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(UNCLASSIFIED TITLE)  
**COMBUSTION TAILORING CRITERIA  
FOR  
SOLID PROPELLANTS**

LOCKHEED PROPULSION COMPANY  
REDLANDS, CALIFORNIA

TECHNICAL REPORT AFRPL-TR-68-133

JULY 1968

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COMBUSTION TAILORING CRITERIA  
FOR SOLID PROPELLANTS

LOCKHEED PROPULSION COMPANY  
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## FOREWORD

This is the third phase report issued under Contract FO4611-67-C-0089. Reported herein is research conducted between 30 April and 1 July 1968. The report was prepared by R. L. Derr and M. W. Beckstead, Engineering Research Department, Lockheed Propulsion Company (LPC). Contributors to this program include N. S. Cohen, R. L. Coates, W. R. Glace, T. L. Mills, C. F. Price, L. L. Stiles, and W. West, all representing Lockheed Propulsion Company.

Work performed under this contract is monitored by the Air Force Rocket Propulsion Laboratory (AFRPL), Edwards, California. The Project Officer is Captain Charles E. Payne, AFRPL/RPMCP.

This report is classified CONFIDENTIAL-NOFORN because it makes reference to advanced propellant formulations and ingredients of interest to the Air Force.

Publication of this report does not constitute Air Force approval of the findings or conclusions. It is published only for the exchange and stimulation of ideas.

Captain C. E. Payne  
AFRPL/RPMCP

## ABSTRACT

The objective of this research program is to develop an analytical model describing the steady-state combustion of solid propellants which is suitably equipped to provide combustion tailoring criteria for both state-of-the-art and advanced propellant formulations. The model will be developed and implemented from experimental data acquired from a dual experimental program. Tests with small samples of propellants will provide experimental data for modeling the propellant combustion process for different ingredient variations, e.g., oxidizer size and concentration, metals. Additional tests in apparatus simulating the environment of a rocket motor will provide experimental data showing the effect of rocket motor environment upon the combustion process. During Phase III, work was directed toward developing the analytical model and modifying aspects of the model that seemed least realistic. A list of propellants was chosen for study in tests to be conducted during the remainder of the research program. An experimental technique was developed for examining the surface of burning propellants. Small samples of a CTPB/AP propellant were burned in a pressurized combustion bomb and extinguished by rapid depressurization of the bomb. The surface structure was examined through a 10X - 80X microscope. Results from tests conducted in apparatus simulating the environment of a rocket motor (slab combustor) and using a CTPB/AP/Al propellant showed that the thermal environment of the combustor reaches a maximum level at a pressure of 200 psia. To learn what effect the thermal environment has upon the propellant burning rate, a technique was developed to directly measure burning rate in the slab combustor. The approach uses a high-speed camera to record the movement of the propellant burning surface with respect to a reference scale.

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## SECTION I

### INTRODUCTION

Past studies of solid propellant combustion were directed toward both experimental and analytical investigations of the combustion process. Experimental studies provided a wealth of information pertaining to steady-state and non-steady-state combustion for a variety of propellants and propellant ingredients. Analytical studies also were performed. However, those studies were not successful in representing the combustion process successfully such that they can account for changes in oxidizer, binder, catalysts, and metal additives in the framework of establishing combustion tailoring criteria.

In April of 1967, the Air Force Rocket Propulsion Laboratory (AFRPL) contracted Lockheed Propulsion Company (LPC) to study the combustion of solid propellants with the objective of providing such criteria. The program includes a comprehensive combustion literature survey to establish the present state of combustion knowledge and provide a baseline for analytical and experimental work.

In the first year, the program included the design and fabrication of combustion research apparatus, representative of a motor amenable to instrumental monitoring of various facets of steady-state and transient combustion. Tests with a series of well characterized propellants were also included in the first year effort to check out experimental apparatus.

In the second year, tests with a series of state-of-the-art and advanced propellant types will be included providing data from the experimental program which will be used to develop and implement an analytical model of the combustion process. Development will progress from a model capable of describing the combustion characteristics of simple propellant systems, i.e., nonmetalized and uncatalyzed, to one that is capable of describing the characteristics of advanced propellant systems, i.e., propellants containing metal additives and energetic binders. In that way, the capability of combustion tailoring will be built into the analytical model.

The two-year combustion research program can be divided into five major tasks summarized as follows:

- (1) Literature survey
- (2) Design, fabrication, and pretest check of combustion research apparatus
- (3) Experimental program with well-characterized propellants
- (4) Experimental program with operational and advanced formulations
- (5) Analysis and formulation of combustion tailoring model.

Results contained herein cover research conducted during the first quarter of the second-year effort.

## SECTION II

### PROGRAM SUMMARY

This report describes work accomplished on a study of the combustion of solid propellants. The overall goal of the program is to develop an analytical model suitably equipped to provide combustion tailoring criteria for both state-of-the-art and advanced propellant formulations. The model will be developed and implemented from data acquired from experimental apparatus simulating the environment of a rocket motor and small sample studies. Accomplishments of the program are summarized in the following subsections.

#### 1. LITERATURE SURVEY

The purpose of the literature survey is to establish the state of knowledge of the combustion of solid propellants, including the role of propellant ingredients, with a view toward providing a baseline and some guidelines for the current effort. The review encompasses theoretical developments, experimental results, and techniques beginning with the state of knowledge as of approximately 1950. The survey was completed during the past report period and published as a separate document (1).

#### 2. ANALYTICAL INVESTIGATION

The analytical model for the combustion of composite solid propellants (based on the published Hermance model) is being developed. The Hermance model has been modified so that it is now based on a more realistic physical description. The modification shifts the emphasis of the model from one based on the heterogeneous reactions to one based on the decomposition of the oxidizer. The model is being programmed for the computer and adapted to an optimization technique to identify the dominant parameters. It is anticipated that from experience gained with the model, it should be possible to identify key steps in the combustion process. This will ultimately allow the qualitative tailoring of burning rates.

#### 3. EXPERIMENTAL INVESTIGATION

##### a. Selection of Propellants for Second Year Effort

A list of propellants was established for experimental studies to be conducted in the second year effort of this research program. Systematic changes in ingredient variables have been incorporated in the list to allow maximum use of the data to develop the analytical model. The list is presented in Table I of Section IV.

b. Study of Burning Surface Structure

A technique was developed to study the surface structure of burning solid propellant samples. Photographs of extinguished surfaces of a nonmetalized rubber-base propellant are presented indicating that the burning surface is somewhat different than that postulated in Hermance's Heterogeneous Reaction Model.

c. Heat Flux Measurements in the Slab Combustor

Total heat flux measurements were made in a slab combustor that simulates the environment of a rocket motor. Nonmetalized and metalized rubber-base propellants were used in tests to determine what level of heat flux prevails in a motor environment and how it influences the combustion process.

d. Burning Rate Measurement in the Slab Combustor

A technique was established to directly measure the burning rate of propellants in the slab combustor. Results are presented for a nonmetalized rubber-base propellant and compared with burning rates measured in a pressurized combustion bomb using small propellant samples.

## SECTION III

## ANALYTICAL INVESTIGATION

In the two previous reports (1, 2), models of steady-state propellant combustion were discussed and the conclusion reached that the Hermance heterogeneous model (3, 4) is the most realistic and versatile of the published models. The basic structure and philosophy of the model were studied in detail for application to conventional propellants, and brief outlines of the model with discussions of its most important aspects were discussed in earlier reports (1, 2). During this report period, a modification was made on one of the least realistic parts of the model.

### 1. PHYSICAL CHARACTERISTICS OF THE CRYSTALLINE OXIDIZER RELATIVE TO THE BURNING SURFACE

In the Hermance model the oxidizer particles are assumed to be spherical and are assumed to regress in such a manner as to remain spherical at all times. Figure 1 is a schematic of this relationship between the oxidizer and fuel showing the crevice that results from the assumption and in which the heterogeneous reaction between the oxidizer products and the fuel is assumed to take place.

During the present report period an experimental program was begun with the intent of determining the nature of the burning surface of a composite propellant. The initial results from this study are presented in Section IV. These results, along with those of other investigators (5, 6), indicate that the interface between the oxidizer and fuel is continuous and a crevice does not form. Therefore, this aspect of the model was modified.

For ammonium perchlorate (AP)-containing propellants, experimental observations indicate that the crystals protrude above the surface at low pressures and at high pressures are recessed below the fuel surface. Thus, using the same statistical analysis developed by Blum and Wilhelm (7), and used by Hermance (3), the following model can be constructed for composite propellants (assuming that the oxidizer is initially spherical). At low pressure, the crystals will protrude above the fuel as shown in Figure 2. It has been shown (7) that for a large number (statistical sample) of randomly packed spheres a plane of intersection (or in this case the fuel surface) will intersect the spheres such that the average diameter of the intersection circle is  $(\frac{2}{3})^{1/2} D_0$ , where  $D_0$  is the original diameter. Statistically, one-half of the crystals will be intersected above their center while the other half will be intersected below their center giving the configuration demonstrated in Figures 2 and 3 (where Figure 3 is the high-pressure configuration).

