

DISCLAIMER NOTICE

THIS DOCUMENT IS BEST QUALITY PRACTICABLE. THE COPY FURNISHED TO DTIC CONTAINED A SIGNIFICANT NUMBER OF PAGES WHICH DO NOT REPRODUCE LEGIBLY.

:

ŕ

<u>A Critical Analysis of</u> <u>Arc Image Ignition of Solid Propellants</u>

Aerospace and Mechanical Sciences Report No. 789

by

M. Summerfield and T. J. Ohlemiller

AFOSR CONTRACT AF 49(638)1267 Project-Task 9711-01

July 1967

Transmitted by:

listo

Martin Summerfield' Principal Investigator

1. Distribution of this document is unlimited.

Oualified requestors may obtain additional copies from the Defense Documentation Center. All others should apply to the clearinghouse for Federal Scientific and Technical Information.

Guggenheim Laboratories for the Aerospace Propulsion Sciences Department of Aerospace and Mechanical Sciences PRINCETON UNIVERSITY Princeton, New Jersey

ACKNOWLEDGEMENT

Carts.

しきんがったい いんかいいいがんかいがく

たのないというともないとない

いいうというというというというないというない

ŝ

2.0

STREET STREET

ă,

÷

This research under Contract AF49(638)1267, Project-Task 9711-01 and for the period 1 October 1965 through 30 September 1966 was sponsored by the Air Force Office of Scientific Research, Office of Aerospace Research, United States Air Force.

Technical Supervisor for this program is Dr. Bernard T. Wolfson, Acting Chief, Propulsion Division, Directorate of Engineering Sciences, Air Force Office of Scientific Research.

Conditions cf Reproduction

Reproduction, translation, publication, use and disposal in whole or in part by or for the United States Government is permitted.

TABLE OF CONTENTS

		rag
Tit	le Page	i
Ack	nowledgement	ii
Tab	le of Contents	iii
Lis	t of Figures	v
Abs	tract	vi
1.	Applicability of the Arc Image Technique	1
2.	Description of Apparatus and Methods of Measuring Ignitability	1
3.	Comparison of Effects of Radiant Energy on Ignition with Other Modes of Energy Input	2
	 a. Modes of Action of Practical Igniters b. Characteristics of Conductive or Convective Energy Input c. Peculiarities of Radiative Energy Input d. Chemical Effects of Radiative Energy Absorption e. Effect of Radiation Depends on Ignition Mechanism f. Radiation May Alter The Normal Mechanism of Ignition g. Radiation May Induce Bubbling, A Non-Representative Phenomenon h. Radiative Ignition May be Much Slower Than Conductive Ignition i. Summary: Radiative Ignition is Very Different From Normal Ignition 	2 2 3 4 5 5 6 6 6
4.	Theoretical Formulations	7
	 a. Exact Theory b. Discussion of Expected Solutions c. Approximate Theoretical Solutions d. Comparison With Other Laboratory Modes of Ignition 	7 10 11 13
5.	Comparison of Theory With Experiment	14
	 a. Comparison With The Experimental Data of Beyer and Fishman b. Comparison With The Experimental Data of Bastress, Niessen and Richardson c. Sensitivity of Radiative Ignition Time to Absorptivity 	<u>1</u> 4 15 17
6.	Conclusions	17
7.	Discussion of Future Studies of Radiative Ignition	18
	a. Theory b. Experiment	18 18

ŝ

TABLE OF CONTENTS - Cont'd.

Page

References	21
Nomenclature	23
Figures	24
Appendixes A. Distribution List B. DD Form 1473	A-1 B-1

-

.

いたいというないというというかんである

10000

.

•

LIST OF FIGURES

and the state of t

Strate Martin Strate State

		Page
1.	Arc Image Furnace With Double Elliptical Mirror Arrangement	24
2.	Typical Spectral Energy Distribution For A Carbon Arc Source	25
3.	Bubbling Induced In a Pure PBAN Polymer Sample by a Low Radiant Heat Flux	26
4.	Comparison of Arc Image Ignition With Other Methods of Ignition	27
5.	Thermal Wave Development For Gas Phase and Heterogoenous Ignition of a Pure Fuel:	29
6.	Thermal Wave Development For Limiting Cases of Small and Large Values of Characteristic Time,	30
7.	Comparison of Theoretical and Experimental Effect of Pressure on Ignition Energy	31
8.	Comparison of Theoretical and Experimental Effect of Radiant Flux Level on Ignition Energy	32
9.	Comparison of Theoretical and Experimental Effect of Radiant Flux Level on Ignition Delay Time	33
10.	Comparison of Theoretical and Experimental Effect of Convec- tive Heat Flux Level on Ignition Delay Time	34
11.	Comparison of Theoretical and Experimental Effect of Radiant Flux Level on Ignition Delay Time	35
12.	Illustration of the Theoretical Effect of Propellant Absorp- tivity on Ignition Delay vs Radiant Flux Level	36

ABSTRACT

The use of the arc image furnace to evaluate the ignition characteristics of solid propellants has become a widespread practice. Data obtained in this mannerare used to predict propellant ignition behavior in a rocket motor. However, the validity of the application of such data to a rocket motor environment has not been well established and is, in fact, subject to question.

In arc image testing, a purely radiative heat input replaces the normal predominantly conductive and convective heat inputs to which a propellant is subjected in rocket motor ignition. However, radiation is of a fundamentally different nature from the molecular heat transfer processes which constitute conduction and convection and thus can interact with the propellant in a significantly different manner. For example, by virtue of its electro-magnetic character, radiation is partially reflected at phase boundaries; the unreflected portion is either transmitted or absorbed in depth, not at the boundary surface. For a composite propellant, this implies that in the arc furnace environment, unlike the motor environment, energy deposition occurs selectively in certain propellant constituents; the temperature distribution can be altered considerably. Ignition delay time becomes sensitive to propellant opacity which can be changed considerably by minor additives. Various further alterations can occur, as well.

1

Interpretation of radiative ignition data requires further experimental and theoretical guidelines. A physical model is proposed which includes several effects peculiar to radiative ignition such as in-depth absorption and pyrolysis. Both extreme cases of a heterogeneous mechanism and a gas phase mechanism are considered. The equations describing these cases are very complex and have not as yet been solved. The solutions to these models will require experimental verification; for this purpose a high power laser appears distinctly preferable to an arc image furnace.

A simpler physical model of radiative ignition is derived on the basis of asymptotic expressions for ignition delay at various radiant flux levels. No statement of the mechanism of exothermic runaway is required. The predictions of the model compare favorably with the radiative ignition data of Beyer and Fishman and of Bastress.

1. Applicability of the Arc Image Technique.

The arc image furnace has gained considerable acceptance in recent years both as a tool for evaluating the ignition energy requirements of solid propellants and also for studying their fundamental ignition mechanism. The common focus in both of these applications is, of course, the ignition of solid propellant rocket motors.

