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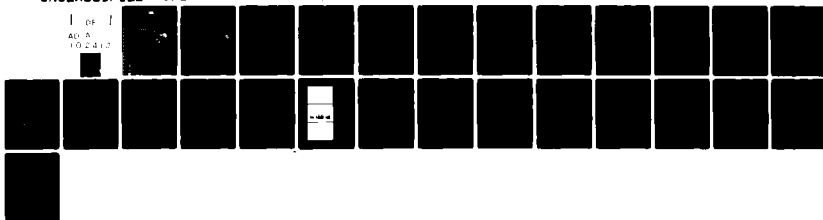
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Composite Propellant Combustion And Transition to Detonation Annual Research Progress Report

N. S. Cohen
L. D. Strand

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February 1981

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Prepared for
U.S. Air Force Office of
Scientific Research
Through an agreement with
National Aeronautics and Space Administration
by
→ Jet Propulsion Laboratory
California Institute of Technology
Pasadena, California

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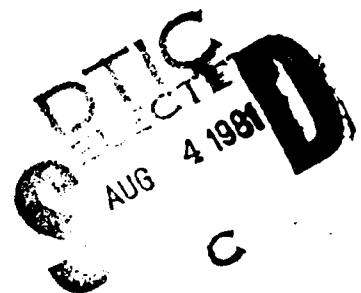
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Annual Research Progress Report

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out features that are not contained in current propellant combustion models. The potential of the apparatus for more detailed future work is discussed. A literature review of combustion response models that attempt to account for the heterogeneity of composite propellants was completed, and a new response function model based upon a preferred frequency concept was formulated.

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ACRONYMS/ABBREVIATIONS

AFRPL	Air Force Rocket Propulsion Laboratory
AIAA	American Institute of Aeronautics and Astronautics
AP	ammonium perchlorate
ASME	American Society of Mechanical Engineers
BDP	Beckstead-Derr-Price model of oxidizer monopropellant combustion
BYU	Brigham Young University
CMDB	Composite-modified double-base propellant
CPIA	Chemical Propulsion Information Agency (at Johns Hopkins University)
DDT	deflagration-detonation transition
FY	fiscal year
HMX	cyclotetramethylene tetranitramine
HTPB	hydroxyl-terminated polybutadiene
JANNAF	Joint Army-Navy-NASA-Air Force
MPa	Megapascal
NWC	Naval Weapons Center
O/F	oxidizer/fuel (ratio)
SAE	Society of Automotive Engineers
SRI	Stanford Research Institute
μ	micron

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SECTION I

RESEARCH OBJECTIVE

1.1 OVERALL OBJECTIVE

The objective of this research program is to further the understanding of composite solid propellant combustion under steady-state conditions and in response to transient stimuli. This understanding is expressed by the development of suitable and descriptive analytical models. Special attention is given to advanced nitramine-containing propellants which are of interest to the Air Force for armament and rocket propulsion applications, and to problems of burn-rate tailoring, combustion instability, and deflagration-detonation transition (DDT) which can result from the use of the nitramine family of propellants.

1.2 SPECIFIC OBJECTIVES FOR FISCAL YEAR 1980

- (1) Complete the development of the improved steady-state combustion model.
- (2) Apply the porous burner apparatus, configured to simulate composite propellants, to study the properties of diffusion flames.
- (3) Apply the closed vessel apparatus to study the burning rates and extinguished surface structures of propellants at very high pressures.
- (4) Formulate an analytical model for the pressure-coupled response function, with specific regard to the effects of composite propellant heterogeneity.

An improved steady-state model was developed and verified for ammonium perchlorate (AP) composite propellants in the course of FY 1979 work. The following tasks were planned to complete this work. First, apply the model to Miller's series of AP propellants (Ref. 1) to provide a more thorough evaluation. Second, apply the model to HMX-containing propellants to verify its generality. Third, develop and incorporate a model for aluminum agglomeration, and apply the model to aluminized propellants to test predictability of agglomeration and burn rate effects. Fourth, incorporate a model of active binder combustion and apply the model to composite-modified double-base (CMDB) propellants.

The porous burner apparatus to study diffusion flames of simulated composite propellants was fabricated, installed, and checked out during FY 1979. Experiments planned for FY 1980 consisted of observations of diffusion flame structure as a function of the heterogeneity of the fuel-oxidizer sources. Similarly, the closed vessel apparatus was installed and checked out; and the high pressure combustion experiments were planned to begin in FY 1980.

