

PROJECT ADMINISTRATION DATA SHEET

ORIGINAL  REVISION NO. \_\_\_\_\_

Project No. E-16-648 GTRI/~~GTR~~ DATE 9 / 6 / 84

Project Director: Dr. Edward W. Price School/~~KMK~~ AE

Sponsor: Morton Thiokol, Inc. Aerospace Group  
110 North Wacker Drive; Chicago, Ill. 60606

Type Agreement: Purchase Order No. 0584

Award Period: From 7/1/84 To 6/30/86 (Performance) 6/30/86 (Reports)

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Funded: \$	<u>50,000</u>	\$ <u>50,000(fixed price)</u>

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Title: "Study of Oscillatory Interaction of Solid Propellant Combustion and Combustor Flow"

ADMINISTRATIVE DATA

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Defense Priority Rating: n/a Military Security Classification: n/a  
(or) Company/Industrial Proprietary: n/a

RESTRICTIONS

See Attached n/a Supplemental Information Sheet for Additional Requirements.

Travel: Foreign travel must have prior approval - Contact OCA in each case. Domestic travel requires sponsor approval where total will exceed greater of \$500 or 125% of approved proposal budget category.

Equipment: Title vests with sponsor

COMMENTS:

This is a fixed price Level-of-Effort Purchase Order.

Advance No. assigned in the amount of \$1,793.



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Project No. E-16-648 School/~~XXX~~ AE

Includes Subproject No.(s) N/A

Project Director(s) E. W. Price GTRC /~~GIT~~

Sponsor Morton Thiokol, Inc.

Title "Study of Oscillatory Interaction of solid propellant combustion and combustor flow"

Effective Completion Date: 6/30/86 (Performance) \_\_\_\_\_ (Reports)

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E-16-698

# GEORGIA INSTITUTE OF TECHNOLOGY

ATLANTA, GEORGIA 30332

SCHOOL OF  
AEROSPACE ENGINEERING

404-894-3000

DANIEL GUGGENHEIM SCHOOL  
OF AERONAUTICS

8 August 1984

Activity Report for  
Morton Thiokol Contract  
"Study of Oscillatory Interaction of Solid Propellant Combustion and Combustor Flow"

Activity during July consisted of the following:

1. Participation in the IR&D meeting at Ogden (in June).
2. Review of literature on microstructure of the combustion zone, and of approximations used by models of perturbation response (review continuing).
3. Review of literature on oscillatory flows, particularly as they pertain to flow systems with mass addition at the walls (review continuing).
4. Preliminary thoughts on what would be required of a physical model to correctly describe the qualitative features of the dynamic response of the combustion zone (Attachment A).
5. Preliminary planning for solution of the equations for axially symmetric oscillating, viscous laminar flow with mass transpiration at the walls (Attachment B).



Attachment A

THE COMBUSTION ZONE  
FOR AP-HC BINDER PROPELLANTS

The following are some preliminary thoughts on the features of the composite propellant combustion zone that must be considered in developing a valid model of dynamic response:

1. The gas phase combustion zone is a disordered array of flamelets; the disorder stems from the disorder of the propellant microstructure; however, the flamelets are individually coupled to the local surface region.

2. The gas phase flamelets behave in a quasi-steady manner to externally imposed disturbances at frequencies below about 5000 Hz. The local surface region responds on a much longer time scale, and the gas phase state follows the perturbed surface state in phase.

3. The local flamelets have a characteristic structure referred to here as the local flame complex. This consists (depending on conditions) of an AP monopropellant flame (including an exothermic surface froth), a diffusion flame, and a kinetically limited leading edge flame (at the leading edge of the diffusion flame). These features of the flame are individually dependent on the local features of the burning surface, and may be qualitatively different depending on surface details, pressure, propellant temperature, and propellant ingredient variables (AP purity, type of binder, particle size).

4. While there is a tendency to describe the local flame complex as being linked to the exposed oxidizer surfaces (which can be described statistically in terms of oxidizer particle size distribution), the complex is also dependent on the state of the surrounding binder and lateral sub-surface heat transfer between oxidizer and binder. Further, the state of the binder is not the same at all points around the oxidizer perimeters, and typically changes as a function of distance burned in the oxidizer particle. Because of these three-dimensional processes, the oxidizer surface is not flat, nor is the oxidizer "flame". Details differ with time during burning of the particle, size of the particle, nature of adjoining particles, pressure, etc. It is not yet clear how to link a statistical description of the flame population to a statistical description of the propellant.



5. A proper description of dynamic response must encompass the dynamic response of local three-dimensional features of flamelet-condensed phase response. Such response would reflect the difference between binder and oxidizer pyrolysis characteristics, the three-dimensional surface geometry and the three-dimensional heat flow. These details must oscillate in response to oscillations in the flame complex, a situation that must induce oscillations in local surface and flame geometry and surfacewise average oscillations in gas composition and flame temperature. Existing models do not describe these local details of combustion zone response, and introduce surfacewise composition oscillations artificially.

6. Attention must be addressed to proper characterization of the burning surface in terms of structural elements that control local flamelet behavior, elements that reflect the propellant microstructure, and that can be summed to give the collective response to flow disturbances. It should not be assumed in advance that the surface elements are AP surfaces with binder peripheries, or even that there is only one class of surface elements to be considered. It seems likely that the central part of large oxidizer surfaces responds in an entirely different way to flow disturbances than does the peripheral region near the surrounding binder, and that the relative importance of such different surface elements is highly dependent on oxidizer particle size, pressure, and even stage in the burning of the AP particle. Only out of such considerations will a proper model for phase-correlated composition oscillations and preferred frequencies emerge.

Attachment B

NUMERICAL SOLUTION OF EQUATIONS FOR AXIALLY SYMMETRIC  
OSCILLATORY VISCOUS FLOW WITH MASS ADDITION

The aim of this effort is to provide a computer program to compute the flow variables and acoustic properties of a rocket motor. In general, this is to be done by modifying an existing computer code which solves the axisymmetric form of the unsteady Navier-Stokes equations. The modifications involve mainly the reformulation of the boundary conditions in the code to simulate those expected in a rocket motor. This reformulation takes place in two steps: the first is an analytic derivation (which is the most lengthy), and the second is code implementation.

In July, the major effort has been the process of obtaining the code from the Air Force Flight Dynamics Laboratory, as well as working on the first step of the reformulation.

To obtain a government code is not complicated, but can be time-consuming if the procedure is not clearly spelled out, as was the case here. After numerous phone calls and several letters, it seems the requirements have been satisfied and the code is to be shipped forthwith.

As for the analytic derivation of the rocket motor boundary conditions, these are:

- 1) Transpiration wall boundaries to simulate the production of gases at the surface.
- 2) A line of symmetry at the centerline of the axisymmetric motor.
- 3) Supersonic outflow at the nozzle exit plane or a sonic boundary at the nozzle throat.

In July, the process of casting these boundary conditions into finite difference expressions has been pursued.

E-16-649

GEORGIA INSTITUTE OF TECHNOLOGY

ATLANTA, GEORGIA 30332

SCHOOL OF  
AEROSPACE ENGINEERING

404-894-3000

DANIEL GUGGENHEIM SCHOOL  
OF AERONAUTICS

20 November 1986

TO: Dr. D. A. Flanigan  
Director, Advanced Technology  
Aerospace Group  
Morton Thiokol, Inc.  
3340 Airport Road  
Ogden, UT 84405

FROM: E. W. Price, Principal Investigator *EWP*

SUBJECT: Monthly Report on Contract P. O. 0584, Amm. 31  
Covering August - November 1986

1. The objective of ongoing work is to develop a more realistic description of the response of propellant combustion to incident pressure disturbances. The approach is to view the propellant combustion zone as an array of flamelets that are coupled individually and locally to underlying heterogeneous microstructure of the propellant surface and solid (for the present, an AP-HC binder propellant is being considered). The flamelets consist of different kinds, each of which is coupled to its own kind of microstructural feature of the surface. Certain areas (centered on AP particles) are dominated by the AP self-deflagration flame. Such surface localities exhibit dynamic response characteristics analogous to pure AP self-deflagration. Around the periphery of AP particles, the combustion response is dominated by a kinetically limited O-F flamelet, which is coupled to the portions of AP and binder surface and subsurface close to the AP-binder contact lines on the surface. Contrary to current models of dynamic response, we believe it is necessary to address these different kinds of flamelets as locally coupled to the surface features that produce them and we are seeking to characterize this locally coupled behavior. The net combustion response is then the sum of the responses of all flamelets present.

2. The primary difference between the present and previous approaches is the characterization of the dynamic response as the sum of the response of different kinds of flamelets. Previous analytical models have considered such flamelets, but have not treated them as locally and independently coupled to their parent surface sources. This locally coupled approach is a more difficult one in that the individual flamelets are not coupled one-dimensionally to the surface, and the associated micro-surface and subsurface are not viewed as one-dimensional. In addition, the problem of characterizing the collective response of the flamelet population is more difficult than in earlier models. However, the limited utility of earlier models indicates that the present more rigorous approach is necessary.



Dr. D. A. Flanigan  
20 November 1986  
page 2

3. In order to limit the scope of the approach to a tractable level, we are treating the burning surface as a combination of two kinds of areas, AP self-deflagration areas (one-dimensional) and binder-oxidizer "interface areas" (interface line, oxidizer and binder surface areas close to the interface). Because we have extensive experience with AP self-deflagration and with combustion of interface areas from previous sandwich burning studies, we are viewing the interface area burning as two-dimensional. The sandwich burning results help us to know how much surface area to allocate to the 2-D interface region, how much to the 1-D self-deflagration of AP. Our efforts are concentrated at present on the problem of nonsteady combustion behavior of the 2-D interface region (with its associated flamelet). A complete analysis appears to be prohibitively difficult, so we are combining

- a) experimental inputs from sandwich burning studies,
- b) exploratory analyses to find out what are the most important processes to include in a model,
- c) more detailed analytical modeling of key processes, and
- d) combination of results to give an approximate model of interface region dynamic response.

4. Along with these modeling efforts, we are exploring the possibility of T-Burner testing of propellants which are tailored to provide certain key data regarding O-F flamelet structure and stability, data which will help in formation of realistic analytical models. The background for this experimental study was provided in last year's report. The goal has also been extended to tests and dynamic response of AP self-deflagration (needed to evaluate the role of the AP self-deflagration areas of the propellant surface).

5. The details of the analysis to date are outlined in the attached progress brief by John Deur.

EWP/ed  
prrpt.th6

cc: R. B. Kruse  
B. B. Stokes

18 November 1986

FROM: J. M. Deur, Graduate Research Assistant

SUBJECT: Combined Monthly Report (August - November) on Contract P. O. 0584,  
Amm. 31

Progress has been made in the following areas:

1. Edgewise Pyrolysis of Laminates. This is the non-steady state extension of the one-dimensional work reported last year. The purpose of this study was to estimate the effects on the acoustic admittance, the appropriate boundary condition to the acoustic analysis of a rocket motor cavity, of mixture ratio oscillations in the surface pyrolysis of an oxidizer-binder slab system. To maintain simplicity, this work was based largely on an existing response model, that of Denison and Baum, with modifications added to account for flame temperature and molecular weight fluctuations stemming from mixture ratio oscillations. Since the results indicate that effects are only substantial for the case of "laboratory" propellants with lower oxidizer loadings, e.g., 80 percent, and since it is reasoned that two-dimensional heat transfer effects in the condensed phase, ignored in the current analysis, would reduce the mixture ratio effect even more, the decision has been made to cancel development of a numerical analysis which would solve the transient heat transfer problem in the condensed phase. However, the completed analysis will be useful for the overall laminate combustion model and is believed to be of sufficient merit, especially in what it has to say to experimentalists dealing with "laboratory" propellants, to warrant submission of a technical paper for the upcoming AIAA Joint Propulsion Conference (see Enclosure A).

2. Perturbation of an Idealized Mixing Region Solution. The classic Burke-Schumann diffusion flame solution is, in reality, a special case of a more general analysis which finds the entire concentration field in the mixing fan of two parallel or, as more usually seen, concentric jets of fuel and oxidizer gases. To date, the parallel jet analysis has been rederived, to ensure familiarity with all assumptions in the original work, and has been linearized following the same technique employed in linearizing the Denison and Baum gas phase analysis. Both the Denison and Baum gas phase rate equation and the Burke-Schumann diffusion solution are found by steady state analyses and, hence, the care taken in pointing out the parallels between their linearizations. The solutions, both steady state and linearized, are being written into a computer program which will be able to map out the results for clearer understanding of the resultant equations. The results of this analysis will be used to study the effects of inflow condition and pressure oscillations on the mixing fan and, thus, the Kinetically Limited Leading Edge Flame (KLLEF) linked to this region.

3. Experimental Combustion Response Study. Some preliminary coordination work has been done to examine what T-burner facilities and equipment are available at Georgia Tech. or how the T-burner at the Huntsville Division might be employed by a Georgia Tech. student. The T-burner is, at present, the device of choice to qualitatively study the predicted breaks in the combustion response in appropriately formulated bimodal propellants. These breaks are the

non-steady counterparts to the breaks in steady state burn rates previously reported by E. W. Price and co-workers. Evidence of the response breaks would be the first clear demonstration of a link between the combustion response and a non-steady aspect of the flame complex, illustrating a mechanism not in any existing model, but likely to be significant to the modeling work currently being conducted at Georgia Tech.

Turning to plans for continuing work, a model for sandwich combustion response should be completed by early June 1987. This model will include the laminate pyrolysis and mixing fan models discussed above. Work is still needed in describing the position and lateral extent of the KLEF in the mixing fan as well as the behavior of the quench limits at the edges of the KLEF in response to the mixing fan oscillations. The question of KLEF behavior when approaching and following detachment must also be addressed. Models of the other combustion regions, the AP flame and final diffusion flame, have to be formulated. As an aside, a simplistic calculation of the combustion response of AP has been performed via the Denison and Baum model and may serve as a preliminary model for the AP combustion zone. Finally, a statistical scheme for combining these component models to describe the global sandwich response has to be devised.

Sincerely,

John Deur  
Graduate Research Assistant  
School of Aerospace Engineering

thio.txt

Enclosures: A. Abstract



## A SIMPLE MODEL FOR DYNAMIC FLAME RESPONSE

J. M. Deur  
and  
E. W. Price

Georgia Institute of Technology  
Atlanta, Georgia

### ABSTRACT

Oscillatory combustion of solid propellants is a persistent problem in rocket motor development programs. This phenomenon results from the complex interaction between oscillatory gas motion in the motor cavity and the propellant's combustion process. While many simplified analyses have been made to describe the gas motion, the combustion, and their interaction with one another, the resulting models have generally failed to provide quantitatively or, in some cases, even qualitatively meaningful results. Ongoing efforts to identify and remedy deficiencies in analytical models have resulted in the realization that the most difficult aspect to adequately, yet simply, model is the dynamic response of the propellant combustion to flow perturbations. This paper is concerned with this problem.

Before discussing the specifics of this paper, a few comments about how and why the above problem has been addressed in the past are in order. This will provide the background and necessary contrast in approach to show why the method currently undertaken is both unique and analogous to the real process of oscillatory combustion. In previous models, it has been common to separate the analyses into two parts, an acoustic analysis of the gas phase and a simple model of the thin combustion zone.

In particular, the combustion model in such a case provides only a boundary condition to the gas phase system. This boundary condition is the acoustic admittance defined, as shown below, as the ratio of velocity to pressure perturbations taken just at the outer edge of the combustion zone.

$$A = \frac{u'/\bar{u}}{p'/\bar{p}} \quad (1)$$

However, to adequately describe the combustion process, the details of what is occurring in this thin region have to be examined. The difficulties with this are that the combustor flow field models used in most analyses are inadequate to describe the gas motions near the wall or, in other words, in the combustion zone and, excepting in very simplified cases which usually exclude any detailed examination of gas oscillations, the combustion zone is very difficult to describe.

The current analysis will attempt to make progress in this area by employing one-dimensional analyses to maintain simplicity, while including multi-dimensional effects in an approximate manner. This will be accomplished by

examining the dynamic response of a particular part of the burning surface which is dominated by the oxidizer-fuel flame, the Kinetically Limited Leading Edge Flame (KLLEF) discussed in previous papers by the second author. This locale includes the surface interface between the oxidizer and binder and sits under the KLLEF which straddles the stoichiometric surface in the gas phase mixing fan of oxidizer and fuel vaporization products. Because of the close proximity of the KLLEF to this region, it is the dominant heat source for the surface pyrolysis and, hence, also dominates the dynamic response of the surface in this region. This is complicated by the heterogeneity effects (due to differences in decomposition kinetics and thermophysical properties) resulting from having both binder and oxidizer surfaces within the confines of this local area. Summarizing, due to the proximity of the KLLEF, there is a localized response governing the behavior of the region near the oxidizer-fuel interface, which is further dependent on the effects of mixture ratio oscillations stemming from the differences in the binder and oxidizer decomposition regions that make up this surface area.