If it is assumed that the portion of the crystal that protrudes above or is recessed below the surface is spherical, but always joining the fuel at the planar fuel surface, then the area of oxidizer that is exposed to reaction

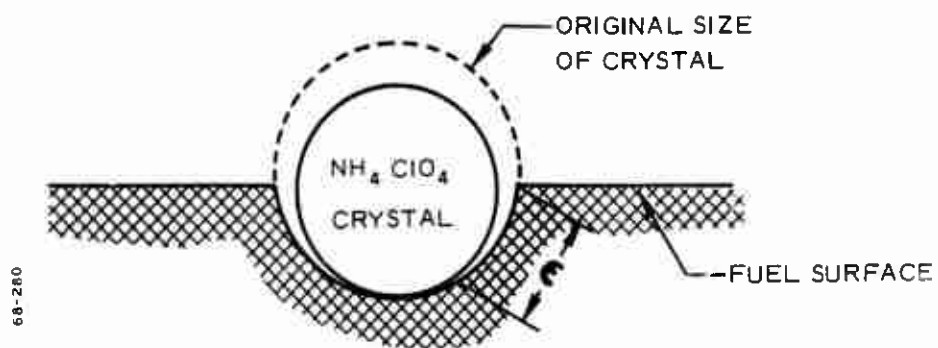


Figure 1 - Oxidizer-Fuel Relationship as Described  
in References 3 and 4

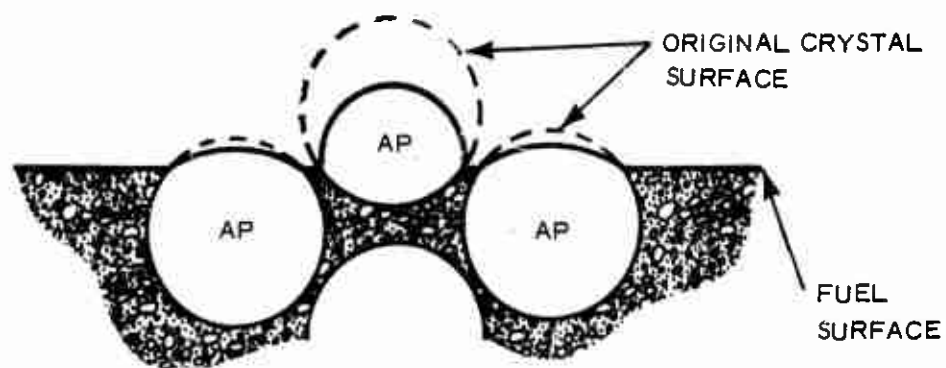


Figure 2 - Low Pressure Relationship Between the Oxidizer Crystal and the Fuel Surface

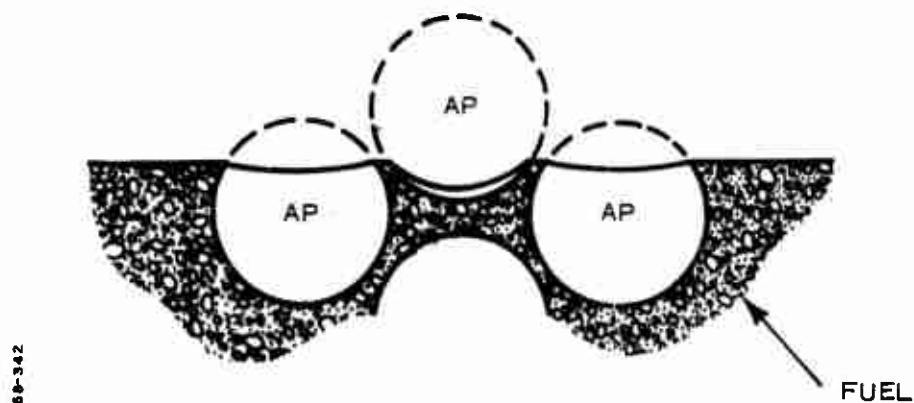


Figure 3 - High Pressure Relationship Between the Oxidizer Crystal and the Fuel Surface

can be calculated. Referring to Figure 4, the surface area of the spherical segment representing the burning AP surface is

$$S_{ox} = 2\pi R|h| \quad (1)$$

Where  $S_{ox}$  is the area of a single oxidizer crystal. From triangle I

$$\begin{aligned} R^2 &= x^2 + (R-h)^2 \\ 2R|h| &= x^2 + h^2 \end{aligned} \quad (2)$$

From triangle II

$$\begin{aligned} (D_o/2)^2 &= x^2 + (D_o/2-L)^2 \\ x^2 &= D_o L - L^2 = L(D_o - L) \end{aligned} \quad (3)$$

Substituting Equations (2) and (3) into Equation (1)

$$S_{ox} = \pi [h^2 + L(D_o - L)] \quad (4)$$

where  $h$  is a measure of the non-planarity of an oxidizer crystal and  $L$  is the distance from the top of the original crystal surface to the fuel surface. From the statistical analysis discussed above, it can be shown that the average diameter of randomly packed spheres at the intersection plane is  $(2/3)^{1/2} D_o$ . Combining this with Equation (3) yields

$$x^2 = \left[ \frac{(2/3)^{1/2} D_o}{2} \right]^2 = \frac{D_o^2}{6} = L(D_o - L) \quad (5)$$

and

$$L = \frac{D_o}{2} \left[ 1 \pm \sqrt{\frac{1}{3}} \right] \quad (6)$$

Combining Equation (5) with Equation (4) gives

$$S_{ox} = \pi D_o^2 \left[ \left( \frac{h}{D_o} \right)^2 + \frac{1}{6} \right] \quad (7)$$

The term involving  $h/D_o$  can be evaluated using the same ignition delay concept employed by Hermance. Thus, referring to Figure 4,

$$h = L - y_{AP}$$

where  $y_{AP}$  is the linear distance that the AP crystal regresses and is equal to the product of the linear burning rate of AP under the given conditions and

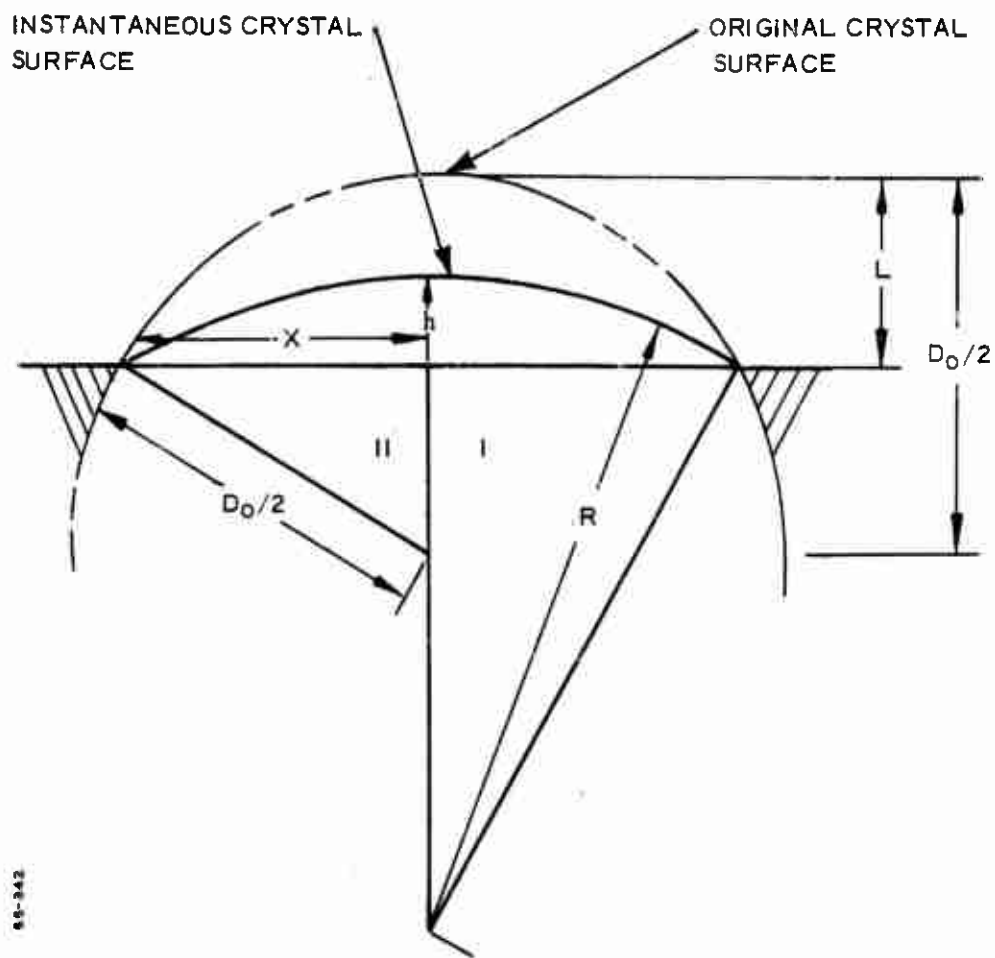


Figure 4 - Geometrical Relationship of the Oxidizer Crystal to the Burning Area

the time that the crystal burns. The burning time of the crystal is equal to the total time necessary for the surface to regress the distance  $L$  minus the ignition delay time. Thus

$$h = L - v_{AP} (t_b - t_{ign}) = L - v_{AP} \left( \frac{L}{r} - t_{ign} \right) \quad (8)$$

where the average regression rate  $r$  has been used to evaluate the total burning time. Equation (8) can be rearranged to give

$$\begin{aligned} \frac{h}{D_o} &= \frac{L}{D_o} \left( 1 - \frac{v_{AP}}{r} \right) + v_{AP} \frac{t_{ign}}{D_o} \\ &= \frac{1}{2} \left( 1 \pm \frac{1}{\sqrt{3}} \right) \left( 1 - \frac{v_{AP}}{r} \right) + v_{AP} \frac{t_{ign}}{D_o} \end{aligned} \quad (9)$$

where the ignition delay time has the form

$$t_{ign} = \frac{K_o D_o^{n+1}}{p^m} \quad (10)$$

Combining Equations (7), (9), and (10), the oxidizer surface area that is undergoing reaction can be calculated. The AP burning rate is calculated from an Arrhenius expression (see subsequent section) instead of the empirical relationship used by Hermance. Because the burning rate of the propellant is involved, this will lead to a transcendental equation in the burning rate that requires an iterative solution.

## 2. THE STEADY-STATE BURNING RATE

The basic equation in the model relates the linear regression rate of the propellant to the mass fluxes because of fuel pyrolysis, heterogeneous reaction and oxidizer decomposition, and the corresponding surface areas [see Equation (10) Ref. 3]

$$r = 1/\rho_p [m_f(S_f/S_o) + m_{ox}(S_{ox}/S_o) + m_{sr}(S_{sr}/S_o)] \quad (11)$$

where  $m$  represents the mass flux and  $S$  the surface area,  $S_o$  representing the total propellant area subject to reaction. The total surface area can be written as [utilizing Equation (7)]

$$S_o = S_f + S_{sr} + N \pi D_o^2 \left[ \left( \frac{h}{D_o} \right)^2 + \frac{1}{6} \right] \quad (12)$$

where  $N$  is the number of crystals per unit area of propellant surface. Both the fuel pyrolysis and the heterogeneous reaction take place on the fuel surface. Thus, if  $S_f$  is the total fuel area exposed to reaction and  $\beta$  is defined as the fraction of fuel area undergoing heterogeneous reaction, then

$$S_{sr} = \beta S_f \quad \text{and} \quad S_f = (1-\beta) S_f \quad (13)$$

On a statistical basis the volume fraction of fuel is equal to the ratio of fuel area to planar propellant area (assuming that the fuel surface is planar). Similarly, the volume fraction of oxidizer is equal to the ratio of the planar intersection area to that of the propellant. Thus

$$1 - v = \frac{S_f}{S_p} \quad \text{and} \quad v = \frac{N \frac{\pi}{4} D'^2}{S_p} \quad (14)$$

where  $D'$  is the average intersection diameter. The number of oxidizer crystals on the surface is

$$N = \frac{6 v S_p}{\pi D_o} \quad (15)$$

Equation (12) can now be rewritten as

$$\begin{aligned} \frac{S_o}{S_p} &= (1-v) + 6v \left[ \left( \frac{h}{D} \right)^2 + \frac{1}{6} \right] \\ &= 1 + 6v \left( \frac{h}{D} \right)^2 \end{aligned} \quad (16)$$

The individual reaction areas can now be determined as

$$\frac{S_{sr}}{S_o} = \beta \frac{(1-v) S_p}{S_o} = \frac{\beta (1-v)}{1 + 6v \left( \frac{h}{D} \right)^2} \quad (17)$$

$$\frac{S_f}{S_o} = \frac{(1-\beta) (1-v)}{1 + 6v \left( \frac{h}{D} \right)^2} \quad (18)$$

$$\frac{S_{ox}}{S_o} = \frac{N S_{ox}}{S_o} = \frac{v \left[ 6 \left( \frac{h}{D} \right)^2 + 1 \right]}{[6v \left( \frac{h}{D} \right)^2 + 1]} \quad (19)$$

Equations (17), (18), and (19) now can be used to evaluate the relative areas upon which the three types of reactions are occurring. The equations are based on all of the oxidizer particles being spherical and the same size. This is extremely close to what would be determined for a unimodal but discrete particle distribution. The equations can be extended to include a bimodal distribution in the same manner utilized by Hermance.