Experiments carried out at JPL (Ref. 2) and elsewhere have shown unique effects of AP size distribution on the combustion response function. Classical theories of combustion response are unable to explain these effects because they assume homogeneous propellants. Demands of future reduced smoke propellants, and the viability of combustion tailoring through control of size distribution, make it imperative that the effects of size distribution be understood. The work planned for FY 1980 consisted of a review of models in the literature and the formulation of a model that would represent mechanisms derived from composite propellant heterogeneity.

SECTION 2

STATUS OF THE RESEARCH EFFORT

2.1 SUMMARY OF PREVIOUS ACCOMPLISHMENTS

A summary of previous accomplishments is presented in Table 1.

The program has been effective in making concrete progress, completing assigned tasks in reasonable times, and being able to respond to Air Force needs by pursuing ideas on a variety of combustion topics. Interchanges with government laboratories and private industry, presentations at technical meetings, and technical papers in journals of the AIAA and publications of CPIA have been numerous and productive as described in previous annual reports. Fruition of one of the ideas resulted in a patent* awarded last year, and the technique is being applied in the development of improved gun propellants.

2.2 CURRENT STATUS

2.2.1 Steady-State Combustion Modeling

The analytical model was evaluated by application to Miller's series of nonaluminized multimodel AP/HTPB propellants (Ref. 1). Results shown in Figure 1 demonstrate the general capability of the model to predict AP particle size distribution effects on burning rate over a wide pressure range. Furthermore, the model calculated reasonable values for the various internal details of the combustion process: surface temperatures, surface structures, flame heights, energy partitioning parameters, and component regression rates.

The model was next evaluated by application to series of HMX composite propellants which were subjects of prior research at JPL (Ref. 3). It was desired to learn whether or not the improvements developed for AP, and applicable to HMX, would impair the former capability of the model regarding HMX propellants. This did not prove to be the case. Changing computer program input constants, as appropriate for HMX, resulted in predictions in good agreement with the former model and experimental burn rate data over a very wide pressure range.

With the model validated for multimodel HMX propellants, the method of treating multicomponent propellants was next evaluated by application to several AP/HMX mixed oxidizer propellants (nonaluminized). The propellants were those used in the T-burner tests in prior JPL work (Ref. 4). Predictions of burn rate were in good agreement with data between 1 and 10 MPa.

*No. 56,245,169, Methods to Achieve Desirable Burning Rate Characteristics in Nitramine Propellants, U.S. Department of Commerce, Patent and Trademark Office.

Table 1. Summary of Previous Accomplishments

Fiscal Year	Contract No.	Accomplishments
1975-1976	AF0SR-ISSA-75-0005 AF0SR-ISSA-76-0006	Development of an analytical model for the steady-state burning of multicomponent composite propellants.
1977	AF0SR-ISSA-77-0001	Development of an analytical model for the transient burning of nitramine propellants in a confined volume. Experiments showing that HMX propellants are less susceptible to low-intermediate frequency combustion instability than AP propellants of equivalent energy and burn rate.
1978	AF0SR-ISSA-78-0004	Development of an analytical model of DDT emphasizing propellant combustion contributions.
1979	AF0SR-ISSA-79-0016	Development of an improved analytical model for the steady-state burning of ammonium perchlorate composite propellants. Experiments showing that the addition of HMX to an energetic binder has no significant effect on susceptibility to low-intermediate frequency combustion instability.

