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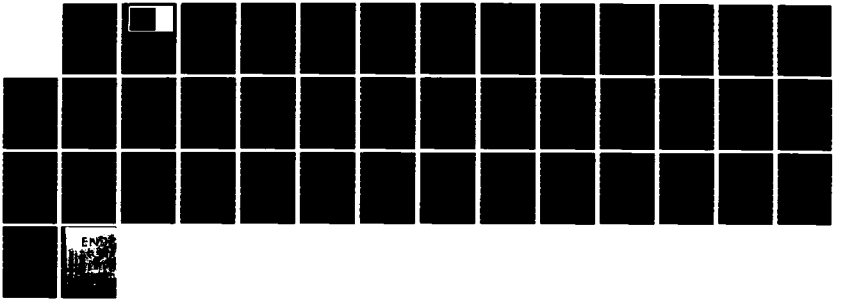

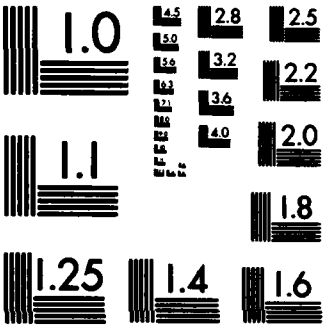

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DROPLET DECOMPOSITION IN A REACTIVE ATMOSPHERE: COMPLETE RESPONSES FOR LARGE ACTIVATION ENERGIES

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DROPLET DECOMPOSITION IN A REACTIVE ATMOSPHERE: COMPLETE RESPONSES FOR LARGE ACTIVATION ENERGIES

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ABSTRACT

A two-step combustion process, consisting of monopropellant (i.e. one-reactant) burning followed by bipropellant (i.e. two-reactant) burning, is considered. The original reactant, namely vapor from a liquid droplet, decomposes into a fuel that combines with oxidant in the surrounding atmosphere. Complete responses $M(D_1,D_2)$ of the evaporation rate M of the droplet to the Damköhler numbers D_1,D_2 of the two reactions are determined in the limit of large activation energies. Conditions under which the response is monotonic or multi-valued (thereby exhibiting auto-ignition and auto-extinction) are identified. Previous conjectures, based on Damköhler-number asymptotics, are found to be mostly correct.

AMS (MOS) Subject Classifications: 80A25, 34E05

Key Words: two-step combustion, monopropellant decomposition, bipropellant burning, burning rate, large activation energies, matched asymptotic expansions.

Work Unit Number 2 - Physical Mathematics

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SIGNIFICANCE AND EXPLANATION

Asymptotic methods are responsible for remarkable advances in combustion theory over the last decade, however they have been largely limited to single-step reactions. This paper uses such methods to investigate an example of multiple-step reactions, which are of greater practical interest.

The following two-step process is considered. Vapor given off by evaporation of a liquid droplet decomposes into a fuel. (This decomposition constitutes the first step of the combustion.) The fuel produced then reacts with the surrounding oxidizing atmosphere in the second step. The burning of a hydrazine droplet in oxygen is an example of such a reaction.

The goal of this investigation is to determine the evaporation rate since it measures the rate of consumption of the reactants, a quantity of primary interest in the applications of combustion theory. This is accomplished by using the method of matched asymptotic expansions based on the physically realistic limit of large activation energies. Evaporation rates which are proportional to the droplet surface area or to its radius are found. In addition, conditions for ignition and extinction are identified.

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DROPLET DECOMPOSITION IN A REACTIVE ATMOSPHERE: COMPLETE RESPONSES FOR LARGE ACTIVATION ENERGIES

H. V. McConnaughey* and G. S. S. Ludford**

1. Introduction

The central question of combustion theory is to determine how fast the reactants are consumed. When there is a single-step reaction this amounts to determining the burning rate M as a function of the so-called Damköhler number D; other parameters may be involved, but they are supposed to be held fixed. For example, consider the bipropellant (i.e. two-reactant) burning of a fuel droplet, in which the liquid in a spherical droplet evaporates at its surface and the vapor is oxidized by the ambient atmosphere in a concentric reaction zone. The Damköhler number depends notably on the rate of chemical reaction, the ambient pressure, and the radius of the droplet; any of these can be used to vary D. The response $M(D)$ is to be found for various values of three additional parameters: the droplet temperature T_s, the ambient temperature T_∞, and the latent heat of evaporation L. The shape of the response curve depends on the (fixed) values of $T_\infty - T_s$ and L.

In general, the analytical determination of $M(D)$ is prevented by the nonlinearity of the governing equations, which persists in the (Arrhenius) reaction term even when simple geometries (such as that of the droplet problem) are adopted. The Arrhenius nonlinearity has been overcome by asymptotic methods, in which the activation energy θ tends to infinity; this is a physically realistic limit since many reactions of interest in combustion do have large activation energies. For the fuel droplet this leads to monotonic, S-shaped, and (for the practically unimportant case $T_\infty < T_s$) C-shaped response

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curves, depending on the values of $T_\infty - T_s$ and L. Similar results are found for the monopropellant (i.e. single-reactant) burning of other droplets, in which the vapor decomposes into products in a concentric reaction zone. (See Buckmaster and Ludford [1, pp. 100 and 127].)

Asymptotic methods are responsible for remarkable advances in combustion theory during the last decade, even though they were largely limited to single-step reactions. A recent account of some of these successes has been given by Buckmaster and Ludford [2]. Nevertheless, the need for a more thorough treatment of multiple-step reactions has been recognized for some time, and isolated efforts are now being replaced by broad action.

The simplest problem of combustion theory is the steady, unbounded plane flame, and it is natural to begin an investigation of multiple-step reactions there. That restricts the discussion to premixed flames, i.e. the combustion of reactants that are already mixed, because plane diffusion flames, in which the reactants are originally separate and must mix by diffusion, only exist when limited in extent. In order to extend the existing discussion to diffusion flames, we have therefore adopted a spherical geometry which, because it is still one-dimensional, introduces a minimum of complications.

The multi-step reaction that we have chosen to examine is a combination of the monopropellant and bipropellant reactions mentioned above that also has practical interest. Vapor from a droplet decomposes at a premixed flame into a fuel (and possibly other products) that is oxidized by the ambient atmosphere at a diffusion flame (Figure 1). The complete response of the burning rate to the two Damköhler numbers D_1, D_2 is determined as the two activation energies θ_1, θ_2 tend to infinity. (An atmosphere hotter than the droplet, i.e. $T_\infty > T_s$, is assumed since the reverse is not of great practical interest.)

A popular experimental study of this type of hybrid combustion investigates the burning of a hydrazine droplet in an oxidizing environment. Variation of the evaporation rate with droplet diameter or with pressure are typically measured (Dykema and Greene [3], Lawver [4], Rosser and Peskin [5], Allison and Faeth [6]). Hybrid droplet combustion is also mentioned by Williams [7, pp. 246-7], who points out the need for theoretical investigation of the process. Our results can hardly be compared quantitatively with such

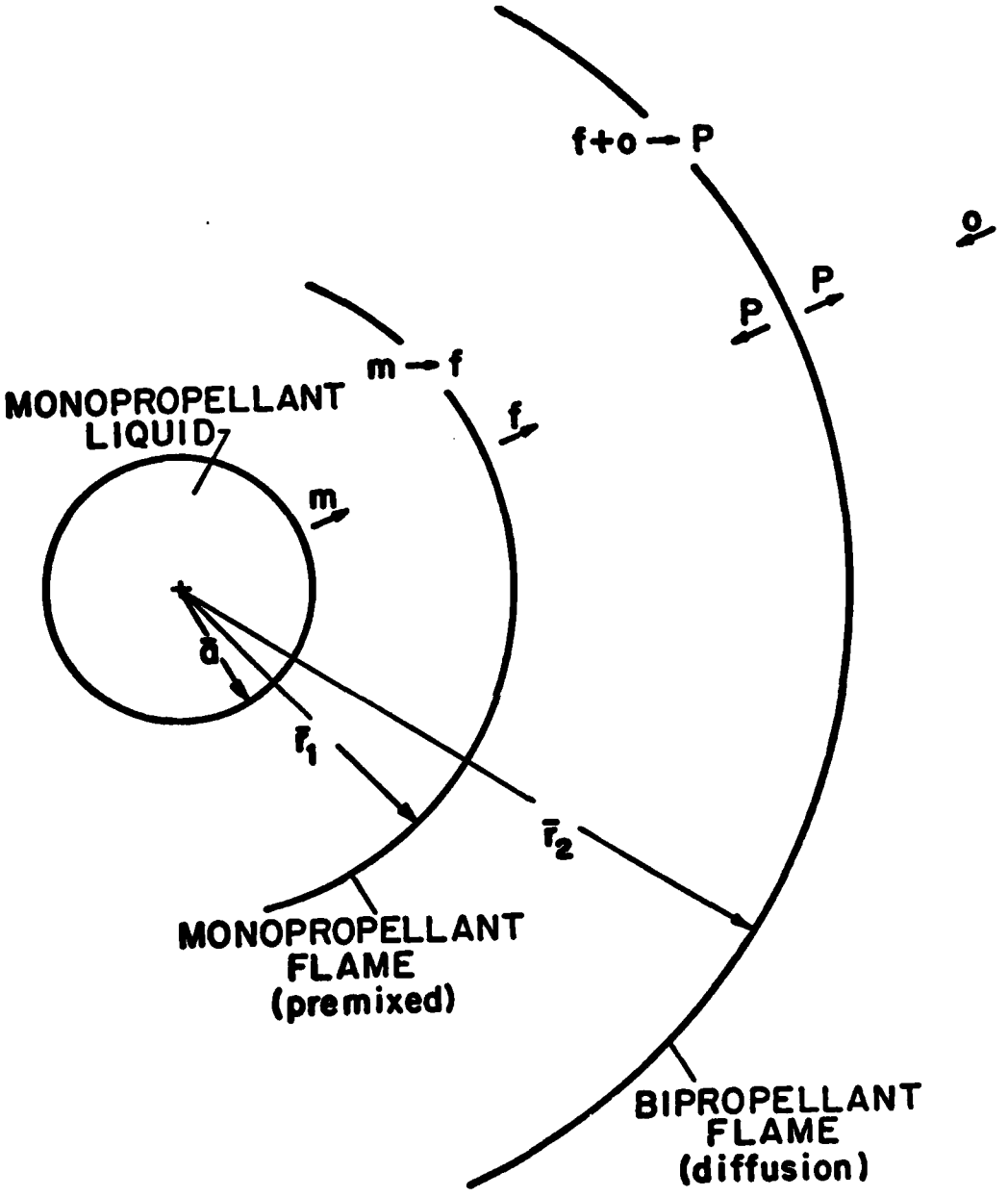

Figure 1. Schematic representation of droplet whose vapor decomposes in a reactive environment. m = monopropellant vapor, f = fuel, o = oxidant, P = products.

experiments, but their qualitative features suggest that more extensive experiments should be made.

Theoretical work on the problem has been done by Fendell [8] and by Buckmaster, Kapila and Ludford [9]. Both these investigations are based on Damköhler-number asymptotics, i.e. large or small D_1 and large or small D_2; hence they can described at most the "corners" of the complete response surface $M(D_1,D_2)$. Fendell identifies the resulting burning-rate formulas, while Buckmaster et al. clarify and add to Fendell's results, suggesting how the valid corner values are connected by the "edges" of the response surface $M(D_1,D_2)$. The latter authors acknowledge the tentativeness of their findings and the need for analysis with finite D_1 and D_2, recognizing that "such analyses can be carried out when the activation energies θ_1 and θ_2 are large, but such a description is quite complicated...".

Details of the complication have been excluded from our account as far as possible. Of the 18 types of asymptotic solutions we describe just 3 typical ones, relegating the supporting analysis (in the main) to impressionistic appendices. However, the tables and figures do give a complete picture of the results and further information can be found in McConnaughey's thesis [10]. Finally, the results are shown to agree with earlier, well established findings, and are compared and contrasted with the conjectures of Buckmaster et al. [9].

2. Formulation

The problem formulation and notation of Buckmaster et al. [9] is adopted. The governing equations in dimensionless form are

$$L(Y_m) = D_1\omega_1,\ L(Y_f) = -D_1\omega_1 + D_2\omega_2,\ L(Y_o) = D_2\omega_2,\ L(T) = -D_1\omega_1 - QD_2\omega_2 \quad , \qquad (2.1)$$

where

$$L \equiv d^2/dr^2 + [(2r-M)/r^2]d/dr,\ \omega_1 = Y_m \exp(-\theta_1/T),\ \omega_2 = Y_f Y_o \exp(-\theta_2/T) \quad ,$$

and the other quantities are defined in the list of symbols. The boundary conditions are

$$r = 1:\ \frac{dY_m}{dr} = M(Y_m - 1),\ \frac{dY_f}{dr} = MY_f,\ \frac{dY_o}{dr} = MY_o,\ \frac{dT}{dr} = ML,\ T = T_s \quad , \qquad (2.2)$$

$$r = \infty; \quad Y_m = 0, \; Y_f = 0, \; Y_o = Y_{o\infty}, \; T = T_\infty > T_s \quad . \tag{2.3}$$

A derivation of this model is sketched by McConnaughey [10], based on a more general development given by Buckmaster and Ludford [1].