Further simplifications result from limiting examination to oxidizer-binder laminates and focusing only on this one region of the surface, leaving for future work the calculation of the response of other burning surface regions such as the regions where AP self-deflagration dominates and the summation of these different responses to provide the overall combustion response as well as extending the results to true propellant combustion.

The greatest simplification in this analysis will be the extensive use of existing one-dimensional combustion response models of the form:

$$R_p = \frac{m'/\bar{m}}{p'/\bar{p}} = \frac{nAB}{AB - (A+1) + k + A/k} \quad (2)$$

where:

$$A = \frac{E}{RT_s^2} (T_s - T_0) \quad (3)$$

$$B = \frac{1}{\sigma_p (T_s - T_0)} \quad (4)$$

and  $k$  is the positive root of:

$$k^2 - k - i\Omega = 0 \quad (5)$$

with:

$$\Omega = \lambda \rho \omega / m^2 c_p \quad (6)$$

It has been noted in the literature that all one-dimensional models which employ the quasi-steady gas phase assumption lead to this two parameter equation, i.e., an expression in terms of an A and B, or some permutation of it.

The acoustic admittance and the pressure coupled response can be related via the mass conservation law and the ideal gas equation as follows:

$$A = R_p - 1 + \frac{T'/\bar{T}}{p'/\bar{p}} - \frac{M'/\bar{M}}{p'/\bar{p}} \quad (7)$$

The remainder of the analysis finds expressions for the temperature and molecular weight terms in the above expression.

The temperature term is composed of two parts, one dealing with the necessity of the flame temperature to oscillate in order to accommodate the lagging thermal response of the condensed phase to oscillatory heat transfer from the gas phase and the second due to flame temperature changes resulting from the fluctuations in the o/f ratio at the surface. An expression for the former effect can be found in the model of Denison and Baum, while an expression for the latter can be obtained by using information relating flame temperature and composition found via a standard thermochemistry program and a perturbation analysis of the definition of the o/f ratio. The combined result is:

$$\frac{T_f'/\bar{T}_f}{p'/\bar{p}} = (R_{p_o} - \eta) \frac{c_{p_c}}{\sigma_p \bar{T}_f c_{p_g}} + \frac{T_f'}{\theta'} \frac{\bar{\theta}}{\bar{T}_f} (R_{p_o} - R_{p_f}) (1 - \bar{\theta}) \quad (8)$$

where the pressure coupled responses of the oxidizer and binder will be determined via the two parameter model. The remaining terms have the usual definitions, except for  $\theta$  which represents the oxidizer mass fraction.

As the Denison and Baum model does not provide for molecular weight fluctuations, the molecular weight response consists only of that portion due to composition fluctuations. The resulting expression is found to be:

$$\frac{M'/\bar{M}}{p'/\bar{p}} = (R_{p_o} - R_{p_f}) \frac{M'}{\theta'} \frac{\bar{\theta}}{\bar{M}} (1 - \bar{\theta}) \quad (9)$$

Calculations made with the above expressions using typical propellant, oxidizer, and binder parameters found in the literature show that the effects of the temperature and molecular weight terms are greatest for propellants with low oxidizer loadings, e.g., 80 percent, and virtually negligible for high loading propellants, i.e., loadings on the order of 90 percent. Further, the molecular weight term is found to be generally negligible. The temperature term, where important, mainly increases the peak response, although slight peak frequency shifts also occur.

One reason for the drop in effect with higher loadings is the nature of the flame temperature dependence on composition. As the mixture ratio approaches



stoichiometric proportions, the flame temperature reaches a maximum. For AP-hydrocarbon systems, this occurs at roughly 90 percent. Thus, at lower loadings, there is a strong positive feedback in flame temperature to composition oscillations which falls off rapidly and eventually becomes negative beyond the stoichiometric point. However, most laboratory propellants are still in the 80 percent loading region, due to the difficulties in mixing higher loading propellants in the lab; hence, the results may be of particular interest to the experimentalist.

E-16-648

Annual Report

Study of Oscillatory Interaction  
of Solid Propellant Combustion and Combustor Flow

by

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Prepared for

Morton Thiokol, Inc.

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June 1985

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## STATEMENT OF THE TECHNICAL PROBLEM

Oscillatory combustion in solid rocket motors is a phenomenon that has persisted as a practical development problem and resisted intensive efforts aimed at quantitative characterization and prediction. Growing understanding of this complex phenomenon has led to clarification of a number of aspects of the phenomenon, but the most crucial aspect remains inadequately measured or modeled. The actual coupling between the propellant combustion and the gas oscillations, the "combustion response", remains poorly measured and poorly modeled in spite of extensive efforts. The experimentalist is confronted with formidable problems of measurement of transient features of a complex and microscopic combustion process at high pressure. The analytical modeler is confronted with representation of this same complex and unmeasured process while limiting himself to a tractable analytical and computational problem. Recent advances in understanding of the steady state combustion of composite AP propellants offer the prospect of improved understanding, modeling, and prediction of oscillatory combustion response. Such advances would significantly reduce the incidence and cost of encounters with combustion instability in future motor development programs.

## OBJECTIVE

The long-term objective of this investigation is to establish realistic physical and analytical models for the dynamic response of composite propellant combustion to incident flow oscillations.

Studies during FY 1985 have been concerned with clarification of the combustion zone microstructure during steady state burning, and with considerations of what features of the combustion zone would be important to oscillatory combustion response (see below).

In FY 1986, the objectives are to complete experimental studies of the "steady state" combustion zone structure; examine the features of the steady state combustion zone that are most likely to be involved in dynamic response; and start modeling dynamic response. It is anticipated that this will lead (in FY 1987) to statistical characterization of a population of responsive components of the combustion zone, with the components of the population being dependent on propellant microstructure, pressure, and characteristics of the gas oscillation (including frequency).

## APPROACH

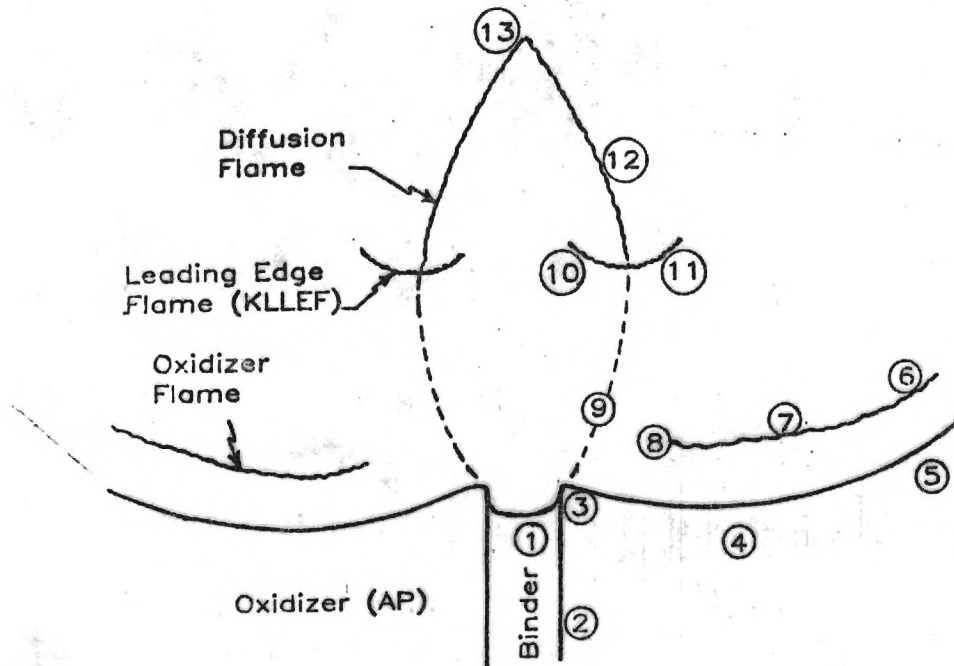
Progress depends on parallel investigations of:

- a) Combustion zone microstructure
- b) Identification of components of the combustion zone that are most responsive to flow disturbances, and the conditions (pressure, frequency, etc.) for large responsiveness
- c) Development of modeling schemes for responsive components, and for their interactions
- d) Development of a statistical scheme to represent populations of responsive components
- e) Development of a statistical scheme combining c) and d) to represent the collective dynamic response of the population to macroscopic flow disturbances

These efforts are concurrent, but the emphasis shifts from a - e with time during the program. Details are discussed below.

### Combustion Zone Microstructure

The combustion zone, including burning surface and heated condensed phase, is being described along lines indicated by recent research on steady state burning of ammonium perchlorate-hydrocarbon binder sandwiches. The sandwich combustion zone is described in the sketch in Fig. 1, which is typical of intermediate pressures (500-800 psi). With propellants, the flames are oriented around the periphery of exposed areas of oxidizer particles of various sizes. The details of the local flame complex differ as a function of position around the periphery of each particle because the adjoining fuel supply is non-uniform. The flame complex that surrounds any particular particle may be determined at its base by the local features of the burning surface at each peripheral point, but the neighboring flames become increasingly interactive further



Principal features of the combustion zone microstructure and processes as suggested by accumulated results. 1. Binder lamina. 2. Interface plane between binder and oxidizer. 3. Oxidizer surface adjoining binder (smooth band). 4. Leading edge of the oxidizer burning front. 5. Oxidizer region that regresses at the normal AP self-deflagration rate. 6. AP flame. 7. Leading edge region of AP flame. 8. Oxidizer flame, modified by the anomalous decomposition in the smooth band (flame may be quenched). 9. Oxidizer-fuel diffusion region, with stoichiometric surface indicated by broken line. 10.-11. Kinetically limited leading edge flame (KLLEF) (fuel-rich and oxidizer-rich sides). 12. Diffusion flame. 13. Tip of diffusion flame.

Fig. 1 Schematic representation of the combustion zone of an AP-Binder-AP sandwich, showing features of the flame complex (typical of 500 psi test conditions).

from the surface due to increased diffusion times and local depletion of the less plentiful reactants. These details are illustrated by the sketch in Fig. 2, which shows the nature of the flame complex as a function of position around the periphery of an oxidizer particle. In the figure, the surface along the periphery of a particle is mapped out in a straight line to aid in display of details. In location (A), the binder is thin and a diffusion flame closes over the binder. Further along there is a fuel excess, and the closure over the binder opens up and tends toward closure over the oxidizer particle. However, such closure is never possible in three dimensions because of the closure over the binder in fuel-lean regions like (A) where AP particles are nearly in contact. Our goal is to describe this flame complex with qualitative accuracy, develop a scenario for its changes as a function of time during burning of particles, and identify inherent instabilities or flow-responsive features of the flame complex. This is being done by experimental observation of burning surface microstructure (quenched sample studies); qualitative correlation of the surface observations with flame configuration through theoretical arguments, and validation of the resulting view of the flame complex by testing its ability to predict measurable singular combustion behavior.

### Responsive Components of the Combustion Zone

The picture of the propellant combustion zone emerging from the studies described above is too complex to expect early success in quantitative modeling. However, any realistic consideration of dynamic interaction with flow disturbances must start with a realistic picture of the combustion zone, even if that picture is only qualitative. Then the present goal is to recognize what aspects of the flame complex are directly responsive to flow disturbances, under what conditions (of pressure, frequency, propellant variables) they are responsive, how they couple to less responsive aspects of the combustion, and the time dependence (phase) of the coupled response. Such qualitative considerations are necessary to determine what aspects of the combustion must be contained in a response function model, and how that judgment depends on propellant variables and test conditions.

To illustrate the foregoing, the exothermic reaction regions in Fig. 1 all have different exposure to flow disturbances, and different reaction rate response. Complete analytical representation of transient response of all of these reaction regions is not currently tractable, and, in the interest of tractability, an approach must

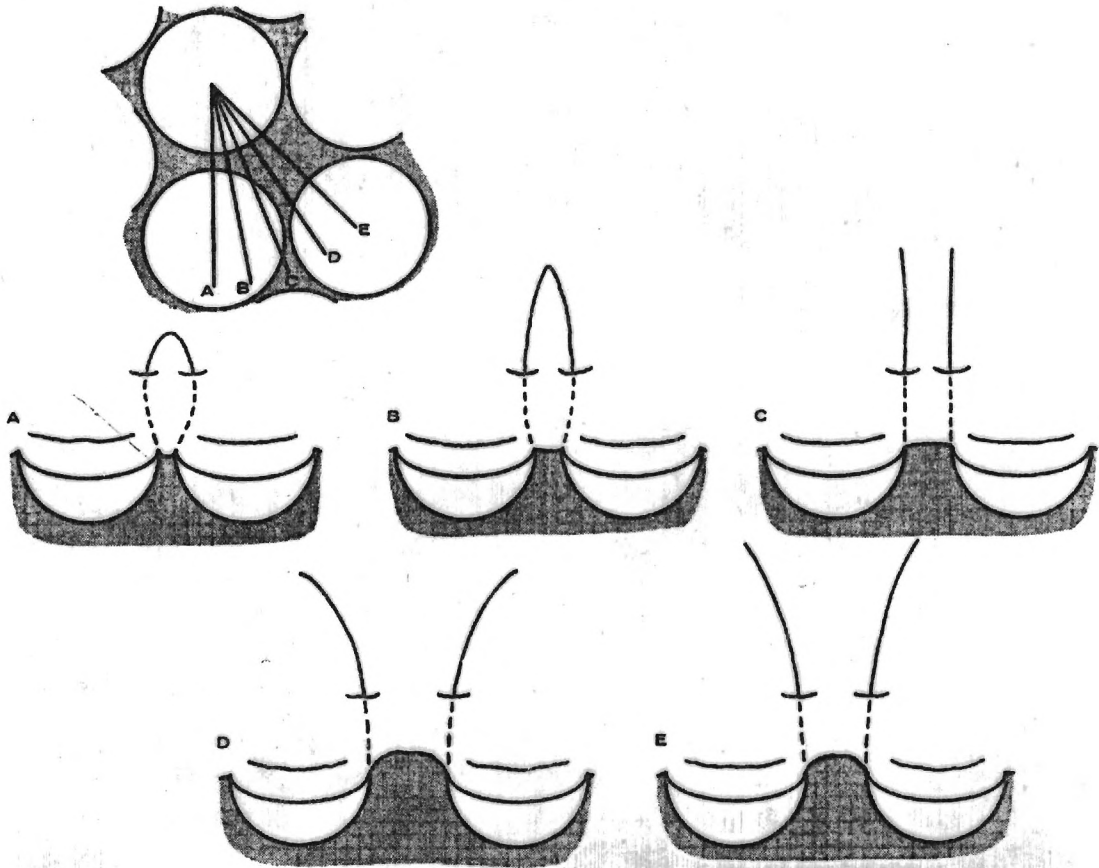


Fig. 2 Illustration of changing flame structure as a function of position around the periphery of a single AP particle (the perimeter of one particle is mapped out as a straight line to aid in displaying flame shape).



be used that chooses a minimum level of complexity of modeling of each that is compatible with its importance to the overall combustion response under prescribed conditions. For example, at low frequency the dynamic response is strongly affected by thermal accommodation in the solid (no matter which exothermic reaction region is considered). Under these conditions accurate three-dimensional modeling of the coupled response of exothermic steps and heat flow in the solid is important. On the other hand, at high frequency (e.g., 10 KHz) it is more important to model the transient response of the gas phase flames and their interactions with each other. As a further example, at low pressure, some of the exothermic steps are probably negligible at all frequencies (e.g., surface and gas flames for oxidizer (AP) particles). In this phase of the approach, such considerations are raised, and calculations are made to determine what level of rigor is required in modeling for each component of the flame complex under various conditions of propellant microstructure and flow environment.

### Modeling Responsive Components

For AP-hydrocarbon binder propellants, the exothermic components of the flame complex are the outer diffusion flame (region 12 of Fig. 1), the kinetically limited leading edge flame (regions 10-11), the AP flame (regions 6,7), and the AP surface froth reaction layer (region 4,5). It is the fluctuation in these flames that produce flow disturbances. Instability results from an organized response of the whole population of such microscopic flames, which can come about when they respond to macroscopic flow disturbances in the combustor. In this part of the study, models of the four exothermic regions will be developed that will allow analytical representation of their dynamic response to flow disturbances. Design of each model will be as simple as is consistent with an objective of correct qualitative description of reality, in order to "contain" the complexity of a model that describes their combined response. However, there are certain minimal requirements that useful models must meet in order to realistically represent perturbation behavior, and the trade-off between realism and mathematical tractability must be regarded as the hardest part of the problem.

### Statistical Representation of Responsive Components

When analytical representation of individual components of the flame have been achieved, one must then develop a description of populations of such flame components, keyed to the microstructure of the propellant. This will require a

statistical scheme, one that cannot be chosen until Items a-c are sufficiently advanced. Previous statistical schemes are not considered to be suitable, because they represent the populations in a way designed to provide local flame contributions to a one-dimensional energy conservation equation, and then calculate the dynamic response from a one-dimensional transient combustion model. In the present approach, the local dynamic response is needed, and that requires that a statistical model describe a population of local flame sites representative of the propellant microstructure.