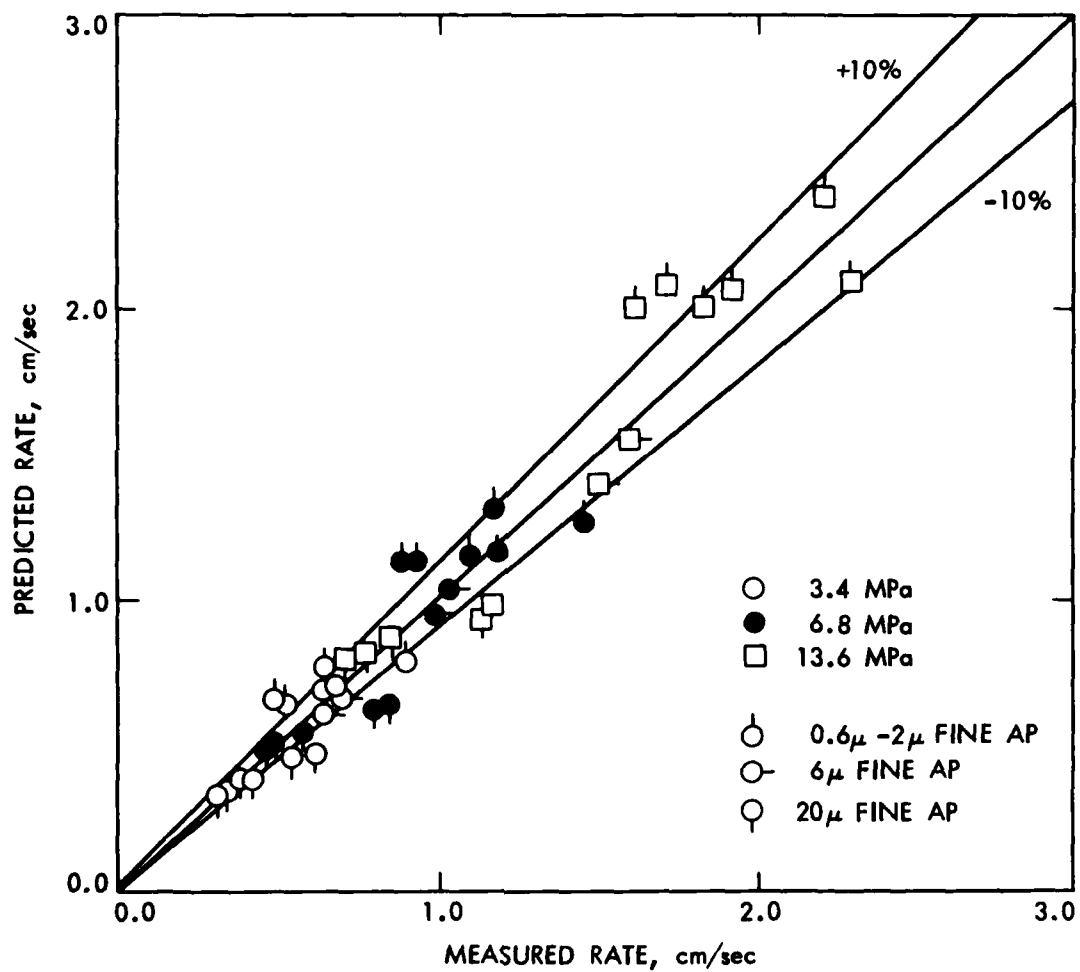


Figure 1. Model Application to Miller's Nonaluminized Propellants.

For energetic binder propellants, the former JPL model (Ref. 3) used the experimental burning rate of the binder as an input to describe the binder contribution to the burning. This has now been replaced by a combustion model for the energetic binder. The model is very similar to one published by Beckstead (Ref. 5), but is incorporated into a composite propellant model rather than standing alone. This model was applied to a series of Hercules composite-modified double-base (CMDDB) propellants, containing AP or HMX, but not containing aluminum. Qualitative effects of composition and pressure were predicted quite well, but quantitatively the predictions of burning rate became high at low pressures. It was concluded that the model for the binder needs to be improved in its treatment of the condensed phase heat release and fizz zone thermochemistry as functions of pressure,

A model for aluminum agglomeration was developed and incorporated into the code, replacing empirical expressions that were developed last year. The model is a form of "pocket model," based upon the premise that the fraction agglomerated is proportional to the amount of aluminum that melts within an effective pocket volume framed by the oxidizer particles. Pocket volumes are a function of the composite propellant microstructure, and are calculated for the fine, intermediate, and coarse sizes of a trimodal distribution. It is assumed that the particles are uniformly dispersed; coarse pockets encompass fine particles situated within. To minimize agglomeration, it is desired that the effective pocket be formed by the finest array. Two criteria are imposed to achieve this condition. First, the flame temperature of the fine pseudopropellant must exceed the aluminum ignition temperature. Second, the size distribution of the fine AP must encompass the aluminum particle sizes that melt. The flame temperature of the fine pseudopropellant depends upon AP/binder allocation with particle size (Refs. 6-8); this pseudopropellant tends to be more fuel rich with decreasing fine size. Aluminum sizes that melt are largely a function of the burn rate, and are determined by a heating analysis in the solid propellant thermal wave; fine aluminum is more likely to be encompassed by fine AP; but, on the other hand, is more likely to melt. To the extent that these two criteria are not met, the next larger pocket determines the effective pocket.