Equations (2.1) can be combined and integrated subject to (2.2) and (2.3) to yield the Shvab-Zeldovich relations

$$Y_m + Y_f - Y_o = 1 - (1 + Y_{o\infty})e^{-M/r} \tag{2.4}$$

$$Y_m + QY_o + T = T_s - L + 1 + (T_\infty - T_s + L - 1 + QY_{o\infty})e^{-M/r} \quad , \tag{2.5}$$

which are valid everywhere. Thus only two of equations (2.1) are needed. We select equations (2.1a) and (2.1d) and the corresponding boundary conditions to form a fourth-order system subject to five boundary conditions. The unknown constant M is thereby determined as a function of the various parameters which appear.

The objective of this work is to describe the behavior of $M(D_1, D_2)$ for all values of its arguments, by considering the independent limits $\theta_1 \to \infty$ and $\theta_2 \to \infty$.

3. Asymptotics and Results

In accordance with the usual strategy of activation-energy asymptotics (Buckmaster and Ludford [1]; Kapila and Ludford [11]), the Damköhler numbers are written

$$D_i = D_i' \exp(\theta_i/T_i), \; i = 1 \text{ or } 2 \quad , \tag{3.1}$$

where T_i is a positive parameter and D_i' is at most algebraic in θ_i. The i^{th} reaction term then contains the expression $D_i' \exp(\theta_i/T_i - \theta_i/T)$ and the limit $\theta_i \to \infty$ confines the associated chemical activity to a thin layer either at temperature $T_{i*} = T_i$ with thickness of order T_i^2/θ_i or at temperature $T_{i*} > T_i$ with thickness of order $\exp[c(\theta_i/T_{i*} - \theta_i/T_i)]$, where $c = 1/2$ or $1/3$. The reactions are negligible outside of these flame sheets, hence the majority of the combustion field is described by the linear system

$$L(Y_m) = L(T) = 0 \quad . \tag{3.2}$$

Matched asymptotic expansions are then used to describe the solution inside and outside the flame zones.

3.1. Solution Categories

The solution obtained by this method depends on the assumed flame configuration and flame types. With the location of the decomposition reaction denoted by r_1 and with the bipropellant flame at r_2, it is found that all possible configurations for which $1 < r_1 < r_2 < \infty$ admit a solution. Whether or not $Y_m(r_1)$, $Y_f(r_2)$ and $Y_o(r_2)$ vanish as $\theta_i \to \infty$ determines the nature of the flames, which is often not unique for a given flame arrangement. Hence, for each configuration, it is necessary to consider separately the various types of flames possible, i.e., partial vs. complete decomposition at r_1 and a complete-burning (or so-called Burke-Schumann) diffusion flame vs. a partial burning flame at r_2. When all possible combinations of flame locations and types are investigated, the cases shown in Table 1 are found to admit solutions of interest. (Other conceivable cases, see McConnaughey [10], are omitted; they correspond to special cases whose treatment has been omitted for simplicity.) The first thirteen categories represent separated flames for which $M = 0(1)$; the next four correspond to merged flames with $M = 0(1)$; the last is the only configuration for $M \gg 1$. The asymptotic analysis of these different possibilities is illustrated in the following sections and associated appendices.

3.2. Separated Flames with $M = 0(1)$

Solution category 6, which is illustrated in Figure 2, is considered here. The reactionless system (3.2) is valid for $1 < r < r_1$, $r_1 < r < r_2$ and $r_2 < r < \infty$; the leading terms of the corresponding asymptotic expansions are

$$1 < r < r_1: \quad Y_m = 1 - e^{M/r_1} e^{-M/r}, \quad T = T_s - L + Le^{M} e^{-M/r} , \tag{3.3}$$

$$r_1 < r < r_2: \quad Y_m = 0, \quad T = T_s - L + 1 + (T_\infty - T_s + L - 1 + QY_{o\infty})e^{-M/r} , \tag{3.4}$$

$$r_2 < r < \infty: \quad Y_m = 0, \quad T = T_s - L + 1 + Q + (T_\infty - T_s + L - 1 - Q)e^{-M/r} ,$$

where conditions and relations (2.2) - (2.5) have been invoked as have $Y_m(r_1) = o(1)$, $Y_f(r_2) = o(1)$, $Y_o(r_2) = o(1)$ and continuity to leading order of Y_m, Y_f and Y_o at r_1 and r_2.

Table 1: Description and categorization of flames which admit a solution of interest.

Solution Category	Description
1	$r_1 = 1 < r_2 < \infty$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = o(1)$
2	$r_1 = 1 < r_2 < \infty$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = O(1)$
3	$r_1 = 1 < r_2 < \infty$; $Y_f(r_2) = O(1)$; $Y_o(r_2) = o(1)$
4	$r_1 = 1 < r_2 < \infty$; $Y_f(r_2) = O(1)$; $Y_o(r_2) = O(1)$
5	$r_1 = 1$; $r_2 = \infty$
6	$1 < r_1 < r_2 < \infty$; $Y_m(r_1) = o(1)$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = o(1)$
7	$1 < r_1 < r_2 < \infty$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = O(1)$
8	$1 < r_1 < r_2 < \infty$; $Y_m(r_1) = o(1)$; $Y_f(r_2) = O(1)$; $Y_o(r_2) = o(1)$
9	$1 < r_1 < r_2 < \infty$; $Y_f(r_2) = O(1)$; $Y_o(r_2) = O(1)$
10	$1 < r_1 < r_2 = \infty$; $Y_m(r_1) = o(1)$
11	$1 < r_1 < r_2 < \infty$; $Y_m(r_1) = O(1)$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = o(1)$
12	$1 < r_1 < r_2 < \infty$; $Y_m(r_1) = O(1)$; $Y_f(r_2) = O(1)$; $Y_o(r_2) = o(1)$
13	$1 < r_1 < r_2 = \infty$; $Y_m(r_1) = O(1)$
14	$1 = r_1 = r_2$
15	$1 < r_1 = r_2 < \infty$; $Y_m(r_1) = o(1)$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = O(1)$
16	$1 < r_1 = r_2 < \infty$; $Y_m(r_1) = O(1)$; $Y_f(r_2) = o(1)$; $Y_o(r_2) = O(1)$
17	$r_1 = r_2 = \infty$
18	$M \gg 1$; $r_1 = 1$; $r_2 = \infty$

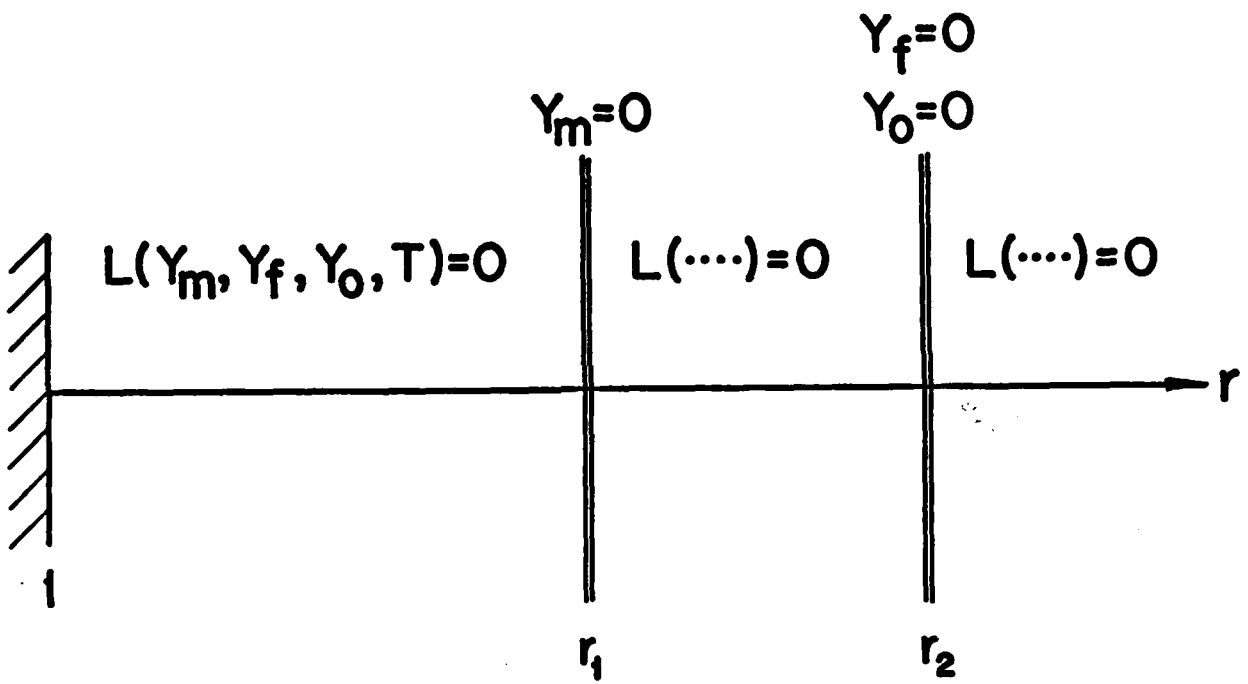

Figure 2. Representation of separated flames. The reaction zones divide the combustion field into three chemically inert regions.

Since $Y_m(r) \neq 0$ for $r < r_1$, the temperature must be less than T_1 in that region, for otherwise the reaction term $D_1^{\prime} Y_m \exp(\theta_1/T_1 - \theta_1/T)$ would not be negligible there. In order for the reaction to become important at r_1 then, we must have $T(r_1) = T_1 > T_s$. Given this and continuity to leading order of T at r_1 and r_2, the solution shows that

$$e^{M/r_1} = \frac{T_\infty - T_s + L - 1 + QY_{O\infty}}{T_1 - T_s + L - 1}, \quad e^{M} = \left(\frac{T_1 - T_s + L}{L}\right) e^{M/r_1}, \quad e^{M/r_2} = 1 + Y_{O\infty} \ .$$

The outer expansions alone have therefore determined M, r_1 and r_2 as functions of various parameters. In order that these results be consistent with the hypothesis $1 < r_1 < r_2$, the following conditions must hold:

$$L > 0; \ T_s - L + 1 < T_1 < T(r_2) \quad \text{or} \quad T(r_2) < T_1 < T_s - L + 1 \ , \tag{3.5}$$

where $T(r_2) = [T_\infty + Y_{O\infty}(T_s - L + 1 + Q)]/(1 + Y_{O\infty}) \equiv T_{BS}$.

It is also necessary to examine the structure of the two flames in order to discover what additional conditions restrict the validity of the above results. Such restrictions amount to existence conditions for solution of the inner problems at r_1 and r_2, where the expansions must match the outer ones. The associated analysis is sketched in Appendix 1 and adds the requirements

$$T_2 < T_{BS}, \ T_1 > T_s - L + \frac{1}{2} \quad \text{when} \quad T_1 > T_{BS} \ .$$

This completes the investigation of category 6. In a manner similar to what just demonstrated, the solution in each of the separated-flame categories 1-13 along with restrictions on its validity are determined. The burning rates are listed in Table 2 and the supporting analysis has been given by McConnaughey [10].

3.3. Merged Flames with $M = O(1)$

The outer problem for merged flames (Figure 3a) is much the same as for separated flames. The analysis of merged-flame structures, on the other hand, is not a straightforward extension of the inner problems associated with separated flames. Both reactions

Table 2: Burning-rate formulas for separated-flame categories in Table 1.

$$M_1 = \ln[(T_\infty - T_s + L - 1 + QY_{O\infty})/(L-1)]$$

$$M_2 = \ln[(T_2 - T_s + L - 1)(T_\infty - T_s + L - 1 - Q)/(L-1)(T_2 - T_s + L - 1 - Q)]$$

$$M_3 = M_1$$

$$M_4 = \ln[(2T_2 - T_\infty - T_s + L - 1)/(L-1)]$$

$$M_5 = \ln[(T_\infty - T_s + L - 1)/(L-1)]$$

$$M_6 = \ln[(T_1 - T_s + L)(T_\infty - T_s + L - 1 + QY_{O\infty})/L(T_1 - T_s + L - 1)]$$

$$M_7 = \ln[(T_1 - T_s + L)(T_2 - T_s + L - 1)(T_\infty - T_s + L - 1 - Q)/L(T_1 - T_s + L - 1)(T_2 - T_s + L - 1 - Q)]$$

$$M_8 = M_6$$

$$M_9 = \ln[(T_1 - T_s + L)(2T_2 - T_\infty - T_s + L - 1)/L(T_1 - T_s + L - 1)]$$

$$M_{10} = \ln[(T_1 - T_s + L)(T_\infty - T_s + L - 1)/L(T_1 - T_s + L - 1)]$$

$$M_{11} = \ln[(2T_1 - T_\infty - T_s + L - QY_{O\infty})/L]$$

$$M_{12} = M_{11}$$

$$M_{13} = \ln[(2T_1 - T_\infty - T_s + L)/L]$$

now occur at the same $O(1)$ location, and the problem becomes analytically intractable, in general, if they are indistinguishable. To circumvent this difficulty, a model of the merged flames is considered in which the two reaction zones are distinct, with thicknesses of disparate orders. The thinner flame is embedded in the broader flame and appears as a discontinuity on the scale of the thicker zone, as shown in Figure 3b. This model was also used by Kapila and Ludford [11]. In the present work, the flames for which the decomposition-reaction zone is the thinner one are referred to as Type A flames; those with a comparatively narrow diffusion flame are called Type B flames.