In the evaluation of propellant ignitability, the arc image furnace is merely a convenient substitute for the motor igniter. As such it facilitates considerably more rapid, better controlled, and less expensive ignition tests than does a motor-igniter combination. However, the validity of this substitution must be subjected to close scrutiny; the disadvantages incurred due to alteration of the ignition mechanism and hence the ignition behavior may well outweigh the advantages gained in using the arc furnace. This is particularly true when the arc furnace is applied to routine evaluation of propellant ignitability characteristics, since the behavior trends thus determined may be qualitatively very different from those displayed in actual motors.

This objection is less relevant when basic ignition mechanism research is concerned, since here one must inevitably abstract and simplify the physical situation in order to perform controlled experiments. Even though the ignition stimulus may be very different, the ignition response can disclose the underlying processes. If the special physical and chemical effects peculiar to the use of an arc image heat source can be identified and taken into account, this tool can be quite valuable in propellant ignition research.

This report offers a discussion of several of these radiant energy effects and their implications with regard to ignition testing and ignition mechanism research.

2. Description of Apparatus and Methods of Measuring Ignitability.

A schematic drawing of the optical arrangement in a typical arc image furnace is shown in Figure 1. The particular mirror (and/or lens) combination may differ considerably from that shown; in addition, various arc lamps may be substituted for the carbons. However, all such arrangements have two factors in common: the radiant energy is spread over a wide spectral region and the beam convergence angle required to achieve high heat fluxes is large. The half angle of convergence may be as high as 70° to 80° , and in the focal region thus produced the flux is a rapidly varying function of position. An example of the type of spectral energy distribution obtained from a carbon arc source is sketched in Figure 2; the occurrence of spikes, the position of the peak, and the detailed shape of the curve will, of course, depend significanly on the composition of the part_cular source used and the power level at which it is run. However, in general the preponderance of the energy will be in the visible and near infrared regions, with a small but perhaps photochemically important fraction in the near ultraviolet.

The usual measurements of practical interest in arc image ignition tests are the ignition delay time and minimum ignition energy as functions of various parameters such as pressure, flux level, propellant composition, environmental gas composition, etc. These two items of data are defined unambiguously only for the go/no-go type of ignition test, in which one varies the exposure time (or some other convenient parameter) until the minimal requirement for ignition and subsequent burning is just met. A second type of test is to measure ignition delay by means of a photocell which detects the instant that radiation is emitted by the incipient flame, while the incident flux is still on. The ignition energy calculated from the delay thus measured is not necessarily the minimum required, and the error on the plus side may be quite large in the very important limit of short delay times. This method has the additional disadvantage that the criterion may be misleading, that the observed intensity of the emitted radiation is probably not indicative of the local gas or surface temperature during ignition. Data obtained by both of these methods have been used to characterize propellant ignitability and to check various theoretical ignition models.

Typical data are shown in Figures 7b, 8b and 9b taken from Reference 1. In order to obtain ignition delay times in the interesting range of 10 to 100 millisec, radiation fluxes of the order of 10 to 100 cal/cm² sec are required, depending on the gas pressure in the test cell. This tends to be somewhat larger than the amount of heat required in the case of convective ignition.

3. <u>Comparison of Effects of Radiant Energy on Ignition With Other Modes of</u> Energy Input.

a. Modes of Action of Practical Igniters.

As was mentioned previously, the standard by which the arc image ignition technique described above must be judged is the rocket motor ignition process. The particular mode (or combination of modes) by which heat is transferred to the propellant grain in a motor is, of course, strongly dependent on what type of igniter is used. Hot particle and gaseous igniters transfer heat predominately by conduction and convection, respectively; radiation is usually of secondary importance with both of these types. Catalysis in the contact region may be important in the case of hot particle igniters. Hypergolic igniters constitute a separate class in that the ignition stimulus derives from the strong surface reactivity of the material at ambient temperature rather than the reactions at elevated temperature.

b. Characteristics of Conductive or Convective Energy Input:

The time variation of the conductive heat transfer rate from a hot particle to the propellant grain is quite complex; it is initially infinite but drops quite rapidly to zero because of the small heat content of the particle. On the other hand, convective heat transfer from a hot gas is more nearly constant with respect to time after a spike at the starting instant. Despite this difference in timewise heat transfer variation and other obvious differences in the resulting gaseous environment adjacent to the grain surface, on a molecular level these two igniter types transfer heat in essentially the same manner, i.e., thermal energy is transferred via collisions to the lattice and molecular vibrational modes of the surface molecules, thus exciting them and leading to their decomposition. The colliding molecules do not discriminate between the binder and oxidizer of a composite propellant, and the heat transfer to these two materials will differ only insofar as their thermal properties differ. The reference standard for ignition is, then, a process in which heat is transferred at essentially the same rate to the surface of both propellant components and high temperature material (gas or solid) is present in the region immediately adjacent to the propellant surface. Various ignition methods which simulate these conductive and convective heat inputs have been used extensively for ignition mechanism research.^{2,3,4}

c. Peculiarities of Radiative Energy Input:

Radiative ignition tests readily duplicate the square-wave like variation of heat transfer versus time characteristic of convective ignition, but, aside from this, the similarities between the radiative ignition stimlus and the predominately conductive and convective ignition stimuli in a rocket motor are few. Radiation incident upon a composite propellant undergoes, first of all, a partial reflection at the surface; the fraction reflected is a function of surface condition, wavelength (and hence propellant composition), and probably oxidizer particle size. Unless the surface is a perfectly diffuse reflector, measurement of the reflectivity applicable to the arc furnace conditions is relatively difficult. Some accurate measurements have been made but usually this factor is either ignored or accounted for only roughly. The net reflectivity is, however, only a part of the problem; the relative reflectivities of binder and oxidizer must also be considered. In general these will be different, and thus, in this first step, there exists a difference in the heat input rates to the two materials.

The situation is further complicated when one considers the fate of the unreflected fraction of the radiation. This fraction is not absorbed at the surface but rather in depth; the depth of penetration is a function of the wavelength-dependent absorptivity of the particular material in question. It is well known, too, that spectral absorption coefficients are sensitive to temperature and to the fraction of the substance that has been cltered photochemically. The absorption may be time-dependent, therefore. Oxidizers such as ammonium perchlorate are largely transparent to visible radiation; many common binders are fairly opaque by comparison. Also, the presence of catalyst particles usually added to the binder increases its effective absorptivity. This difference causes further divergence in the relative heat input rates to binder and oxidizer. The situation approaches that in which the oxidizer particles, instead of being heated by the flux on their exposed surface, are heated around their immersed surface by conduction from the binder. With respect to homogeneous, nitrocellulose case propellants, it is well known that without an opacifier, radiative

ignition can occur far inside the propellant grain. The probable net effect of these factors is a significant retardation in the rate of appearance of oxidizing species in the gas immediately adjacent to the exposed surface and a possible forcing cf sub-surface reactions which would not occur under normal convective heat flux exposure.

d. Chemical Effects of Radiative Energy Absorption:

The absorption of radiation in depth is itself a vory complex process. In general, radiation in the visible or ultraviolet regions is absorbed by means of electronic excitations. The objective of adding the radiant energy is, of course, equilibrium excitation of lattice and molecular vibrational modes (thermal excitation). However, this is only one of the possible consequences of the electronic excitat.ons; the most likely alternative result is the breaking of atomic bonds. Obviously the bonds most susceptible to radiative rupture need not be the same as those most susceptible to thermal disruption; therefore, the possibility exists that radiation can significantly alter the propellant degradation mechanism. In practice, it is doubtless true that thermal and radiative degradation proceed concurrently. The extent to which radiation changes the degradation path is strongly dependent on the particular materials involved and the radiant intensity at the critical wavelengths. Bastress has investigated this possibility for one propellant composition cont.ining PBAA by the use of a series of bandpass filters in conjunction with an arc furnace. Within experimental error, no systematic dependence of ignition delay on wavelength was found for this composition; it should be emphasized, however, that this result cannot be generalized and possible photochemical effects should be kept in mind whenever a new propellant composition is tested. It should be noted here that if direct radiative (i.e., photochemical) degradation dominated, the degradation rate at a given point in the solid would be largely a function of the local intensity and not the local temperature.

