_b >> T_a$, the dependence of A on T_b is slight and can be ignored with the result that

$$R \frac{d \ ln \ d_{cr}}{d(1/T_b)} = Q \approx \frac{E}{2} - R\bar{T}_b \tag{13}$$

where $\bar{T}_b$ is a mean oxidizer temperature in the temperature range of interest and Q is an apparent activation energy. The measured value of Q for ignition of hexadecane drops in hot air is 7 kcal/mole,[7] corresponding to a value for E of 15 kcal/mole.

C. Model II: Reaction Rate, a Function of Position and Temperature

Analysis of Model I revealed the dependence of δ_{cr} on the dimensionless temperature difference between the hot and cold boundaries of the reactive spherical shell. The study also revealed that the δ_{cr} of importance in determining the drop critical initial size was that corresponding to a solid sphere, $\delta_{cr} = 3.32$. The model, however, is unreal because it does not include the effect of concentration gradients associated with the diffusion of fuel away from an evaporating drop. If the reaction rate $\dot{q}$ varies explicitly with fuel concentration C_f, with oxidizer concentration C_{0x}, as well as with temperature T, the concentration gradients will affect the conditions required for criticality.

The effect of variable reactant concentration can be simulated by including in the model a rate of heat release given by

$$\dot{q} = AC_f C_{0x} e^{-E/RT} \quad . \tag{14}$$

In the physical situations of interest the dimensionless boundary temperature $\theta_0 << 0$. By analogy to the slab problem,[4] for $\theta_0 << 0$, the maximum reaction rate occurs in the vicinity of the hot boundary. In that region the concentration of oxidizer is approximately equal to the value in the ambient atmosphere and can be absorbed into the pre-exponential term A, giving

$$\dot{q} = A^1 C_f e^{-E/RT} \tag{15}$$

where

$$A^1 = AC_{0x} \quad .$$

The reaction rate in Model II is assumed to be of the form given in Eq. (15). The boundary conditions are $T = T_a$ at $r = a$ and $T = T_b$ at $r = b$. The element that differentiates Model II from Model I is the dependence of the reaction rate on position through the fuel concentration C_f. Because the variation of C_f with position is intended to simulate the results of diffusion, the concentration C_f is required to satisfy the equation $\nabla^2 C_f = 0$ with the boundary conditions $C_f = C_0$ at $r = a$ and $C_f = 0$ at $r = b$. That is,

$$\frac{C_f}{C_0} = \frac{\left(\frac{1}{r} - \frac{1}{b}\right)}{\left(\frac{1}{a} - \frac{1}{b}\right)} \quad . \tag{16}$$

In terms of the variable y, Eq. 16 becomes

$$\frac{C_f}{C_0} = \frac{y(1 - y_0)}{y \cdot (1 - y)} \tag{17}$$

where $y_0 = 1 - (a/b) = 1 - z_0$. Now let

$$\delta = \frac{Eb^2}{\lambda RT_b^2} A^1 C_0 e^{-E/RT_b} \quad . \tag{18}$$

Then Eq. 2 becomes

$$\frac{d^2\theta}{dy^2} - \frac{2}{1-y}\frac{d\theta}{dy} = -\delta e^{\theta}\frac{y(1-y_0)}{y_0(1-y)} \qquad (19)$$

with boundary conditions

$$\theta = 0 \quad \text{at} \quad y = 0$$

$$\theta = \theta_0 \leq 0 \quad \text{at} \quad y = y_0 \qquad (20)$$

As before, the problem is to determine as a function of θ_0 and z_0 the maximum value of δ, δ_{cr}, that permits solution of Eq. 19. The range of variation of θ_0 and z_0 is the same as in the analysis of Model I.

Determination of specific values of δ_{cr} was done in the same manner as in the analysis of Model I with the results shown in Table III.. Comparison of the data in Tables I and III reveals that the inclusion of a concentration gradient has resulted in: (1) a marked increase in δ_{cr} for fixed θ_0 and z_0, (2) a minimum in δ_{cr} as a function of z_0 for fixed θ_0, and (3) a rapid increase in δ_{cr} as z_0 decreases towards zero from the value corresponding to the minimum in δ_{cr}. The cited minimum value is quite evident for small values of θ_0. However, it occurs at a value of z_0 that decreases with decreasing θ_0, and therefore it is not apparent in Table III for large $|\theta_0|$. The rapid increase in δ_{cr} with decreasing z_0 for z_0 less than the value corresponding to the minimum in δ_{cr} was not investigated in detail.