A third kind of merged flame is also considered. In this Type C flame, both reactions are spread over the same $o(1)$ interval at r_*, but the structure problem becomes tractable by assuming that the right and left side of the fuel equation (2.1b) are of disparate orders. This requires that Y_f be small and that the right side dominate equation (2.1b). The situation thus described is a decomposition reaction in which very little fuel is produced relative to the amount of monopropellant present, and whatever fuel is produced is immediately consumed by the bipropellant reaction.

The kinds of structure problem which result from the models described above are illustrated in Appendices 2 and 3, which consider the structures for solution category 15. The outer solution and structure results for that category are as follows.

Assume that the monopropellant and bipropellant reactions both occur at $r = r_*$, with $1 < r_* < \infty$, and that $Y_m(r_*) = o(1)$, $Y_f(r_*) = o(1)$, $Y_o(r_*) = O(1)$. The $O(1)$ solution of (3.2) which satisfies these assumptions and conditions (2.2) - (2.5) is then

$$1 < r < r_*: \quad Y_m = 1-e^{M/r_*}e^{-M/r}, \quad T = T_s-L+Le^{M}e^{-M/r} \quad , \tag{3.6}$$

$$r_* < r < \infty: \quad Y_m = 0, \quad T = T_s-L+1+Q+(T_\infty-T_s+L-1-Q)e^{-M/r} \quad . \tag{3.7}$$

In order for the decomposition reaction to take place at r_*, the temperature there must be T_1, with $T_1 > T_s$. It follows that

$$e^{M/r_*} = \frac{T_\infty-T_s+L-1-Q}{T_1-T_s+L-1-Q}, \quad e^{M} = \left(\frac{T_1-T_s+L}{L}\right)e^{M/r_*} \quad . \tag{3.8}$$

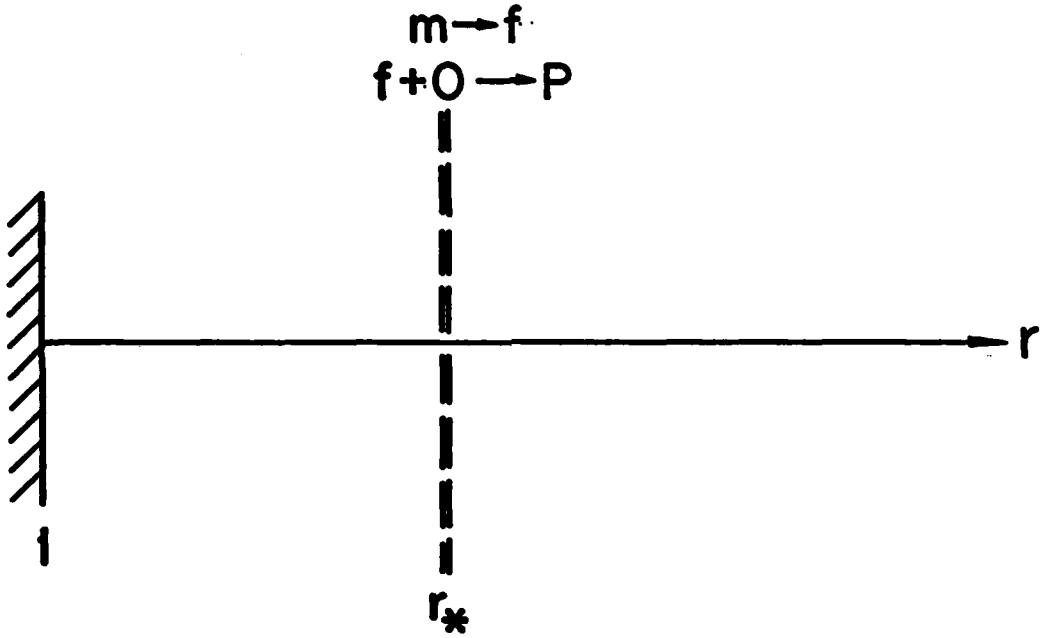

Figure 3a

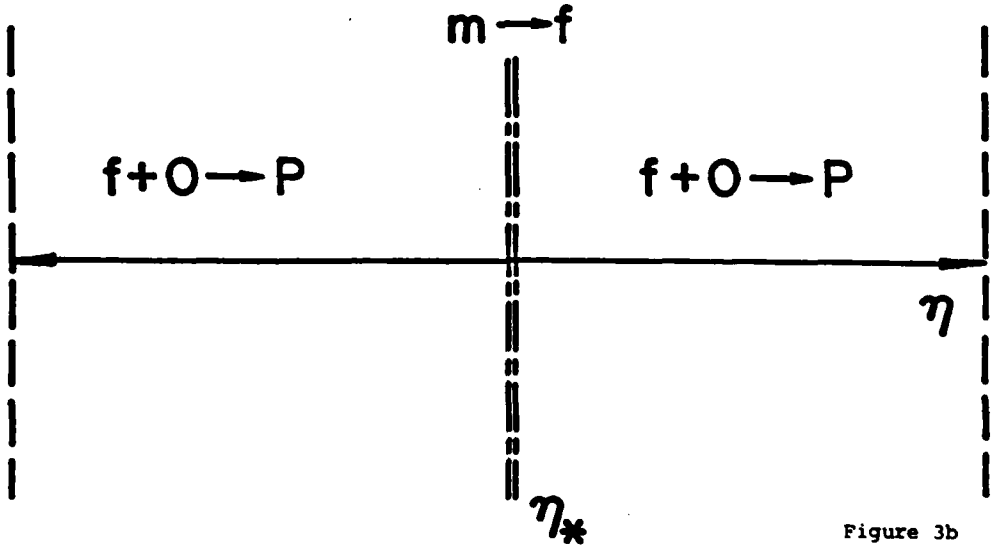

Figure 3b

Figure 3. Representation of merged flames. (a) Appearance on the O(1) scale: a single discontinuity at r_*; the two reaction zones are indistinguishable. (b) Type A structure: the thinner, decomposition flame appears as a discontinuity at η_* on the scale of the bipropellant-reaction zone.

Consistency with the hypothesis and nonnegative mass fractions requires either

$$L > 0 \quad \text{and} \quad \max(T_s, T_s - L + 1 + Q, T_{BS}) < T_1 < T_\infty$$

or

$$L > 0 \quad \text{and} \quad T_\infty < T_1 < \min(T_s - L + 1 + Q, T_{BS}) \quad .$$

Since Y_f can vanish for $r \neq r_s$, the bipropellant reaction can occur at r_s if T_2 does not exceed T_1.

The flame structures must now be analyzed to uncover additional constraints on the validity of the above results. Appendices 2 & 3 treat the case where T_2 is less than T_1, hence only Type A and Type C flames are considered. (Type B flames do not exist for $T_2 < T_1$, as may easily be shown.) It is found that the results (3.6) - (3.8) are valid for

$$L > 0;\ T_2 < T_1;\ \text{and } \max(T_s, T_s - L + 1 + Q, T_{BS}) < T_1 < T_\infty \text{ or}$$
$$\max[T_\infty, T_s - L + (1+Q)/2] < T_1 < \min(T_s - L + 1 + Q, T_{BS}) \quad . \tag{3.9}$$

An approach similar to that just described yields the results shown in Table 3 for merged-flame categories 14-17. Details of the analysis are given by McConnaughey [10].

Table 3: Burning-rate formulas for merged-flame categories in Table 1

$$M_{14} = \ell n[(T_\infty - T_s + L - 1 - Q)/(L - 1 - Q)]$$
$$M_{15} = \ell n[(T_1 - T_s + L)(T_\infty - T_s + L - 1 - Q)/L(T_1 - T_s + L - 1 - Q)]$$
$$M_{16} = M_{13}$$
$$M_{17} = \ell n[(T_\infty - T_s + L)/L]$$

3.4. M >> 1

For $M \gg 1$, relations (2.4) and (2.5) become

$$Y_m + Y_f - Y_o = 1,\ Y_m + QY_o + T = T_s - L + 1 \quad , \tag{3.10}$$

for $r < \infty$ while equations (3.2) imply that Y_m and T are constant outside of the flames, which is incompatible with the boundary conditions (2.2). There must therefore be

a reaction zone at $r = 1$. In addition, equation (3.10a) can only be true if $Y_o(r) = 0$ everywhere, since the sum $Y_m + Y_f + Y_o$ cannot exceed unity and $Y_o > 0$ (by definition of mass fraction). But this violates the condition $Y_o = Y_{o\infty}$ at $r = \infty$, so that there must also be a layer at $r = \infty$. Clearly the remote reaction must be the bipropellant reaction, while the decomposition reaction occurs in the surface layer and entirely consumes the monopropellant there. Hence,

$$Y_m(r) = 0, \quad T(r) = T_s - L + 1 \quad \text{for} \quad 1 < r < \infty \ .$$

The analysis of the reaction zones is given in Appendix 4 and leads to

$$M = M_{1S} = \sqrt{2D_1'}\,\theta_1^{-1}(T_s - L + 1)^2 \exp[\theta_1(T_s - L + 1 - T_1)/2T_1(T_s - L + 1)] \tag{3.11}$$

which is valid for

$$0 < L < 1 \quad \text{and} \quad T_1 < T_s - L + 1 \ . \tag{3.12}$$

4. Discussion of Results

4.1. Description of Results

Considering the limit of large activation energies yields an $O(1)$ burning rate M in solution categories 1-17. The dimensional evaporation rate $\bar{M}$ thus varies with the dimensional droplet radius $\bar{a}$, i.e.

$$\bar{M} = k\bar{a} \ , \tag{4.1}$$

where k is a constant with appropriate units. Solution category 18, on the other hand, gives $M \sim \sqrt{D_1}$ for sufficiently large T_1^{-1} in which case

$$M = k\bar{a}^2 \ . \tag{4.2}$$

Note that this is only possible for $L < 1$, i.e. when the heat of decomposition exceeds that of vaporization, but in practice this is almost invariably the case of interest (Ludford, Yannitell and Buckmaster [12]).

It is well established that (4.1) holds for pure bipropellant burning and for decomposition of small droplets in an inert atmosphere, while (4.2) holds for simple decompositional burning of relatively large drops (Williams [13]). Experimental evidence of the relation between $\bar{M}$ and $\bar{a}$ for the hybrid combustion problem considered in this

work, however, is limited and inconclusive. Investigations of the decompositional burning of a hydrazine droplet in oxygen do suggest, though, that (4.2) is satisfied for all but the smallest droplets, when (4.1) holds (Dykema and Greene [3]; Rosser and Peskin [5]; Allison and Faeth [6]).

An increase in droplet diameter results in an increase in the reaction rates D_1 and D_2. (Recall that it is the variation of M with D_1 and D_2 that is sought here.) Definition (3.1) indicates that the response $M(D_1,D_2)$ may be described qualitatively by the behavior of $M(T_1^{-1},T_2^{-1})$. The latter is deduced from the results reported in (3.11), Table 2 and Table 3 by allowing T_1 and T_2 to vary in the burning-rate formulas while all other parameters are held fixed.

Different responses $M(T_1^{-1},T_2^{-1})$ are found in each region of the parameter plane shown in Figure 4. The domain of the response associated with selected regions is shown in Figures 5. Figures 5a-5c show that $M(T_1^{-1},T_2^{-1})$ is a single-valued function when $T_\infty - T_s$ is positive and greater than $-L+1+Q$. The resulting response surfaces are found to be continuous and monotonically increasing with T_1^{-1} and T_2^{-1}, as illustrated in Figures 6. For any point on these surfaces, the flame temperatures do not exceed the ambient temperature T_∞ and heat is gained from the atmosphere; the decomposition at r_1 is complete and all fuel is consumed at r_2.