The mass flux terms for the three reactions are

$$m_f = \rho_f A_f \exp (-E_f/RT_g) \quad (20)$$

$$m_{ox} = \rho_{ox} A_{ox} e^{\left(\frac{-E_{ox}}{RT_s}\right)} \quad (21)$$

$$m_{sr} = c_1 \left(\frac{T_s}{M_g}\right)^{1/2} \left(\frac{M_c}{R_c T_s}\right) e^{-\frac{(E_{ox} + E_{sr})}{RT_s}} \quad (22)$$

where  $A$  is the frequency factor,  $E$  is the activation energy, and  $T_s$  is the surface temperature.

Equations (20) and (22) are the same as Equations (17) and (23) in Reference 3. Equation (21) is introduced here as being more consistent than the manner utilized in Reference 3 to evaluate the oxidizer mass flux. Now, combining Equations (17) through (22) with Equation (11), the burning rate of the propellant can be calculated as a function of the surface temperature and the several parameters that are involved. The surface temperature is determined by the same technique used by Hermance and is subject to the same advantages and disadvantages as his technique.

The series of equations developed in this section were incorporated into the computer program that has been used in the past (2). The program is being debugged and results are expected soon. The changes that have been made will cause a major change within the results of the model. The emphasis has been shifted from the heterogeneous reaction to the oxidizer decomposition. Whereas in the Hermance model, the calculated burning rate was completely dominated by the heterogeneous reaction and the parameters involved with it, in the present modification the decomposition of the oxidizer has the major influence on the overall burning rate. From a phenomenological point of view, the latter appears more realistic and closer to what actually occurs in a burning propellant.

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## SECTION IV

## EXPERIMENTAL INVESTIGATION

## (U) 1. GENERAL DISCUSSION

The experimental studies conducted in the research program can be divided into two parts. The first considers the study of small propellant samples to gain information directly applicable to development of the analytical model. Tests include:

- Measurement of steady-state burning rates in a pressurized combustion bomb
- Thermal profile measurements beneath the burning surface using microthermocouples
- Microcinematography studies of the burning surface in a pressurized window bomb
- Microscopic studies of propellant surfaces extinguished after steady-state combustion is established

The second part of the experimental studies considers the effect of motor environment upon the combustion of solid propellants. These tests are conducted in a propellant slab combustor representative of a motor environment which has been described in previous reports (2, 8).

Tests in this part of the experimental study include:

- Measurement of steady-state burning rates in a motor environment
- Measurement of the heat flux level in a motor environment
- Measurement of the weight fraction of unburned metals in the exhaust products of the slab combustor
- Characterization of the  $L^*$  instability in a motor environment.

In the following discussion, propellants selected for study in the aforementioned experiments are listed. Difficulties in establishing a binder system compatible with each ingredient of interest has delayed progress in initiating the experiments. Plans for this report period included testing a series of propellants wherein a carboxy-terminated polybutadiene (CTPB) propellant would serve as the control propellant. However, it was found that this binder system is not compatible with all of the ingredients included in the propellant list. As a result, efforts were directed toward selecting a new control binder.

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(U) Following the discussion pertaining to propellants to be studied, two tests developed during the past report period are outlined and results using a metalized composite propellant (AP/rubber-base/aluminum) and a nonmetalized version of the same propellant are presented. The metalized composite propellant formulation is given in Reference 10 and will hereafter be referred to as Mod AGC. The nonmetalized version will be referred to as Mod AGC-NOAL.

## 2. PROPELLANTS TO BE STUDIED IN SECOND YEAR

(C) A list of propellants was established for experimental studies to be conducted in the second year effort of this research program. The list includes propellants ranging from simple nonmetalized systems to those systems containing advanced oxidizer, energetic binders, and thermogens related to advanced propellant systems, as presented in the following tabulation.

| <u>Propellant No.</u> | <u>Formulation</u> |
|-----------------------|--------------------|
| 835-2                 | PU/AP              |
| 835-3                 | PU/KP              |
| 835-4                 | PU/HMX             |
| 835-5                 | Saturated PU/HAP   |
| 835-6                 | PU/AP/Al           |
| 835-7                 | PU/AP/LMH-1        |
| 835-8                 | PU-TVOPA/AP        |
| 835-9                 | PU-TVOPA/AP/Al     |
| 835-10                | PU-TVOPA/AP/LMH-1  |

(C) The objective of the research program is to develop an analytical model for combustion tailoring of propellants. In organizing a list of propellants for experimental studies it is necessary that the list is consistent with anticipated future propellant development trends and with the approach deemed most logical for the development of the analytical model. Thus, because the first step in the model development is to establish the effect of changing the oxidizer type, size, and concentration, simple propellant systems uncomplicated by metals and other additives were chosen for the first propellant types (835-2, 835-3, 835-4, 835-5). The oxidizers chosen were AP, KP, HMX, and HAP. The variation in size and concentration are established according to the size-concentration matrix shown in the following tabulation

| <u>Oxidizer Size</u> | <u>Oxidizer Concentration</u> |               |            |
|----------------------|-------------------------------|---------------|------------|
|                      | <u>High</u>                   | <u>Medium</u> | <u>Low</u> |
| 200 microns          |                               | X             |            |
| 50 microns           | X                             | X             | X          |
| 9 microns            |                               | X             |            |

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(U) Three unimodal grind sizes at 9, 50, and 200 microns were chosen to yield size effects and three different concentration levels (depending upon the oxidizer type) were chosen to yield concentration effects.

(C) The second step in developing the analytical model is to include the effect of metals in the combustion process. Propellants chosen to provide experimental data for this step (835-6 and 835-7) contain AP for the oxidizer with aluminum and LMH-1 for the metals.

(C) The third step in developing the model is to include formulations considered to be advanced systems. The propellants chosen for this phase of the model development (835-8, 835-9, and 835-10) contain a TVOPA-plasticized PU binder and AP for the oxidizer. Metal additives include aluminum and LMH-1.

(U) In preparing the list of propellants, an attempt was made to select propellant formulations that have a low hazard risk and a low probability of batch disposal because of incompatibility between ingredients leading to unsatisfactory cures. In addition, because this is a combustion research program and not a propellant development program, formulations already established at LPC were preferred over those requiring test batches. With these constraints, the selection of conventional nonmetalized and advanced propellants for the list posed no problem. However, the additional criterion that a systematic ingredient variation be incorporated in the list, commensurate with demands of the analytical program, introduced a situation wherein compatibility between ingredients became a problem.

(C) Originally, it was planned to use a CTPB/AP propellant as a control in the propellant series. The CTPB binder was to be used in formulations containing the different oxidizers (AP, KP, HMX, and HAP) and a PU system was to be used with formations containing TVOPA and LMH-1. However, it was learned from past studies conducted at LPC (9), that the CTPB/HAP propellant was not a compatible combination. Thus, a new control propellant was deemed necessary for the list.

(C) The choice was a PU/AP propellant (R-18 prepolymer with isocyanate-triol cure). This binder system is compatible with ingredients in all but one of the formulations listed. The exception is the propellant containing HAP. For that propellant, a saturated PU binder (Telagen S prepolymer with isocyanate cure) was found to yield compatibility with HAP. Small batches of the Sat-PU/HAP propellant were mixed to confirm this.

### (U) 3. STUDY OF THE BURNING SURFACE STRUCTURE

The purpose of this study is to provide necessary information for formulating a realistic model of solid propellant combustion encompassing a number of oxidizer types. In Hermance's original model for composite propellants (containing AP), it was postulated that the oxidizer crystal

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burned in such a manner that a "cusp" formed at the crystal-binder interface as discussed in Section III. To check the validity of this assumption, or any other assumption regarding the surface structure, it was decided to develop a technique to study the surface of each of the propellants included in the propellant list.

The approach deemed most acceptable for this study involved extinguishing burning samples of propellants by rapid depressurization. This technique allows examination of the surface structure with a high power microscope and the advantage of high magnification photography without complications arising from combustion products issuing from the burning surface and movement of the propellant surface. In the following discussion, the technique is described in detail and results are presented from tests using the Mod AGC-NOAL propellant.

The extinguishment test apparatus was fabricated from the slab combustor and the pintle apparatus described in Reference 8. The free volume of the slab combustor was reduced to 27 in.<sup>3</sup> and a mount for a small propellant sample ( $\frac{1}{4}$  by  $\frac{1}{4}$  by  $\frac{3}{4}$  inches) was installed in such a manner that the burning surface could be oriented parallel or perpendicular to the axis of the  $1\frac{1}{2}$ -inch diameter nozzle port. The nozzle port was sealed to allow the combustor to be pressurized by driving a plug mounted on the pintle shaft into the nozzle port. Combustion termination was detected by an IRC model 49-0614 photocell mounted directly on a plexiglas window viewing the entire length of the propellant sample.

Operation of the extinguishment test apparatus consist of sealing the nozzle port with the pintle plug, pressurizing the combustor to the desired test pressure, and igniting the propellant strand. After steady-state combustion is attained, the pintle plug is withdrawn, releasing the chamber gases and thereby extinguishing the propellant sample by rapid depressurization. The pressure in the combustor during combustion remains essentially constant by virtue of the small propellant sample size and the large combustor free volume.