This disruptive eifect of radiation on the normal degradation process is probably more serious for the polymeric binder than for the ammonium perchlorate, since the latter is largely non-absorbing in the visible and near infra-red (up to 2μ). The effect on the binder can be prevented to some degree by the addition of absorbing solid particles in order to stop the radiation near the surface and make the energy enter by heat conduction. The particles must be small enough to prevent further distortion of the already disturbed temperature profile and large enough to be more than mere scattering centers. If these two requirements can be met, the particles then not only help prevent abnormal degradation but also prevent deep penetration of the radiation into the propellant. The situation cannot, of course, be completely corrected and indeed one must ask whether the particles themselves will alter the polymer degradation or otherwise affect the ignition process.

Some idea of the physical heating situation is obtainable from the following facts. In 100 millisec a thermal wave penetrates 0.1 mm into a typical solid propellant. A propellant with a radiation attenuation coefficient of 100 cm⁻¹ allows the incident flux to penetrate to a depth of 0.1 mm. Obviously, the picture is not simply the application of heat to the surface followed by the penetration of this heat to the interior by conduction, as is usually believed. For a case of ignition in 10 millisec, the contradiction is even more striking.

e. Effect of Radiation Depends on Ignition Mechanism:

The radiative effects considered thus far come into play the instant that the radiation strikes the sample. During the induction period the preferential heating of the binder and the penetration of radiation below the propellant surface merely cause an inefficient utilization of the incident energy and thus tend to extend the induction period. However, the effect of these factors during the latter portions of the ignition lag and the effect of photochemical absorption and other factors must be considered in the light of what may be the actual ignition mechanism in more normal circumstances.

At present, there are two theories which have had reasonable success in explaining propellant ignition behavior; they differ primarily in their hypotheses as to the position and nature of the reactions that ultimately lead to ignition. Neither of these, the heterogeneous nor the gas phase theory, has as yet been applied to the most interesting case, i.e., convective ignition, primarily because of its mathematical complexity; however, the basic ideas of each theory as applied to this case can be stated quite simply. In the heterogeneous theory, ⁶, ⁷, the convective heat transfer to the the heterogeneous theory, propellant surface raises its temperature (subsurface heating occurs purely by conduction during the induction period). Subsequently, the heated oxidizer begins to decompose, liberating gaseous oxidizing species; binder gasification, if it occurs, is of secondary importance. The oxidizing species are adsorbed on the immediately adjacent binder surface where they react exothermically with the fuel; these reactions may occur both at the propellant surface and immediately below at the heated binder-oxidizer interfaces. The heat thus produced further aids the oxidizer decomposition and thus by a bootstrapping process ultimately leads to ignition of the propellant. In the gas phase theory, the heat-up process is assumed to be the same but now binder gasification is as important as oxidizer gasification. Gaseous oxidant and fuel species diffuse from the surface and intermingle and react in the hot gas environment above the propellant surface. The heat produced by this reaction is conducted in part to the propellant surface, raising the volatility, and thus leading to the same general kild of bootstrap process. This hot gas environment is highly in trumental in aiding the exothermic reaction between these two species and thus affects the speed of ignition.

f. Radiation May Alter The Normal Mechanism of Ignition:

Keeping in mind that either one or both of these mechanisms is the key process in motor ignition, one can look at how radiation may shift the relative likelihood of their occurrence or otherwise alter the ignition process. The high degree of radiation penetration through the oxidizer crystals creates the possibility of abnormal hot spots at the back of these crystals; subsequent gasification and reaction at these points, whether heterogeneous or gas phase is largely independent of the propellant environment if the propellant is fairly rigid. Thus the response of the ignition delay to environment may differ in motor tests and arc image tests, particularly if the gas phase mechanism dominates. Photochemical absorption produces highly reactive free radicals; in so doing, it not only alters the polymer degradation process, but also introduces the possibility of heterogeneous or solid phase reactions that would not normally occur. Again, this possibility is a strong function of the binder type.

g. Radiation May Induce Bubbling, A Non-Representative Phenomenon:

In-depth absorption of radiation in the binder has another rather drastic effect. Since there is a conductive heat loss from the hot surface to the cold gas adjacent to it in the usual test arrangement, the maximum binder temperature will be at some depth below the surface; consequently, the maximum binder pyrolysis rate will also occur below the surface. The result of this is vigorous bubbling in the liquefied binder layer at the surface'; a rather extreme case of this is illustrated in Figure 3 which shows a pure PBAN sample exposed to a radiative flux of 10 cal/cm²sec. Such bubbling is, of course, more severe for polymers with low thermal stability. Not only do these bubbles obscure the position of the surface, they also totally disrupt the temperature and concentration profiles in the gas by ejecting small jets of fuel vapor when they burst. These bubbles may have no particular effect on a heterogeneous mechanism (except to the extent that they block adsorption), but they may strongly affect a gas phase mechanism. In the latter case they have the beneficial effect of aiding the oxidant-fuel mixing process; however, the fuel jets also cause convective cooling of the reactive species. The net result of these opposing influences is extremely difficult to predict, but it appears that bubbling can change a gas phase ignition into a very haphazard process.

h. Radiative Ignition May Be Much Slower Than Conductive Ignition:

Finally, there is the effect of differing temperatures of the faseous environment. As was mentioned previously, in the motor ignition event, the environment adjacent to the propellant surface consisted either of hot gases or hot solid particles which heat the adjacent gases; the environmental gas in arc ignition tests is almost invariably at room temperature. If the ignition mechanism is heterogeneous, the substitution means only a slight increase in the ignition delay due to the small heat loss to the gas. However, if a gas phase mechanism dominated in the convective heating case, it could be considerably retarded or even prevented in the cold environment of the arc image test unless conditions are such that the gas phase reaction will occur very close to the surface.

i. Summary: Radiative Ignition is Very Different From Normal Ignition:

In summary, the substitution of an arc image heat source for the normal motor igniter introduces the possibility of extensive physical and chemical changes in the ignition process. Selective reflection

and absorption of the radiant energy can cause a considerably modified temperature distribution in the propellant, yielding reactions at different positions and possibly different types than those occurring in a motor. Bubbling and the presence of a cold gas environment can cause additional modifications in the ignition mechanism, particularly if it normally occurs in the gas phase. Figure 4 summarizes all of these peculiar features of radiative ignition and contrasts them with the features of various other ignition methods. The net effect of these various factors will obviously depend strongly on the particular propellant being tested, but the conclusion seems inescapable that routine use of the arc image furnace to evaluate propellant ignition characteristics is potentially very misleading. This discussion has, of course, been largely qualitative since adequate data for quantitative assessment of the various factors are lacking. The fact that radiation does indeed cause very real changes in propellant ignition behavior & s been demonstrated experimentally by Bastress, et al, * ; their results are shown in Figures 10b and 11b. Comparison of these results for radiative and convective heating indicates that radiation causes a somewhat different response to variations in heat flux level and also causes a considerable change in the relative ignitabilities of the four propellants. Opacifying a propellant by adding Al makes it easier to ignite by radiation, as our discussion indicates. The same addition of Al makes it slower to ignite by convection because of increased thermal conductance of the propellant. An unaluminized propellant is slow to ignite by radiation (by a factor of 29 primarily because of transparency. This highlights the point that radiative ignition can be very different in character from the predominantly convective and conductive modes of ignition employed in practical rocket motors.