When $0 < T_\infty - T_s < -L + 1 + Q$ is satisfied, the function $M(T_1^{-1},T_2^{-1})$ is multiple-valued over part of the domain, as illustrated by Figures 5d-5f. The flame temperatures are no longer bounded above by T_∞ and heat is now lost to the ambient. Decomposition at r_1 is complete in Regions IV-VII; it is only partial in Regions VIII-XIV for certain values of T_1 and T_2. In Regions IV and V, the fuel is entirely oxidized at r_2, while portions of the responses associated with Regions VI-XIV correspond to partial fuel-burning diffusion flames.

The multivaluedness of $M(T_1^{-1},T_2^{-1})$ is manifested by the appearance of folds in the response surface, as seen in Figures 7, and corresponds to the phenomena of auto-ignition and auto-extinction of one or both reactions. The lower turnaround point in a constant-T_1 (or T_2) cross-section of the surface is the ignition point at which the solution jumps

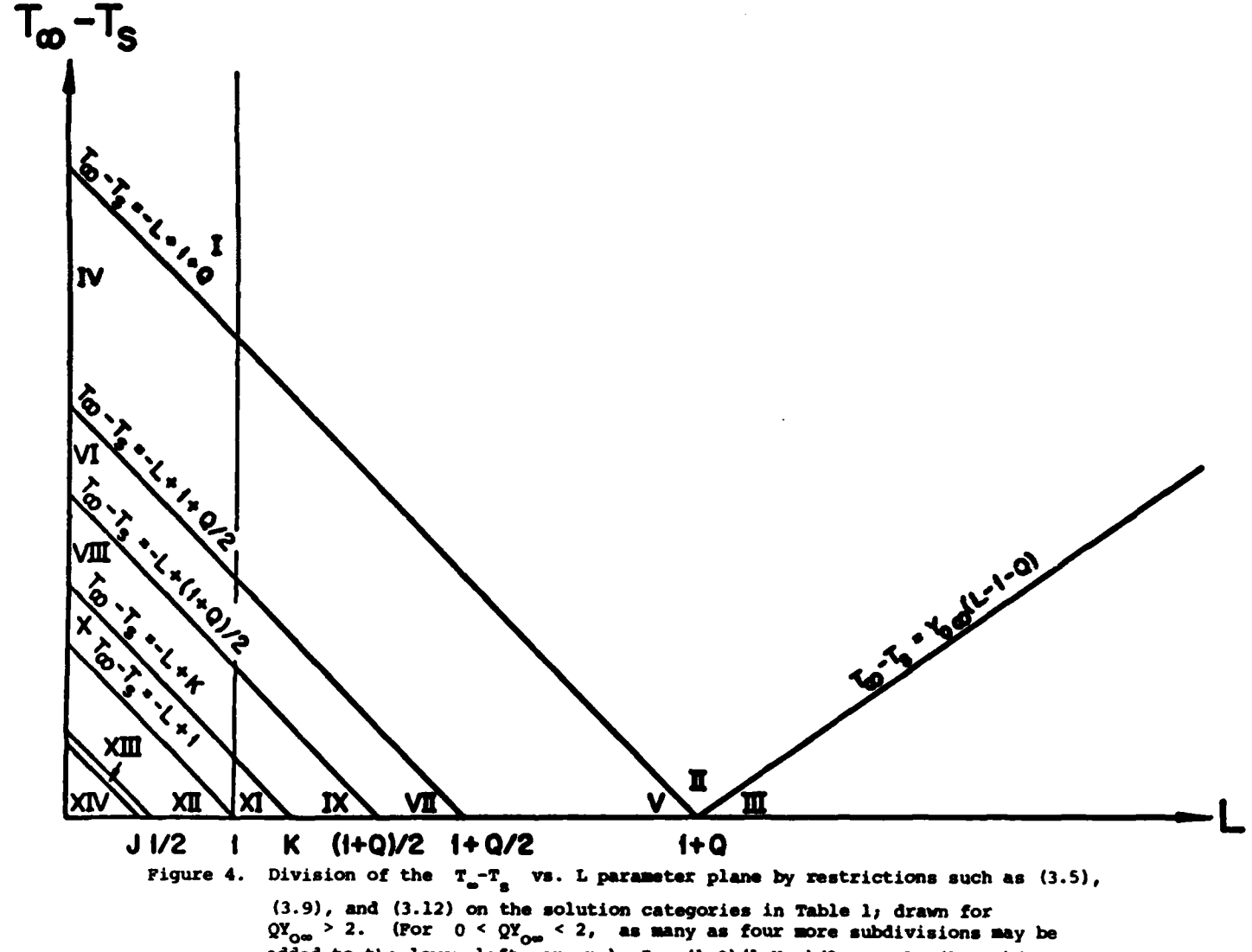

Figure 4. Division of the $T_\infty - T_s$ vs. L parameter plane by restrictions such as (3.5), (3.9), and (3.12) on the solution categories in Table 1; drawn for $QY_{O\infty} > 2$. (For $0 < QY_{O\infty} < 2$, as many as four more subdivisions may be added to the lower left corner.) $J = (1+Q)(1-Y_{O\infty})/2$, $K = 1+Q(1-Y_{O\infty})/2$.

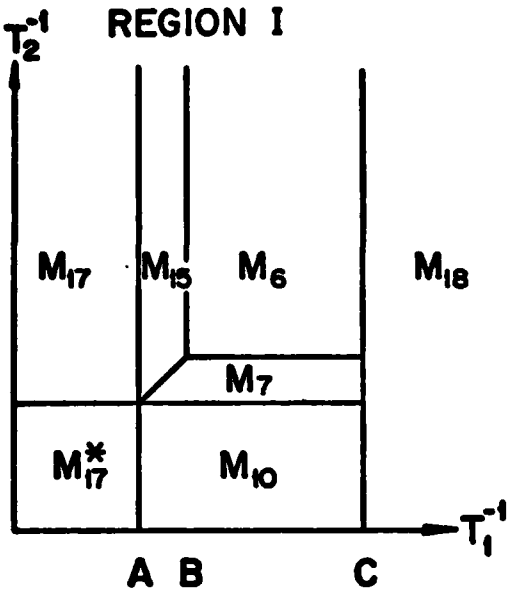

Figure 5a

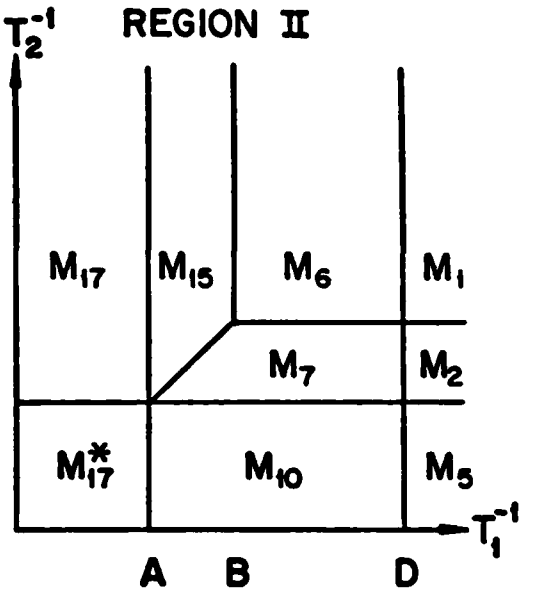

Figure 5b

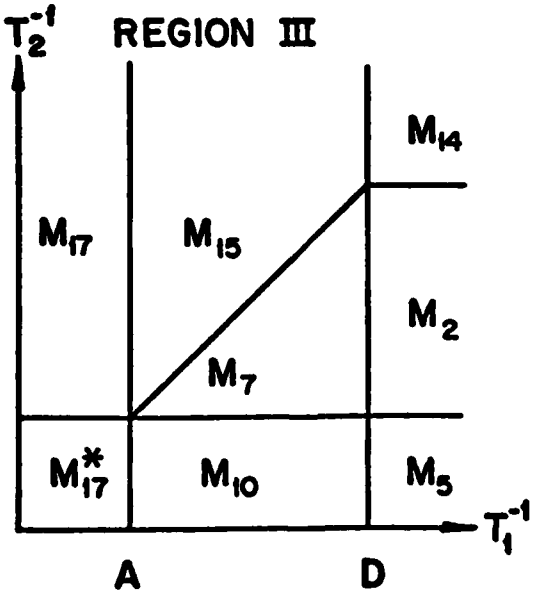

Figure 5c

(See caption on page 19.)

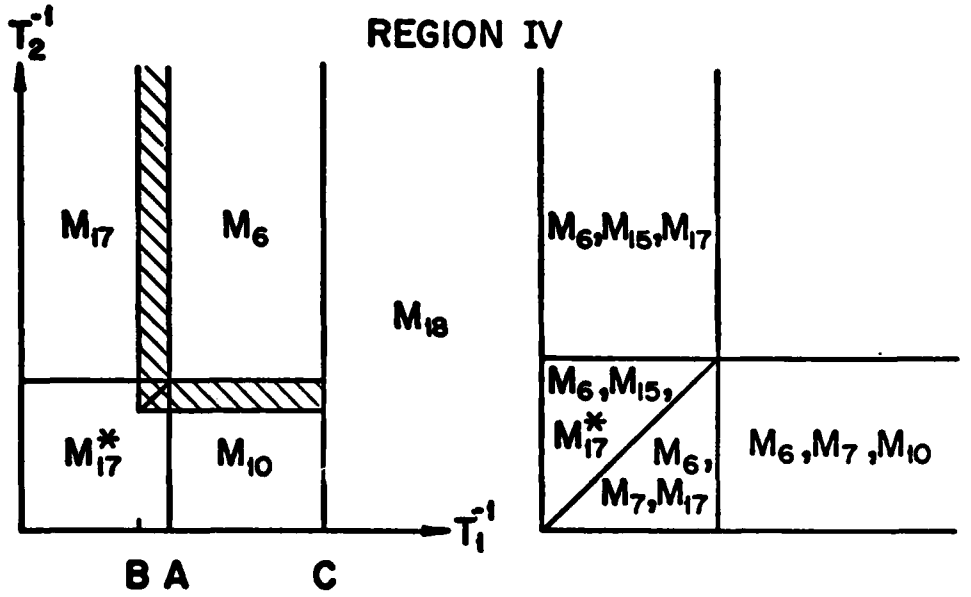

Figure 5d

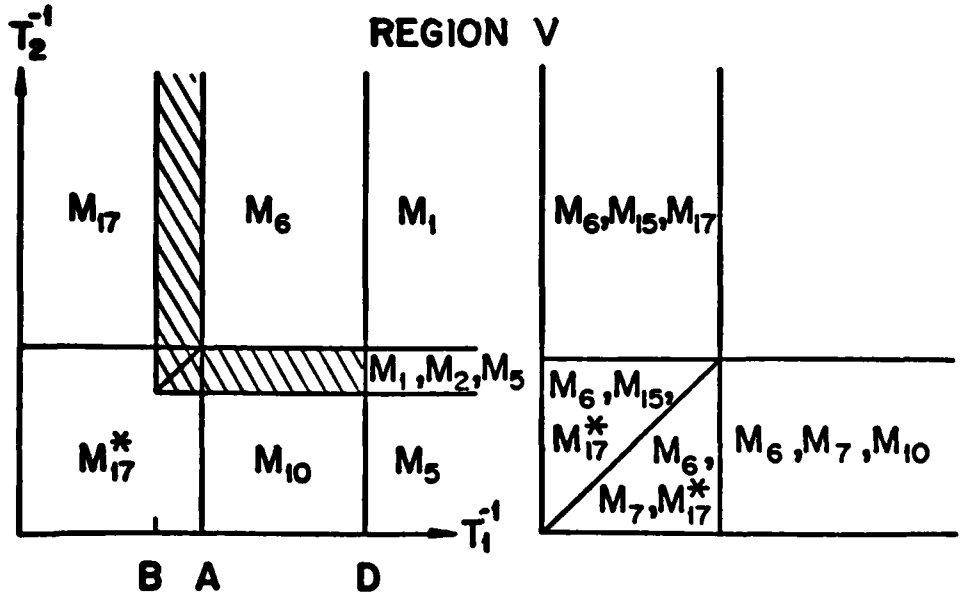

Figure 5e

(See caption on page 19.)