Tests were conducted in the test apparatus using the Mod AGC-NOAL at initial combustor pressures of 1 atmosphere, 250, 450, 500, and 600 psia. The samples were examined through an American Optical Microscope (Spencer type Binocular Bench Scope) with 10-80x magnification. Attention was given to exposed oxidizer crystals and the possibility of a cusp existing between the exposed oxidizer crystal and surrounding binder.

A photograph of the extinguished surface of a propellant sample burning at 1 atmosphere is shown in Figure 5. The overall view shows extinguished crystals projecting above the propellant binder surface. Microscopic examination of the surface as presented in Figure 6 shows that no cusp exists around the oxidizer crystals. In addition, the surface of exposed crystals is similar in shape to the original crystal and the surfaces are smooth with no discontinuities where the crystals join the propellant binder surface.

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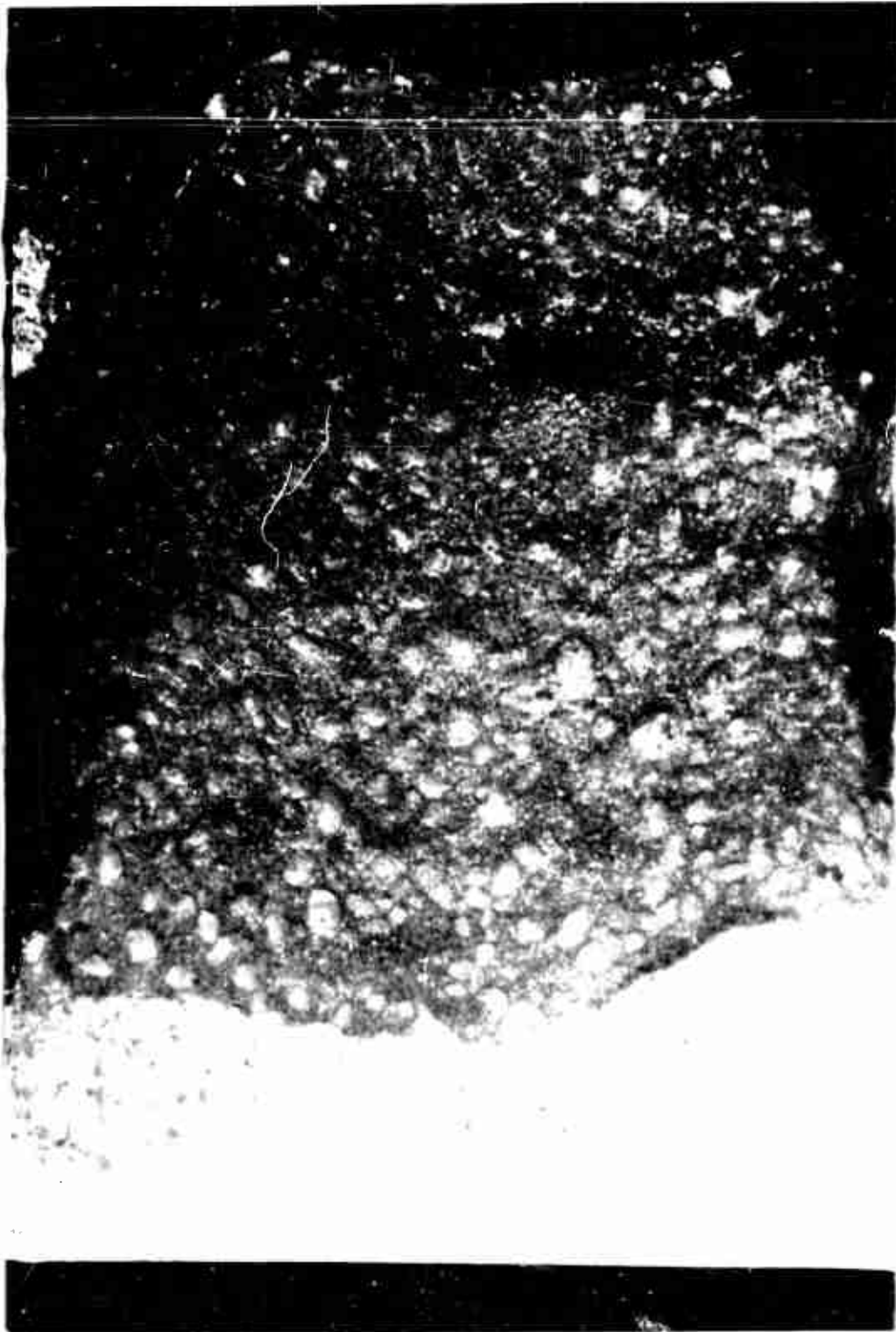


Figure 5 Photograph of Extinguished Propellant Surface  
( $P_{\text{initial}} = 1 \text{ atm}$ , Propellant = Mod AGC-NOAL)

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Figure 6 Microphotograph of Extinguished Propellant Surface  
( $P_{\text{initial}} = 1 \text{ atm}$ , Propellant = Mod AGC-NOAL)

Tests conducted at higher pressures exhibited a very interesting phenomenon. At initial burning pressures of 250 and 450 psia, exposed crystals again were found to project above the propellant binder surface. Microscopic investigations of the surface showed, however, that the shape of the exposed crystals was not similar to the original crystal shape as noted in the tests conducted at 1 atmosphere. Instead, the crystal projected only at its center as depicted in Figure 7. A microphotograph demonstrating this crystal configuration is shown in Figure 8. Again no cusp is observed at the edge of the oxidizer crystal.

At higher pressure tests, the oxidizer crystals did not project above the fuel surface, resulting in a relatively flat surface with some indentations at crystal sites. A photograph showing the complete extinguished surface of a strand burning at a pressure of 528 psia is shown in Figure 9. Microphotographs of the same surface are shown in Figure 10. Again, no cusp can be observed at the edge of the oxidizer crystal.

The highest pressure examined was 600 psia. Tests at this pressure showed that the crystals again project above the surface as seen in Figure 11. However, one important difference was noted when the strands were examined under a microscope. At this pressure, the small dimple in the center of the exposed crystal surface appeared to be hollow. Microphotographs of such crystals are shown in Figure 12. The presence of the hollow dimple was not restricted to isolated crystals, but instead was generally true of oxidizer particles exposed on the surface as noted from a careful examination of the overall view of a strand extinguished at 600 psia as shown in Figure 11. Again, a close examination of the crystal edges showed no cusp as assumed in Hermance's model.

There are several important observations that can be made from the results of these extinguishment tests. First, it appears obvious that there is no basis for Hermance's assumption that a cusp appears at the oxidizer crystal edge. Thus, the revision of the model discussed in subsection III 6 of this report appears necessary to eliminate this shortcoming in the model. Second, the oxidizer crystals appear to burn more rapidly than the surrounding binder at high pressures thereby resulting in smaller values of the parameter  $h$  (defined in Section III) than would be realized at low pressures. Third, the presence of the hollow dimple on the exposed crystals appear to indicate a melt on the crystal surface during combustion.

Several additional comments on these observations appear necessary to qualify their validity. It was mentioned previously that a photocell was employed to detect combustion termination in the combustor. In some tests, it was noted that after the photocell indicated that detectable radiation had decreased because of extinguishment, small light levels still were detected by the photocell. This would indicate, then that the pressure at which the samples were burning when depressurization was initiated is not necessarily the pressure at which the sample extinguished. However, the fact remains that there is definitely an effect of varying the pressure before or during depressurization of the combustor. This problem will be examined in more detail.