4. Theoretical Formulations

a. Exact Theory

Theoretical solutions for radiative ignition, supported by appropriate diagnostic experiments, are needed to serve as a guide for those who are using the arc image furnace in ignitability measurement programs. A complete theoretical accounting for all of the factors discussed above would lead, of course, to an essentially intractable mathematical model. As with any problem of such complexity, one must construct simplified models to facilitate solutions which then can be compared with experiment. A simplified experimental analog of propellant ignition which has proven to be quite valuable in past work is the ignition of a pure solid fuel in an oxidizing gaseous atmosphere; when applied to the present case of radiative ignition, this analog retains several of the essential features of arc image ignition of propellants. With regard to the possible sites and types of the important exothermic reactions that constitute ignition, it is necessary to examine at least both extreme cases of purely gas phase and purely h)geneous reaction; either or both of these mechanisms is probably operative under various conditions of propellant ignition.

With regard to the fate of the incident radiant energy in both models, it is desirable that the theory correspond as closely as possible in this respect to the experiments designed to verify it. Thus, it should be borne in mind that reflection at the fuel surface is a function of both angle of incidence and wavelength, as is the refraction of the radiation which penetrates the surface; the effective absorptivity and hence the depth of penetration is also a function of these two factors. The relative importance of these effects is, of course, dependent on the particular experimental apparatus; however, it is essential that radiation penetration in depth be accounted for in some manner. Since photochemical effects are of potential importance, it is necessary to allow for them in the analysis; the differences in ignition behavior resulting from the extreme cases of pure thermal and pure photochemical absorption of the radiation can then be examined.

The following assumptions are made in constructing the mathematical models for both mechanisms. The densities of the gas and solid are constant; the thermal properties of gas and solid are constant; the mass diffusivities of all gaseous species are constant and equal; the chemical reactions are such that the number of moles of products is approximately equal to the number of moles of reactants; the molecular weights of all gaseous species are constant and equal. These assumptions are common in combustion theory; they allow a significant reduction in the complexity of the mathematics.

In the gas phase model it is assumed that the exothermic reactions which lead to ignition are adequately represented by a single overall reaction rate which has an Arrhenius temperature dependence and is proportional to the product of the local oxidizer and fuel concentrations. It is further assumed that the polymer vaporizes in depth; as was pointed out previously, this factor can be important since the maximum solid temperature is below the surface. However, in the mathematics that follows, bubble formation is ignored, mainly because we do not know how to include the process of bubbling properly. In defense of this decision it may be noted that the rupturing of the surface by bubbling seems to be pronounced only under conditions of relatively long duration, low intensity ignitions. For rapid ignitions of opaque propellants, only a thin layer is heated and violent bubbling seems not to occur. The gas resulting from pyrolysis in depth is assumed to emerge from the surface without resistance and without gross break-up of the solid layer, perhaps by flow through many minute cracks and by diffusion through the ultrathin layer.

The equations describing the gas phase model are then:

Solid:

$$\frac{\partial T}{\partial t} = K_{s} \frac{\partial^{2} T}{\partial x^{2}} - V_{s} \frac{\partial T}{\partial x} - \left(\frac{Q_{vap} Z_{s}}{C_{s}}\right) e^{-E_{s}/RT} + \left(\frac{1}{\rho_{s}C_{s}}\right) \frac{\partial T_{x}}{\partial x} + \left(\frac{p Q_{pc} T_{x}}{\rho_{s}C_{s}}\right)$$
Gas:

$$\frac{\partial T}{\partial t} = K_{q} \frac{\partial^{2} T}{\partial x^{2}} - V_{q} \frac{\partial T}{\partial x} + \left(\frac{Q_{chem} Z_{q}}{\rho_{s}C_{q}}\right) C_{ox}C_{i} \cdot e^{-E_{s}/RT}$$

$$\frac{\partial C_{ox}}{\partial t} = D \frac{\partial^{2} C_{ox}}{\partial x^{2}} - V_{q} \frac{\partial C_{ox}}{\partial x} - N Z_{q} C_{ox}C_{F} e^{-E_{q}/RT}$$

$$\frac{\partial C_{F}}{\partial t} = D \frac{\partial^{2} C_{F}}{\partial x^{2}} - V_{q} \frac{\partial C_{F}}{\partial x} - Z_{q} C_{ox}C_{F} e^{-E_{q}/RT}$$

and the boundary conditions are:

$$T(x_{10}) = T_{o} \qquad T(\stackrel{t}{=} \cdots_{j} \stackrel{t}{t}) = T_{o} \qquad T_{gas}(o,t) = T_{solid}(o,t)$$

$$C_{ox}(x_{10}) = C_{ox}^{\infty} \qquad C_{ox}(too_{j}t) = C_{ox}^{\infty} \qquad \lambda_{g}(\stackrel{\partial T}{\partial x})_{o,t} = \lambda_{s}(\stackrel{\partial T}{\partial x})_{o,t}$$

$$C_{F}(x_{10}) = O \qquad C_{F}(too_{j}t) = O \qquad C_{ox} \forall_{g} = D \stackrel{\partial Z_{sox}}{\partial x}$$

$$\dot{m}_{F} = C_{F} \forall_{g} - D \stackrel{\partial Z_{F}}{\partial x}$$

The nomenclature is explained in the table at the end of this report. In the first equation, the fourth term is that due to polymer pyrolysis in depth; this pyrolysis is assumed to be a one-step, zero-order, gasification reaction. The fifth term in this same equation is that due to radiation absorption in depth and the sixth is that due to photochemical heat release. It should be noted that these two terms are coupled in that they both draw upon the same energy source; this implies that an empirically determined polymer absorptivity for a given wavelength cannot be assigned purely to one or the other process without further information regarding their relative proportions at that wavelength. The mass flow rate m which appears in the above equations is the net mass flux resulting from the in-depth pyrolysis, that is

$$\dot{m}_{F} = \int_{0}^{0} (p_{s} z_{s} e^{-E_{s}/RT(x)} + \beta p I_{x}) dx$$

It is assumed here that fuel vaporization results both from the thermal degradation of the polymer and photochemical degradation as well.