### Collective Dynamic Response

A combination of detailed response dynamics (c) and statistical characterization of flame site population (d) will provide the means to calculate the macroscopic response to disturbances, as a collective response of individual sites in the flame population.

## PROGRESS

Progress to date is in three general areas.

1. Experimental investigation of combustion zone microstructure (AP-hydrocarbon systems).
2. Exploration of the potential of computational solution of the Navier-Stokes equation to determine the nature of oscillatory gas motion near the burning surface (i.e., at the site of the combustion zone).
3. Review of existing statistical combustion models and existing "response function" models, with a view toward construction of more realistic models.

As indicated in the Approach, initial effort has been heaviest on Item 1 (which corresponds to Item (a) of the Approach). Item 2 is a component of Item (c) of the Approach, and has been carried as far as contemplated in the proposal (and is now the basis of separate proposals for more detailed work). Item 3 is related to items (d) and

(e) of the Approach, and results to date have led to a decision to abandon all present statistical models and use a scheme that describes collective combustion response in terms of the combined dynamic response of individual flame sites in a population characteristic of the propellant microstructure.

### Combustion Zone Microstructure

These studies evolved from an ONR-funded study of combustion of AP-polymer sandwiches, which has led to an improved understanding of the flame complex that occurs in these systems. The present studies have been concerned with translation of this improved understanding to the context of a heterogeneous propellant, and to exploration of important aspects of propellant combustion that are not simulated in sandwich burning. Using theory and experiments, the following conclusions have been reached:

1. For characterization of oscillatory behavior, it is better to characterize the flame complex at any point on the surface as a local complex coupled to the oxidizer-binder interfaces rather than to the oxidizer particles.

2. The diffusion flames are usually viewed as being locally closed over the oxidizer particles. In the interface-based description, complete closure is impossible, as it will be over the binder regions where oxidizer particles are in near contact, but will transition to open flames further around the periphery of the oxidizer particle where adjoining binder areas are larger.

3. When considering a range of pressures and particle sizes, the nature of the flame complexes will change in qualitative ways (e.g., in a bimodal propellant, the flame complex associated with fine oxidizer particles will be entirely different than that with coarse particles).

4. The burning surface configuration of oxidizer particles and interface regions changes continuously as a function of time during burning of the particles and size of the particles. This has important implications for any scheme for averaging in statistical representations of combustion.

Some experimental results related to the foregoing arguments are shown in Fig. 3 and 4. Figure 3 shows the features of a burning surface (two pressures), and Fig. 4 shows AP particles that have been partially removed from the burned propellant surface to permit correlation of the surface features with the "time during burning" of the particle.

In Fig. 3 the differences in combustion zone structures at widely different pressures are suggested by the conspicuous differences in the burning surface. The propellant was one with bimodal particle size distribution. At low pressure the samples show surface areas consisting of protruding 400  $\mu\text{m}$  AP particles, and surface areas of binder/fine AP mixture; at low pressure the burning is controlled by the areas of binder/fine AP mixture and an as-yet-undetermined pilot flame complex in these areas. The coarse AP is pyrolyzed by the pilot flame complex, and "outer" O-F flames. At higher pressure, the self-deflagration flames of the large AP particles cause the AP to regress at a rate comparable to, or higher than the mean propellant burning rate and the AP particles have a "dished out" surface profile. The point to be made here is that the flame complex has very different characteristics at these different pressures, and one would expect very different dynamic response to flow disturbances. Likewise, the flame structure over the fine AP particles is entirely different than over the coarse particles, and at low pressure the two flame structures are strongly coupled. At both pressures, the surface features suggest that the flame complexes are three-dimensionally coupled with a three-dimensional thermal field in the solid, a feature that implies that dynamic response cannot be realistically represented by a one-dimensional representation of surface energy flux and heat flow in the solid.

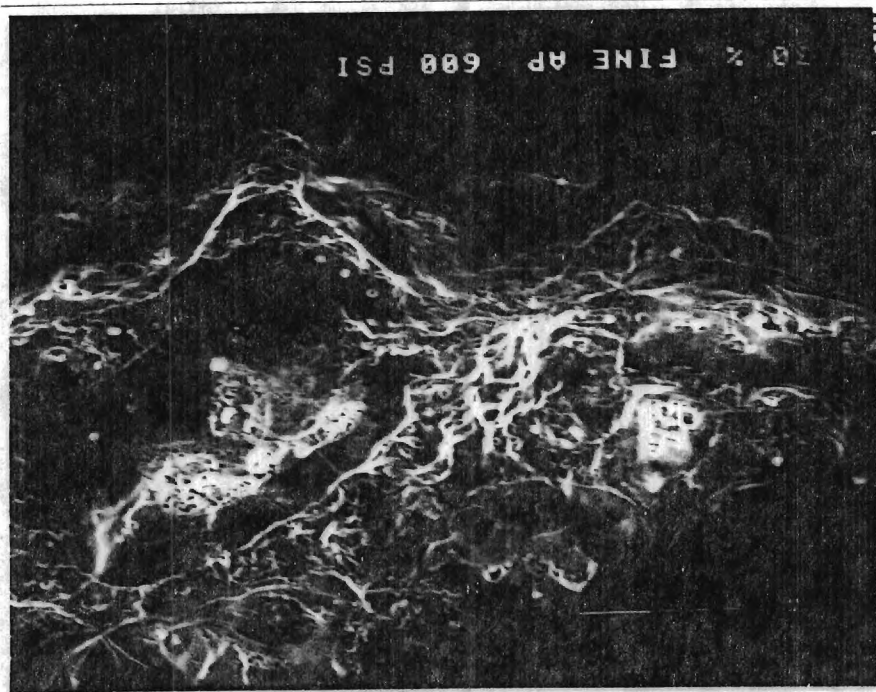
In Fig. 4, the burning process of individual AP particles is seen to be one of progressive change. In part A, a particle is shown that has started to burn at three different sites. At this point, surface features suggest that quasi-steady self deflagration is already approached at these sites. In part B, the particle is half burned and has become relatively flat, protruding some at the edges where we have argued that the AP is pyrolyzing rather than self deflagrating. The central region has the appearance of AP self deflagration. In part C, the particle is 80/90% burned, and is convex downward. Thus the surface configuration has experienced a progressive change during burning.



Fig. 3 Comparison of features of the burning surface at low and intermediate pressure (propellant has 70/30 ratio of 400  $\mu$ m and 49  $\mu$ m AP, in an 87.5/12.5 ratio of AP and PBAN binder).

a) Quenched from 60 psi

b) Quenched from 600 psi



(a)





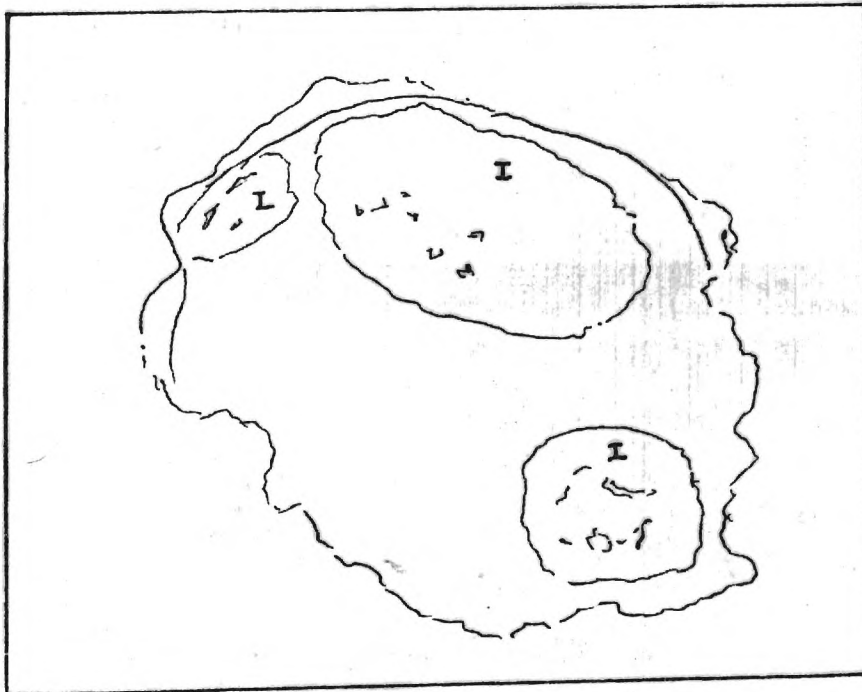
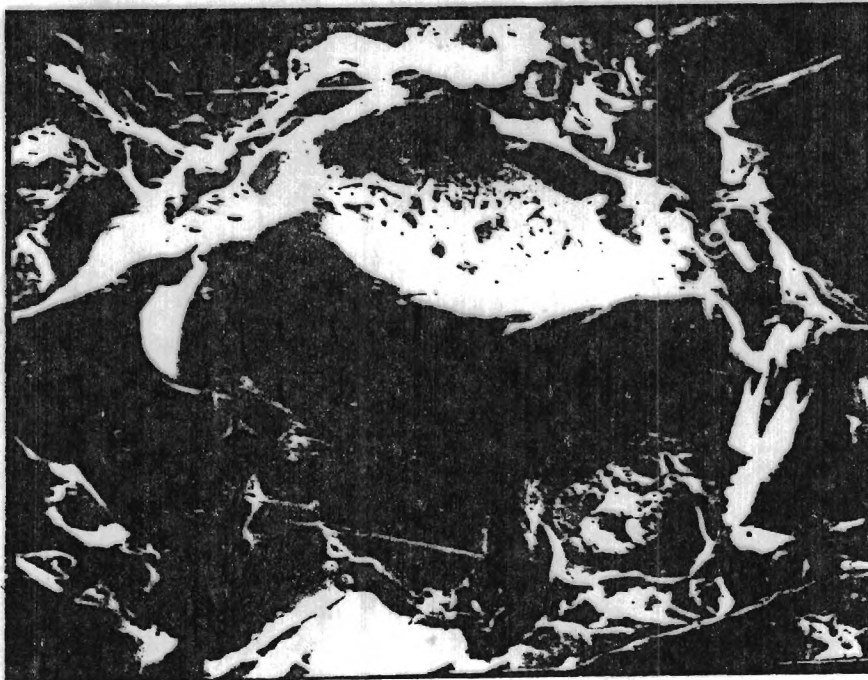
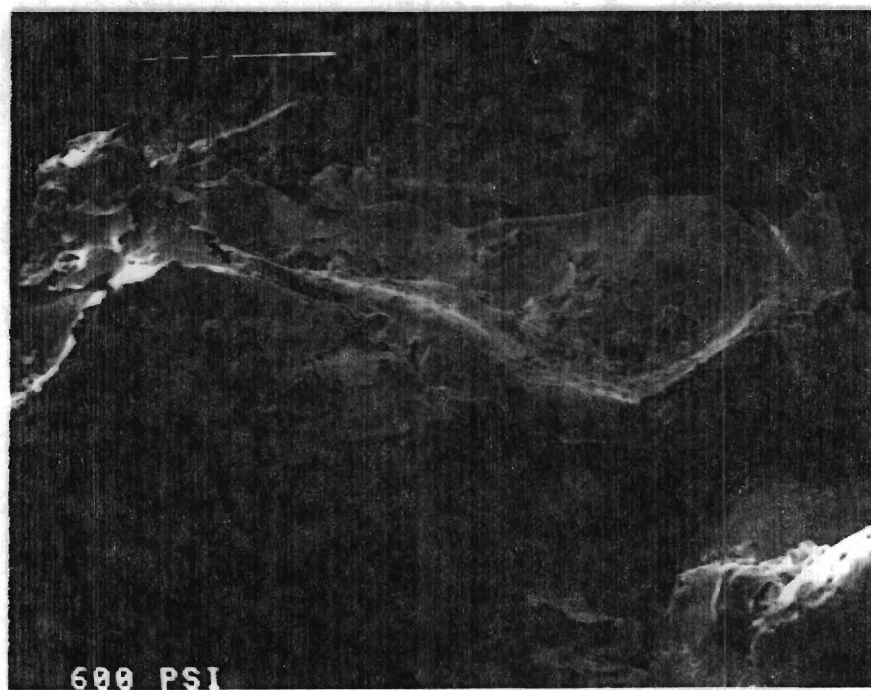


Fig. 4 AP particles quenched at different times during burning (same propellant as Figure 3, quenched from 600 psi).

a) Oxidizer particles, partially separated from the surrounding matrix. Example is early in burning of the particle, and shows three areas that have started to burn, denoted by "I" in the sketch. The particle is 400  $\mu\text{m}$  diameter.



(b)



(c)

Fig. 4 AP particles quenched at different times during burning (same propellant as Figure 3, quenched from 600 psi).

- b) Quenched about halfway through burning;
- c) Quenched late in burning of particle.

### Solution of Navier-Stokes Equation for Oscillatory Flow with Mass Addition

The goal of this effort is to develop a proposal for an investigation leading to computation of oscillatory gas motions near the burning surface of a cylindrical charge during axial mode oscillations of the combustor. The approach was to replace the combustion by direct mass transpiration and calculate the mean and oscillatory flow field assuming viscous, nonturbulent flow. A numerical method was to be chosen that would permit realistic modeling of the near-wall behavior. A steady state solution would first be obtained for a given mass transpiration rate. Then a specific, constant amplitude oscillation of mass flow rate would be imposed over a selected circumferential section of the wall, and the growth of oscillations in the system would be calculated for several frequencies and a first resonant axial mode frequency would be identified. The nonsteady flow field would then be calculated in detail over a few cycles of oscillation, allowing the near-wall motion to be related to the mainstream motions. Of particular interest is the shearing motion near the surface, and its phase relative to the mainstream (axial) velocity. The results would also show the effect of the mean flow on the acoustic pressure field near the walls. Such information is essential to realistic modeling of combustion zone response to mainstream oscillation.

Progress to date consists of development of the above concept for the investigation, exploration of computational methods that a) are compatible with the no-slip, mass addition boundary condition, b) will permit computation with relatively high spatial resolution near the walls, and c) will be compatible with available computers. A computer program was written that will generate a suitable coordinate grid for this particular problem (Fig. 5). In addition, a program which performs a numerical integration of the Navier-Stokes equations using a well-known procedure has been obtained from the Air Force Flight Dynamics Laboratory. The procedure has second-order accuracy in both space and time and has successfully captured the natural vortex shedding (Karman vortex sheet) from a circular cylinder in an incompressible flow as well as the pressure oscillations from an open cavity in a compressible free stream. The method thus appears capable of capturing the above oscillatory flow behavior. A proposal was submitted to ARO and Army BMD for continued work on this problem. No further work is planned on the present contract.

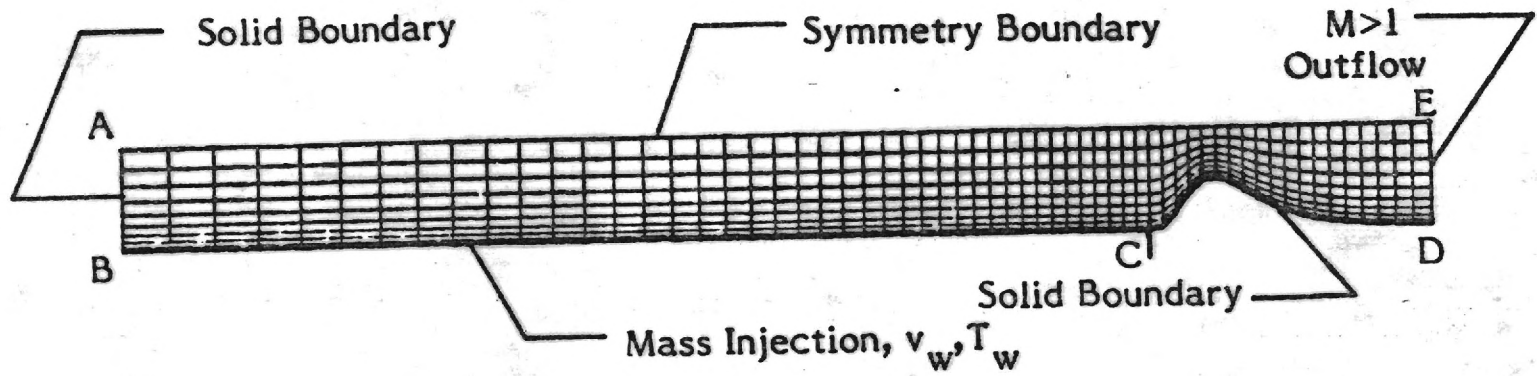


Fig. 5 Illustration of grid spacing arrangement for viscous flow computations.