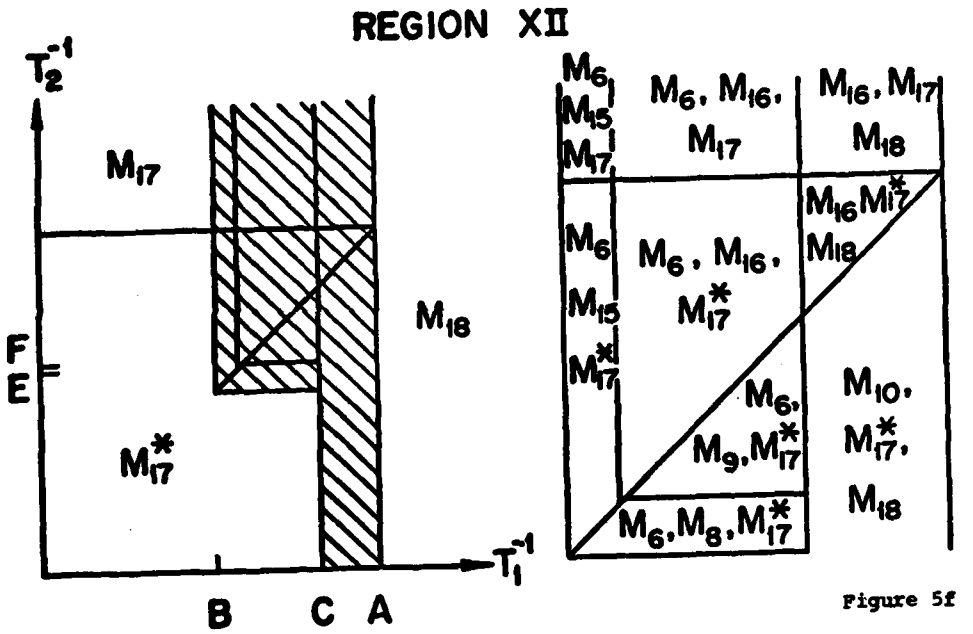

Figure 5f

Figure 5a-f. Burning-rate formulas which hold in Regions I, II, III, IV, V, XII and their respective domains; shaded areas are detailed at right. $A = T_\infty^{-1}$, $B = T_{BS}^{-1}$, $C = (T_s - L + 1)^{-1}$, $D = T_s^{-1}$, $E = [T_s - L + (1+Q)/2]^{-1}$, $F = (T_\infty + QY_{O\infty}/2)^{-1}$, * = valid only for $\theta_1 >> \theta_2$.

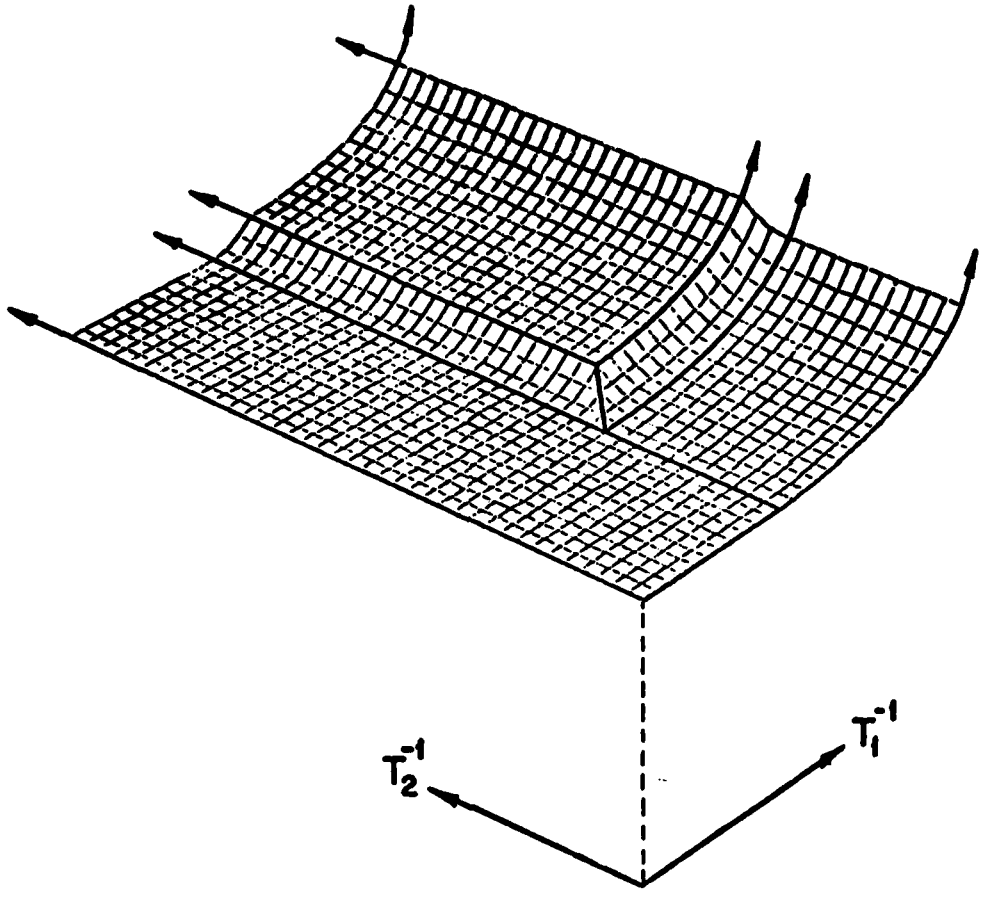

Figure 6a. (See caption on page 22.)

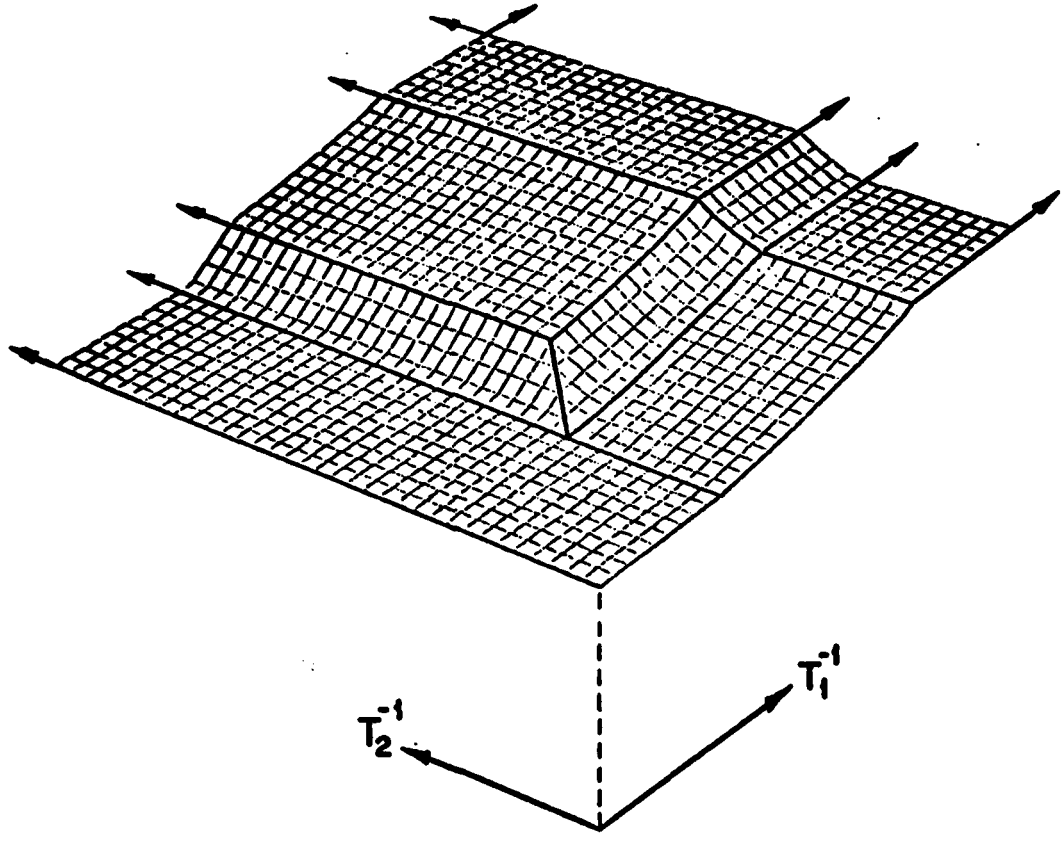

Figure 6b. (See caption on page 22)

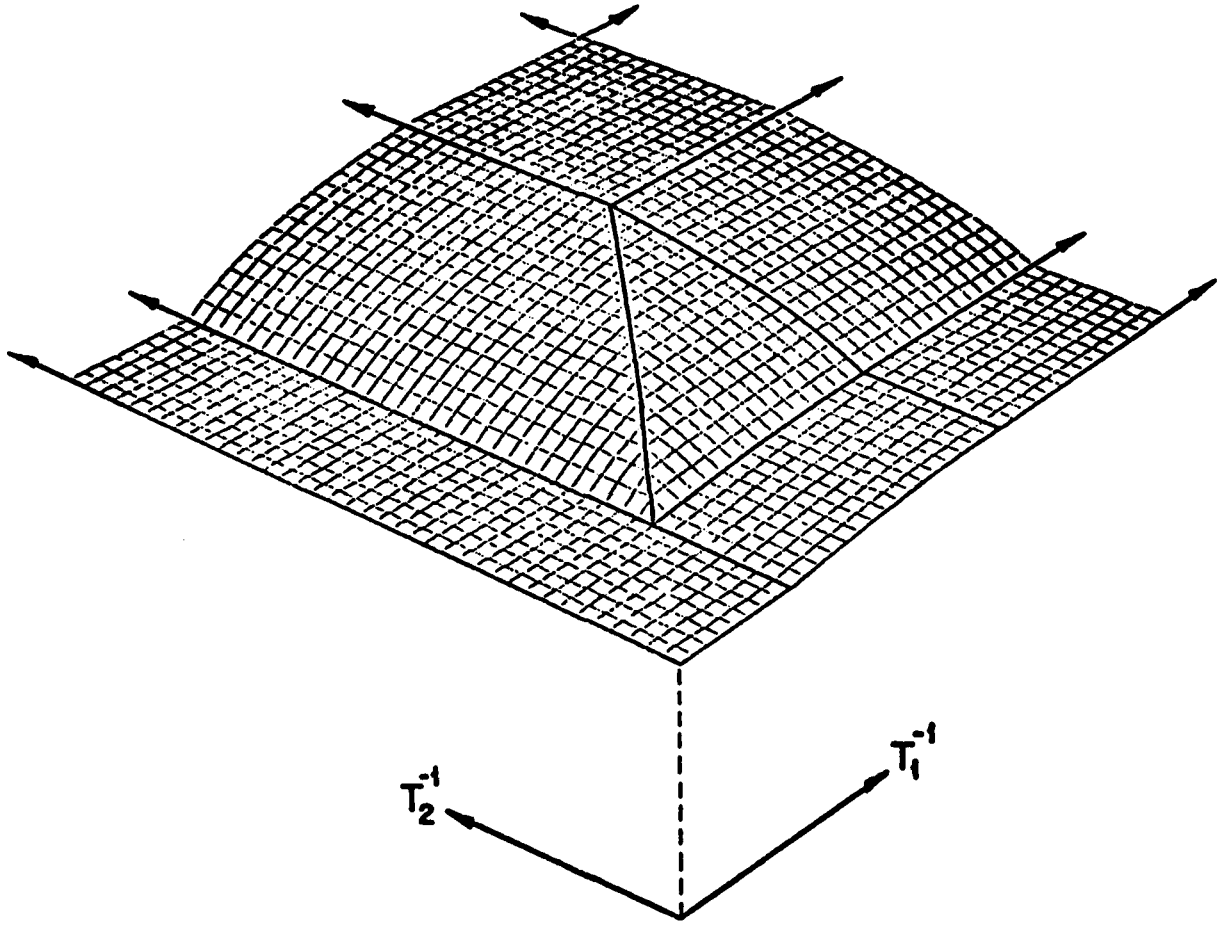

Figure 6c

Figure 6a-c. Response surfaces in Regions I-III respectively.