The fraction of aluminum that agglomerates is in itself of interest for the development of advanced high energy propellants for upper-stage and space motor applications. It is also used in the model for its effect on burning rate. Qualitatively, agglomeration tends to decrease burn rate because of the energy absorption involved and because there are fewer fine particles available to burn sufficiently close to the propellant to contribute heat feedback. Quantitatively, however, the effect is generally found to be small; the effect of aluminum on burn rate is largely a matter of what the aluminum replaces in the propellant.

The agglomeration model was applied to a series of Aerojet propellants which were the subject of an agglomeration study (Ref. 9). Calculated results are shown in Figure 2. There is no attempt to quantitatively compare the model with data because the model does not calculate a size distribution of agglomerates, and because the

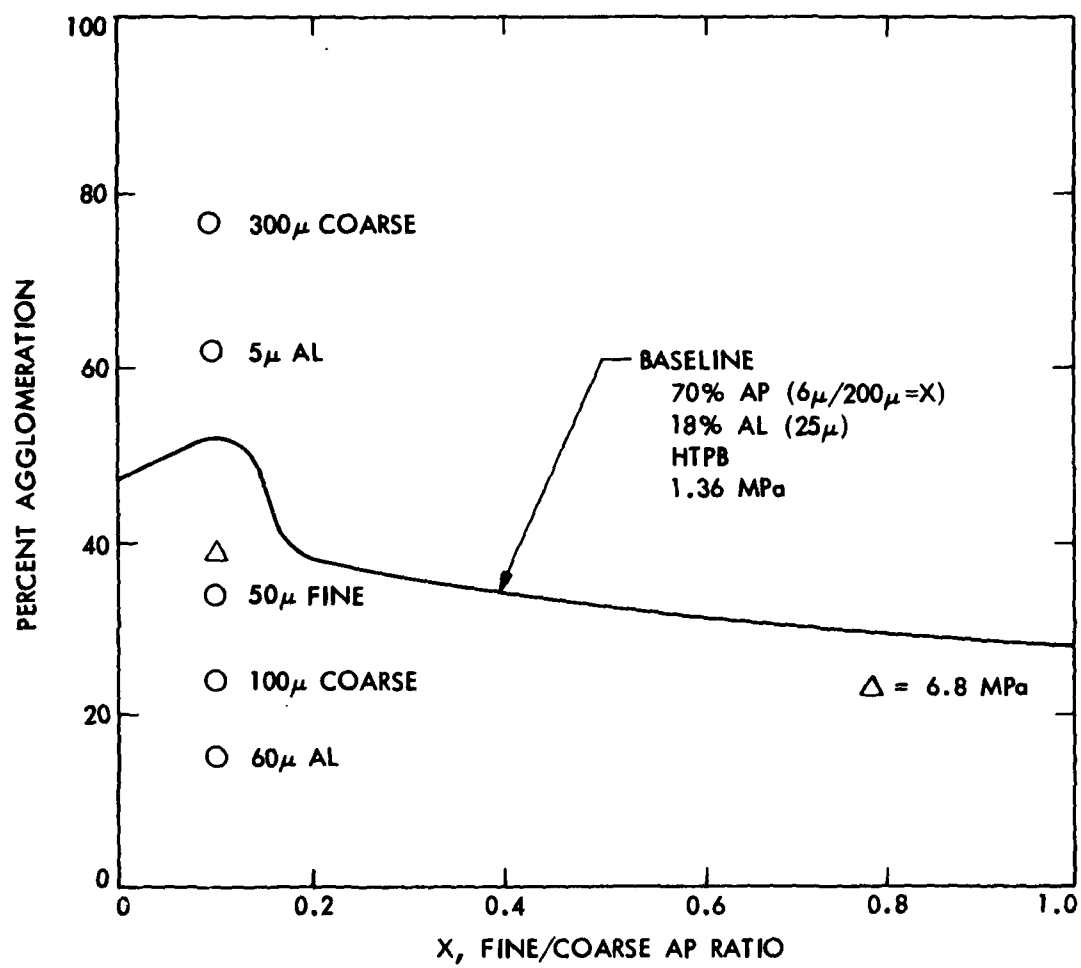


Figure 2. Pocket Model Calculations for Aerojet Propellants.