### Review of Existing Models: Conclusions and Plans

Considerable time has been spent in review of existing response function models and comparison with our current understanding of combustion zone structure. We have reached several conclusions that figure in future efforts.

1. No one tractable model is suitable over a wide range of propellant variables, pressure, etc.
2. For AP-polymer propellants, the multiple flame complex is significantly different than that used in the Beckstead-Derr-Price model (see Fig. 6).
3. Realistic response function modeling (AP-polymer propellants) needs to describe gas phase response as locally coupled to surface features and local multidimensional heat flow.
4. The local flame complex (Fig. 7) contains certain features that are expected to be particularly responsive to flow disturbances, features not considered in current models. These features (labeled A,B,C in Fig. 7) are quench regions at the lateral extremities of transverse flamelets, and response is postulated to include oscillation in lateral extent of those flamelets.
5. Consistent with 3) above, it is believed to be necessary to consider variation of the local flame complex as a function of peripheral distance around the exposed surfaces of each AP particle (i.e., we do not believe it is realistic to model the flame as burning of a circular AP surface surrounded by a uniform ring of binder). The real situation does not permit complete closure of the diffusion flame over the AP particles, as the diffusion flame sheets tend to occur locally over the binder in peripheral locations where the binder is thin. The dynamic response of such a three-dimensional flame complex is not likely to be well simulated by an axially symmetrical flame assumed in current models. It will still be necessary to identify the flame complex with individual oxidizer particles because it is the known oxidizer size distribution that characterizes the propellant heterogeneity; but a more realistic description of the local flame is needed.



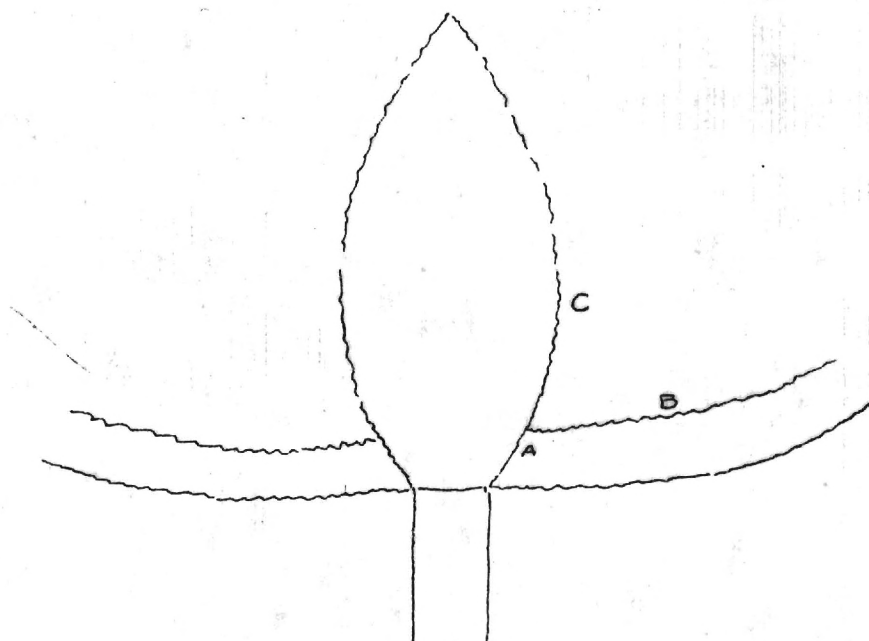


Fig. 6 Sketch of flame complex used by BDP model. (A) designates "primary" kinetically controlled O-F flame; (B) designates AP flame; (C) designates O-F diffusion flame.

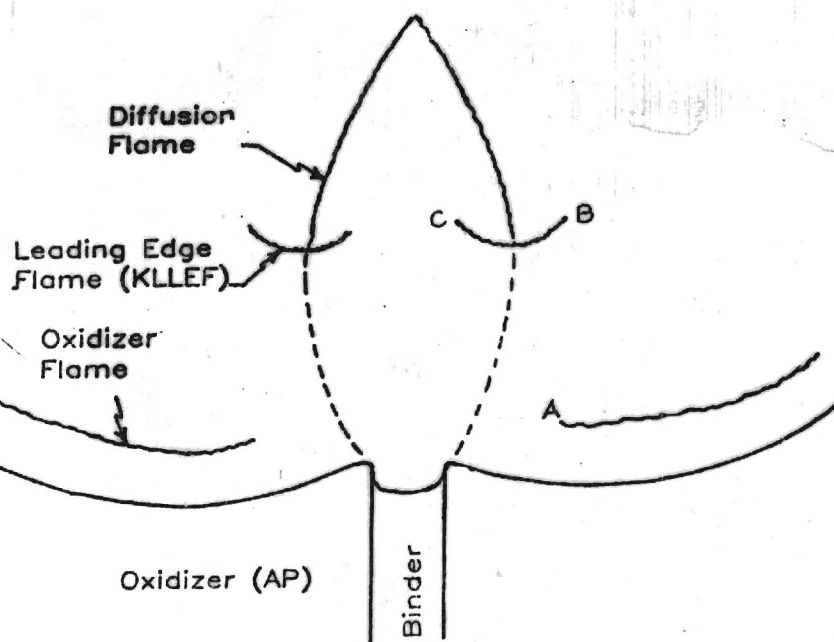


Fig. 7 Quench limits in flame complex of Figure 1. (A) designates lateral limit of AP flame; (B) designates lateral limit of KLLEF on oxidizer-rich side; (C) designates lateral limit of KLLEF in fuel-rich side.

6. Assumptions in present models regarding geometry of the exposed oxidizer surfaces do not take into consideration the burning history of real AP particles (i.e., the models assume the shapes of the surfaces and generally replace the real population by an average one). In practice, the character of the exposed oxidizer surface is strongly dependent on pressure, particle size, and time during burning of the particle. The local multiple flame complexes are correspondingly dependent. It is not clear how well these realities can be modeled, but it seems clear that the population of flame complexes that are perturbed in a modeling scheme should reflect such features of the real burning surface unless they can be shown to be unimportant.

A good analysis of combustion-flow interaction should be as realistic as possible within practical limits. The practical limits are set by 1) the state of knowledge, 2) computational capacity, and 3) the importance of the end objective. We do not know yet what will emerge as a suitable compromise of those three factors. We are in a much better situation relative to 1) than at the time of development of previous models, but uncertain as to how much of the improved knowledge can or should be included in new models. Computational capability has reached the point where cost of computer time is the dominant computational limitation, although lack of "user friendliness" has been a serious problem with past programs and deserves more attention. Regarding importance of the end product, we believe that the goal should be a family of models designed to fit different combinations of oxidizer particle size, pressure range, O-F ratio, catalyst content, etc. These may be subroutines in a master model and computer program, but that option depends on a continued perception of need, which can only be assessed as the model develops and experimental results from burner tests and developments demonstrate need and relevance.

E-16-648

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DANIEL GUGGENHEIM SCHOOL  
OF AERONAUTICS

13 June 1986

TO: Dr. D. A. Flanigan  
 Director, Advanced Technology  
 Aerospace Group  
 Morton Thiokol, Inc.  
 3340 Airport Road  
 Ogden, Utah 84405

FROM: E. W. Price, Principal Investigator

SUBJECT: Annual Report on Contract P. O. 0584, Amm. 31

This letter is to report progress on Morton Thiokol Contract P. O. 0584, Amm. 31, entitled, "Study of Oscillatory Interaction of Solid Propellant Combustion and Combustor Flow."

Current studies are on the following topics:

1. Analysis of the edgewise pyrolysis of laminates.
2. Preparation of a proposal for an experimental evaluation of a critical feature of combustion zone response to pressure oscillation.

Details of completed work are contained in Enclosures A and B, and summarized briefly below along with ongoing work.

1. Edgewise pyrolysis of laminates: The initial phase of this study was to determine the steady state pyrolysis of a laminate of two different materials (e.g., fuel and oxidizer) under simplistic assumptions of heating (no reaction except surface pyrolysis, no lateral heat flow, and a uniform heat source moving with the pyrolyzing surface). The objective was to provide some idea of the relative temperatures and locations of the lamina surfaces, and how they depended on pyrolysis kinetics and energetics, properties of the solids, and pyrolysis rate. It is the results of this analysis that are summarized in Enclosure A. We are now looking at the problems modified by allowance for nonsteady conditions and heat transfer between laminae (two-dimensional in the solid phase). The objective is to be able to calculate surface profiles for a specified surface heating situation, and calculate the oscillations in profile and ingredient product flows under oscillatory heating conditions. The imposed surface heating is intended to simulate flame oscillations, but is being kept geometrically simple for the present. The results will help in estimating flame response; the ultimate goal is to develop a model for a coupled flame-pyrolyzing solid system.

2. Proposal for Experimental Combustion Response Study: This proposal (Enclosure B) is concerned with certain features of the flame complex of a composite propellant that are considered to be important to combustion response, but that are absent in any existing models. The goal is to demonstrate experimentally that the postulated flame behavior is important (or not), in order to guide realistic modeling of combustion response. The work is advanced as a separate proposal because the proposed experimental work is beyond the scope of the present contract, and well suited as a project at Morton Thiokol. It is suggested that funding be sought for this work as a joint Morton Thiokol-Georgia Tech project. It should be noted that the original concept for this work evolved from Navy-sponsored studies of combustion zone microstructure. In that work it was predicted from theoretical arguments regarding flame structure that the burning rate of certain propellants should show abnormally high sensitivity to pressure and oxidizer particle size for certain pressure-particle size ranges. It is also likely that the dynamic combustion response will be very large under these same conditions of particle size and pressure. In the earlier study, the predicted trend in steady state burning rate was verified experimentally, supporting the original mechanistic argument. The more important effect on the response function has not been verified yet. The present proposal is to conduct a series of T-Burner tests designed to evaluate the trend of the pressure-coupled response function over the critical range of pressure and particle size already determined by the steady state burning rate tests. Successful completion of the tests will, for the first time, demonstrate a clear link between combustion response and a specific "unstable" aspect of the flame complex; verification of this heretofore unrecognized mechanism is needed for flame modeling work currently in progress on the present contract.

### 3. Plans for Continuing Work


Our long-term plan is to develop a model of oscillatory combustion that is more realistic than present ones, based on the kind of flame complex developed from the sandwich burning and bimodal propellant studies. In the condensed phase, we are working now on a two-dimensional transient model for heat transfer and surface pyrolysis, using the 1-D model reported here as an aid to determining how the 2-D problem should be set up. We are looking for simplistic descriptions of the mixing fans that are present in the flow away from ingredient interfaces, descriptions that remain realistic enough to serve as a basis for a description of KLEF location and lateral extent (i.e., how much of the O-F mixture is consumed in the KLEF, how close is it to the surface, and how do these features of the KLEF oscillate?). A realistic model must describe the conditions under which the KLEFs detach, and describe their nonlinear oscillatory behavior when they are near detachment conditions. Other aspects of the flame complex are not being examined at present because their transient behavior is less obscure (AP flame), or less important (diffusion flame). Finally, the global response must be represented as the collective response of the population of flame complexes from the population of propellant microstructures on the burning surface. The statistical schemes for getting from local flame response to global combustion



Dr. D. A. Flanigan  
13 June 1986  
page 3

response are viewed as fundamentally straightforward, and we are preoccupied with the more difficult job of understanding and modeling the population of local microflame complexes, and modeling the dynamic response of the members of the population.

Sincerely,

  
Edward W. Price  
Regents' Professor  
School of Aerospace Engineering

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prcpt.th2

Enclosures:     A. Paper: "Steady State One-Dimensional Pyrolysis of  
                  Oxidizer-Binder Laminates"  
                  B. Proposal for Experimental Combustion Response Study

# AIAA'86

**AIAA-86-1446**

## **Steady State One-Dimensional Pyrolysis of Oxidizer-Binder Laminates**

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and  
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Georgia Institute of Technology  
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**AIAA/ASME/SAE/ASEE 22nd Joint  
Propulsion Conference**

June 16-18, 1986/Huntsville, Alabama

STEADY STATE  
ONE-DIMENSIONAL PYROLYSIS  
OF OXIDIZER-BINDER LAMINATES

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and

E. W. Price\*\*

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Abstract

The steady state burning of a solid propellant is modeled to determine the protrusion of one ingredient relative to the other on the burning surface and to determine the difference in temperature of the oxidizer and fuel surfaces. The propellant is represented by a laminate solid (alternating layers of oxidizer and binder), burning edgewise to the layers. The analysis assumes that the surface is heated by a constant-temperature planar heat source, that heat flow is one-dimensional, and that each lamina pyrolyzes on its edge according to an Arrhenius rate law and heat of phase change. Sources for input parameters are discussed, choices made, and the corresponding relations among lamina heights and lamina surface temperatures are presented. The implications of the results relative to the role of subsurface heat flow between ingredients are examined and related to experimental results from combustion studies of ammonium perchlorate-polymer sandwiches. The results indicate that subsurface lateral heat flow between ingredients is a significant factor in combustion.

Nomenclature

Symbols

A	pre-exponential or pre-factor
$c_p$	specific heat
$E_s$	activation energy
L	heat of vaporization
m	mass flow rate
r	regression rate
R	gas constant
s	standoff distance
T	temperature
x	distance into condensed phase
$\rho$	density
$\lambda$	thermal conductivity

Subscripts

f	flame, fuel
g	gas phase
o	initial propellant conditions
ox	oxidizer
s	surface

Introduction

In modeling the combustion of composite solid propellants, it is generally necessary to consider the heterogeneity of the solid and the difference

in pyrolysis behavior of the constituent ingredients. However, the geometrical complexity of the propellant and the burning surface on the micro-scale is so great that it is difficult to measure, describe or calculate the behavior of the individual ingredients or their interactions. Most analytical models consider chemical heterogeneity only in the gas phase and neglect surface and subsurface effects such as non-flat surfaces with non-uniform temperatures or heat flow between ingredients below the surface.

Some insight can be gained into the effect of a non-homogeneous solid on surface and subsurface processes by going to a simpler distribution of reactants such as a "laminate propellant" made up of alternate layers of oxidizer and fuel (Fig. 1). When burned edgewise to the laminae, the surface will regress with a two-dimensional configuration, with easily pyrolyzed ingredient laminae recessed between laminae of the less easily pyrolyzed ingredient. Extensive experimental studies have been made of edge-burning sandwiches of binder laminated between sheets of ammonium perchlorate<sup>(1)</sup>, which show the surface profiles that actually occur. However, full two-dimensional analytical representation of burning of such systems is still very difficult, and there is a need for simple analyses that provide explanation of qualitative aspects of the sandwich burning and aid in the tough decisions about what must be included in more rigorous models. The present analysis represents the simplest model of laminate burning that can be made.

Suppose the laminae are separated by partitions that are perfect insulators, and the flame complex is replaced by a planar, constant temperature heat source that advances at a constant speed in the direction of the laminae interface plane. The laminae are heated by conduction from the "flame" and vaporize at the surface of the lamina edges, which recede at the same velocity as the advancing heat source plane. Then one can ask two practical questions of this system that are of interest in real burning laminates:

1. How do the surface temperatures of the two kinds of laminae compare?
2. What are the relative positions of the lamina edge surfaces?

From the results, one can reason qualitatively about how the surface profile will differ when lateral heat transfer is present and how those differences will change when different binder lamina thicknesses are considered. This reasoning will, in turn, support and guide the interpretation of experimentally observed surface profiles and guide in development of two-dimensional steady and non-steady models of laminate pyrolysis and burning. This, in turn, should be helpful in

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modeling combustion of more geometrically complex systems such as heterogeneous propellants.

### Analysis

Figure 1 shows the arrangement of the problem under consideration. It consists of one-dimensional pyrolysis of homogeneous slabs, but is pictured as an alternating series of thin slabs in order to help visualize pairs of solutions corresponding to adjoining laminae of different material. Mathematically, all the slabs (laminae) obey the same pyrolysis law, but with different values of physical and kinetic parameters. Each slab in the laminate has the same surface regression rate, i.e., the surface has achieved a steady state of regression and profile, and is heated by the same planar heat source. The laminae are otherwise assumed to be insulated from each other, and each establishes its own distance between surface and heat source independently according to its pyrolysis characteristics. The surface energy balance for a lamina (either oxidizer or fuel) will be specified as:

$$\lambda_g \left( \frac{dT}{dx} \right)_{o+} = m [ L + c_p (T_s - T_o) ] \quad (1)$$

where  $\lambda_g$  is the gas phase thermal conductivity,  $m$  is the mass flow rate per unit area leaving the surface,  $L$  is the heat of vaporization,  $c_p$  is the condensed phase specific heat,  $T_s$  is the surface temperature, and  $T_o$  is the initial condensed phase temperature.