Table III

CRITICAL VALUES OF δ: Model II

	$\theta_0 = 0$	$\theta_0 = -2$	$\theta_0 = -4$	$\theta_0 = -6$	$\theta_0 = -8$	$\theta_0 = -10$	$\theta_0 = -14$
$z_0 = 0.05$	50.48	66.29	83.01	100.80	119.76	139.96	184.32
$z_0 = 0.125$	29.09	46.06	66.83	91.52	120.33	153.46	233.65
$z_0 = 0.25$	24.73	47.27	79.77	123.40	179.35	248.87	433.60
$z_0 = 0.50$	37.35	89.26	181.88	327.61	538.19	825.10	1673.72
$z_0 = 0.75$	122.62	337.63	781.56	1562.96	2788.89	4563.9	10174.7
$z_0 = 0.90$	707.39	2078.43	5114.69	10764.0	19992.4	33746.3	78559.5

D. Model II: Drop Ignition

The existence of a minimum δ_{cr} for fixed θ_0 affects the initial droplet size required for ignition. A drop trajectory in the δ-z_0 plane must intercept the δ_{cr}-z_0 curve for ignition to occur. As shown in Fig. 3, the critical initial droplet size is that size corresponding to the trajectory tangent to the δ_{cr}-z_0 curve. Because, in practical cases, $\theta_0 << 0$, the point of tangency will occur at values of z only slightly greater than zero. Unlike the situation represented by Model I, the δ_{cr} of interest is greater than 3.32 and increases with decreasing θ_0.

Because the δ_{cr} that determines drop critical size varies with θ_0 and occurs at values of $z_0 > 0$, deduction of the temperature variation of d_{cr} is more difficult than it was in the case of Model I. However, in the cases of interest, $\theta_0 << 0$ and the critical droplet trajectory is tangent to the criticality curve at a value of z_0 very close to zero. Consequently, it may be assumed that the value of b at the point of tangency is proportional to d_{cr}. The definition of δ, Eq. 9, then leads to

$$d_{cr} \propto \left(\frac{\lambda R}{EA^1 C_0}\right)^{1/2} \delta_T^{1/2}\, T_b e^{E/2RT_b} \tag{21}$$

where δ_T, the value of δ_{cr} at the point of tangency, will be very close to the minimum value of δ_{cr} for a given θ_0. If T_a, C_{0x}, and C_0 are held constant, then the apparent activation energy Q is given by

$$R\frac{d\ ln\ d_{cr}}{d\left(\frac{1}{T_b}\right)} = Q = \frac{E}{2} - R\bar{T}_b + \frac{R}{2}\frac{d\ ln\ \delta_T}{d\left(\frac{1}{T_b}\right)} \tag{22}$$

a relation differing from Eq. 13 in the third term on the right. That term can be re-expressed in the form

$$\frac{R}{2}\frac{d\ ln\ \delta_T}{d\left(\frac{1}{T_b}\right)} = \frac{R}{2}\frac{d\ ln\ \delta_T}{d\theta_0}\frac{d\theta_0}{d\left(\frac{1}{T_b}\right)} \tag{23}$$