experimental definition of fraction agglomerated is arbitrary and not standardized. It was hoped that the model would be able to show and explain the major trends. The curve in Figure 2 shows the effect of varying the fine/coarse oxidizer ratio of a baseline propellant with the pressure held constant. The effects of other changes in variables are illustrated at one fine/coarse ratio. Aluminum agglomeration is shown to be reduced by (1) reducing the size of the coarse AP (the dominant variable), (2) increasing the size of the fine AP, (3) increasing the aluminum particle size, and (4) increasing pressure. These trends are all in qualitative agreement with the Ref. 9 data.

Finally, the model was applied to Miller's series of aluminized multimodel AP/HTPB propellants (Ref. 1). Results could be placed in two categories. First, where the fine AP size was 6 microns or more, or the coarse AP size was 200 microns or less, burning rate predictions were as good as those shown in Figure 1. These propellants are referred to as "ordinary." Second, where the fine AP was 2 microns or less and the coarse AP was 400 microns, in the same propellant, the predictions were very poor. These propellants are referred to as "extraordinary." They behave as though the fine AP were absent and, in fact, it was found that the model was able to predict their burning rates fairly well by the artificial procedure of omitting the fine AP from the computations. Miller has reported that the same phenomenon occurs in nonaluminized propellants, but requires a more extreme formulation. Thus the phenomenon originates with the AP and binder, and is merely aggravated by the presence of aluminum. It is hypothesized to result from very low flame temperatures in the fine pseudopropellant, occurring with extremes in particle size and high coarse/fine ratio, in accordance with AP/binder allocation theory. It is also hypothesized that aluminum helps to bring it about, at less extreme conditions, by acting as a heat sink in the fine AP/binder matrix. Attempts to model this mechanism in such a way as to be able to predict the behavior of both ordinary and extraordinary propellants on a consistent basis were not successful.

In conclusion, the steady-state modeling work as an outgrowth of the 1978 JANNAF Workshop has been completed as scheduled. For the most part, it has been successful. A lengthy paper was prepared and presented at the 17th JANNAF Combustion Meeting. Primary areas for future work are considered to be improvements in the model for energetic binders, and a model that would bridge the gap between ordinary AP composite propellants and those containing extremes of particle sizes.

2.2.2 Porous Burner Experiments

A series of experiments were conducted with the porous burner apparatus. Porous plates designed to simulate monomodal coarse, monomodal fine, and bimodal oxidizer composite propellants were utilized. Fuel gases are metered through the porous plate to represent binder. Oxidizer gases are metered through ports arranged in the porous plate to represent oxidizer particles. The system is described elsewhere (Ref. 10), except for new plates and manifolding acquired for this program. The new plates provide different simulation geometries, and the manifolding affords independent flow control and/or the use of two different oxidizer gases. The gases used for these tests were ethane, air, oxygen,

and oxygen-air mixtures. Independent variables were flow rate, O/F ratio, and pressure. Diffusion flame structures were observed visually and photographically. The most interesting results were acquired with the bimodal oxidizer porous plate.

There was much evidence in the diffusion flame structures to show that fine oxidizer ports tend to operate more fuel-rich than coarse ports in a bimodal arrangement. At highest oxidizer flow rates, the coarse port flames tended to bend toward the fuel (i.e., away from the ports) whereas the fine port flames were columnar in nature. This indicates that the coarse ports were oxidizer-rich whereas the fine ports were near stoichiometric. At lower oxidizer flow rates, the coarse port flames became columnar whereas the fine port flames began to bend towards the oxidizer port centerlines and eventually close over them (forming a parabolic flame). This indicates that the coarse ports were near stoichiometric and the fine ports were becoming fuel-rich. At still lower rates, both sets of flames closed over their respective oxidizer ports but the fine port flames exhibited more carbon emission. Although flame heights were larger over the coarse ports, in accordance with diffusion requirements, the coarse port flames commenced closer to the surface of the porous plate. This indicates a shorter reaction distance with the coarse port, which may be attributed to a higher temperature and, in turn, to operating closer to stoichiometry. Flame limits encountered by independently varying fuel and oxidizer flow rates, the relative sensitivity of the fine port flames to O/F ratio, and effects of substituting oxygen for air, all indicated that the fine port flames were more fuel-rich. Examples of photographs acquired are shown in Figure 3.