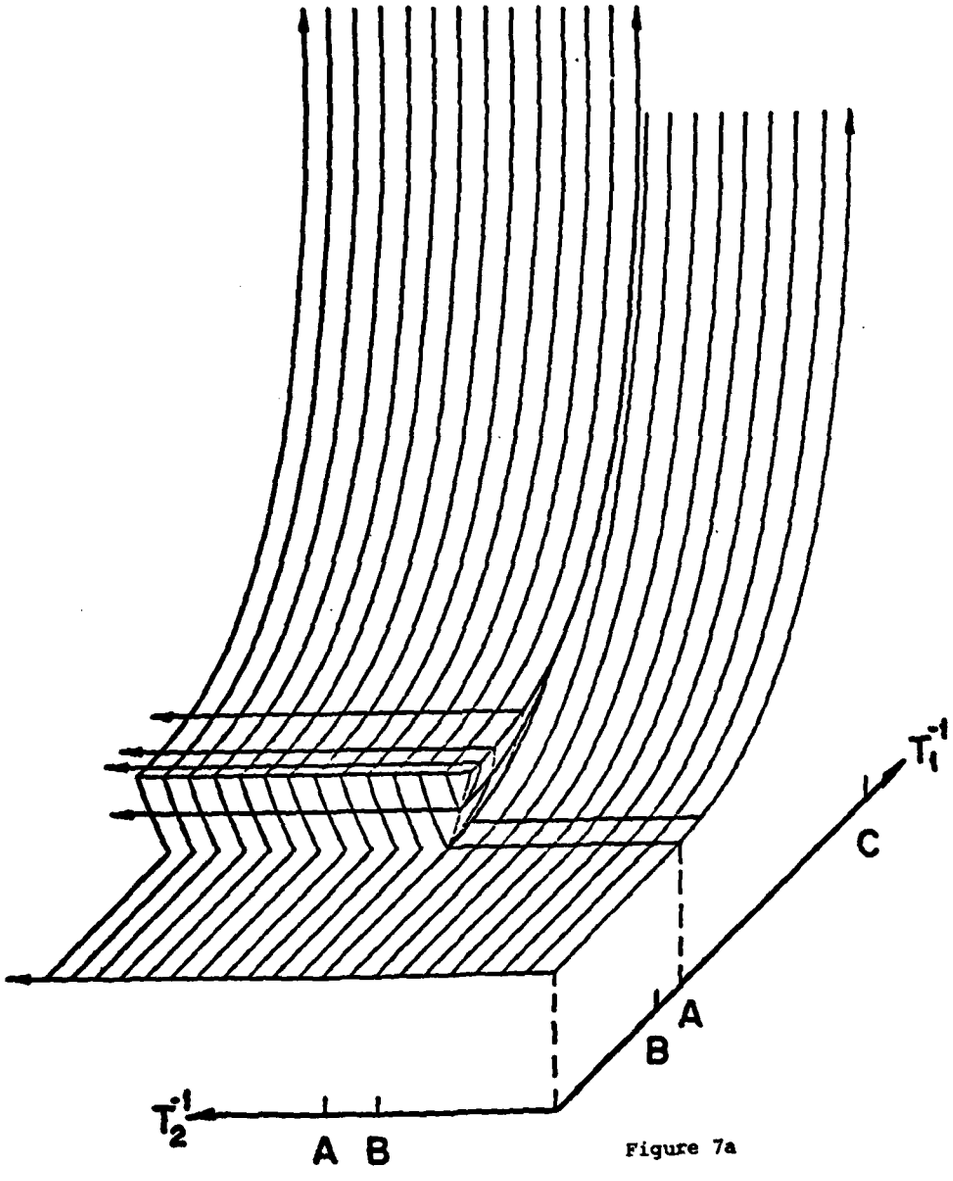

Figure 7a

Figure 7a. (See caption on page 25.)

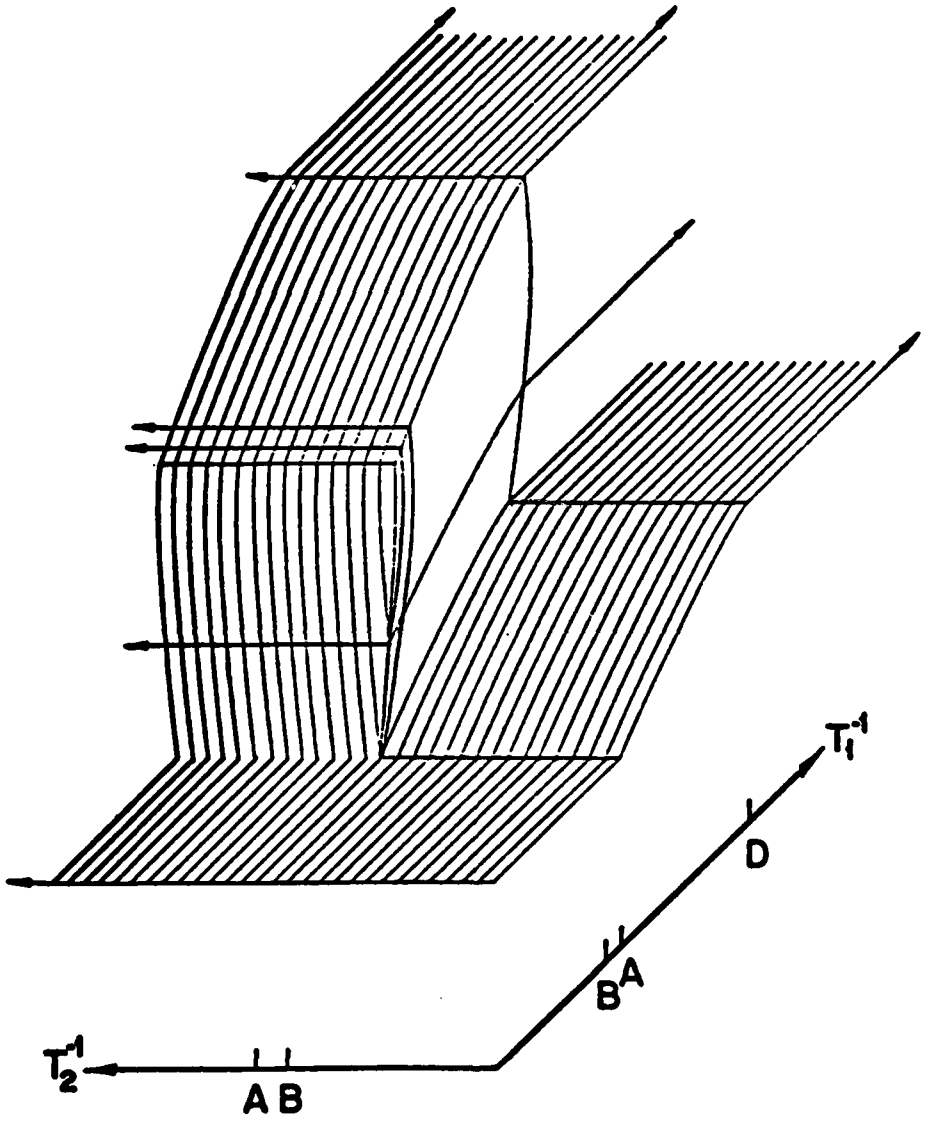

Figure 7b. (See caption on page 25.)

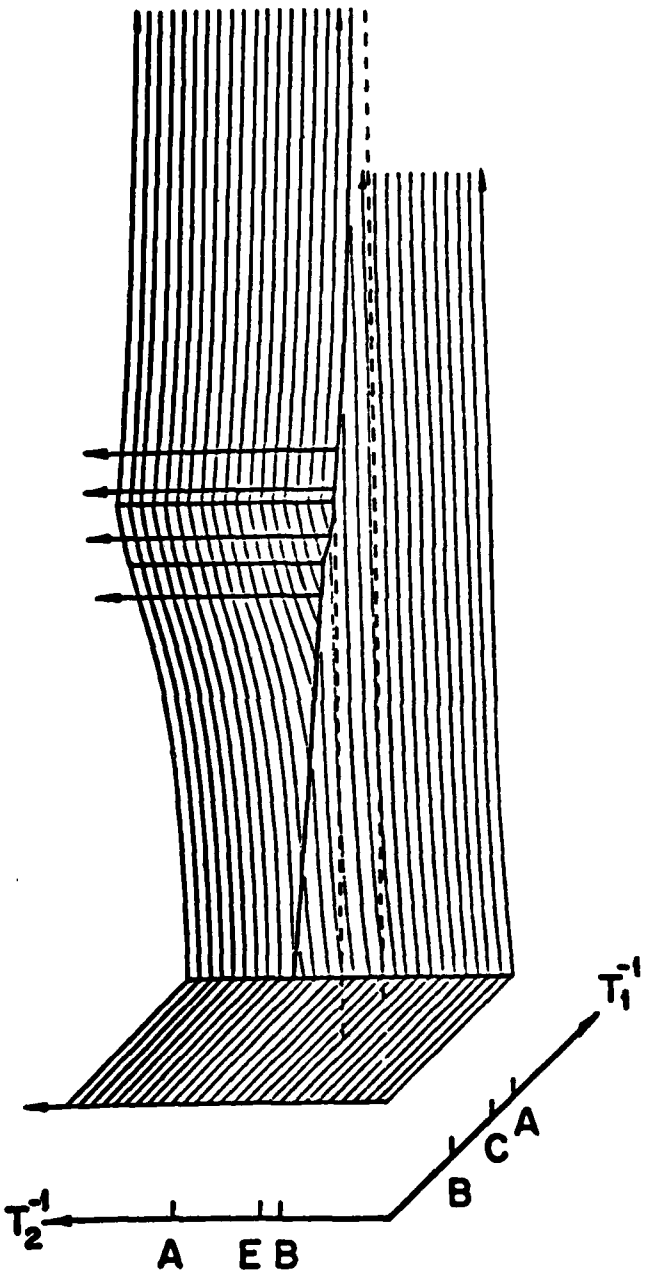

Figure 7c

Figures 7a-c. Response surfaces in Regions IV, V and XII respectively.

from a nearly extinguished state up to an ignited state as T_2^{-1} (or T_1^{-1}) is increased through its local maximum. The upper turnaround point is the extinction point where the solution jumps from an ignited state to a weakly reactive, nearly extinguished state as T_2^{-1} (or T_1^{-1}) is decreased through its local minimum. The middle level, which is bypassed, is believed to be unstable since it implies a decreasing burning rate with increasing reaction rate or pressure (Buckmaster and Ludford [1]; Kapila [14]).

The responses in Regions IV, VI, VIII and X differ only in their middle level, hence the realizable responses in these regions are the same. Similarly, Regions V, VII, IX and XI share responses which are effectively the same, as do Regions XII-XIV. Figures 7a and 7b thus indicate for Regions IV-XI the possibility of ignition and extinction of both reactions simultaneously or of the bipropellant reaction alone. Figure 7c shows that where ignition and extinction occur in the response of Regions XII-XIV, they occur in both reactions, ignition being possible only when the diffusion flame is remote.

4.2. Comparison with Related Works

Three "edges" of the response surfaces found here represent previously addressed problems, so that related results may be compared.

The bipropellant reaction is found to be remote for $0 < T_2^{-1} < \min(T_{BS}^{-1}, T_\infty^{-1})$; the burning rate is then controlled by the decomposition reaction alone. The corresponding edge of the response surface therefore describes pure monopropellant decomposition. That problem has been considered by Liñán [15] and by Ludford, Yannitell and Buckmaster [12] in the limit of large activation energy. For $T_s < T_\infty$, the work of Ludford et al. produces the burning rates M_5, M_{10}, M_{13}, M_{17} and M_{18} in the same regions of the $T_\infty - T_s$ vs. L parameter plane and with the same domains as found in the present work for $T_2^{-1} < \min(T_{BS}^{-1}, T_\infty^{-1})$. Similarly, there is agreement between the associated response curves $M(D_1)$ obtained by Ludford et al. [12] and the constant-T_2 cross-sections of the response surfaces found in this work for $T_2^{-1} < T_{BS}^{-1}$. Liñán [15] obtains the above expressions for M, but does not specify the parameter values for which each is acceptable.

Next, it may be noted that for $T_1 < T_s$ and $L > 1$, the decomposition flame is sitting on the droplet surface and the problem in $1 < r < \infty$ is that of an ordinary fuel drop at temperature T_s, but with latent heat $L-1$. Appropriately modifying the results of Law [16], obtained via activation-energy asymptotics, yields burning rates M_1, M_2, M_4 and M_5; the corresponding response curves $M(D_2)$ are consistent with the constant-T_1 cross-sections of the response surfaces found here for $L > 1$ and $T_1 < T_s$.

Finally, the results of Buckmaster, Kapila and Ludford [9] are considered. As mentioned earlier, these authors used Damköhler-number asymptotics in their treatment. They were thus able to determine only the three "corners":

$$\lim_{D_1,D_2\to\infty} M(D_1,D_2),\quad \lim_{D_2\to 0}\,[\lim_{D_1\to\infty} M(D_1,D_2)],\quad \lim_{D_1\to 0}\,[\lim_{D_2\to\infty} M(D_1,D_2)]$$

of the response surface. The expressions M_1, M_5, M_{14} and M_{17} were obtained. Formula M_{18} was also found by considering the limit $\theta_1 \to \infty$ for $M \gg 1$. More precisely, they found

$$\lim_{D_1,D_2\to\infty} M(D_1,D_2) = \begin{cases} M_{18} & \text{for } L < 1 \\ M_1 & \text{for } 1 < L < 1 + Q + Y_{O\infty}^{-1}(T_\infty - T_s) \\ M_{14} & \text{for } L > 1 + Q + Y_{O\infty}^{-1}(T_\infty - T_s) \end{cases}$$

$$\lim_{D_2\to 0}\,[\lim_{D_1\to\infty} M(D_1,D_2)] = \begin{cases} M_{18} & \text{for } L < 1 \\ M_5 & \text{for } L > 1 \end{cases}$$

$$\lim_{D_1\to 0}\,[\lim_{D_2\to\infty} M(D_1,D_2)] = M_{17}\ ,$$

in agreement with the corresponding corners of the responses found in the present work. Discrepancies exist, however, between the nature of the curves connecting these corners as conjectured by Buckmaster et al. and the curves dictated by the finding of the present work, as illustrated in Figures 8.