The temperature gradient in the gas at the surface of a lamina will be approximated by the simple conductive heat transfer expression:

$$\left( \frac{dT}{dx} \right)_{o+} = \frac{T_f - T_s}{s} \quad (2)$$

where  $s$  is the distance from the heat source plane to the lamina surface in steady state (the effect of convection is neglected for mathematical simplicity). The requirement of steady state also implies that the surface regression rates of adjoining laminae be equal, so:

$$r_{ox} = m_{ox}/\rho_{ox} = m_f/\rho_f = r_f \quad (3)$$

where we now distinguish where necessary between adjoining laminae as oxidizer and fuel ( $\rho$  is the density of the solid).

The mass flow rates from the laminae are assumed to depend on surface temperature according to an Arrhenius rate law:

$$m = A e^{-E/RT_s} = \rho r \quad (4)$$

where  $A$  and  $E$  are kinetic constants for the

surface decomposition process and  $A$  and  $E$  are different for oxidizer and fuel.

From the foregoing equations, an expression can be obtained for the distance between a lamina edge surface and the heat source (two forms of the equation are useful):

$$s = \frac{\lambda_g}{A} \frac{T_f - T_s}{L + c_p (T_s - T_o)} e^{E/RT_s} \quad (5a)$$

$$\frac{\rho c_p}{\lambda_g} sr = \frac{T_f/T_o - T_s/T_o}{L/c_p T_o + T_s/T_o - 1} \quad (5b)$$

where  $T_f$ ,  $T_o$ ,  $r$ , and  $\lambda_g$  are parameters in the problem;  $L$ ,  $c_p$ ,  $A$  and  $E_s$  are properties of the lamina material; and  $T_s$  is to be determined by another equation.

From Eq. (3) and (4), an equation for the surface temperature can be found:

$$T_s = \frac{E}{R \ln (A/r\rho)} \quad (6)$$

Since we are particularly interested in relative values of  $s$  and  $T_s$  for different laminae, the following form of Eq. (5) is useful (where the subscripts "ox" and "f" are added to distinguish between oxidizer and fuel laminae):

$$\frac{s_{ox}}{s_f} = \frac{(T_f - T_{sox})}{(T_f - T_{sf})} \cdot \frac{L_f + c_{pf} (T_{sf} - T_o)}{L_{ox} + c_{pox} (T_{sox} - T_o)} \cdot \frac{A_f e^{-E_f/RT_{sf}}}{A_{ox} e^{-E_{ox}/RT_{sox}}} \quad (7)$$

where the last factor is also  $\rho_f/\rho_{ox}$  in the present problem in which  $r_f = r_{ox}$ . Equation (7) gives the ratio of surface-flame standoffs as a function of  $T_{sf}$  and  $T_{sox}$ . From Eq. (6), the difference in lamina surface temperature is

$$T_{sox} - T_{sf} = \left[ \frac{E_{ox}}{\ln (A_{ox}/r\rho_{ox})} - \frac{E_f}{\ln (A_f/r\rho_s)} \right] \frac{1}{R} \quad (8)$$

The difference between surface-flame distances is

$$s_{ox} - s_f = s_f \left( \frac{s_{ox}}{s_f} - 1 \right) \quad (9)$$

where  $s_{ox}/s_f$  is given by Eq. (7).

It is Eqs. (5b), (8) and (9) that are of particular interest, because of the nature of related experimentally determined information. Specifically, interrupted burning tests on edge-burning laminates<sup>(1)</sup> provide the opportunity to measure  $s_{ox}-s_f$  for various combinations of oxidizer and binder and various combustion pressures. Regarding  $T_{sox}-T_{sf}$ , there is general consensus on approximate values for  $T_{sox}$ .<sup>(2)</sup> Values for  $s$  and for  $T_{sf}$  are more controversial, and there is a particular need to know values for  $T_{sox}-T_{sf}$  in order to determine the importance of considering a more rigorous representation (2-D) of heat flow in the solid phase, i.e., remove the assumption of insulation between laminae. Before examining these issues, attention is temporarily diverted to the question of choice of reasonable values of the parameters in the problem, i.e., values for  $L$ ,  $c_p$ ,  $A$ ,  $E_s$ , and  $T_f$ .

### Choice of Parameters

In the present study, the differences  $s_{ox}-s_f$  and  $T_{sox}-T_{sf}$  are of primary interest and may be viewed as the dependent variables of the problem. The objective is to determine the plausible range of these variables. It will be assumed that the oxidizer is ammonium perchlorate, because there is substantial data available about AP decomposition and self deflagration and substantial data on combustion of AP-polymer sandwiches. Reasonable estimates can be made for values of  $c_p$ ,  $\rho$  and  $\lambda_g$  of both AP and polymeric binders. Values of  $T_s$ ,  $L$ ,  $A$  and  $E$  are matters of continuing debate, with some experimental data available but of uncertain relevance. The value of  $r$  (range of values) has been adequately measured in combustion experiments as a function of variables such as pressure, so that a relevant range is known; however, relevant combinations of  $r$ ,  $T_s$ ,  $A$  and  $E$  for Eq. (4) or of  $L$ ,  $r$  and  $s$  for Eq. (5) have not been established. The heats of phase change,  $L$ , are subject to some reasonable (but broad) limiting arguments, but not to accurate measurement (partly because the concept of a simple heat of phase change is an oversimplification of the surface vaporization process). In the following, comments are made on limits of knowledge of parameters and on rationales for choice of reasonable combinations of values for the present objectives. On the basis of these rationales, the subsequent section will examine plausible ranges of  $s_{ox}-s_f$  and  $T_{sox}-T_{sf}$ . Because there is a better consensus on the values of  $E$ ,  $T_s$  and  $L$  for the oxidizer than for the fuel, the first comments will be on "oxidizer values".

### Values of Parameters for Ammonium Perchlorate

Studies on AP decomposition and deflagration show a range of values of  $E_{ox}$  that centers on 30.5 kcal/mol (see Table 1 and its references). Burning rate in the pressure range 3-10 MPa is 0.3 to 1.0 cm/sec<sup>(1,3)</sup>. A value of  $T_{sox}$  of around

870° K is plausible in this range<sup>(2)</sup>, but measurements are not good enough to provide  $T_{sox}$  as a function of  $r$ .  $A_{ox}$  is not measured independently of  $E_{ox}$ ,  $T_{sox}$  and  $r$ , and is highly sensitive to choice of  $E_{ox}$  and  $T_{sox}$  (see later).  $L$  represents a surface gasification that can range from dissociative sublimation at low pressure (endothermic) to exothermic decomposition in a surface melt-froth layer.<sup>(4-6)</sup> A pure endothermic sublimation would exhibit a value of  $L$  of 495 cal/g.<sup>(10)</sup> The exothermic decomposition could have a value of  $L$  of -1075 cal/g if the complete heat of reaction of the AP were released at the surface.<sup>(4,5)</sup> Some guidance in proper choice of a value of  $L_{ox}$  within the above broad limits can be obtained from results of Ref. 1 (discussed in Appendix A of the present paper). It is argued that the mode of AP decomposition changes as a function of distance (on the burning surface) from the binder lamina, with a corresponding change in  $L_{ox}$  from around 0 cal/g near the binder lamina to -90 cal/g at a location about 50  $\mu$ m out from the binder lamina (details dependent on pressure). These values are estimated in Appendix A by choosing typical values of sandwich burning rate ( $r$  of 0.5 cm/sec) and flame standoff distance ( $s$  of 50  $\mu$ m) corresponding to sandwich burning tests at 3.5 MPa.

### Kinetic Parameters

Equation (4) can be written as:

$$A/r\rho = e^{E/RT_s} \quad (10)$$

which is shown graphically in Fig. 2. Compatible values of  $(A/r\rho)$  and  $E/RT_s$  are thus on the curve in this figure. A point on the curve labelled OXIDIZER is typical of AP with  $\rho_{ox}$  of 1.95 g/cm<sup>3</sup>,  $r$  of 0.5 cm/sec,  $T_{sox}$  of 870° K,  $E_{ox}$  of 30.5 kcal/g. For this point  $A/r\rho$  of  $4.89 \times 10^7$  and  $A_{ox}$  of  $4.77 \times 10^7$  g/cm<sup>2</sup> sec. For the binder lamina, assuming the same value of  $T_s$  as for the oxidizer and a value of  $E_f$  of 16.7 kcal/mol,  $(A/r\rho)_f$  of  $1.62 \times 10^4$  and for the above value of  $r$  and the value of  $\rho_f$  in Table 1,  $A_f$  of 9617 g/cm<sup>2</sup> sec. In the quenched sandwich tests<sup>(1)</sup> it was noted that the binder is generally recessed between the adjacent AP laminae, which suggests that  $T_{sf} < T_{sox}$  (this is confirmed by a two-dimensional analysis in Ref. 7). Thus, the value of  $A_f/\rho_f r$  noted above (that was based on  $T_{sox}$  of  $T_{sf}$ ) is probably too low, and the range of plausible values of  $A_f/\rho_f r$  indicated in Fig. 2 includes substantially higher values than that noted above. It should be noted in the figure that a range of plausible values of  $E_f/T_{sf}$  is indicated that



reflects not only uncertainty of  $T_{sf}$ , but also uncertainty in the value of  $E_f$  for any particular binder and differences among binders. This results in an enormous range of uncertainty in  $A_f/\rho_f r$ . However, the necessity to satisfy Eq. (4) for a value of  $r$  that is often experimentally known provides a means to assure choice of reasonable combinations of  $A_f$  and  $E_f$ .

### Results and Discussion

The present analysis describes the relative heights and surface temperatures of unlike laminae during steady pyrolysis, neglecting the heat flow between unlike laminae. Using two reasonable combinations of parameters for AP-polymer laminates, the relation between  $s_{ox}-s_f$  and  $T_{sox}-T_{sf}$  is shown for several values of  $L_f$  in Figs. 3a and 3b. The conditions in Fig. 3a were chosen to correspond to a sandwich burning experiment at 3.5 MPa. Choosing reasonable values of  $L_f$ , this figure shows that if the binder and oxidizer pyrolyze with approximately equal standoff distance from the heat source (as observed in sandwich burning experiments at this pressure), the surface temperatures of the oxidizer laminae are appreciably higher than those of the binder. If one were to allow for lateral heat flow, the binder near the oxidizer would be heated by lateral subsurface heat flow, while the oxidizer near the interface would be cooled. This would cause the oxidizer to protrude, and cause the binder to be recessed in the region of the contact plane (Fig. 4a). The width of the affected region would be of the same order as the thermal wave thickness in the one-dimensional pyrolysis. Further, if the binder lamina thickness were comparable to the thermal wave thickness, the entire lamina would be recessed (Fig. 4b). This is consistent with observations in tests on AP-polymer sandwiches at pressures in the 2-5 MPa range. (1)

In Ref. 1, it was proposed on the basis of several observations and arguments that the protrusion of the AP was enhanced by a local shift in the AP decomposition toward a less exothermic process. The present analysis (Fig. 3b) indicates that a less exothermic or an endothermic process would indeed lead to a protrusion of AP relative to the binder, along with a lowering of the difference in surface temperature. In other words, the shift in local nature of the AP decomposition, which was postulated in Ref. 1 to be a consequence of the lowered surface temperature, further enhances the local protrusion of the AP near the contact plane. Determination of the quantitative details would require a two-dimensional analysis.

At higher pressure, the extent of heat release at the AP surface becomes greater ( $L_{ox}$  shifts to larger negative values). Referring to Eq. (5b) or to Fig. 5, if:

$$L_{ox} = -c_{pox} (T_{sox} - T_o) \quad (11)$$

$s_{ox}$  goes to infinity. Physically, this corresponds to the situation where the burning rate  $r$  is maintained with a surface temperature  $T_{sox}$  by the surface heat release alone, i.e., without the O-F flame. This situation is apparently reached at around 7 MPa with sandwiches. At higher pressure, the sandwich burning rate is the same as that of pure AP, implying that the O-F flame is no longer contributing to heating of the leading front in the AP. The limiting value of  $L_{ox}$  reached at 7 MPa must be around -198 cal/g (based on Eq. (11) and  $T_{sox}$  of 870° K). The burning rate continues to increase with pressure in the manner of pure AP. It is significant that, at these higher pressures, the AP adjoining the binder lamina protrudes increasingly with pressure (Ref. 1, Fig. 4c), reflecting a continued heat loss to the binder lamina, lowered AP surface temperature (relative to that at the leading front) and presumably a less exothermic decomposition at the AP surface (compared to the leading front). The O-F flame is increasingly distant from the leading front of the AP, consistent with the behavior of  $s_{ox}$  in Fig. 5 when Eq. (11) applies. While the details cannot be calculated from the present model, the relative location of oxidizer and binder surfaces (and relative temperatures) in the neighborhood of the contact plane are better estimated by Fig. 3b than 3a, because Fig. 3b is based on a value of  $L_{ox}$  reflecting the retarded AP decomposition in that region.

### Appendix A: Combustion Zone Structure and Size

Considerable information on combustion zone structure and characteristic dimensions is presented and can be inferred from Ref. 1. In this reference, results of studies of edge-burning of AP-polymer-AP sandwiches are reported. Surface profiles obtained from quench tests are described, and a flame complex is inferred from consolidation of a combination of experimental results and theory. Fig. A-1 is reproduced from that reference and shows certain features of the combustion zone that pertain to choice of relevant values of parameters in the present simplified model. In the tests leading to the figure, the thickness of the binder lamina was typically 50  $\mu$ m. The oxidizer adjacent to the binder protruded roughly 10  $\mu$ m above the binder, and the surface then sloped off roughly 10  $\mu$ m over a lateral distance of 25-50  $\mu$ m. (These features depended quantitatively on pressure, with the numbers here typical of 3.5 MPa). Further from the binder, the AP surface slopes upward and regresses perpendicularly to its surface at a rate characteristic of AP self-deflagration (indicating that influence of the O-F flame drops off beyond the recessed region). The "leading front" in the AP represents a region where the combined effect of AP exothermic decomposition and heat flux from the O-F flame maximize the regression rate. This requires that the O-F flame be close to this part of the AP surface, and yet far from the AP surface 100  $\mu$ m further out (where the AP rate is not enhanced by O-F flame heat). This would only be possible if the part of the O-F flame responsible for the AP rate enhancement were a flame of limited lateral extent, located close enough to the surface to heat the

enhanced rate region and yet exert little effect 100  $\mu\text{m}$  further out from the binder. From an examination of the geometry, this would require that the effective part of the O-F flame be located about 50  $\mu\text{m}$  above the surface. This portion of the O-F flame was characterized in Ref. 1 and in Fig. A-1 as a kinetically limited leading edge flame "KLLEF", part of the O-F diffusion flame, and was pictured as a localized flame sheet oriented "parallel" to the surface. Since it is desired that the present analysis be pertinent to this micro-domain of the edge-burning laminate, the flat heat source in the present analysis takes the place of this KLLEF.

In Ref. 1, it was argued that in the region of protruding AP immediately adjacent to the binder lamina, the AP seemed to be decomposing without the surface froth characteristic of exothermic surface processes. In the present paper, this leads to consideration of a range of values for heat of vaporization of the oxidizer, ranging from positive values (endothermic decomposition) to negative values (exothermic decomposition).

To examine further, Eq. (5b) was used to show how  $L_s$  and  $T_s$  are related for a given burning rate (see Fig. 5). Referring to this figure, flame standoff of 50  $\mu\text{m}$  with an oxidizer surface temperature of 870° K corresponds to a value of  $L_{ox}$  of -90 kcal/g. This seems to be a reasonable combination of  $s$ ,  $T_{sox}$  and  $L_{ox}$  for the AP at the "leading front". The AP closer to the binder lamina typically protrudes 5-10  $\mu\text{m}$  relative to the leading edge ( $s_{ox}$  is less) and presumably is at a somewhat lower surface temperature (i.e., the surface regression perpendicular to itself is less than at the leading front, so the reaction rate must be less, which implies lower temperature).<sup>\*</sup> In Fig. 5, these two changes imply that  $L_{ox}$  shifts to higher values, possibly positive (endothermic decomposition). In Fig. 5 a value of  $s_{ox}$  of 33  $\mu\text{m}$  with  $T_s$  of 820° K gives a value of  $L_{ox}$  of 0 cal/g.

Referring to the binder, a value of  $s_f$  of 50  $\mu\text{m}$  seems reasonable from results of sandwich burning tests (i.e., the recessed binder is typically at a level comparable to the AP leading front). There is some controversy about the value of  $T_{sf}$ , but a value of  $L_f$  of 220 cal/g is reasonable (6,14,15). However, using Fig. 5 with a combination of  $s_f$  of 50  $\mu\text{m}$  and  $L_f$  of 220 cal/g, the resultant surface temperature is found to be an unrealistic 376° K. To obtain a more accept-

\* The full explanation of the sandwich profile in Ref. 1 involved two-dimensional heat transfer, which permits the surface of the protruding AP to be closer to the flame and yet have a lower surface temperature. The one-dimensional analysis cannot provide the correct combination of  $s$ ,  $T_s$  and  $L$  for such a situation, but it provides a basis for an estimate.

able temperature, e.g., 870° K,  $L_f$  must be lowered to a value of 43 cal/g. More generally, comparing the standoff distance of the AP leading edge to that of the binder for the above value of  $r$ , the surface temperature of the fuel is always less than the surface temperature of the oxidizer. Indeed, in sandwich burning tests at higher pressure, the heat of self-deflagration of the AP is increasingly released at or near the surface, corresponding to large negative values of  $L_{ox}$  in the present analysis, and the leading front of the AP proceeds far ahead of the fuel lamina and immediately adjoining AP.