In the heterogeneous model, it is assumed that ignition is the result of exothermic reaction of adsorbed oxidizer molecules with the surface molecules of the polymer. This is represented by a reaction which is zero-order with respect to the fuel, n-th order with respect to the oxidizer and which has an Arrhenius temperature dependence. Polymer vaporization is ignored and no further reactions are considered. The equations describing the model are:

Solid:

$$\frac{\partial T}{\partial t} = K_{s} \frac{\partial^{2} T}{\partial x^{2}} + \left(\frac{1}{\rho_{s}C_{s}}\right) \frac{\partial T_{x}}{\partial x} + \left(\frac{p Q_{re}T_{x}}{\rho_{s}C_{s}}\right)$$
Gas:

$$\frac{\partial T}{\partial t} = K_{s} \frac{\partial^{2} T}{\partial x^{2}}$$

$$\frac{\partial C_{ex}}{\partial t} = D \frac{\partial^{2} C_{ex}}{\partial x^{2}}$$

with the boundary conditions

ĸ

ч ф т

\$

$$T(x_{10}) = T_{o} \qquad \lambda_{q} \left(\frac{\partial T}{\partial x}\right)_{o,t} + Q_{chex,i} C_{ox}^{n} Z e^{-E/RT(o)} = \lambda_{s} \left(\frac{\partial T}{\partial x}\right)_{o,t}$$

$$C_{ox}(x_{10}) = C_{ox}^{\infty} \qquad D \frac{\partial C_{ox}}{\partial x} = C_{ox}^{n} Z e^{-E/RT(o)}$$

$$T(t_{ox}, t) = T_{o} \qquad T_{qxs}(o, t) = T_{solits}(o, t)$$

Here again radiation absorption in depth and photochemical heat release are included. The primary heat release term, i.e., the heterogeneous reaction term, appears in the surface heat balance.

The objective of the analysis is to solve the system of equations for the temperature distribution as a function of time and then to compute the time required from the start of the exposure until the temperature in the gas phase boundary layer (1st model) or on the surface (2nd model) runs rapidly upward. This is the ignition delay. Obviously, it can be evaluated only by defining in advance some criterion (critical temp. level, critical rate of rise, etc.) of runaway. It is the broader objective of the analysis to find the dependence of the ignition delay on radiation intensity, pressure level, physical properties, chemical properties, etc. These are the desired solutions.

Note that both of the mathematical models are highly non-linear and involve complicated couplings, and thus they are amenable to solution only by numerical methods. How we, various characteristics of the desired solutions can be anticipated by purely physical reasoning; these are discussed in the next section.

b. Discussion of Expected Solutions

The manner in which ignition develops in the gas phase model for a pure fuel is similar to that for a propellant as discussed previously. However, in the present case, the oxidizer is available in the gas from the start; consequently, as soon as the fuel begins to vaporize and diffuse into the gas, exothermic redox reactions commence. Since the gas is heated only by conduction from the hot fuel surface, the site of the most intense reaction will be in a thin gas layer right at the surface. (In the case of a composite propellant, the site of most intense reaction may be slightly off the surface, where the state of mixedness of the two reacting vapors is more advanced). The resulting feedback effect and subsequent runaway are analogous to those for a propellant. The thermal wave development for this model is sketched in Figure 5. The discontinuous change in radiant intensity across the surface is due to reflection; the effects of radiation penetration are discussed below. Again, note that the conductive heat loss to the gas coupled with the indepth absorption of radiation causes the maximum solid temperature to occur slightly below the surface.

The thermal wave development for the heterogeneous model is also sketched in Figure 5. Here, in theory, reaction commences immediately since it occurs on the surface which has a finite temperature at time zero. From there, the reaction rate and the oxidizer diffusion rate are equal; the surface heat release at first supplements and then overshadows the external energy flux as the reaction rate grows and ignition finally ensues.

The qualitative effects of absorptivity on thermal wave development have been discussed to some extent by Bastress ^ and Price' the basic ideas are treated more precisely here and the expected results are illustrated in Figure 6. The parameter which determines the type of behavior to be expected is the characteristic time ($\sqrt{\kappa^2 K}$); this derives directly from consideration of the relative thermal wave thicknesses due to conduction and in-depth absorption. In the top part of Figure 6 is shown the case of a very opaque fuel (high lpha) for which the ignition delay is much greater than the characteristic time. This approaches the limit of infinite absorptivity in which, obviously, all of the non-reflected radiation is absorbed at the fuel surface; energy penetrates below the surface only by conduction and hence it alone determines the thermal wave thickness in the solid. The incident radiation is thus utilized in the most efficient manner possible and the thermal lag portion of the ignition delay is minimized. The bottom part of Figure 6 illustrates a case which tends toward the opposite extreme, i.e., a transparent fuel (low \ltimes) having an ignition delay much less than the characteristic time. As the absorptivity tends toward zero, energy deposition occurs to an increasing depth in the solid. The resulting temperature gradient is so shallow that conduction plays no significant part in determining its shape. The radiant flux is called upon to heat a considerable mass of solid fuel, and hence the thermal lag is greatly extended; the efficiency of energy utilization tends to zero. It should be noted that this dominance of absorptivity in determining thermal wave shape exists also in the case of large but finite absorptivity for sufficiently small times since the energy penetrates to a finite depth instantaneously. Figure 5 serves to illustrate the intermediate case in which ignition delay and characteristic time are of the same order; both conduction and in-depth absorption influence the thermal wave development. Note that the same qualitative effects here attributed to absorptivity variations also can result from thermal diffusivity variations, although the sensitivity is less. In practice, of course, diffusivity is much less variable than is absorptivity; for propellants, the latter is strongly dependent on the particular formulation being tested.

c. Approximate Theoretical Solutions

The ideas discussed above can be extended and used in constructing an approximate theory of radiant ignition. First, however, one must account for processes other than the thermal lag in the solid; in both ignition models the processes of reactant diffusion and chemical reaction require a finite time to elapse. Furthermore, this time is a complex function of the various ignition conditions. For the present simplified analysis, however, it will be assumed that these processes are adequately represented by a characteristic time which depends only on the pressure, i.e.,



Then, in the asymptotic case of very high incident radiant flux, the surface temperature rises almost instantly to the level T* at which vigorous pyrolysis takes place (gas phase model) or at which vigorous surface reactions begin (heterogeneous ignition model). Some limiting mechanism, perhaps spalling or boiling, prevents the surface temperature from rising much higher. The thermal lag in the solid, in this case, is very small and the chemical-diffusion time comprises the major portion of the ignition delay.

$$t_{ign} \simeq t_{co}$$

For an incident flux less than the above case but still relatively high, falling on a propellant with a finite absorptivity, the thermal lag tends to dominate the ignition delay. However, since the ignition delay is still relatively short, there is very little spreading of heat, and it is in-depth radiation absorption rather than conduction that determines the time required for the surface to reach the temperature T*:

$$t_{ign} = \frac{1}{\alpha} \left[\frac{\rho c \Delta T^*}{T_{\alpha} (1-r)} \right]$$

This expression is the result of a simple heat balance based on the fact that the equivalent, uniform temperature, thermal wave thickness in the solid is $1/\alpha$ when absorption dominates. For simplicity, we are going to dismiss photochemical teactions for the present and we will use the concept of a fixed auto-ignition temperature; the validity of this concept is subject to question but for the purposes of deriving an approximate equation it is adequate.