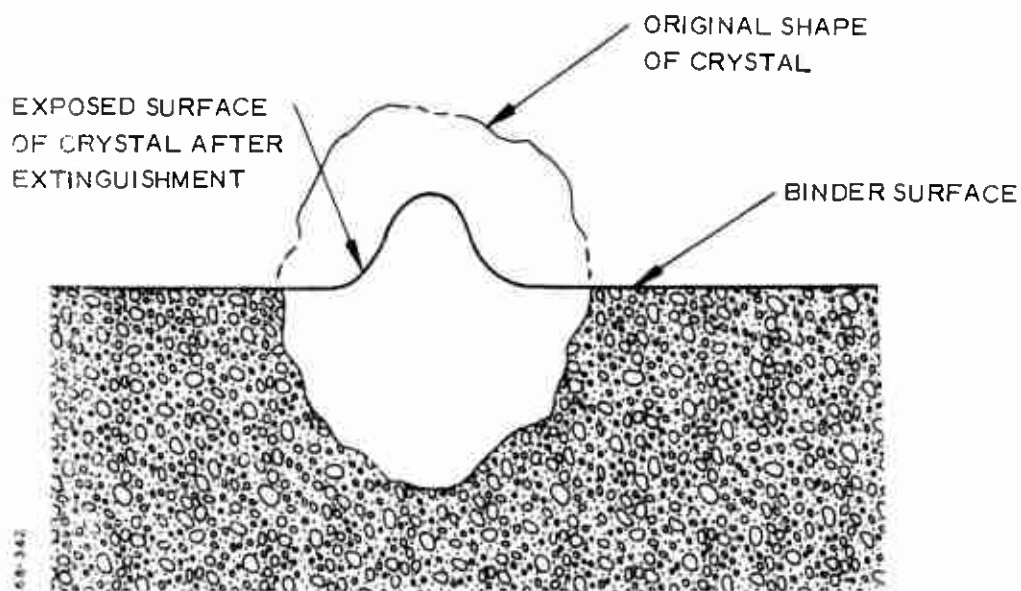


Figure 7 - Schematic of Oxidizer Crystal  
after Extinguishment

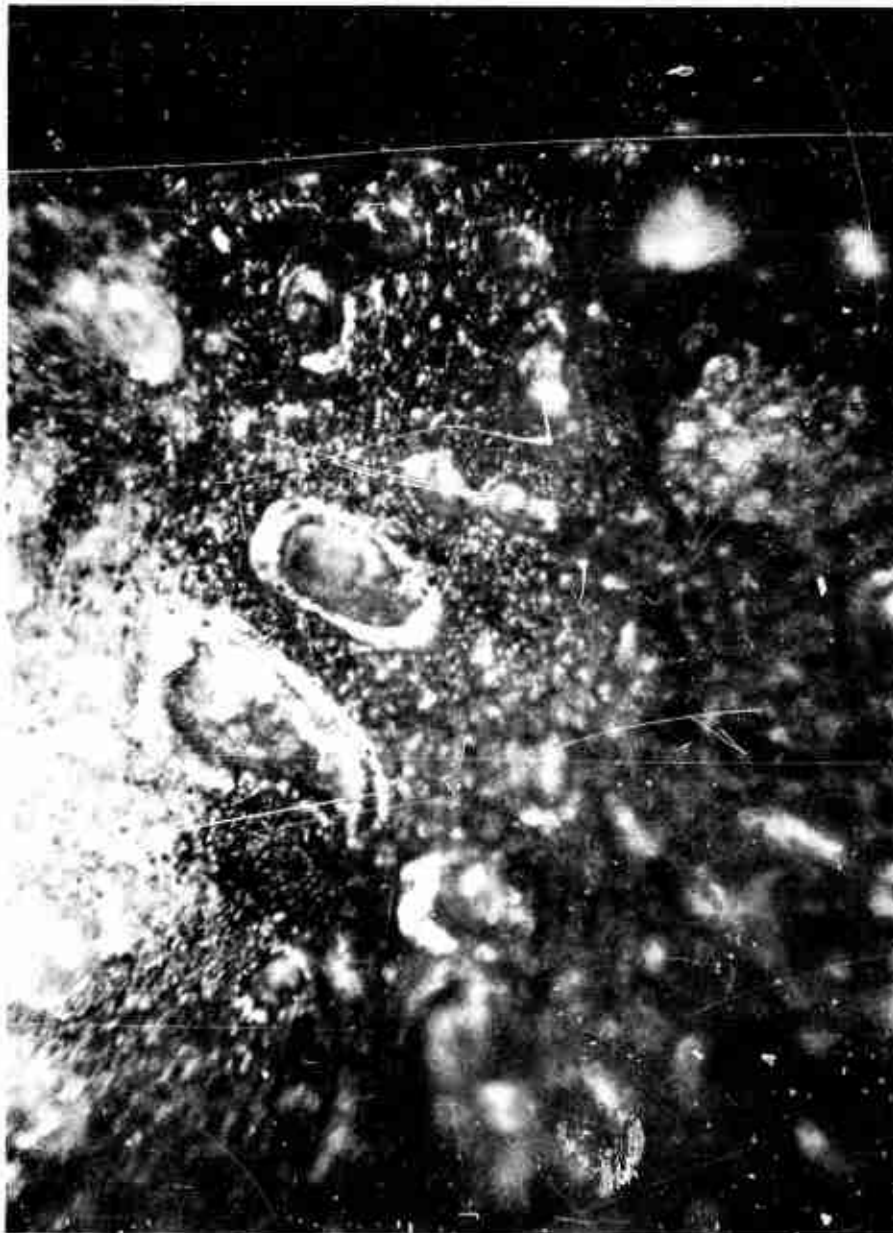


Figure 8 Microphotograph of Extinguished Propellant Surface Showing Center Projection of Oxidizer Crystals (Propellant = Mod AGC-NOAL)



Figure 9 Photograph of Extinguished Propellant Surface  
( $P_{\text{initial}} = 528$  psia, Propellant = Mod AGC-NOAL)

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Figure 10 Microphotographs of Extinguished Propellant Surface  
( $P_{\text{initial}} = 528 \text{ psia}$ , Propellant = Mod AGC-NOAL)



Figure 11 Photograph of Extinguished Propellant Surface  
( $P_{\text{initial}}$  = 600 psia, Propellant = Mod AGC-NOAL)

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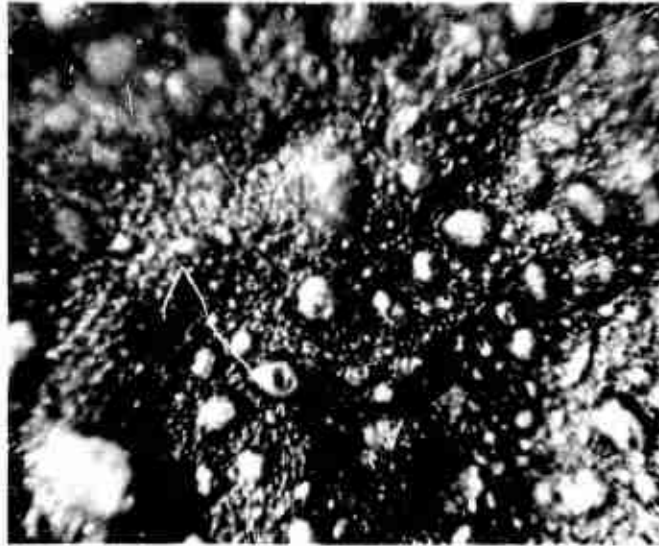


Figure 12 Microphotographs of Extinguished Propellant Surface  
( $P_{\text{initial}} = 600$  psia, Propellant = Mod AGC-NOAL)

The possibility that the AP crystals undergo melting during combustion at the higher pressures is intriguing. Studies with single AP crystals at NWC by Hightower (6 and 10) have reported a liquid layer on the AP surface during deflagration. Studies indicating a liquid layer on AP crystals when burned in a binder matrix have been reported by Jensen (11).

#### (U) 4. HEAT FLUX MEASUREMENTS IN THE SLAB COMBUSTOR

The objective of the heat flux measurements in the slab combustor is to provide a qualitative understanding of the gas-cloud imposed convective and radiative heat transfer level in a motor environment and its effect upon the combustion process of solid propellants. Results presented in the previous report period (2) for the Mod AGC propellant indicated that the heat flux was largely radiative because the measured heat flux in the slab combustor correlated better with pressure as shown in Figure 13 than mass flow rate between the propellant slabs. It was also reported that the heat flux data exhibited a large degree of scatter at pressures greater than 150 psia in the combustor. Two possible origins of the scatter were advanced:

- The emissivity of the calorimeter sensing face was changing due to deposition on the sensing face.
- Inhibitor failure was occurring on the propellant slab close to the calorimeter face.

In an effort to check the validity of the heat flux data and the two premises, tests were conducted in the combustor using the Mod AGC propellant with two total heat flux calorimeters mounted in the combustor. The calorimeters were located opposite each other such that their outputs would be the same if the aforementioned premises were not important. The sensing surfaces of the calorimeters were cleaned and coated before each test with camphor-black to ensure that the initial emissivity was the same for both calorimeters.

The results of such a test are presented in Figure 14. It can be observed that the two heat flux levels are not precisely the same throughout the test; however, it is important to note that variations in heat flux are in phase for the two calorimeter outputs. This would indicate that there is probably a change in the surface conditions of the calorimeters during the ignition process due to ignition material depositing on the sensing surfaces. However, after the ignition process, the fluctuations in heat flux appear real and not due to sensing surface changes.

Tests with the Mod AGC-NOAL version of this propellant yielded similar findings. Results of a test using this propellant is shown in Figure 15. Again the two calorimeters are not in precise agreement; however, changes in flux levels are observed to be in phase.