#### Acknowledgement

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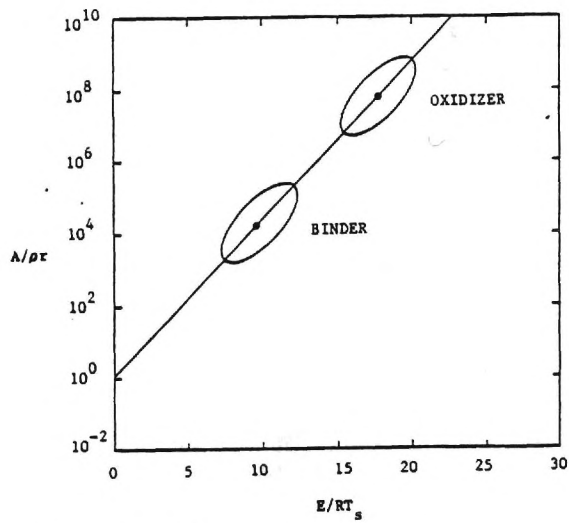
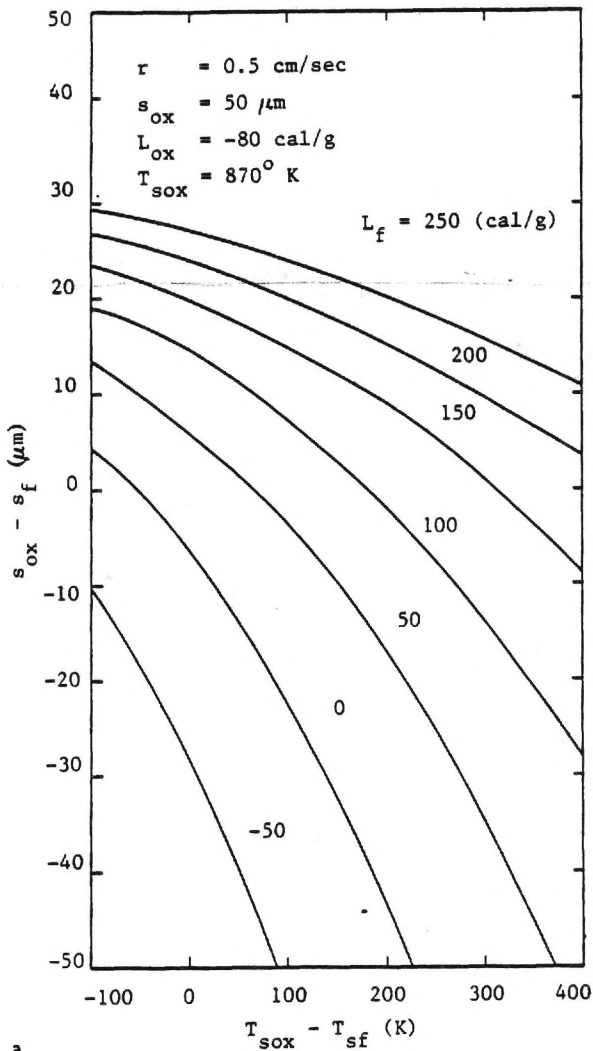
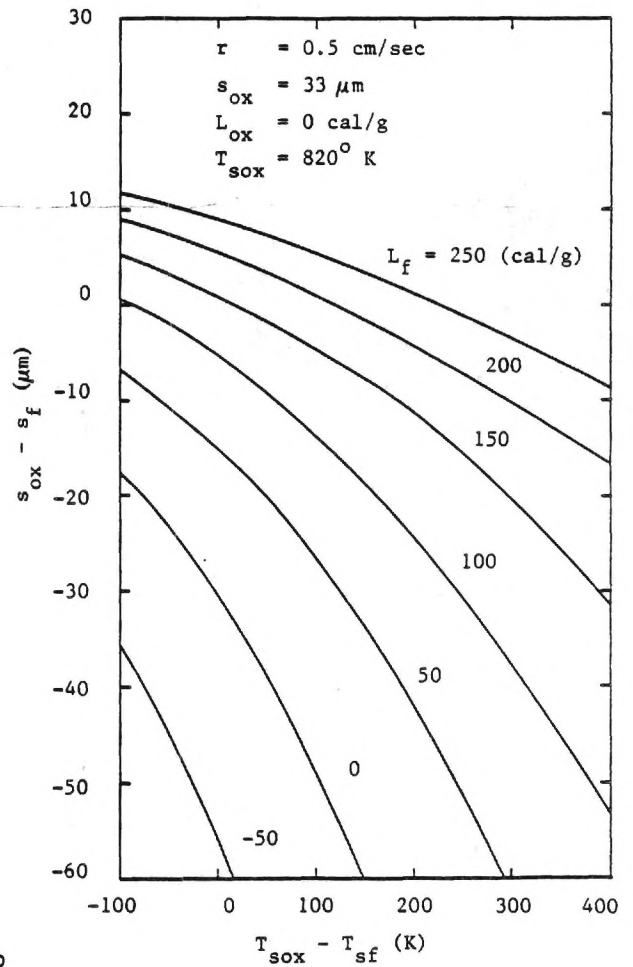


Fig. 2 Range of kinetic parameters.

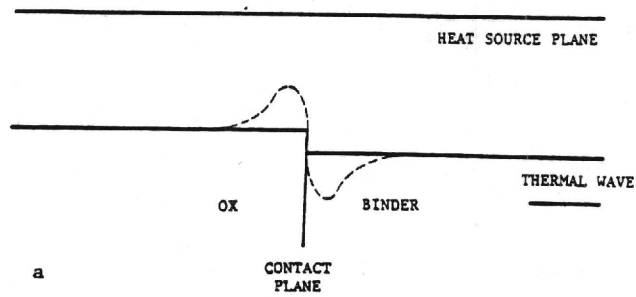


a

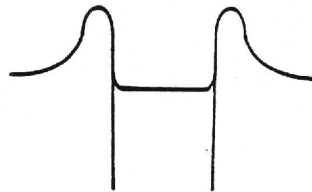


b

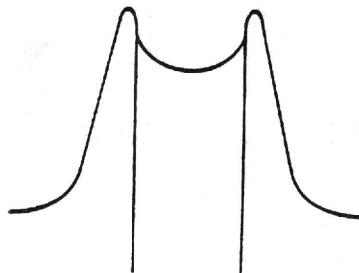
Fig. 3 Relation between lamina surface heights and lamina surface temperatures.  
 a) Conditions typical of the AP "leading front" in sandwich burning.  
 b) Conditions typical of AP near the binder lamina in sandwich burning.



a



b



c

Fig. 4 Laminate profiles.  
 a) Solid line is surface without lateral heat flow. Broken line is postulated surface when lateral heat flow occurs.  
 b) Surface for thin binder lamina when lateral heat flow occurs (Ref. 1).  
 c) Surface of thin binder sandwich burning at high pressure (e.g., 10 MPa) (Ref. 1).



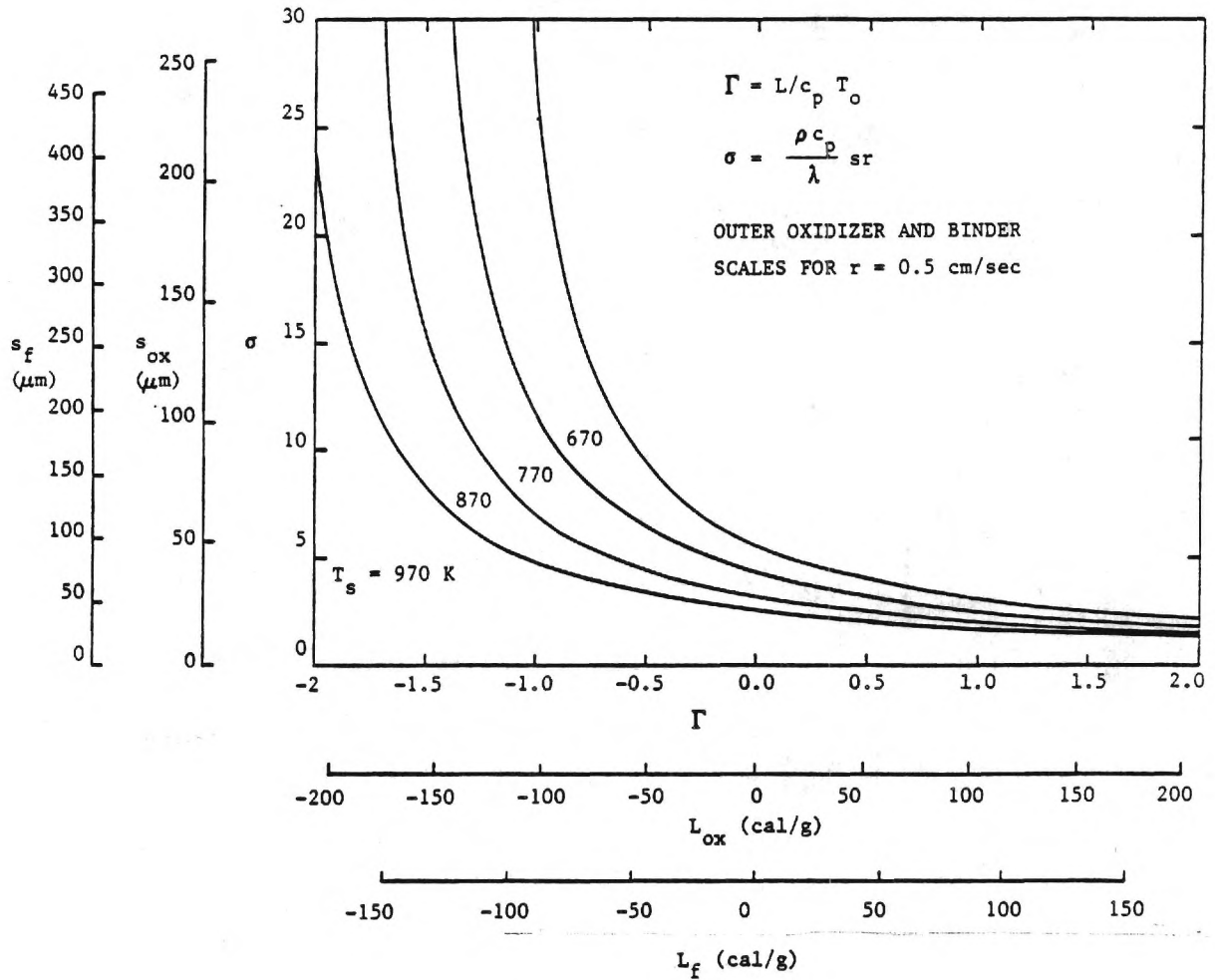


Fig. 5 Generalized standoff distance vs heat of phase change.

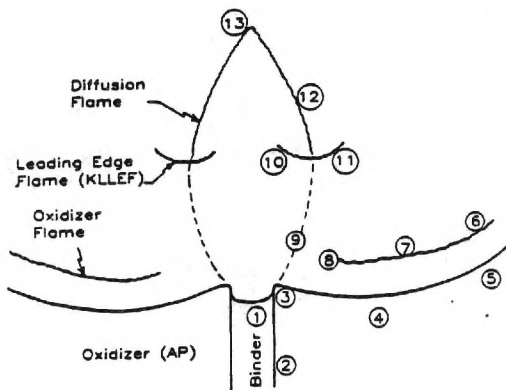


Fig. A-1 Principal features of the combustion zone microstructure and processes as suggested by accumulated results: 1, binder lamina; 2, interface plane between binder and oxidizer; 3, oxidizer surface adjoining binder (smooth band); 4, leading edge of the oxidizer burning front; 5, oxidizer region that regresses at the normal AP self-deflagration rate; 6, AP flame; 7, leading edge region of AP flame; 8, oxidizer flame, modified by the anomalous decomposition in the smooth band (flame may be quenched); 9, oxidizer-fuel diffusion region, with stoichiometric surface indicated by broken line; 10-11, kinetically limited leading edge flame (KLEEF) (fuel-rich and oxidizer-rich sides); 12, diffusion flame; 13, tip of diffusion flame.

## Enclosure B

### A Proposal for a Critical Experiment Linking Combustion Zone Microstructure and Pressure-Coupled Response Function

#### INTRODUCTION

This proposal is for an experimental evaluation of an hypothesis regarding the role of a specific feature of the combustion zone of a solid propellant in determining the dynamic response of combustion to flow disturbances. The origin of the hypothesis is in fundamental studies of the flame complex of composite AP-hydrocarbon binder propellants (Ref. B-1). The uniqueness of the proposal is that:

1. The hypothesis predicts a singular transition in measurable combustion response that should be readily distinguishable from other effects.
2. The conditions for transition are predicted qualitatively by the fundamental studies, and by already completed steady state burning studies designed specifically to test the underlying mechanisms.
3. Demonstration of singular transition in combustion response would provide a further stringent test of the underlying mechanistic argument, and establish the qualitative importance of a specific aspect of the flame complex that is suspected of being critical to dynamic response.
4. If the outcome of tests confirms the hypothesis, it would be the first time that features of dynamic combustion response had been linked back to a specific feature of the combustion zone flame complex. In addition, it would establish that a specific feature of the flame complex, absent in current analytical models but expected to be critical to dynamic response, is indeed a primary factor in determining combustion response.

In the following, some background on the status of combustion theory for composite propellants is presented, and it is noted that the present proposal represents a much more fundamental development, and one that is likely to yield decisive results in a technical area that is famous for the speculative state of knowledge. The technical basis for the present work is then reviewed, and the proposed study is outlined.

## BACKGROUND

### Status of Theory of Combustion of AP Composite Propellants

Research on solid propellant combustion has always used an indirect approach to understanding of combustion mechanisms. This approach typically uses analytical models or qualitative arguments regarding controlling mechanisms, based on combustion theory and basic information about ingredients. The models are the basis for hypotheses regarding the global combustion behavior such as burning rate, deflagration limits, ignitability, and their dependence on such variables as pressure, temperature, formulation variables, etc. To the extent that the models are successful in explaining the dependence of global combustion variables on the independent variables, the models are deemed to be validated. To the extent that no more basic proofs of theory can be made (because of inability to make direct observations of microflame behavior), we tend to classify our "successful" models as proven theory, and apply the detailed features of the models to new problems. In retrospect, speculative models that were originally cautiously based on detailed mechanistic features that were acknowledged to be speculative, have now become accepted theories without any substantive direct validation of the detailed features. They are accepted because they have been used successfully to correlate global combustion trends. To the extent that the models do not successfully predict global experimental trends a priori, they have usually been "fixed" by adjustment of parameters or insertion of new features whose neglect is deemed to be responsible for the deviant global behavior.

Scientifically, this whole process is a very shakey business, which is not subjected to truly objective evaluation. Not only are the detailed features of the model never established on a sound basis (because of experimental difficulty), but advances in understanding of the detailed features that do occur are only reluctantly adopted or actively opposed because they require re-structuring of the body of analytical modeling and rationalization of global trends that have become dogma. The "free parameters" in the models, that have been adjusted to make the models fit the global combustion behavior, stand for real processes such as reaction kinetics and diffusion coefficients, but minimal effort is made to determine these parameters from better fundamental studies or

to force the parameters to conform to fundamental determinations. Indeed, relatively little financial support has been available to these more fundamental studies, compared to the self-reinforcing pursuit of model adaptation to global combustion trends. While our efforts have evolved analytical-computational models that correlate certain experimentally established trends of global combustion variables, we have no assurance that other models would not do equally well, and little inclination to seek models that adapt and evolve to accommodate new fundamental results. We rarely use the models to predict the outcome of tests designed specifically to disprove the models, or to predict singular results, as yet unobserved, that can be related to specific features of the model.

Whether our existing models are soundly based or not, we are more notable for our efforts to correlate as much data as possible, than for our examination of the reasons for deviant behavior, or for our efforts to define the limits of the models and reasons for the limits.

From the viewpoint of the present proposal, it is extremely important that analytical models correctly represent the detailed features of the combustion zone, because we are asking for correct representation of the dynamic response. The existing "global fitted" models have been tailored to give correlation of experimental steady state burning rate data, but dynamic response depends on very different features of the flame complex than does steady state burning rate. In spite of this, existing models for dynamic response are based on the same flame complex and burning surface descriptions that have been used for steady state burning rate models. Such models should not be expected to provide anything more than a crude description of dynamic response, even after juggling of free parameters. The models do not contain any representation of the dynamic interaction of different components of the flame complex, multidimensional response of the surface and solid, or coupling among these features of the combustion. It should be no surprise that the correlation between measured pressure-coupled combustion response and model predictions is poor (Ref. B-2). There is no reason to expect the correlation to get better unless a more realistic model of the flame complex is adopted. The present proposal is part of a study aimed at providing and validating such a model.