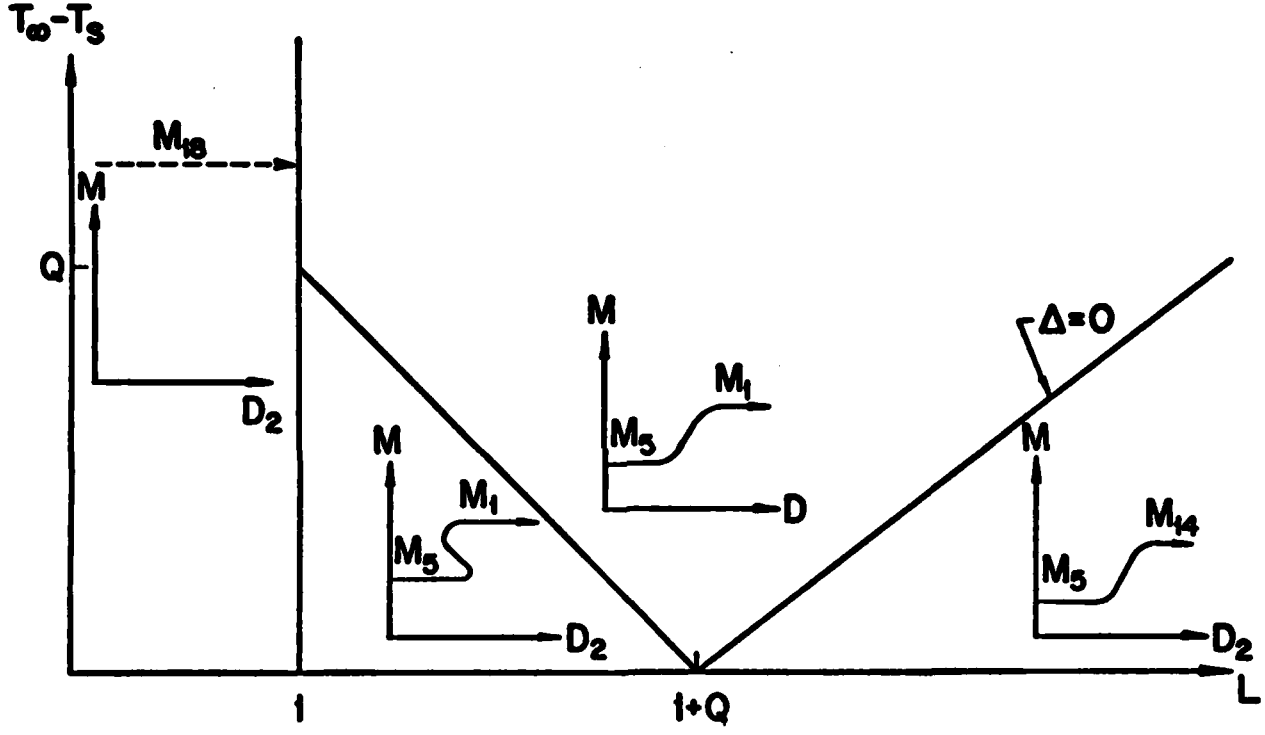

Figure 8a. Results for $\lim_{D_1 \to \infty} M(D_1, D_2)$ obtained by Buckmaster, Kapila and Ludford [9]; these are consistent with the present findings. $\Delta = T_\infty - T_s + Y_{O\infty}(1+Q-L)$.

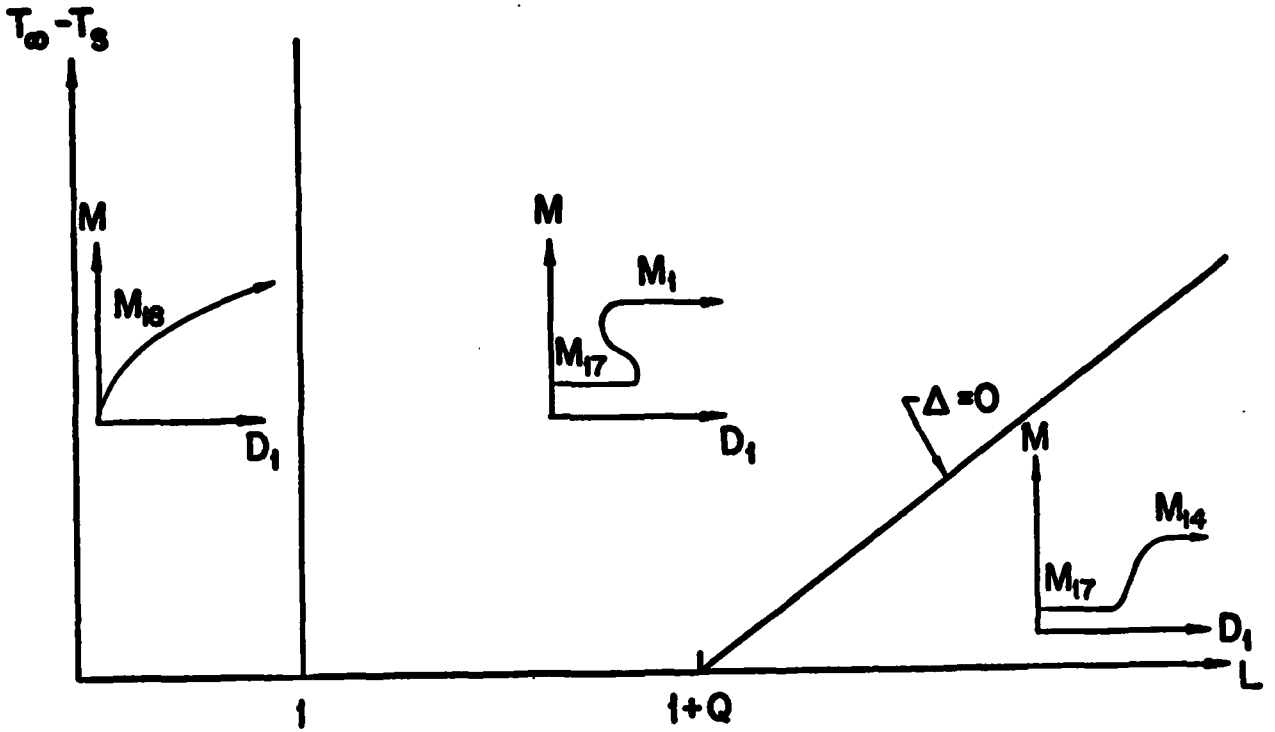

Figure 8b. Behavior of $\lim_{D_2 \to +\infty} N(D_1, D_2)$ conjectured by Buckmaster et al. [9].

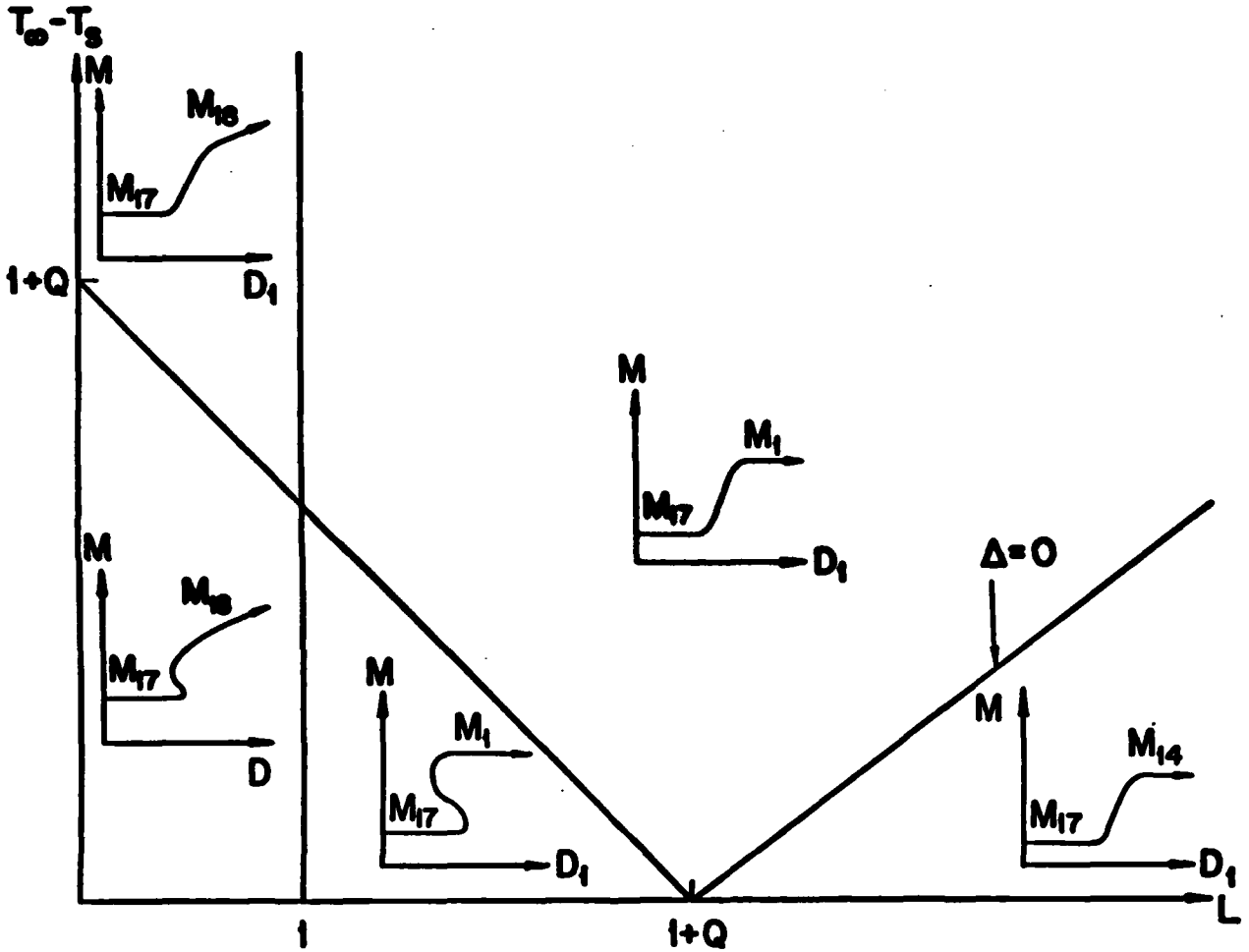

Figure 8c. Behavior of $\lim_{D_2 \to \infty} M(D_1, D_2)$ implied by the $T_2^{-1} > T_\infty^{-1}$ edge of the response surfaces $M(T_1^{-1}, T_2^{-1})$ determined in the present work.

Appendix 1: Structure Analysis for Category 6

Decomposition-Flame Zone

The problem within the decomposition flame, given by (2.1a,d), (3.3) and (3.4) is

$$d^2x_m/d\xi^2 = -d^2x_t/d\xi^2 = x_m\tilde{D}_1\exp x_t \quad ,$$

$$dx_m/d\xi \to -M/r_1^2,\ dx_t/d\xi \to M(T_1-T_s+L)/r_1^2 \quad \text{as} \quad \xi \to -\infty \quad ,$$

$$dx_m/d\xi \to 0,\ dx_t/d\xi \to M(T_1-T_s+L-1)/r_1^2 \quad \text{as} \quad \xi \to +\infty \quad ,$$

where

$$r = r_1 + \varepsilon\xi,\ \varepsilon = T_1^2/\theta_1 \ll 1,\ \tilde{D}_1 = \varepsilon^2 D_1' = O(1) \quad ,$$

$$Y_m = \varepsilon x_m(\xi) + o(\varepsilon),\ T = T_1 + \varepsilon x_t(\xi) + o(\varepsilon) \quad .$$

This may be rewritten as

$$\frac{2d^2x_m}{dy^2} = x_m\exp(\alpha y - x_m),\ \frac{dx_m}{dy} \to -1 \quad \text{as} \quad y \to -\infty,\ \frac{dx_m}{dy} \to 0 \quad \text{as} \quad y \to +\infty \quad , \qquad \text{(A1.1)}$$

where $y = M\xi/r_1^2 + \alpha^{-1}[a_0 + \ell n(2\tilde{D}_1 r_1^4/M^2)]$, $\alpha = (T_1 - T_s + L - 1)$ and a_0 is an undetermined constant. Boundary-value problem (A1.1) has been treated numerically by Liñán [17] whose results indicate that a solution exists provided

$$T_1 > T_s - L + 1/2 \quad \text{if} \quad x_m + c > 0 \quad \text{as} \quad \xi \to +\infty \quad ,$$

$$T_1 > T_s - L + 1 \quad \text{if} \quad x_m \to 0 \quad \text{as} \quad \xi \to +\infty \quad .$$