Consider finally the asymptotic case of low radiant flux. Lere the ignition delay is long and conduction is the dominant factor in determining the thermal wave shape and hence the delay time. Proceed ing as above, one finds that the equivalent thermal wave thickness is

· Ż(π K+t)^½

The ignition delay, therefore, is given by

$$t_{igh} = \frac{\pi}{4} K \frac{\left[p C \Delta T^* \right]^2}{\left[T_0 (1-r) \right]^2}$$

Note that the sensitivity of ignition delay to radiant flux level is much stronger here than in the preceding case.

In the general case of any incident flux level, all of the above processes play a part in determining the ignition delay; approximately, then, we may add the three progressively asymptotic formulas:

$$t_{ign} \approx t_{cD} + \frac{1}{\alpha} \left[\frac{p C \Delta T^*}{I_o(1-r)} \right] + \frac{\pi}{4} K \left[\frac{p C \Delta T^*}{I_o(1-r)} \right]^2$$

Similarly, the measured ignition energy is given by

$$E = I_{t_{in}} \simeq I_{t_{co}} + \frac{1}{2} \left[\frac{\rho C \Delta T^{*}}{(1-r)} \right] + \frac{\pi}{4} K \left[\frac{\rho C \Delta T^{*}}{(1-r)} \right]^{2} \left(\frac{1}{I_{o}} \right)$$

Obviously, the straightforward addition of these three delay times constitutes a further simplification since, in any real case, the processes may interact and overlap appreciably. However, as will be shown, the simple expression derived above exhibits many of the same qualitative behavior trends as do experimental ignition data.

It is instructive to cast the equation for the overall ignition delay into dimensionless form. Define a dimensionless flux F and a dimensionless ignition delay C:

$$Y = \frac{t_{ign}}{t_{c0}} + \frac{t_{c0}}{\pi K} = \left(\frac{4}{\pi} \frac{t_{c0}}{K}\right)^{1/2} \left(\frac{1-Y}{\rho C \delta T^*}\right) I_{o}$$

Then the equation for the ignition delay takes the form:

$$\Upsilon = 1 + \left(\frac{4}{\alpha^2 \pi t_{co} K}\right)^{1/2} + \frac{1}{F} + \frac{1}{F^2}$$

d. Comparison With Other Laboratory Modes of Ignition

It is interesting to note that the above expression can, with minor modification, be applied equally well to other ignition modes. Note, however, that in so doing, one cannot retain the same value for the chemical-diffusion time since this may be influenced by the experimental conditions.

For end wall ignition in a shock tube, sample heating occurs purely by conduction from the shock-heated gas, thus the second term in the expression drops out. Furthermore, the sample surface reaches a sufficiently high temperature instantaneously for chemical and diffusion processes to commence immediately; thus the third term drops out also so that

$$t_{ign} \simeq t_{cD}$$

However, in contrast to radiant ignition, the gas adjacent to the sample surface is hot; if ignition occurs in the gas phase, the chemical reaction rates in particular, will be considerably increased by the high temperature. On the other hand, if ignition occurs via heterogeneous surface reaction, the high gas temperature affects t only insofar as it increases the oxidizer diffusion rate.

For convective ignition, the second term in the expression again drops out. Here, however, the thermal lag in the solid is again significant so that one has

$$t_{ign} = t_{c0} + \frac{T}{4} K \left[\frac{\rho C \Delta T^*}{T_o} \right]^2$$

I is now to be interpreted as the convective heat flux from the hot gas to the surface.

The comments made above regarding the high temperature gas effects apply here as well. In addition, there are the possible complicating effects of the flowing gas which could conceivably influence both ignition mechanisms and hence affect t_{CD}. In the convective ignition tests that have been reported in the literature so far, the second term has been at least 10 to 100 times larger than the first. Therefore, it can be said that such ignition tests have really been mainly tests of the mode of heating; the physico-chemical factors that affect t_{CD} have been obscured.

It is instructive to compare the three methods of ignition on the basis of the preceding dimensionless equation for $\mathcal C$.

In end-wall shock tube experiments only the first term survives, because F is virtually infinite in the usual exposure conditions. In the convective ignition experiments reported so far, the value of F is about 10^{-1} , and the third term is the only one that is important.

In radiation ignition experiments, the numerical coefficient of the middle term is of the order of unity (with perhaps a factor of ten either way) and F values have ranged from about 1 to 10 or 20. Therefore, in radiation ignition experiments it is possible to obtain results at low flux values that resemble convective ig..ition data and results at high flux values that resemble shock tube data. However, the role of the second term obviously depends on the magnitude of the numerical coefficient, and this depends in particular on the absorptivity α and on the reaction time t_{CD}; the presence of the second term can upset all comparisons with non-radiative ignition experiments.

Many of the physical and chemical complexities of real ignition are lumped into t and their further clarification awaits the solution of models such as those proposed in Section 4a. Nevertheless, it is of interest to compare the approximate theory derived above with experimental data; this is done in the following section.

5. Comparison of Theory With Experiment

a. Comparison With the Experimental Data of Beyer and Fishman

The arc image furnace data of Beyer and Fishman¹ comprise a fairly extensive survey of the effects of various experimental parameters on the radiative ignition behavior of a composite propellant in an inert atmosphere. In order to compare these data with the predictions of the approximate theory, the following numerical values wire chosen for the various constants in the expression.

$\rho = 1.6 \text{ g/cm}^3$	$K = 1 \times 10^{-3} \text{cm}^{\prime}/\text{sec}$
C = 0.3 ca1/g	$\propto = 100 \text{ cm}^{-1}$
r = 0.2	$\Delta T^{\star} = 300^{\circ} C$

The above values are considered to be fairly typical of composite propellants; with regard to the choice of ΔT^* , Beyer and Fishman

actually reported a somewhat lower value, however, Sutton and Wellings¹⁴ give values that are higher. The theory itself serves as a guide in the choice of a value for the chemica.-diffusion time, t_{CD} ; as stated in Section 4c, as the radiant flux tends to large values, the ignition delay time approaches t_{CD} . On the basis of the Beyer and Fishman data (see Fig. 9b), a value of t_{CD} equal to 35 msec at one atmosphere was chosen; again note that t_{CD} is assumed to vary inversely with pressure. (Note: expt'1. data are for AP/polysulfide samples in N₂).

The experimental and theoretical curves are compared in Figures 7, 8, and 9. Figure 7 shows the variation of ignition energy with pressure for various incident flux levels. It should be borne in mind that the comparison cannot be exact because the theory gives the ignition delay under continuous radiative exposure, whereas the Beyer-Fishman data were taken in start-stop experiments with the actual ignition occurring after the radiation was stopped. Nevertheless, at high and intermediate flux levels, the qualitative similarity between experiment and theory is seen to be quite good. At low pressures, the chemical-diffusion term in the theory dominates and the ignition energy is a strong function of pressure; at higher pressures this term tends to zero, the pressure-independent thermal lag terms become increasingly important, and the slope tends toward zero. At low flux levels and low pressures, however, the slope of the theoretical curve is markedly less than that found by Beyer and Fishman; this is the regime of long ignition delays and it is conceivable that various new phenomena enter (perhaps bubbling) that are not included within the framework of the approximate theory.