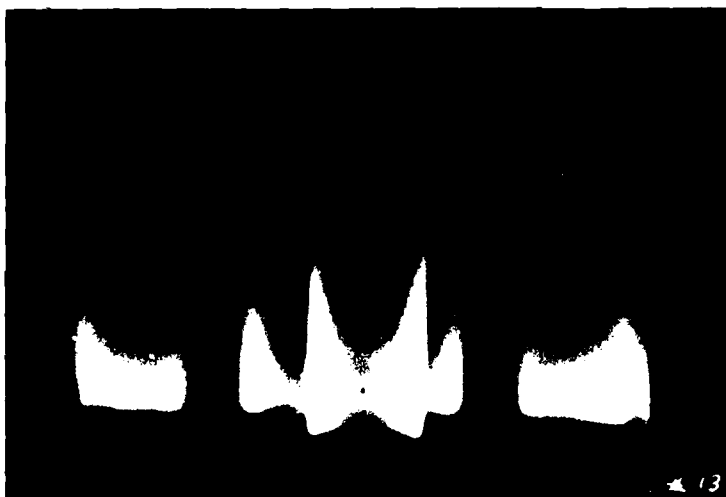
It is not surprising that the fine port flames tend to operate more fuel-rich. Given a fixed interstitial spacing between oxidizer ports, and particular values of specific flow rates, a proportionally greater amount of fuel will be associated with the fine ports. Analytical models of composite propellant burning have attempted to represent this, but the allocation factors are uncertain. Pursuing this line of experiments could help to resolve this problem.

Two interesting effects were noted when oxygen was substituted for air. For a given set of flow rates, the flame structure took on a less fuel-rich appearance (O/F ratio increased, as expected). Second, the porous plate became red-hot in the region of the cluster of fine oxidizer ports; this confirms the higher temperature and heat feedback that would be expected with oxygen, and indicates that there is more heat feedback with the fine ports. By analogy to propellants, fine particle size systems would burn faster unless the fuel allocation became such that the reduced temperature would overcome the reduced diffusion length. This may have happened with Miller's special class of propellants (cf., subsection 2.2.1), and could be tested with suitably configured burners and heat-sensing devices. It is also possible to use gases that are more representative of propellants, but this was not done during FY 1980.

Another purpose of this work was to look for flame interactions between flames of adjacent ports. Although physical overlap or



OXIDIZER RICH
(DIVERGENT FLAME)



STOICHIOMETRIC
(COLUMNAR FLAME)



FUEL RICH
(PARABOLIC FLAME)

Figure 3. Porous Plate Burner Flames - Coarse Bimodal Ethane - Air.

intermingling between adjacent flames was not observed, there were two features worth noting. First, where a fine port is adjacent to a coarse port, the fine port flame is nested under the near-surface region of the coarse port flame such that there could be heat feedback from the coarse flame to the fine flame. Second, where adjacent fine ports are sufficiently close together, the adjacent parabolic flames are joined at their bottoms by a nearly planar flame over the interstitial fuel. This feature provides a more vivid mechanism for direct heating of binder in propellants, and is not predicted by current diffusion flame theory.

There is much potential for future work with this apparatus to help understand the role of diffusion flames in composite propellant burning. Instrumentation could be added to measure temperatures, velocities, species concentrations, and heat flux. The burner could be upgraded to provide for mean and oscillatory cross-flows to simulate erosive burning and velocity-coupling. The apparatus has been demonstrated to be a viable research tool, and some interesting information has begun to be acquired. However, future plans will be affected by priorities and directions provided by the Air Force.

2.2.3 High Pressure Closed Vessel

Problems and delays were encountered in the procurement of an energetic binder HMX propellant which was considered to be most appropriate for high pressure combustion studies regarding DDT. Inert binder HMX propellants can be processed at JPL, but energetic binder propellants require an outside source. As an alternative source to Hercules Inc., the Air Force Rocket Propulsion Laboratory agreed to furnish propellant samples. However, propellants were not received in time for testing during this report period. Plans were made to process inert binder propellants. Considerable testing is planned for the coming year.