Diffusion-Flame Zone

The following holds in the diffusion flame.

$$Y_m = \delta b_0 + o(\delta),\ T = T_{BS} + \delta y_t(\eta) + o(\delta) \quad ,$$

$$d^2y_t/d\eta^2 = -\delta Q D_2' Y_f Y_o \exp(\theta_2/T_2 - \theta_2/T_{BS}) \quad , \qquad \text{(A1.2)}$$

$$y_t \sim b_2\eta + b_3 \quad \text{as} \quad \eta \to -\infty,\ y_t \sim b_1\eta + b_4 \quad \text{as} \quad \eta \to +\infty \quad ,$$

where $\eta = (r - r_2)/\delta$, $\delta \ll 1$, b_0 is a nonnegative constant, $T_2 < T_{BS}$, $b_1 = M(T_\infty - T_s + L - 1 - Q)\exp(-M/r_2)/r_2^2$, $b_2 = M(T_\infty - T_s + L - 1 + QY_{O\infty})\exp(-M/r_2)/r_2^2$, b_3 and b_4 are

undetermined constants, and Y_f and Y_0 are given by (2.4) and (2.5). Consider $T_2 < T_{BS}$. Then (A1.2) may be written as

$$d^2y/dx^2 = x^2-y^2,\ y \sim \pm x \ \text{ as } \ x \to \tilde{+}\infty \ , \tag{A1.3}$$

where

$$y(\eta) = y_t(\eta) - (b_1+b_2)\eta/2-(b_3+b_4)/2,\ x = (b_2-b_1)\eta/2+(b_3-b_4)/2 \ ,$$

and where the small parameter δ has been chosen according to

$$\delta^3 = [(b_2-b_1)/2]^2(D_2')^{-1}\Omega \exp(\theta_2/T_{BS}-\theta_2/T_2) \ .$$

Physical considerations require

$$y < -|x| \ \text{ for all } \ x \ . \tag{A1.4}$$

Equation and boundary conditions (A1.3) constitute the standard structure problem for a Burke-Schumann flame which has been shown by Holmes [18] to possess exactly two solutions, only one of which satisfies (A1.4).

Appendix 2: Structure Analysis for Category 15; Type A Flames

Diffusion-Flame Zone

The appropriate expansions in the bipropellant-reaction zone include

$$r = r_* + \delta\eta,\ Y_m = \delta y_m(\eta) + o(\delta),\ T = T_1+\delta y_t(\eta)+o(\delta) \ ,$$

where $\delta^{-2} = D_2'Y_{o*}\exp(\theta_2/T_2-\theta_2/T_1)$, $Y_{o*} = Y_o(r_*)$, $\theta_1 >> \exp(\tilde{c}\theta_2)$ with $\tilde{c} > 0$, $y_t < 0$ for $\eta < \eta_*$, $y_t = 0$ at η_*, $y_t < 0$ or $Y_m = 0$ for $\eta > \eta_*$ with η_* the location of the decomposition reaction. The function $y_m(\eta)$ thus satisfies

$$\frac{d^2y_m}{d\eta^2} = o(1) \ \text{ for } \ \eta \lessgtr \eta_*,\ \frac{dy_m}{d\eta} \to -\frac{M}{r_*^2} \ \text{ as } \ \eta \to -\infty,\ \frac{dy_m}{d\eta} \to 0 \ \text{ as } \ \eta \to +\infty \ .$$

Continuity of y_m, y_t and $d(y_m+y_t)/d\eta$ at η_* is required, therefore

$$y_m = c_0-M\eta/r_*^2 \ \text{ for } \ \eta < \eta_*,\ y_m = c_0-M\eta_*/r_*^2 \ \text{ for } \ \eta > \eta_* \ ,$$

$$y_t(\eta_*^-) = y_t(\eta_*^+),\ \frac{dy_t}{d\eta}(\eta^-) = \frac{dy_t}{d\eta}(\eta_*^+) + \frac{M}{r_*^2} \ , \tag{A2.1}$$

where the constant c_0 is undetermined. The equation for y_t becomes

$$d^2y_t/d\eta^2 = y_t - c_1\eta - c_2 \quad \text{for} \quad \eta < \eta_*, \quad d^2y_t/d\eta^2 = y_t - c_3\eta - c_4 \quad \text{for} \quad \eta > \eta_* \quad , \quad (A2.2)$$

where $c_1 = M(T_1 - T_s + L)/r_*^2$, $c_3 = M(T_1 - T_s + L - 1 - Q)/r_*^2$, $c_4 = (c_1 - c_3)\eta_* + c_2$ and c_2 is determined by matching higher-order terms. The solution of (A2.2) must satisfy (A2.1) as well as

$$dy_t/d\eta \to c_1 \quad \text{as} \quad \eta \to -\infty, \quad dy_t/d\eta \to c_3 \quad \text{as} \quad \eta \to +\infty \quad .$$

It follows that

$$y_t = c_5 e^{\eta - \eta_*} + c_1\eta + c_2 \quad \text{for} \quad \eta < \eta_*, \quad y_t = c_5 e^{\eta_* - \eta} + c_3\eta + c_4 \quad \text{for} \quad \eta > \eta_* \quad ,$$

where $c_5 = -QM/2r_*^2$. The requirement $y_t < 0$ for $\eta < \eta_*$ is therefore met provided $T_1 > T_s - L + Q/2$.

Decomposition-Flame Zone

In the monopropellant reaction at η_*, the following is valid.

$$r = r_* + \delta\eta_* + \varepsilon\xi, \quad Y_m = \delta y_{m_*} + \ldots + \varepsilon x_m(\xi) + o(\varepsilon), \quad T = T_1 + \varepsilon x_t(\xi) + o(\varepsilon) \quad ,$$

where $\varepsilon = T_1^2/\theta_1$, $y_{m_*} = y_m(\eta_*)$, and the components of Y_m preceding εx_m are constants.

For $y_{m_*} \neq 0$, the problem for $x_t(\xi)$ is

$$\frac{d^2x_t}{d\xi^2} = -\tilde{D}_1 e^{x_t}, \quad \frac{dx_t}{d\xi} \to c_5 + c_1 > 0 \quad \text{as} \quad \xi \to -\infty, \quad \frac{dx_t}{d\xi} \to -c_5 + c_3 < 0 \quad \text{as} \quad \xi \to +\infty \quad ,$$

where $\tilde{D}_1 = \varepsilon\delta y_{m_*} D_1' = 0(1)$. Integration shows that $c_1 = -c_3$.

For $Y_m = o(\varepsilon)$ at η_*, the structure problem is equivalent to (A1.1) with $\alpha = T_1 - T_s + L - 1 - Q/2$.

It follows that a Type A merged flame may exist for category 15 if $\theta_1 \gg \exp(\tilde{c}\theta_2)$ with $\tilde{c} > 0$, and if $\max(T_s - L + 1 + Q/2, T_2) < T_1 < T_\infty$ or $\max[T_\infty, T_s - L + (1+Q)/2, T_2] < T_1$.

Appendix 3: Structure Analysis for Category 15; Type C Flames

The problem in the Type C flame zone for category 15 is

$$\frac{d^2x_m}{d\xi^2} = \tilde{D}_1 x_m \exp[c_3\xi + d_0 - (1+Q)x_m], \quad \frac{dx_m}{d\xi} \to -\frac{M}{r_*^2} \text{ as } \xi \to -\infty, \quad \frac{dx_m}{\delta\xi} \to 0 \text{ as } \xi \to +\infty ,$$

where $Y_m = \varepsilon x_m(\xi) + o(\varepsilon)$, $T = T_1 + \varepsilon x_t(\xi) + o(\varepsilon)$, $\xi = (r-r_*)/\varepsilon$, $\varepsilon = T_1^2/\theta_1$, $\tilde{D}_1 = \varepsilon^2 D_1' = 0(1)$ and d_0 is an undetermined constant. This may be rewritten in the form of (A1.1) and a solution thus shown to exist provided $T_1 > \max[T_\infty, T_s - L + (1+Q)/2]$ or $T_s - L + 1 + Q < T_1 < T_\infty$.

The small amount of fuel produced in the decomposition reaction is $Y_f = \varepsilon D'_1 x_m e^{x_t}/[D_2' Y_{O_*} \exp(\theta_2/T_2 - \theta_2/T_1)]$, which is indeed $o(\varepsilon)$ since $T_1 > T_2$ and $\theta_2 >> 1$.

Appendix 4: Structure Analysis for Category 18

Surface Layer

The region adjacent to the droplet is described by $\zeta = \gamma^{-1}(r-1)$ with $\gamma << 1$, and the burning rate M is of the form $M = \gamma^{-1}N$ where $N = 0(1)$. Within this layer, the following holds.

$$Y_m = m(\zeta) + o(1), \quad T = t(\zeta) + o(1), \quad t = T_s - L + 1 - m ,$$

$$\frac{d^2m}{d\zeta^2} - N\frac{dm}{d\zeta} = \gamma^2 D_1' m \exp(\theta_1/T_1 - \theta_1/t); \quad \frac{dm}{d\zeta} = N(m-1) = -NL, \; m = 1-L \text{ at } \zeta = 0 .$$

Let ζ_* label the smallest value of ζ at which $t(\zeta_*) = t_* = \max[t(\zeta)]$; take $\gamma = [D_1' \exp(\theta_1/T_1 - \theta_1/t_*)]^{-1/2}$ and require $T_1 < t_*$. Then the decomposition reaction occurs at $r = 1 + \gamma\zeta_*$ and

$$m = 1 - \exp[N(\zeta - \zeta_*)] \text{ for } \zeta < \zeta_*, \; m = 0 \text{ for } \zeta > \zeta_*, \; \zeta_* = -N^{-1}\ell nL .$$

The structure problem at ζ_* is given by

$$\frac{d^2x_m}{d\xi^2} = \tilde{D}_1 x_m \exp(-x_m), \quad x_m \sim -N\xi \text{ as } \xi \to -\infty, \; x_m \sim 0 \text{ as } \xi \to +\infty ,$$

where $\xi = (\zeta - \zeta_*)/\varepsilon$, $\varepsilon = t_*^2/\theta_1$, $Y_m = \varepsilon x_m(\xi) + o(\varepsilon)$, $T = t_* - \varepsilon x_m(\xi) + o(\varepsilon)$ and

$\tilde{D}_1 = \varepsilon^2 D_1' = 0(1)$. Therefore, $N = \sqrt{2\tilde{D}_1}$ which specifies N. This result is valid provided $L < 1$ and $T_1 < T_s - L + 1$.

Remote Region

The coordinate of the remote combustion field is $R = \gamma r$, and the bipropellant reaction is located at R_*. The temperature equation, given by

$$d^2T/dR^2 + [(2R-N)/R^2]dT/dR = -\gamma^{-2}QD_2'Y_fY_O\exp(\theta_2/T_2-\theta_2/T) \quad ,$$

is reactionless for $R \neq R_*$ and for any $\theta_1 >> 1$, $\theta_2 >> 1$ provided the product Y_fY_O vanishes for $R \neq R_*$. The bipropellant-reaction zone is consequently a Burke-Schumann diffusion flame with $Y_O = 0$ for $R < R_*$ and $Y_f = 0$ for $R > R_*$. Continuity at R_* fixes the value of R_*.

The structure problem at R_* has the form of (A1.3), hence a solution exists and no additional restrictions need be imposed.

List of Symbols

dimensionless quantities:

Y_A	mass fraction of species A; A = m, f or o
T	temperature
D_i	Damköhler number, varies with $\bar{a}^{2} p^{i}$; i = 1 or 2
θ_i	activation energy
Q	ratio of the heat of combustion of the bipropellant reaction to that of decomposition
M	evaporation rate ("burning rate")
r	radial coordinate
L	latent heat of evaporation
D_i'	$= D_i \exp(-\theta_i/T_i)$
T_i	positive parameter characterizing the magnitude of D_i

dimensional quantities:

$\bar{a}$	radius of droplet
p	pressure
$\bar{M}$	evaporation rate, varies with $\bar{a}M$

subscripts:

m	monopropellant
f	fuel
o	oxidant
1	pertaining to decomposition reaction
2	pertaining to bipropellant reaction
s	surface value
∞	ambient value

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A two-step combustion process, consisting of monopropellant (i.e. one-reactant) burning followed by bipropellant (i.e. two-reactant) burning, is considered. The original reactant, namely vapor from a liquid droplet, decomposes into a fuel that combines with oxidant in the surrounding atmosphere. Complete responses $M(D_1,D_2)$ of the evaporation rate M of the droplet to the Damköhler numbers D_1,D_2 of the two reactions are determined in the limit of large activation energies. Conditions under which the response is monotonic or multi-valued (thereby exhibiting auto-ignition and auto-extinction) are identified. Previous conjectures, based on Damköhler-number asymptotics, are found to be mostly correct.

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