Figure 8 shows the variation of ignition energy with incident flux for various pressure levels. Again the qualitative similarity of theory to experiment is good except at low pressures and flux levels. The significant point to be noted here is that the theory does predict the existence of a minimum in the ignition energy curve. The required energy at first decreases with increasing flux as the conductive contribution to the thermal wave thickness shrinks. As the flux is further increased, however, this effect is first counteracted and then overcome by the energy contribution during the chemical-diffusion lag.

Figure 9 shows the variation of ignition delay time with incident flux for various pressure levels; the comparison is again favorable. At very low fluxes, the ignition delay is quite long; conduction dominates the thermal lag and hence, the ignition delay; the limiting slope of the curve is minus two. At slightly higher fluxes the effect of the in-depth absorption term is felt and the slope begins to decrease. Finally as the flux approaches large values, the flux-independent, chemical-diffusion term begins to dominate and the curves approach asymptotically the value to t_{CD} corresponding to each pressure level.

b. Comparison With The Experimental Data of Bastress, Niessen, and Richardson

The radiative and convective ignition data of Bastress, et al, 10 have been mentioned previously; here they are used as a further check

on the predictions of the approximate theory with regard to radiative ignition and also as a check on its predictions with regard to convective ignition. The data of these authors comprise ignition delay measurements on both aluminized and non-aluminized propellants; thus an attempt was made to include the effects of aluminum addition in the theoretical calculations. All of the propellant physical properties are doubtless affected by this additive but the greatest changes occur in K and α . The following values were assumed for these parameters:

No Aluminum	ca. 15% Aluminum
$K = 1 \times 10^{-3} \text{ cm}^2/\text{sec}$	$K = 3 \times 10^{-3} \text{ cm}^2/\text{sec}$
$\alpha = 25 \text{ cm}^2$	$\alpha = 100 \text{ cm}^2$

The change in thermal diffusivity, K, is based on measured values ; the assumed change in absorptivity, \propto , seems to be a reasonable and perhaps conservative estimate. Wit^L regard to the choice of the chemical-diffusion time, t_{CD}; for convective ignition the experimental data indicate only that it is small. A value of 2 msec at 5 atm. was assumed. For the radiative ignition calculations, the same value of t_{CD} used previously was assumed; thus for a pressure of 5 atm., t_{CD} = 7 msec. A larger value seems reasonable for radiative ignition.

The theoretical and experimental curves for both convective and radiative ignition are shown in Figures 10 and 11. The theory again exhibits many of the same qualitative trends as the experimental data. The agreement between the two for low convective heat fluxes (Fig. 10) is hardly surprising; in this range t CD is negligible and the theory reduces to the same type of expression as that used by Bastress and others' to correlate their experimental data. (Note, however, that the curve for one of the PBAA/AP/A1 propellants does not fit this expression). At higher convective fluxes, the lower theoretical curve (unaluminized propellant) exhibits a decreasing slope due to the increasing importance of the flux-independent chemical-diffusion term. This tendency is not shown in the experimental data and if the experimental data are really firm, this may indicate that the assumed value of t_{CP} is too large.

Both the theoretical and experimental radiative ignition curves (Fig. 11) exhibit a lesser slope than those for convective ignition. In the theory, this effect is caused by the presence of the in-depth absorption term. Note especially the <u>inversion</u> in the relative ignitabilities of the aluminized and non-aluminized propellants exhibited by both the theoretical and experimental curves due to the substitution of radiation for convection. Again, this serves to illustrate the considerable alterations which ensue from this substitution.

Further data on the comparison of convective and radiative ignition are presented by Jensen and Cose¹⁵. These data are of particular interest because the convective ignition times were obtained from the flame-spreading interval of an actual rocket motor with a pyrogen igniter. Although it appears that the convective flux history varied from point to point in the motor and the flux was not constant at any point, the ignition delay times based on average fluxes agree well with arc furnace data for a propellant with nearly the same composition (PBAN/AP/A1). This is not too surprising since all of the data fall in the region where the thermal lag dominates the ignition delay (the mechanism of runaway is of little importance here); the presence of aluminum in the propellant effectively opacifies it so that a radiative heat source has about the same heating efficiency as a convective heat source.

c. Sensitivity of Radiative Ignition Time to Absorptivity

The inversion in relative ignitability noted above and the large changes in absolute ignitability of propellants such as those found by Bastress are probably due to the effects of in-depth absorption, when radiation is substituted for conduction. The effect of absorptivity on ignition delay, within the framework of the approximate theory, is illustrated in Figure 12; the numerical values of the parameters are the same as those used in the previous section. Obviously, the absorptivity is a very important parameter; the ignition delay decreases by as much as a factor of ten as α goes from to infinity. Note in Figure 12 that the sensitivity to 25 cm changes in α is much higher in the region of small and moderate values of \propto (since $t_{igi} \neq$). For sufficiently small values of α , the indepth absorption term dominates the ignition delay; the slope of the curve in Figure 12 tends toward 1/2. For very large values of α , this term is negligible; at low fluxes, the conductive term dominates and the slope tends toward one; at high fluxes, t_{CD} dominates and the slope goes to zero.

Propellants with no opacifying additives probably have effective absorptivities in the range of 25 cm⁻¹ and are thus poor candidates for radiative ignition tests. Those propellants containing various opaque additives may have effective absorptivities as high as a few hundred cm⁻¹ and are therefore not greatly affected by this parameter, i.e., their ignitability will not be significantly decreased by indepth absorption, except at high pressure. However, the other cautionary statements made previously regarding the possible adverse effects of radiation still apply.

6. <u>Conclusions</u>

Although the arc image ignition technique is today widely accepted as a means of evaluating propellant ignition behavior the validity of its use for this purpose is far from being well established. As we have indicated, the substitution of a radiant energy input for a conductive or convective input opens the door to a wide variety of new physical and chemical effects which may have a considerable influence on the ignition process. If arc image data are to be meaningful as a basis for rocket igniter design, one must either be assured that the various perturbing effects are negligible in the arc furnace tests or, since this is not likely to be the case, one must be able to correct the data. We have outlined the initial steps needed both for developing these theoretical corrective guidelines and an overall understanding of the radiative ignition process. However, until further experimental data and theoretical solutions are available, considerable caution is necessary in the use of the arc image furnace. At this moment, it seems to us that radiative ignition is interesting for research purposes but is potentially quite mileading when applied to routine propellant ignitability testing.