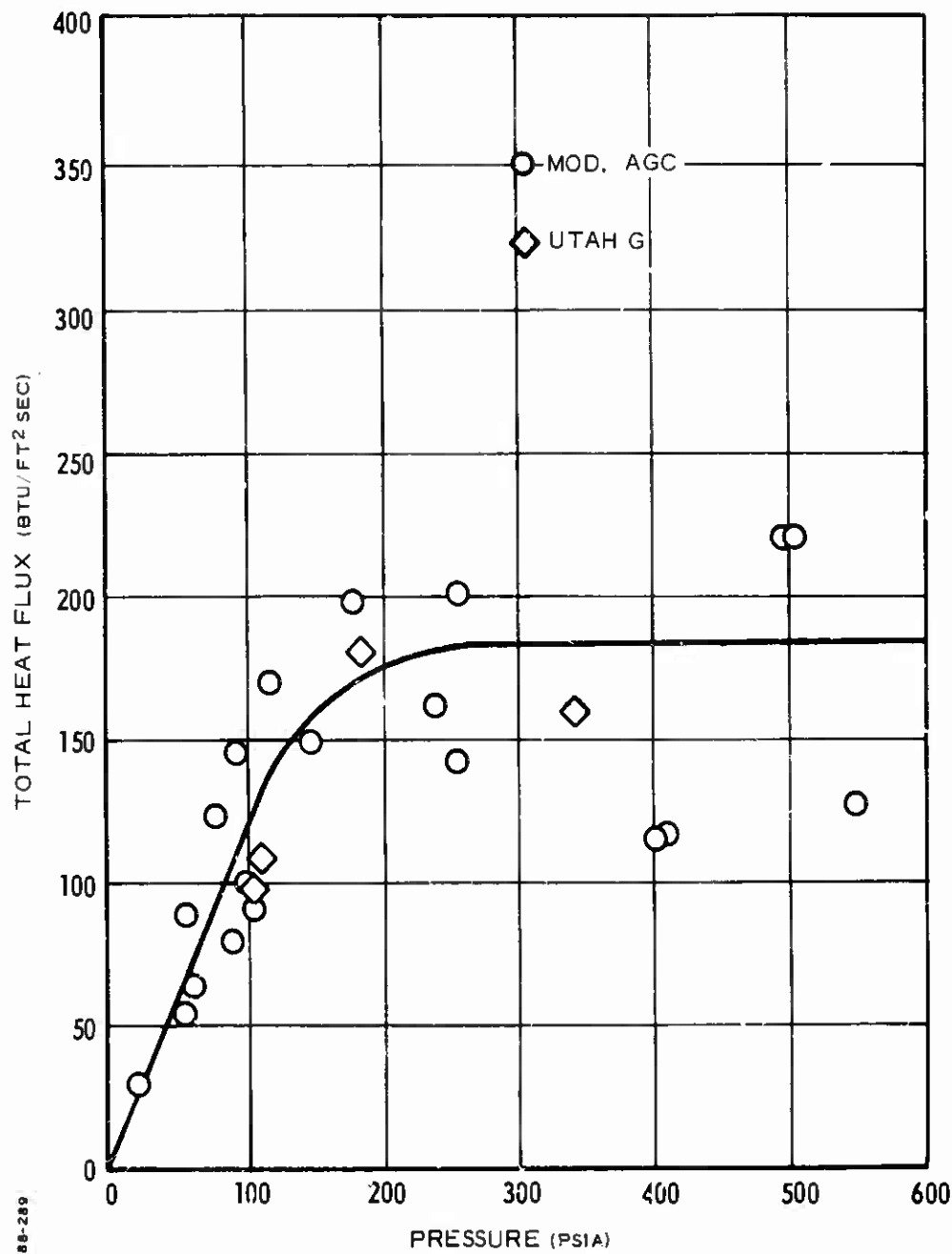


Figure 13 - Total Heat Flux Correlated with Pressure

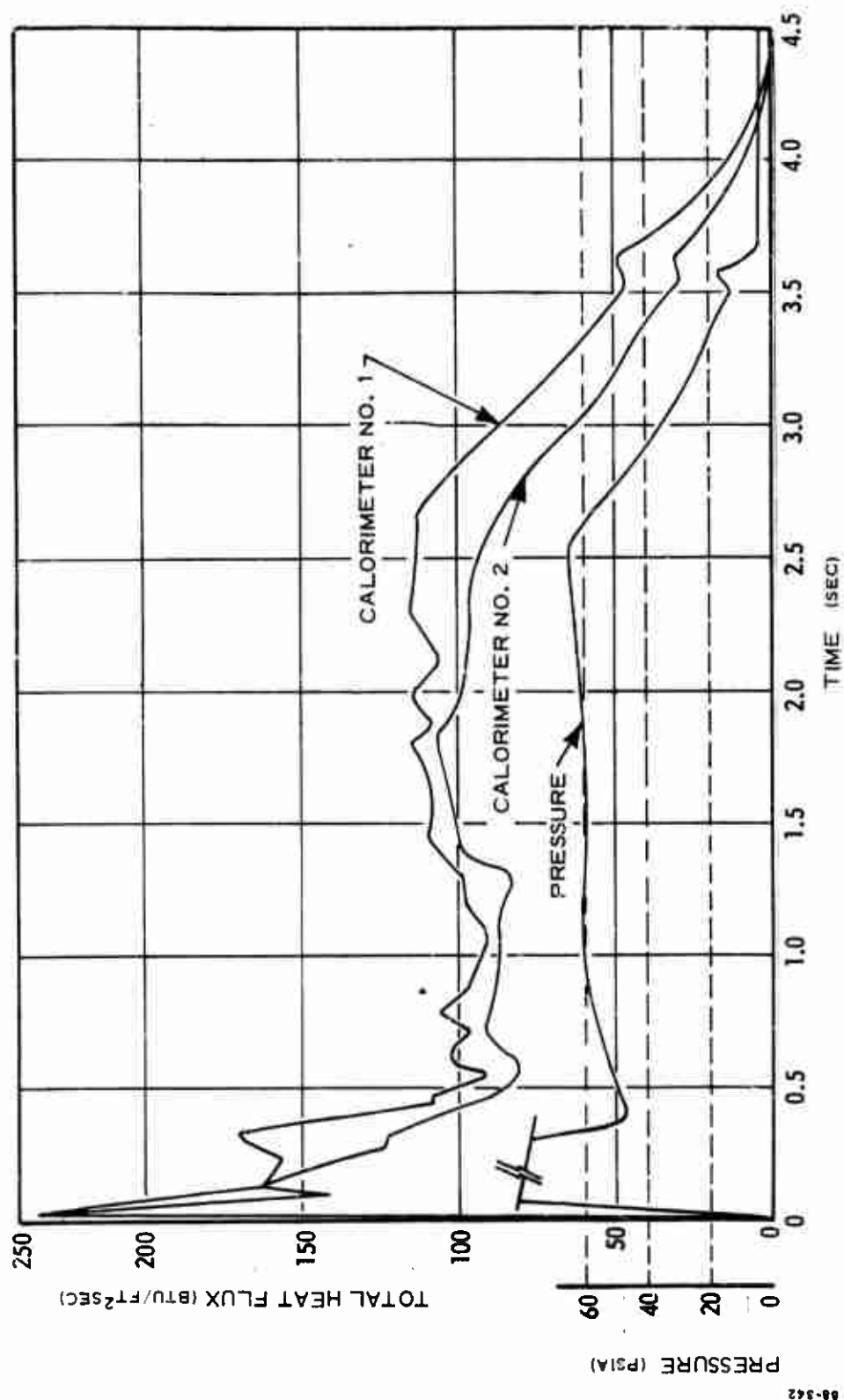


Figure 14 Heat Flux Measurements with Two Calorimeters (Propellant: Mod AGC)

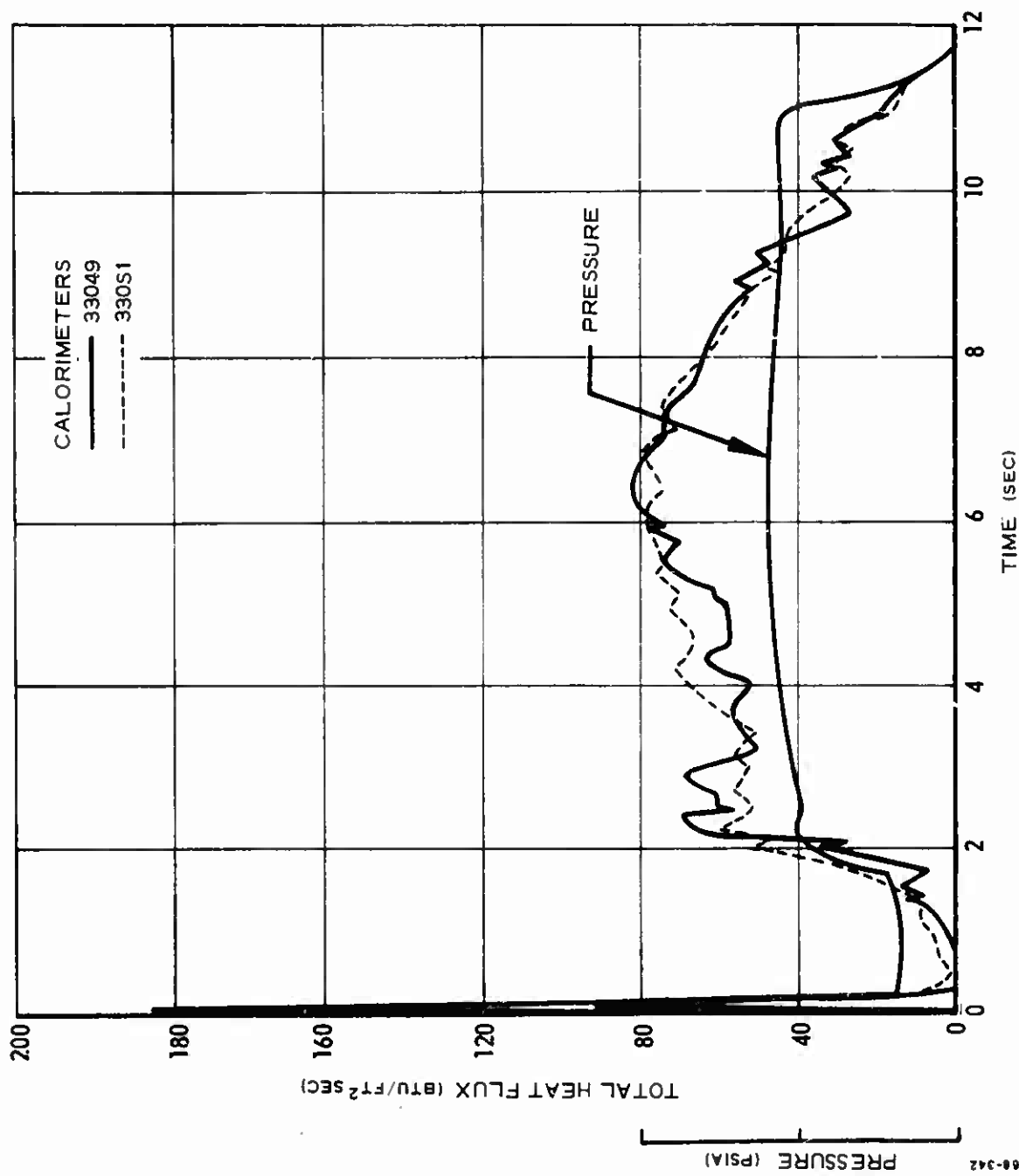


Figure 15 Total Heat Flux Measured with Two Calorimeters (Propellant = Mod AGC-NOAL)

In an effort to relate the measured heat flux levels to the effect of the thermal environment upon the combustion process of solid propellants, the data obtained from the Mod AGC tests were analyzed by a straightforward, simplified calculation comparing the measured heat flux to that required to raise the temperature of the propellant from its initial value to that found at the surface. Assuming an average total absorptivity for the propellant surface, the maximum radiant heat flux absorbed by the surface is

$$Q_{\text{rad}} = \frac{\alpha_{\text{prop}}}{\epsilon_{\text{cal}}} Q_{\text{cal}}$$

where

$\alpha_{\text{prop}}$  = total absorptivity of propellant surface  $\approx 0.8$

$\epsilon_{\text{cal}}$  = emissivity of calorimeter sensing surface = 0.96

$Q_{\text{cal}}$  = indicated heat flux received by the calorimeter

The heat flux necessary to heat the propellant to the temperature found at the propellant surface is

$$Q = \rho_p r c_p (T_s - T_o)$$

where

$\rho_p$  = propellant density  $\approx 0.06 \text{ lbm/in.}^3$

$r$  = propellant burning rate

$c_p$  = propellant specific heat  $\approx 0.27 \text{ Btu/lbm-}^\circ\text{R}$

$T_s$  = surface temperature  $\approx 550^\circ\text{C}$

$T_o$  = initial temperature  $\approx 20^\circ\text{C}$

Plotting the ratio  $Q_{\text{rad}}/Q$  against motor pressure yields a measure of the importance of the thermal environment upon the combustion process. Such a plot for the Mod AGC propellant is shown in Figure 16. At low pressures the effect of the thermal environment would appear to be small. However, as the pressure increases, the indication is that the contribution becomes more important. At pressures greater than 200 psig, the effect of the thermal environment appears to decrease. This occurs because the heat required by the propellant continues to increase while the measured heat flux remains constant; thus, a pressure threshold effect is indicated wherein the motor thermal environment is less important at pressures greater than the pressure where  $Q_{\text{rad}}/Q$  becomes a maximum (hereafter referred to as threshold pressure).