## Research Leading to the Present Proposal

As a result of extended studies of the edge-burning of oxidizer-fuel laminates (Ref. B-1), it was established that the combustion zone of an AP-hydrocarbon binder "sandwich" looked something like the sketch in Fig. B-1. Tests were run on sandwiches that were prepared with "tapered" binder laminae, and it was found that as burning progressed from the thick edge toward the thin edge, samples quenched spontaneously at some binder thickness characteristic of the test pressure (Fig. B-2). Interpretation of this result is explained with the aid of Fig. B-3, which shows how the flame complex changes with decreasing binder thickness. Referring to the flames on one side of the line of symmetry in Fig. B-1 and B-3, the oxidizer-fuel flames consist of a kinetically limited portion ("KLLEF") that stands in the mixing region of the oxidizer and fuel vapors, and a diffusion flame that trails outward from the KLLEF until a point is reached where the fuel vapor is all consumed. This is the point on the line of symmetry, where the symmetrically located diffusion flames "close" over the fuel flow. According to classical diffusion flame theory, the diffusion flames correspond to surfaces of stoichiometric mixture ratio, and we refer to the closure tip as the "stoichiometric" tip. Because the binder is very thin, the samples are oxidizer-rich, and the region above the stoichiometric tip is oxidizer-rich.

Now referring to Fig. B-3, as binder thickness decreases, the stoichiometric tip moves in closer to the surface, and a larger portion of the fuel vapor is consumed in the KLLEFs. A point is reached where the stoichiometric tip occurs below the expected KLLEF location (all the mixture burns in the KLLEF). However, the KLLEF location now corresponds to a location that is oxidizer-rich (and increasingly so with increased distance from the surface). This is a very unstable region for the KLLEF because the flame temperature, which is a maximum at stoichiometric mixture ratio, is less at increasing distances above the stoichiometric tip. Thus there is no stable location for a KLLEF beyond the tip, and the tip has retracted so far toward the surface that the usual conditions for a KLLEF are gone. Thus, there is a critical binder thickness below which a KLLEF will not sustain. At low pressure, where the AP will not deflagrate on its own, the sandwich flame simply goes out for lack of a stable flame complex (Fig. B-2). Because the normal location of premixed flames is pressure-dependent, one expects the binder thickness threshold for loss of the KLLEF to be pressure dependent, in

a way suggested in Fig. B-3 (i.e., spontaneous quench at higher pressure is delayed to smaller binder thickness). Such a trend is decisively evident in Fig. B-2 at low pressures, but other factors come into play in this experiment at higher pressures (see Ref. B-1). (For the present work, the low pressure trend is expected to be preserved to higher pressure in propellant burning; see later.) Under those conditions where the kinetically limited leading edge flame (KLLEF) stands very close to the stoichiometric tip, a modest disturbance in gas motion would be likely to cause major disturbances in the KLLEF, including possible momentary quenching. In a sandwich, the quench may be more than momentary, but in the propellant situation discussed below there would be other flames nearby to provide "re-light", and the KLLEF might be prone to flickering. This postulated instability of the KLLEF is the aspect of the flame complex that we seek to demonstrate in the proposed study.

The flame complex in Fig. B-1 and B-3 has its counterpart in propellant burning. The question can be posed as to whether or not the kinetically limited flame can be established in the primary O-F mixing regions above the oxidizer-binder contact surfaces. In a propellant with continuous particle size distribution, the problem and its answer must be posed in statistical terms. We will avoid this complication by considering a propellant consisting of two widely different oxidizer particle sizes. Fig. B-4 suggests the nature of the flame complex for such a propellant. In some locations in the flame complex (site A), conditions are similar to the thin-binder sandwich discussed above, with closure of the stoichiometric surface over the binder. However, propellants are generally fuel rich, so there are locations (site B) where the stoichiometric surfaces are positioned over the oxidizer particles, and may even be partially closed over them (complete closure is probably rare because of the presence of site A type conditions somewhere along the particle periphery). For the present discussion, the important point to be made is that, if the width of exposed surface features (binder areas or oxidizer areas) is small enough, the closure of stoichiometric surfaces in adjoining mixing fans may sometimes occur too close to the surface for KLLEFs to be established (site C in Fig. B-4), creating local conditions similar to the thin-binder quench limit for sandwiches. In the propellant, which is typically fuel-rich, closure is predominantly over the oxidize particles, and stoichiometric tip height is related primarily to oxidizer particle size. In the studies described below, advantage was taken of two properties of formulation of the bimodal propellants:

1. The fine particles were chosen to be small enough so that, at some convenient pressure, stoichiometric tip closure on small particles would occur too close to the surface for KLLEFs to be established at low pressure in the primary mixing fans.
2. The ratios of ingredients were chosen to assure that surface areas with fine AP particles would be fuel rich, thus assuring closure over the fine particles and fuel rich conditions above the stoichiometric tip.

The latter condition was imposed to cause maximum destabilization of KLLEFs under conditions where the KLLEFs are near the stoichiometric tips.

In order to pursue the question of importance of "loss of the KLLEF" to propellant combustion, a propellant formulation was proposed that was designed to manifest the "loss" by a change in mean burning rate. A family of AP-PBAN propellants was made with bimodal AP particle size distribution. The coarse AP was nominally 400  $\mu\text{m}$ , and the fine size was varied with nominal sizes of 18, 49 and 82  $\mu\text{m}$ . The bimodal size distributions result in propellant microstructure sketched in Fig. B-5. Large oxidizer particles are "loosely packed" with intervening volumes filled with a fine AP-binder mixture. The mass fraction of coarse to fine particles and oxidizer to binder ratio were chosen so that the regions of fine AP-binder mixture would be fuel rich. This would result in closure of the stoichiometric surfaces over the small oxidizer particles, and a fuel-rich condition above the stoichiometric tips, as proposed in 1 and 2 above. Thus it was reasoned that at sufficiently low pressure, the KLLEFs on small particles would be "lost" in the same way as depicted for sandwiches in Fig. B-3. Since the fine-particle flames are particularly important in determining burning rate, it was reasoned that burning rate would drop off rapidly with decreasing pressure in the pressure range where fine particle KLLEFs were being lost. This effect would be evident at higher pressure when the fine AP particles were smaller. Such a pattern of burning rate trends, if unambiguously manifested, would be so distinctive as to be a relatively sound validation of the underlying mechanistic argument, and would also establish the conditions under which loss of the KLLEFs occurred (important for design of later tests of the effect of KLLEF stability on dynamic combustion response).

Burning rate tests were run on the family of bimodal propellants, and the results are shown in Fig. B-6. The predicted trend in burning rate is clearly manifested in the results, and constitutes as decisive a proof of the postulated "loss of the KLLEF" mechanism as one could hope to get, short of direct observation.



## PROPOSED INVESTIGATION

It is proposed that measurements of pressure coupled response function be made on three propellants similar to those used in the steady state burning rate studies (Fig. B-6). From those studies, it appears that a pressure range of 100 to 500 psi would be appropriate to span the range from stable small particle KLLEFs to detached small particle KLLEFs. A program of test variables is proposed in Table B-1. The objective of the tests is to determine whether approach to KLLEF detachment (as determined by the steady state tests, Fig. B-6) leads to sharp increase in response function. This is illustrated by Fig. B-7, which shows response function vs pressure (postulated here for the sake of description). Two curves are shown as possible trends. Curve 1 corresponds to a relatively stable flame complex both above and below the KLLEF detachment pressure, and Curve 2 corresponds to unstable conditions at all pressures below the KLLEF detachment pressure. We do not know enough about the flame complex to predict whether Curve 1 or Curve 2 is more appropriate; but the mechanistic argument requires that a change occur when detachment conditions are approached. The experiments will settle this point, and also show what change occurs (e.g., Curve 1 or Curve 2). It is proposed that the response function measurements be made in T-Burner tests with propellant discs in the ends (pressure coupled tests).

In the foregoing, reference has been made to "loss of the KLLEF", which is viewed as a crucial event relative to burning behavior. In a burning sandwich, it leads to quench. In a bimodal propellant it leads to decrease in burning rate under special conditions demonstrated here. However, the propellant situation poses the question of where the KLLEF goes when it is "lost". We do not intend to answer this question fully, but only note that we have been referring to loss of the KLLEF as a flame identifiable with a specific oxidizer particle or primary O-F mixing fan. We have been discussing situations where the flame is not likely to stabilize immediately above the stoichiometric tip, so that loss of the KLLEF in the above sense corresponds to delay of exothermic reactions to a site considerably further from the surface, some site where favorable ignition conditions exist because of thermal and mass diffusion from other surface or flame elements (e.g., associated with larger particles).



It is the inherent instability of the KLLEF near the stoichiometric tip that we wish to stress, as it is anticipated that the dynamic response to disturbances will be large under conditions of mean pressure and particle size that give a large population of KLLEFs that are near stoichiometric tips. Indeed, this situation may be a major contributor to oscillatory combustion response for all composite propellants, and may explain the widely reported conspicuous dependence of response function on mean pressure (which is not explained by current theories). However, we choose to test propellants that are designed to exhibit KLLEF detachment effects to a maximum degree under distinctive conditions, so as to pose an unambiguous test of the KLLEF detachment role in determining dynamic response.

Up to this point, nothing has been said here about the oscillation frequency for T-Burner tests. Our present understanding of the dynamic behavior of "detaching KLLEFs" is negligible, and one might hope that the dependence of test results on oscillation frequency will give some clues beyond the general result that KLLEF detachment is important (a result that would already be established by the effects of pressure and particle size on combustion response function). The first question, of course, is: What frequency should be chosen for the test series to determine the trend of response with mean pressure? An obvious choice would be a frequency for which response functions usually exhibit a maximum, typically a non-dimensional frequency of around

$$\Omega = \kappa\omega/\bar{r}^2 = 2\pi \kappa f/\bar{r}^2 = 12$$

The value of  $\Omega = 12$  is suggested by computations using perturbation theories such as Dennison and Baum, in which the maximum is governed by the dynamic response of the thermal wave in the condensed phase to oscillations in surface heating by the perturbed flame. Initial tests in the present study will be made at a frequency close to this value, which is estimated to be around 1100 Hz for the pressures and burning rates of interest. Following the tests with pressure as the primary variable, a specific pressure and propellant will be chosen for a series of tests with frequency as the independent variable. Tests will be run from 500-5000 Hz. It is important to stress that testing for frequency dependence of the response function goes beyond the issue of verifying the importance of KLLEF detachment. It probes the question of how the KLLEF behaves

during a cycle of oscillation. The behavior embodied by the concept of KLLEF detachment is not amenable to small perturbation analysis, at least relative to individual KLLEFs. Once a KLLEF detaches during part of an oscillation cycle, it either goes out or retreats to a more remote location. The pyrolysis of the surface and mixing flow associated with that KLLEF experiences a complicated transient adjustment involving falling surface temperatures and pyrolysis rate, slower outward convection of vapors, and accumulation of mixed fuel and oxidizer vapors in the region where the KLLEF was previously located. We do not know whether the KLLEF "re-ignites" in this region later in the cycle of oscillation, or simply propagates back in from an outer location. There may be some conditions under which the KLLEF does not recover at all during a cycle (in which case the average burning rate may be depressed during oscillations). Tests for response function vs frequency (and perhaps vs amplitude) will provide some clues as to details of KLLEF behavior.

#### References

- B-1. Price, E. W., J. K. Sambamurthi, R. K. Sigman and R. R. Panyam, "Combustion of Ammonium Perchlorate-Polymer Sandwiches," Combustion and Flame, Vol. 63, pp. 381-413, 1986.

Table B-1  
 Matrix of Test Conditions  
 Constant frequency tests (- 1100 Hz)

Propellant (87.5% AP/12.5% PBAN)	Test Pressures (MPa)			
70% 400 $\mu\text{m}$ AP + 17.5% 17.5 $\mu\text{m}$ AP + 12.5% PBAN	1.5	2.1	2.6	3.5
70% 400 $\mu\text{m}$ AP + 17.5% 49.0 $\mu\text{m}$ AP + 12.5% PBAN	1.0	1.7	2.0	2.3
70% 400 $\mu\text{m}$ AP + 17.5% 82.5 $\mu\text{m}$ AP + 12.5% PBAN	0.7	1.0	1.4	1.8

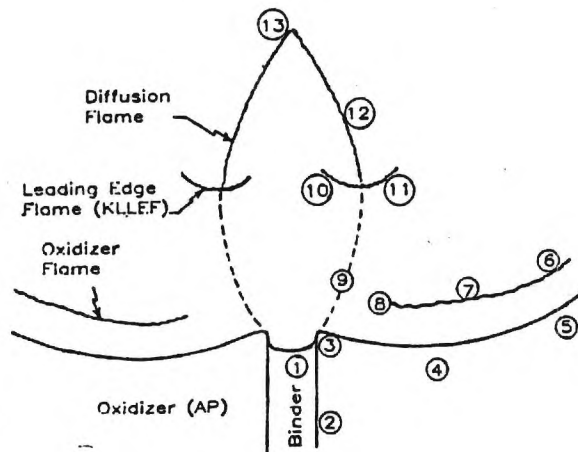


Fig. B-1 Principal features of the combustion zone microstructure and processes as suggested by accumulated results: 1, binder lamina; 2, interface plane between binder and oxidizer; 3, oxidizer surface adjoining binder (smooth band); 4, leading edge of the oxidizer burning front; 5, oxidizer region that regresses at the normal AP self-deflagration rate; 6, AP flame; 7, leading edge region of AP flame; 8, oxidizer flame, modified by the anomalous decomposition in the smooth band (flame may be quenched); 9, oxidizer-fuel diffusion region, with stoichiometric surface indicated by broken line; 10-11, kinetically limited leading edge flame (KLLEF) (fuel-rich and oxidizer-rich sides); 12, diffusion flame; 13, tip of diffusion flame.

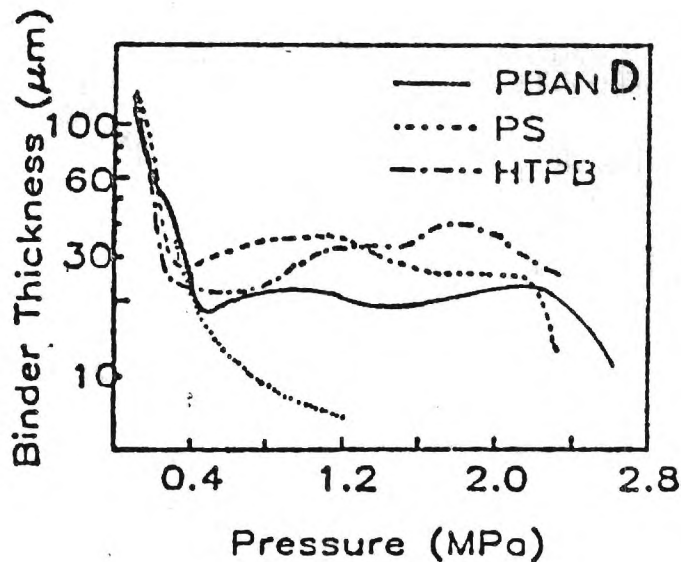


Fig. B-2 Relation of minimum binder thickness to pressure for self-sustained burning of AP polymer sandwiches. Quench is attributed to retraction of the stoichiometric tip below the standoff location of the kinetically limited O-F flame (see Fig. B-3). Minimum binder thickness was determined by spontaneous quench of sandwiches with tapered binder laminae burning at constant pressure.