2.2.4 Response Function Modeling

A survey paper on response function models that specifically attempt to account for the heterogeneity of composite propellants was prepared and presented at the AIAA/SAE/ASME 16th Joint Propulsion Conference. The survey was the initial task for the modeling performed under this program. The paper discussed the deficiencies of the existing models and areas of needed work.

Formulation of the new response function model was completed. It is a linearized analysis of the pressure-coupled response function, containing a postulated mechanism for the effects of AP particle size. The investigation of non-linear combustion effects and, eventually, velocity-coupling, is planned for some future time.

The postulated mechanism is a "preferred frequency" type of mechanism, in which characteristic dimensions in the solid propellant give rise to periodic fluctuations as a result of the normal regression of the surface during burning. The characteristic dimensions are relatable

to the AP size distribution. Idealized packed-bed geometries involving spherical particles can be constructed to show the periodicities for theoretical purposes. Real propellants, however, contain distributions of sizes and shapes of non-spherical particles which are believed to afford a broad spectrum of frequencies with less pronounced peaks. It is planned in future work to experimentally characterize the spectra of propellants. Periodic fluctuations in pressure, temperature, and gas phase composition have been measured in the course of steady-burning (Refs. 11-13), and have been associated with AP particle size. Further, the fluctuations appear to enhance acoustic or nonacoustic instabilities at coincident frequencies (Refs. 2, 11, and 14). Thus the periodic structure of the heterogeneity is considered to be ordered on the macroscopic scale. It is represented in the model by form functions which are summations of Fourier components.

The heterogeneity is implemented in the model via fluctuations in the propellant formulation, which have two principal effects: fluctuations in the flame temperature and fluctuations in the net surface heat release applied at the boundary. Fluctuations in the thermal properties of the solid can be neglected (Ref. 15).

The work of Hamann (Ref. 16) was used to formulate the surface boundary condition of heat flux in the perturbation analysis. Hamann derived algebraic expressions for perturbed quantities based upon the BDP model of steady-state burning. Hamann's work was selected for convenience, and is not expected to have qualitative effects on the response function. The perturbed BDP model in and of itself does not give rise to preferred frequency behavior; it merely alters the quantitative results obtainable from classical theories which assume a homogeneous propellant. What is desired now is some realistic model of the gas phase. More up-to-date models, such as the steady-state model discussed in subsection 2.2.1, could be used for that purpose, but are more complicated and would require numerical methods to obtain the perturbation coefficients. At this stage, the Hamann approach is considered adequate to test the concept.

The analysis boils down to expressions for perturbed burn rate in terms of perturbed pressure and perturbed AP concentration. A "total response function" is expressed as the sum of a "homogeneous response function" (classical theory) and a "heterogeneous response function." The former component describes combustion response in the absence of heterogeneity, and the latter describes combustion response in the absence of pressure fluctuations. Note that, in the framework of this model, there is no such thing as "steady-state;" rather, the combustion of composite propellants is always at least a noise arising from the heterogeneity form functions. The two components of the combustion response will couple only for that segment of the heterogeneity which coincides with the pressure perturbation frequency (e.g., acoustic mode of a cavity). In some (perhaps most) cases, the contribution of the heterogeneity will be weak; but strong contributions can be expected at acoustic frequencies which correspond to significant peaks in the heterogeneity form function.

SECTION 3

TECHNICAL JOURNAL PUBLICATIONS

The following publications appeared in the AIAA Journal during the past year:

- (1) Cohen, N. S., "Review of Composite Propellant Burn Rate Modeling," J. AIAA, Vol. 18, No. 3 (March 1980), pp. 277-293.
- (2) Cohen, N. S. and Strand, L. D., "Analytical Model of High Pressure Burning Rates in a Transient Environment," J. AIAA, Vol. 18, No. 8 (August 1980), pp. 968-972.
- (3) Strand, L. D., and Cohen, N. S., "Effect of HMX on the Combustion Response Function," J. of Spacecraft and Rockets, Vol. 17, No. 6, (November-December 1980), pp. 566-568.