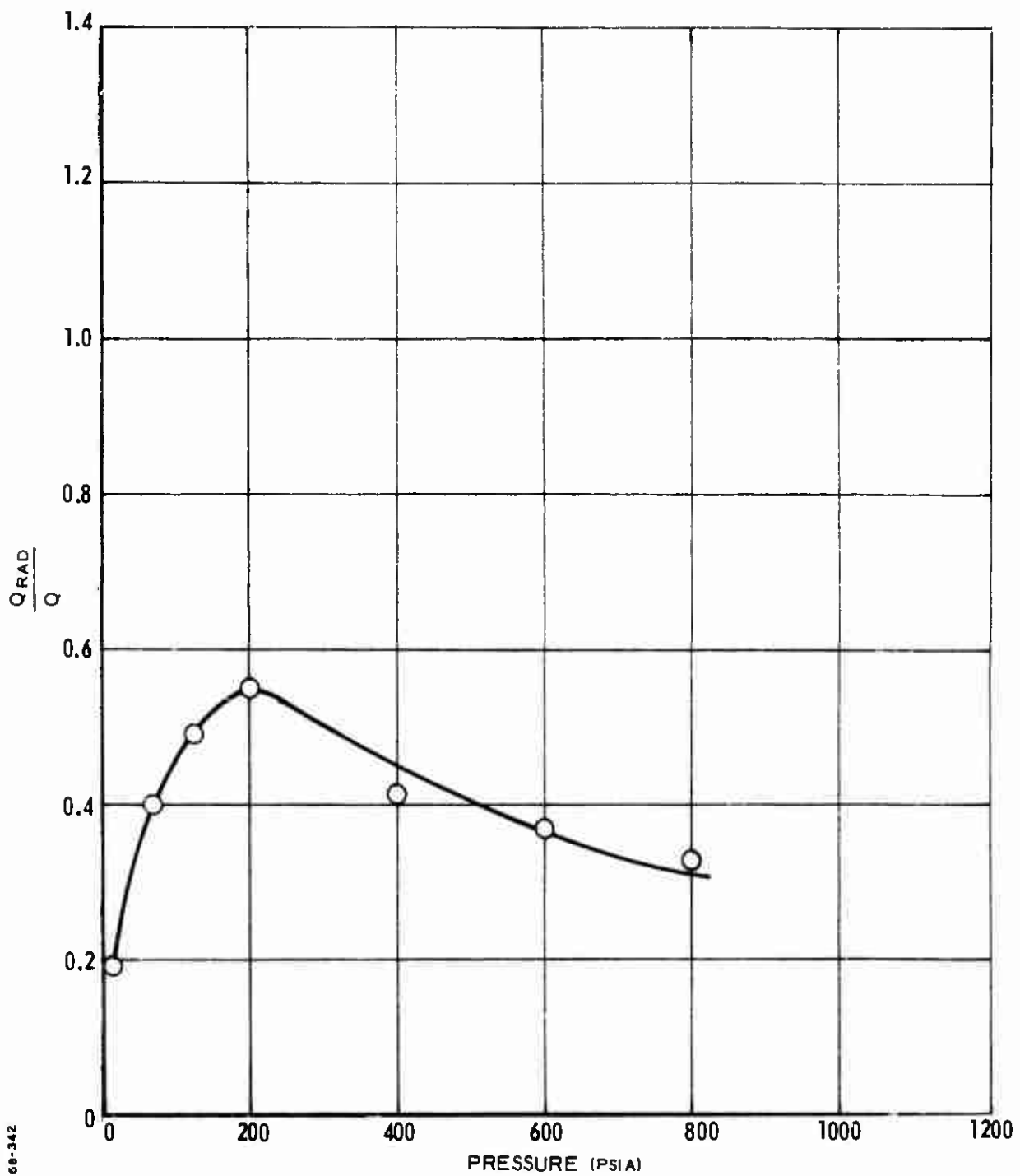


Figure 16 - Contribution of Motor Thermal Environment to Heating of Propellant

It should be emphasized that these results should not be construed as a conclusion of the effect the thermal environment of the motor has on the combustion process. Instead, the results are intended only to introduce a comparison of the measured heat flux to one that is associated with the combustion process. If there is a threshold effect from the thermal environment, its presence should be detectable from measurements of in-motor burning rates. For this reason, efforts were directed toward developing a technique for such a measurement. This technique is described in the subsequent section.

#### (U) 5. BURNING RATE MEASUREMENTS IN THE SLAB COMBUSTOR

Measurement of propellant burning rates in a pressurized bomb using small propellant samples provides an economical and practical means of characterizing the burning rate of propellants over a range of pressures. As mentioned previously, this characterization provides an important set of data for developing the analytical model. However, knowledge of the thermal environment developed by propellants in a rocket motor and the effect of that environment upon the burning rate cannot be gained from small propellant tests. However, direct measurement of the burning rate apparatus simulating the environment of a rocket motor does provide information regarding the effect of the thermal environment. For this reason, a technique was developed for burning rate measurements in the slab combustor.

##### a. Method of Approach

The technique developed for in-motor burning rate measurements employs a high-speed motion picture camera for recording the movement of the burning propellant surface with respect to a reference scale located inside the slab combustor. The reference scale is etched on the inside surface of a 2-inch diameter plexiglas window mounted in the sidewall of the combustor. The scale is in contact with the side of the propellant slab as shown in Figure 17.

The dimensions of the propellant slab are 6 by 2 by 0.625 inches. The propellant slab is bonded to a 6 by 2 by 0.25-inch steel plate and inhibitor is applied to all surfaces except the 6 by 2-inch burning surface and the surface covered by the window. The surface of the window is protected by a thin layer of optically clear vacuum grease (Celvacene grease) to allow an unobstructed view of the burning surface and discourage burning between the window and propellant side.

The high speed camera is equipped with a timing light. Thus, the burning rate is measured by dividing the distance between the reference scale lines (0.1 in.) by the time it takes for the propellant to burn through this distance.

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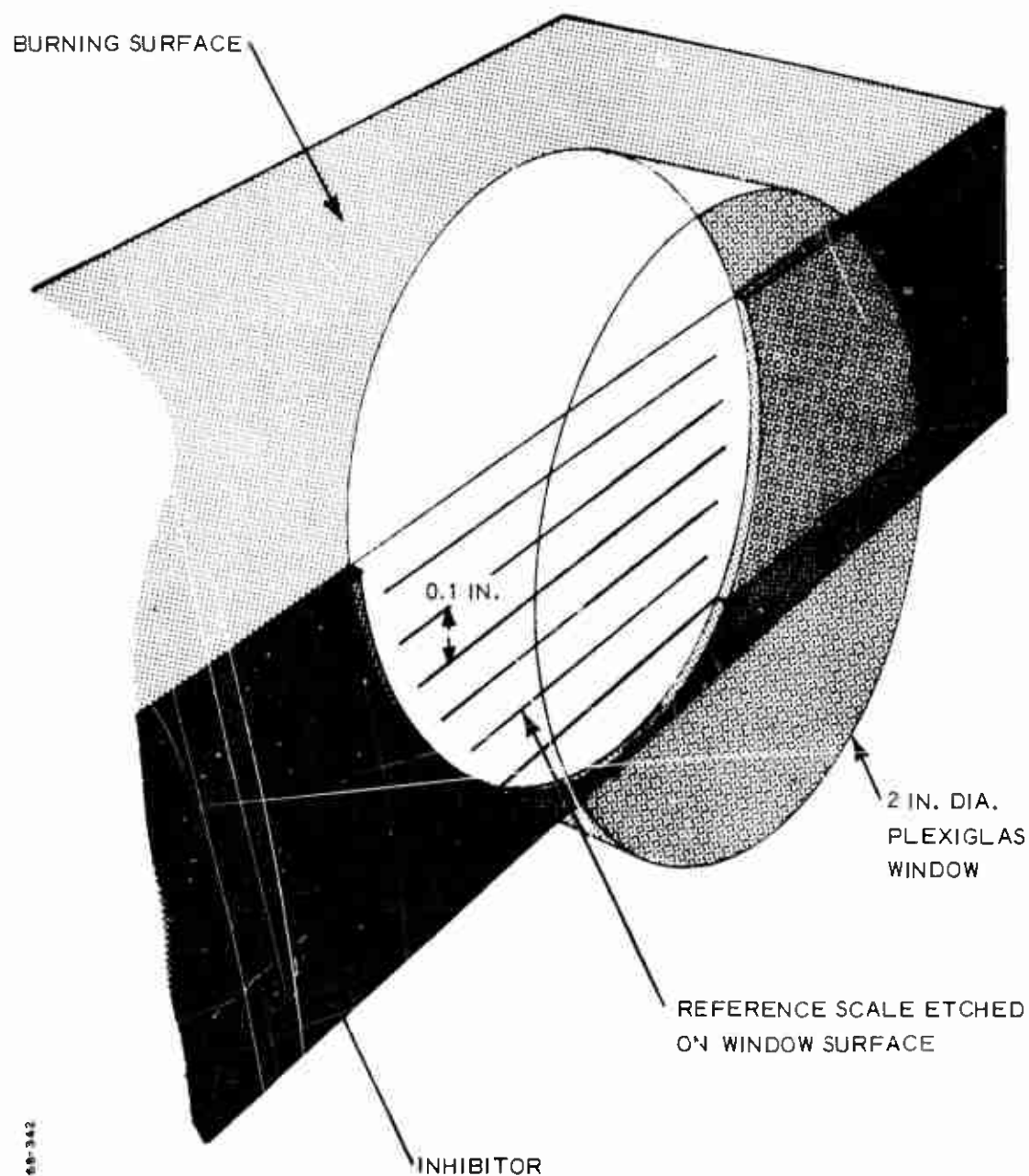


Figure 17 - Propellant Slab and Window with Reference Scale

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### b. Results

The burning rate measurement technique was developed using the Mod AGC-NOAL propellant. Tests were conducted at pressures of 45, 80, 250, and 900 psia. Values of in-motor burning rate measured from these tests are shown in Figure 18. For comparison, the burning rate for the same propellant measured in a pressurized combustion bomb using small propellant samples is included in this figure.

A detailed inspection of the film records showed that the burning surface receded parallel to the reference scale allowing minimum error in the burning rate calculation because of surface slant. The accuracy of the measurement is about  $\pm 2$  percent.

### c. Discussion of Results

Comparison of the in-motor and small sample burning rates shows the in-motor rates to be about 10 percent greater than the small sample rates. This difference is quite obviously not due to motor environment effects alone. The technique of measurement must contribute to the differences observed. However, the results of the heat flux measurements reported in the previous section indicate that the level of the thermal environment is not negligible when compared to the heat flux required to raise the propellant temperature to that of the burning surface. Thus, the conclusion that the technique alone accounts for the 10 percent difference is not valid either. Not evident in these data is the threshold pressure effect described in the previous section. One obvious reason for the absence of this effect is that the propellant used to develop the burning rate technique did not contain aluminum. Results from future tests using propellants containing metals will be examined for the threshold pressure effect. Until that time, results from all past tests will be analyzed for the threshold effect.