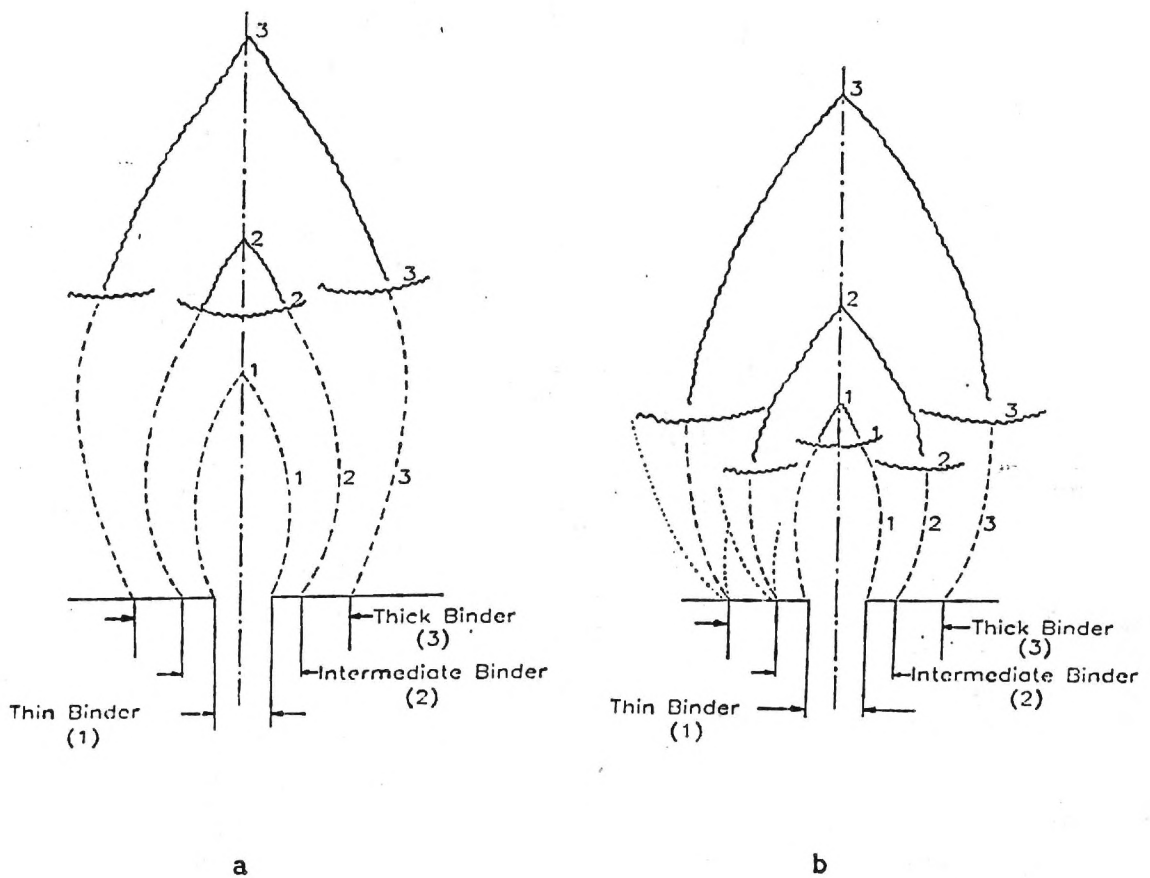


Fig. B-3 Sketches of stoichiometric surfaces (dotted curves) and O-F flames for sandwiches with different binder thicknesses (sketches for different thicknesses are "nested" within each other for easy comparison). Parts A and B are for different pressures. Note that for the thin binder, low pressure case, the stoichiometric regions are limited to locations too close to the surface to sustain the O-F flame (conditions further out are too oxidizer-rich to support the flame and the sample won't burn).



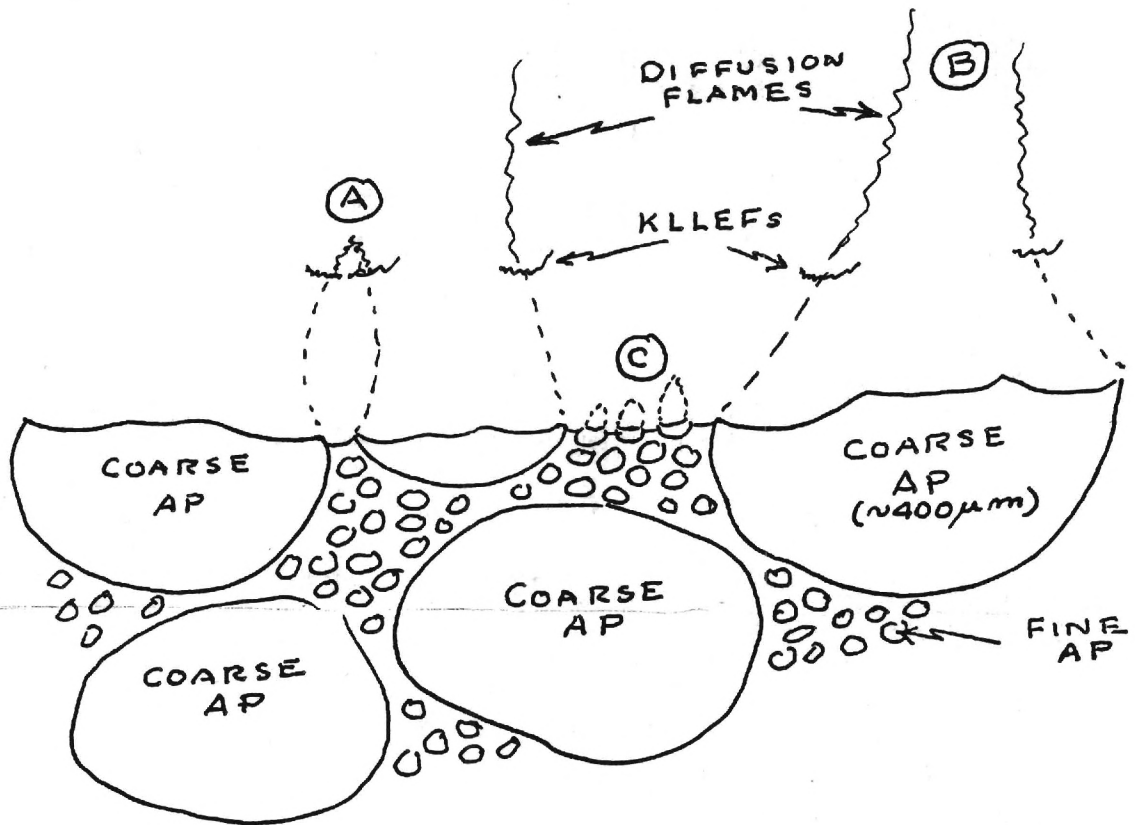
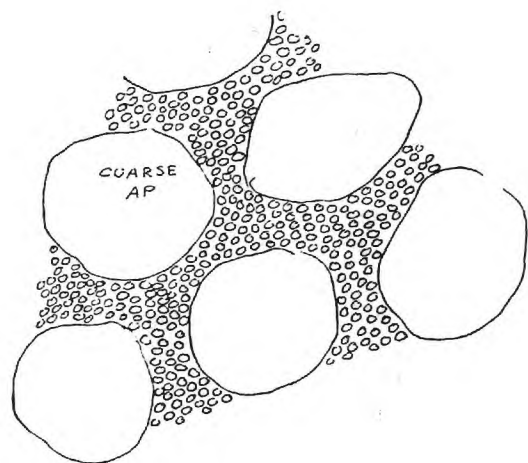
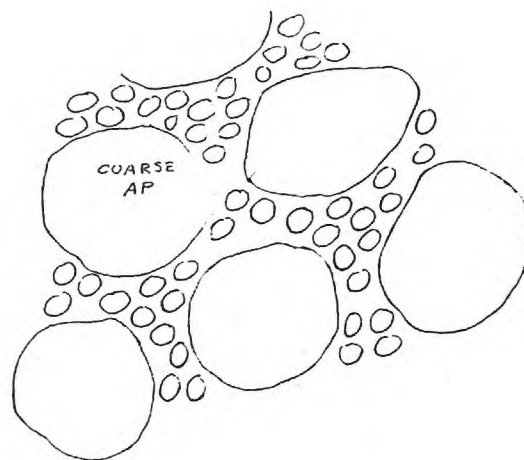


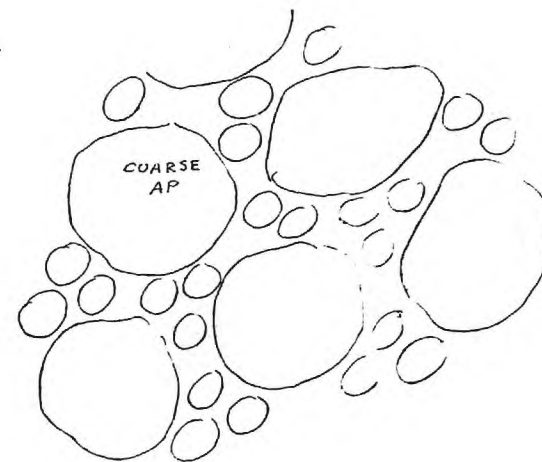
Fig. B-4 O-F flame complex above the surface of a bimodal propellant. Dotted curves are stoichiometric surfaces in micro-mixing fans above oxidizer-binder interfaces.



A) Very small size fine component gives most fine particles surrounded by other fine particles in a fuel-rich environment. Fine particles cannot support individual O-F flamelets at lower pressure because the vapor mixture becomes fuel-rich too close to the surface for flamelets to be established (Site C in Fig. B-4).



B) Intermediate size fine component. Individual O-F flamelets are sustained to lower pressure because individual stoichiometric surfaces extend further from the surface than in A. In addition, more fine particles have large neighbors and see less excess fuel on sides adjoining large particles, a condition conducive to flame-holding.



C) Relatively large size fine component. O-F flamelet holding on in individual particles continues to quite low pressure.

Fig. B-5 Sketch of microstructure of bimodal oxidizer propellants, all with "loosely packed" coarse AP. These sketches approximate the propellants used in the tests in Fig. B-6, which show transitions in burning behavior with changing pressure that are believed to reflect transitions from fine particles burning with individual O-F flamelets (high pressure) to fine particles burning with more remote premixed O-F flame (low pressure). The sketch pictures only two particle sizes in each sketch, but a burning surface would show particles with more varied sizes corresponding to different stages of burning of particles, and some variation of actual particle size. This causes the transitions in burning rates in Fig. B-6 to be less abrupt with pressure change than would be the case if the surfaces were as depicted in the sketches.

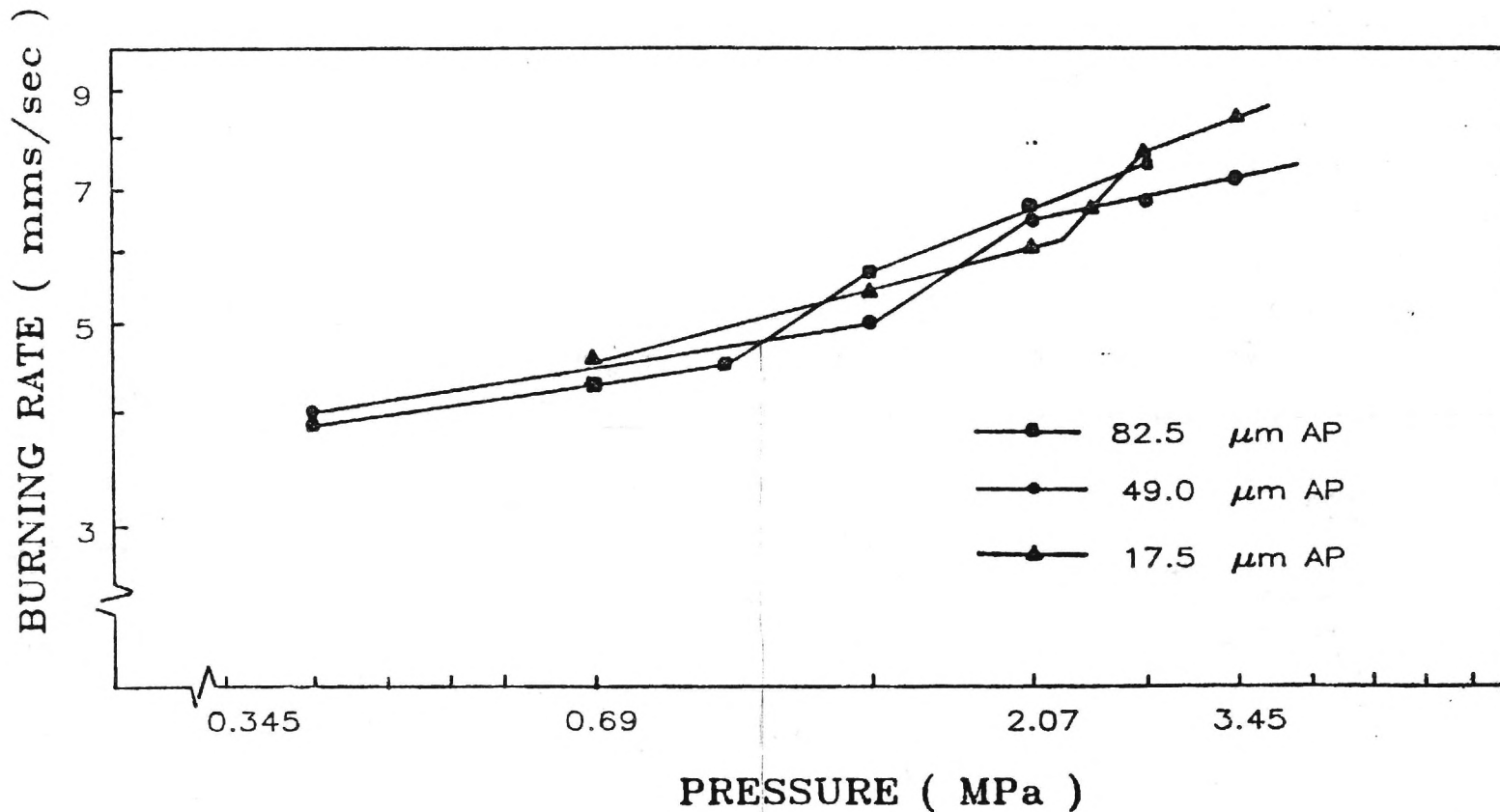


Fig. B-6 Burning rates of three bimodal AP propellants having 400  $\mu\text{m}$  coarse AP and different fine AP sizes (indicated in the figure). Rapid drop-off in rate with pressure in the mid-pressure range is believed to be due to "loss of" the individual near-surface O-F flamelets on the fine particles, because flamelets tend to stand further from the surface at lower pressure and eventually withdraw beyond the stoichiometric tips where fuel-rich conditions are unfavorable for the flamelets (See Fig. B-3).

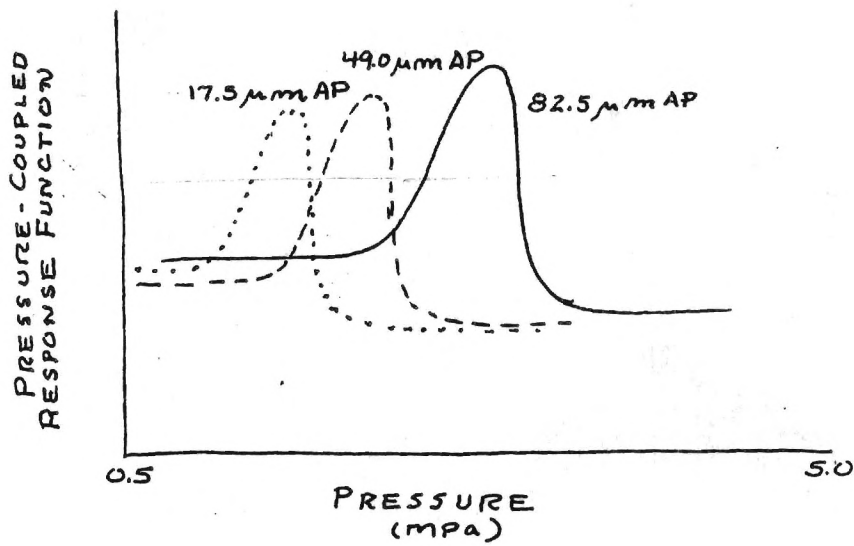
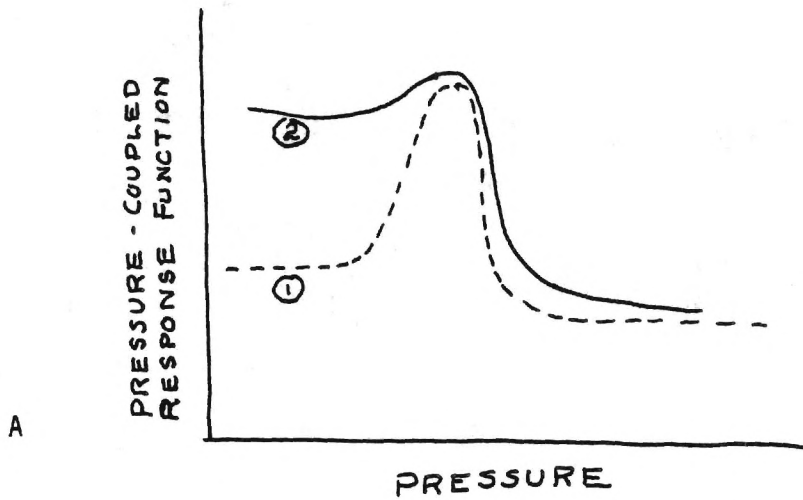


Fig. B-7 Expected trend of pressure-coupled response function as a function of pressure for bimodal propellants (based on a presumption that dynamic response will be larger under conditions close to the threshold for holding-nonholding of individual O-F flamelets on fine particles). Part A shows possible dependence on pressure. Part B shows how the  $R_p$ -p function would be expected to change with particle size of the fine component of the AP.