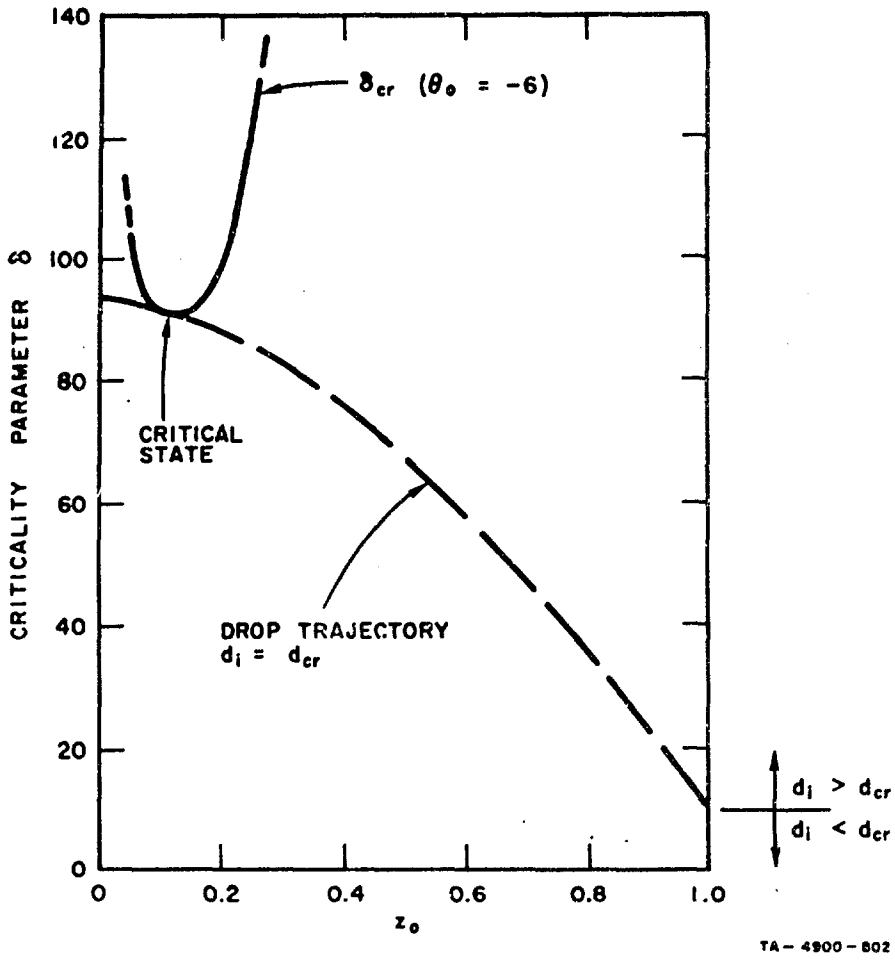

FIG. 3 DROP TRAJECTORY IN $\delta - Z_o$ PLANE: MODEL II, $\theta_o = -6$

The derivative of θ_0, with T_a constant, gives

$$\frac{d\theta_0}{d\left(\frac{1}{T_b}\right)} = \frac{E}{R}\left(2\frac{T_a}{T_b} - 1\right) \tag{24}$$

and may be either positive or negative depending on the specific values of T_a and T_b. From the data in Table III for values of θ_0 that show a minimum in the δ_{cr}-z_0 curve a value for the term $d\ ln\ \delta_T/d\theta_0$ can be estimated as about 5×10^{-2}. Consequently, the contribution of the third right-hand term in Eq. 22 to the overall activation energy is approximately

$$\frac{E}{2}\left(2\frac{T_a}{T_b} - 1\right) \times 5 \times 10^{-2} = Q' \tag{25}$$

In many cases of interest, $(T_b - T_a)/T_b \approx 1$ and

$$Q^1 \approx -5 \times 10^{-2} \frac{E}{2} \tag{26}$$

In such cases Q' makes only a small contribution to Q and can be ignored. In other cases, the absolute value of $|2(T_a/T_b) - 1|$ may be substantially less than one and the contribution to Q will be even less important. The result is that both Models I and II lead to a value

$$Q \approx \frac{E}{2} - R\bar{T}_b \quad . \tag{27}$$

III SUMMARY

The results of the analyses of both Models I and II, when applied to drop ignition, predict the existence of a critical initial diameter, d_{cr}, such that a drop with an initial diameter $d_i > d_{cr}$ will ignite prior to complete evaporation, whereas a drop with $d_i < d_{cr}$ will merely evaporate. For equivalent conditions, d_{cr} for Model I will be substantially less than d_{cr} for Model II. Both models predict that d_{cr} will decrease with increasing T_b with an overall activation energy Q approximately equal to

$$Q \approx \frac{E}{2} - \bar{R}T_b \tag{28}$$

Model I, while formally interesting, does not include the concentration gradients that are an inherent part of the physical problem. Consequently, the usefulness of the Model I analysis lies primarily in determination of the dependence of δ_{cr} on θ_0 and Z_0. Model II includes a variation of fuel concentration with distance from the source and therefore more closely resembles the physical situation. The analysis, however, is not general because of the form assumed for the reaction rate. Inclusion of a fuel concentration variation has two effects. The criticality parameter δ, as defined in Eq. 18, now explicitly depends on C_{0_x} and C_0. Because C_{0_x} and C_0 appear in the pre-exponential factor, the effect is minor and similar for both fuel and oxidizer. A variation in either C_{0_x} or C_0 can be nullified by a much smaller variation in T_b. The second effect is more important. The pronounced increase in δ_{cr} resulting from the variation of C_f with distance from the drop is an indirect consequence of using a reaction rate proportional to C_f. A reaction rate proportional to a power of $C_f > 1$ would presumably enhance the effect. Consequently, the reaction order with respect to fuel is probably an important parameter in determining critical drop size. The importance of the reaction order with respect to oxidizer is much less because the maximum reaction rate occurs near the hot boundary where the concentration of oxidizer is nearly the same as that in the ambient atmosphere and is not sensitive to diffusion.

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13. ABSTRACT

The conditions required for the spontaneous ignition of a volatile fuel drop suddenly exposed to a hot oxidizing environment have been investigated theoretically using a simplified model of the physical situation. Analysis of the model, a reactive spherical shell with unequal boundary temperatures, defined the conditions consistent with thermal stability of the shell. Two types of reaction rate were used, one with a simple Arrhenius form, and a second explicitly involving radial distance as well as a temperature dependence in Arrhenius form.

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