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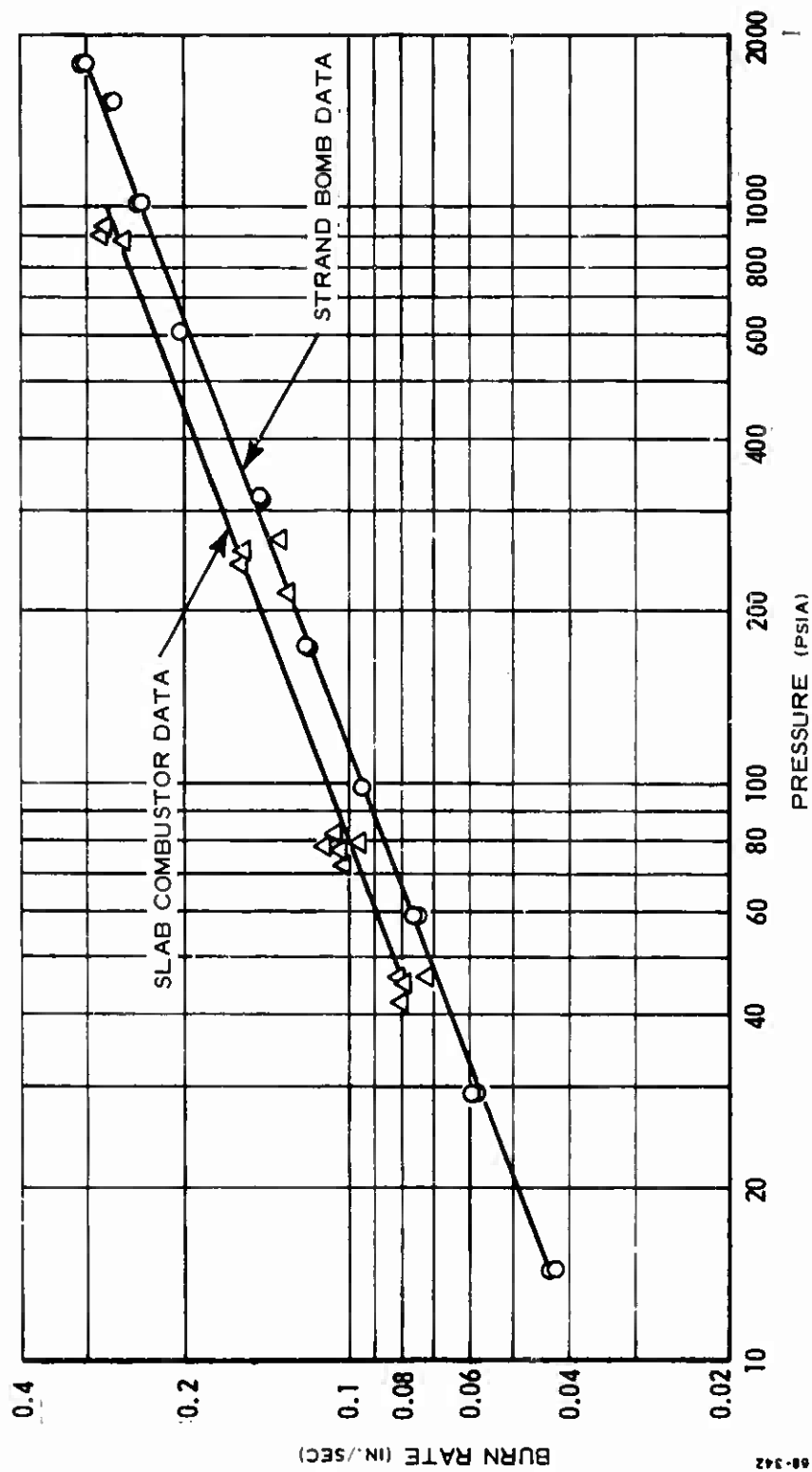


Figure 18 In-Motor Burning Rates for Mod AGC-NOAL Propellant

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835 Phase III

## SECTION V

### PLANS FOR THE NEXT PHASE

#### 1. ANALYTICAL INVESTIGATION

Future efforts on the analytical portion of the program will be directed mainly towards further development of the model. It appears likely that the gas phase analysis of the original Hermance model will need to be revised to account for the heterogeneous structure of the flame. The modifications that are needed to produce a realistic but consistent model will be made.

The present analysis (and future efforts) will be programmed for the computer and adapted to run on the optimization program that was discussed in the most recent report (2). The latter will allow the critical parameters to be identified and their effect studied for a minimal amount of effort.

#### 2. EXPERIMENTAL INVESTIGATION

In the subsequent report period, the propellant formulations dealing with changes in oxidizer type, concentration, and grind size will be tested. Experiments with small samples of the formulations 835-2, 835-3, 835-4, and 835-5 will be conducted. Experiments in the slab combustor will be conducted using formulation 835-3. Tests also will be initiated to study the effect of aluminum and LMH-1 on the combustion process through the use of propellant formulations 835-6 and 835-7.

## NOMENCLATURE

|                  |                                                                                         |
|------------------|-----------------------------------------------------------------------------------------|
| A                | Arrhenius frequency factor                                                              |
| $C_1$            | conversion constant                                                                     |
| $C_p$            | heat capacity of propellant                                                             |
| $D_o$            | original oxidizer diameter                                                              |
| E                | activation energy                                                                       |
| h                | defined in Figure 4                                                                     |
| $K_o$            | oxidizer ignition delay parameter [see Equation (10)]                                   |
| L                | the distance from the top of the original crystal to the burning surface (see Figure 4) |
| M                | molecular weight                                                                        |
| m                | pressure exponent in $t_{ign}$ [see Equation (10)]                                      |
| m                | mass flux associated with propellant components                                         |
| N                | number of oxidizer crystals per unit area of propellant surface                         |
| n                | diameter exponent in $t_{ign}$ [see Equation (10)]                                      |
| P                | pressure                                                                                |
| Q                | heat flux to raise propellant to surface temperature                                    |
| $Q_{rad}$        | radiant heat absorbed by propellant surface                                             |
| $Q_{cal}$        | indicated heat flux received by calorimeter                                             |
| R                | gas constant                                                                            |
| R                | defined in Figure 4                                                                     |
| r                | propellant burning rate                                                                 |
| $S_g S_{og} S_p$ | surface area, total reacting area, total planar area                                    |
| $S_f$            | total fuel area                                                                         |
| $t_{ign}$        | ignition delay time for an AP crystal                                                   |

|                  |                                                                                                |
|------------------|------------------------------------------------------------------------------------------------|
| $t_b$            | characteristic total time of exposure of AP crystal<br>(ignition delay time plus burning time) |
| $T_s$            | propellant surface temperature                                                                 |
| $T_o$            | initial propellant temperature                                                                 |
| $v_{ap}$         | linear burning rate of AP                                                                      |
| $x$              | defined in Figure 4                                                                            |
| $a_{prop}$       | total absorptivity of propellant surface                                                       |
|                  | fraction of fuel area undergoing heterogeneous reaction                                        |
| $\epsilon_{cal}$ | emissivity of calorimeter sensing face                                                         |
| $v$              | volume fraction of oxidizer in propellant                                                      |
| $\rho$           | density                                                                                        |
| $\rho_p$         | propellant density                                                                             |

#### Subscripts

|    |                            |
|----|----------------------------|
| ox | refers to oxidizer         |
| sr | refers to surface reaction |
| f  | refers to fuel             |

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Security Classification

| DOCUMENT CONTROL DATA - R & D                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |                                                                             |                                                                                                                                                                                         |
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| 1. ORIGINATING ACTIVITY (Corporate author)<br>Lockheed Propulsion Company<br>A Division of Lockheed Aircraft Corporation<br>P. O. Box 111, Redlands, California                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                                                                             | 2a. REPORT SECURITY CLASSIFICATION<br>Confidential Noform                                                                                                                               |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                                                                             | 2b. GROUP<br>4                                                                                                                                                                          |
| 3. REPORT TITLE<br>Combustion Tailoring Criteria for Solid Propellants                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           |                                                                             |                                                                                                                                                                                         |
| 4. DESCRIPTIVE NOTES (Type of report and inclusive dates)<br>Phase III Report, covering period 30 April to 1 July 1968                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           |                                                                             |                                                                                                                                                                                         |
| 5. AUTHOR(S) (First name, middle initial, last name)<br>Derr, Ronald L., Beckstead, M. W.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                                                             |                                                                                                                                                                                         |
| 6. REPORT DATE<br>July 1968                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      | 6a. TOTAL NO. OF PAGES<br>44                                                | 7b. NO. OF REFS<br>11                                                                                                                                                                   |
| 8a. CONTRACT OR GRANT NO.<br>F04611-67-C-0089                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    | 9a. ORIGINATOR'S REPORT NUMBER(S)<br>LPC Report 835 Phase III               |                                                                                                                                                                                         |
| b. PROJECT NO.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   |                                                                             |                                                                                                                                                                                         |
| c.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               | 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) |                                                                                                                                                                                         |
| d.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                                                             |                                                                                                                                                                                         |
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| 11. SUPPLEMENTARY NOTES                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          |                                                                             | 12. SPONSORING MILITARY ACTIVITY<br>Air Force Rocket Propulsion Laboratory<br>Research and Technology Division<br>Air Force Systems Command<br>United States Air Force, Edwards, Calif. |
| 13. ABSTRACT<br>The objective of this research program is to develop an analytical model describing the steady-state combustion of solid propellants which is suitably equipped to provide combustion tailoring criteria for both state-of-the-art and advanced propellant formulations. The model will be developed and implemented from experimental data acquired from a dual experimental program. Tests with small samples of propellants will provide experimental data for modeling the propellant combustion process for different ingredient variations. Additional tests simulating the environment of a rocket motor will provide experimental data showing the effect of rocket motor environment upon the combustion process. During Phase III, work was directed toward developing the analytical model and modifying aspects of the model. A list of propellants was chosen for study in tests to be conducted during the remainder of the research program. A technique was developed for examining the surface of burning propellants. Small samples of a CTPB/AP propellant were burned in a pressurized combustion bomb and extinguished by rapid depressurization of the bomb. The surface structure was examined through a 10X - 80X microscope. Results from tests conducted in apparatus simulating the environment of a rocket motor (slab combustor) and using a CTPB/AP/A1 propellant showed that the thermal environment of the combustor reaches a maximum level at 200 psia. To learn what effect the thermal environment has upon the propellant burning rate, a technique was developed to directly measure burning rate in the slab combustor. The approach uses a high-speed camera to record the movement of the propellant burning surface with respect to a reference scale. |                                                                             |                                                                                                                                                                                         |

| 14. KEY WORDS                                                                                                                     | LINK A |    | LINK B |    | LINK C |    |
|-----------------------------------------------------------------------------------------------------------------------------------|--------|----|--------|----|--------|----|
|                                                                                                                                   | ROLE   | WT | ROLE   | WT | ROLE   | WT |
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