The following paper is being considered for AIAA Journal publication:

Cohen, N. S., "Response Function Theories that Account for Size Distribution Effects - A Review," AIAA Paper 80-1123.

It is planned to submit two abstracts for the AIAA/SAE/ASME 17th Joint Propulsion Conference. The abstracts will be based upon the steady-state modeling work.

CPIA Publications are listed in subsection 5.1.

SECTION 4

PROFESSIONAL PERSONNEL

The Principal Investigator for this program is Mr. Leon D. Strand of the Energy and Materials Research Section (M/S 122/123), Jet Propulsion Laboratory/California Institute of Technology, 4800 Oak Grove Drive, Pasadena, California 91109 (telephone 213-354-3108). His co-investigator is Dr. Norman S. Cohen, Norman Cohen Professional Services, 858-A Pine Avenue, Redlands, California 92373 (telephone 714-792-8807).

Mr. Strand has overall program responsibility and specific responsibility for the experimental work performed. Dr. Cohen works under a subcontract, and is responsible for the analytical model developments. Both are members of the AIAA Technical Committee on Propellants and Combustion.

SECTION 5

INTERACTIONS (COUPLING ACTIVITIES)

5.1 PRESENTATIONS

The following presentations have been made under this research contract:

- (1) Cohen, N. S., "Workshop Report: HMX Propellant Combustion Modeling," 16th JANNAF Combustion Meeting (CPIA Publication 308, Vol. III, December 1979), pp. 219-240.
- (2) Cohen, N. S., and Strand, L. D., "Composite Propellant Combustion and Transition to Detonation," 1980 Joint AFOSR/AFRPL Rocket Propulsion Research Meeting, Lancaster, CA (March 1980).
- (3) Cohen, N. S., "Response Function Theories That Account for Size Distribution Effects - A Review," AIAA Paper 80-1123, AIAA/SAE/ASME 16th Joint Propulsion Conference, Hartford, CT (July 1980).
- (4) Cohen, N. S., and Strand, L. D., "A Model for the Burning Rates of Composite Propellants," 17th JANNAF Combustion Meeting, Hampton, VA (September 1980); publication pending.

5.2 INTERCHANGE FUNCTIONS WITH GOVERNMENT LABORATORIES AND CONTRACTORS

The Lockheed Missiles & Space Co. (Dr. G. Lo) continued to express interest in our studies of HMX propellant combustion as an aid to their own in-house work in progress at their Palo Alto Research Laboratory. Discussions with Dr. Lo concerned the role of HMX decomposition in the combustion of HMX propellants. Considerable interest was expressed in the models of burn rate and aluminum agglomeration revealed at the 17th JANNAF Combustion Meeting; advance copies of the paper were requested by Dr. W. Schmidt (Aerojet), Dr. W. Brundige (Thiokol), Dr. M. Beckstead (BYU, and consultant to Hercules), R. Foster (Hercules) and Prof. J. Osborn (Purdue). The agglomeration model is being used by Thiokol on several Air Force Rocket Propulsion Laboratory programs dealing with improved upper stage and space motor propellants. Discussions were also held with Thiokol (Drs. D. Flanigan and W. Brundige) on the burning rate characteristics of HMX-containing propellants. Both Dr. Lo and Dr. Flanigan are interested in methods to catalyze HMX propellants.

In a different vein, discussions were held with A. Juhasz (Army Ballistics Research Laboratory) and B. Moy (Air Force Armament Laboratory) on nitramine particle size selection for gun propellants. Effects of AP particle size and HMX content on combustion instability were discussed with Thiokol (Dr. R. Kruse, R. Hessler, S. Folkman, and L. Sayer), General Dynamics (D. Taylor and A. Messner), and NWC (Dr. R. Derr and J. Crump) in regard to Navy and Air Force development programs. Periodically, AFRPL (W. Roe and J. Levine) and NWC (T. Boggs and C.

Price) were kept informed of our work, and interesting developments were quickly relayed to colleagues (M. Beckstead of BYU, M. King of Atlantic Research, R. Miller of Hercules, and W. Brundige of Thiokol).

Finally, discussions were held with M. Cowperthwaite (SRI) on the possibility of combining, in a future joint effort, the JPL combustion model for DDT with the SRI shock formation model for DDT.

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