7. Discussion of Future Studies of Radiative Ignition

a. <u>Theory</u>

The strong need for further theoretical and experimental analysis of radiative ignition has been indicated previously. However, as has been shown, composite propellant ignition via radiation is extremely complex and presents a very formidable challenge. The problem is thus best attacked in a step-wise fashion. The mathematical models proposed in Section 4 for both heterogeneous and gas phase ignition of a pure fuel form the basis for a logical first step in a detailed analysis. These models incorporate the effects of a cold environmental gas, in-depth radiation absorption, photo-chemical reaction, and, in a simplified way, in-depth fuel vaporization all of which also appear in propellant ignition. These models thus facilitate the study of several factors peculiar to radiative ignition of propellants. However, quite obviously, these models cannot predict all characteristics of actual propellant ignition via radiation. For example, the exclusion of an oxidizing component from the solid phase precludes the possibility of exothermic redox reactions below the surface such as may occur in tests with real propellants. It also precludes the possibility of any added exothermic term due to oxidizer decomposition. Furthermore, the model substitutes one dimensional heat transfer and di fusion processes for what must necessarily be three dimensional processes in actual propellant ignition. These and other added complications of real propellant ignition are best incorporated in the second step, however, so that the problem can be analyzed piecewise, thus facilitating the isolation of the more important factors.

Despite the considerable mathematical simplification afforded by the substitution of a pure fuel for the propellant, the equations of the models are non-linear and not amenable to analytic solution. Parametric studies of the solutions must thus be performed with the aid of a digital computer. Parameters of particular interest in these studies are radiant flux le'el, pressure, oxidizer concentration, fuel absorptivity, fuel volatility and reactivity, and oxidizer reactivity. The dependent variable is, of course. ignition delay time. The validity of the theoretical models is to be checked by comparing the results of these parametric studies with similar experimental test series.

b. Experiment

There are two possible radiation sources that could be used in the experimental program an arc image furnace or a laser. Some of the problems associated with the arc furnace have been alluded to previously. First there are problems of a geometric origin; high fluxes can only be produced by using large beam convergence angles which require very precise sample positioning. Because of this steep convergence the angular dependence of both surface reflection and in-depth absorption must be accounted for. Second, the arc is a polychromatic radiation source; in-depth absorption thus cannot be represented by a simple exponential decay of intensity but must instead be represented by an expression which is the result of integrating over the spectral band using empirically determined absorption data. Third, there is the fact that the arc radiation lies in the same spectral region normally used to detect ignition and thus tends to hinder this detection.

The recent development of the high power carbon dioxide laser has provided a radiation source of power comparable to the arc furnace but without many of its problems. The nearly collimated nature of the laser beam permits focusing at much shallower convergence angles (i.e., the source intensity is much higher); the angular dependence of reflection and in-depth absorption can probably be ignored without introducing appreciable error. The light is monochromatic (10.6μ) ; in-depth absorption thus follows Beer's law (although the temperature dependence of the monochromatic absorption coefficient must still be accounted for). Finally, the laser wavelength is far removed from the spectral region of ignition detection and thus introduces no interference.

The laser is not without its own problems, however; the primary difficulty is connected with the radial flux distribution in the beam. A laser consists of an oscillating cavity containing a medium capable of light amplification; this cavity can only oscillate in certain specific modes or combinations of modes and each mode produces a fixed type of beam flux distribution. For example, the simplest (fundamental) mode produces a beam whose flux distribution is Gaussian (bell-shaped); higher radial modes yield concentric ring patterns.

Now, the ignition models, as written, are one-dimensional; multidimensional versions of the models would add nothing to the understanding of the fundamental processes of ignition and would, in fact, only make solution of the models more difficult. Since the theoretical and experimental results are to be compared, however theory and experiment should correspond as closely as possible. The experiments should thus be devised so as to approach the one-dimensional ideal as closely as possible. This can be done by <u>uniformly</u> irradiating the fuel sample over a sufficiently large area (which can be estimated from the physical properties of the fuel and environmental gas). The difficulty is obtaining this uniform irradiation with a laser.

Various techniques of overcoming this difficulty present themselves Perhaps the most obvious is a straightforward, brute-force approach. The central portion of a Gaussian distribution is nearly flat; one could irradiate the sample uniformly to within \pm 5% by using the central 10% of the beam. However, this requires a laser whose output is ten times larger than the power actually utilized. More economical solutions to the problem appear possible.

Despite the radically different nature of the laser, experimentation with it is basically similar to arc image testing though perhaps somewhat simpler. The other basic elements of the apparatus, a shutter, sample environment concrol system, and ignition detection system are essentially unmodified. As indicated, the experimental program is to consist of parametric studies analogous to those to be done on the theoretical models. These results will provide a considerably increased insight into the ignition process and the qualitative effects peculiar to a radiative heat input; as such they will form a firm basis from which to attack the problem of propellant ignition by radiation.

References

- 1. Beyer, R. S., and Fishman, N., "Solid Propellant Ignition Studies with High Flux Radiant Energy as a Thermal Source", in <u>Progress in Astro-</u> nautics and Rocketry, Vol. I, Solid Propellant Rocket Research, (Academic Press, New York, 1960), p. 673
- Altman, D., and Grant, A. F., "Thermal Theory of Solid Propellant Ignition by Hot Wires", <u>Fourth Symposium on Combustion</u>, (Williams & Wilkins Co., Baltimore, 1953), p. 158
- Baer, A. D., Ryan, N. W., and Salt, D. L., "Propellant Ignition by High Convective Heat Fluxes", in <u>Progress in Astronautics and Rocketry, Vol. I,</u> <u>Solid Propellant Rocket Research</u>, (Academic Press, New York, 1960), p. 653
- McAlevy, R. F., Cowan, P. L., and Summerfield, M., "The Mechanism of Ignition of Composite Solid Propellants by Hot Gases", in <u>Progress in Astro-</u> <u>nautics and Rocketry, Vol. I,</u> (Academic Press, New York, 1960), p. 623
- 5. Bastress, E. K., <u>et al</u>, "Solid Propellant Ignition Studies", <u>Third Quart-</u> <u>erly Progress Report</u>, December, 1963, Arthur D. Little, Inc.
- 6. Anderson, R., Brown, R. B., Thompson, G. T., and Eberling, R., "Theory of Hypergolic Ignition of Solid Propellants", presented at the AIAA Heterogeneous Combustion Conference, Palm Beach, Florida, December, 1963 (AIAA Preprint 63-514)
- Anderson, R., Brown, R. B., Shannon, L. J., "Ignition Theory of Solid Propellants", presented at the AIAA Solid Propellant Rocket Conference, Palo Alto, California, January, 1964, (AIAA Preprint 64-156)
- 8. Hermance, C. E., "Ignition of the Reaction Field Adjacent to the Surface of a Solid Propellant", Princeton University, Department of Aerospace and Mechanical Sciences, Ph.D. Thesis, November, 1963
- Proto taken from arc image ignition film presented by L. J. Shannon of United Technology Center at OSR Propulsion Contractor's Meeting, Palo Alto, California, August, 1966
- Bastress, E. K., et al, "Ignition of Solid Propellants by Various Modes of Heat Transfer", paper presented at the ICRPG/AIAA Solid Propulsion Conference, Washington, D. C., July, 1966
- Bastress, E. K., et al, unpublished work, Arthur D. Little, Inc., Cambridge, M'ss., 1964.
- Price, E. W., <u>et al</u>, "Theory of Ignition of Solid Propellants" <u>AIAA J. 4</u>, July, 1966
- Carslaw, H. S. and Jaeger, J. C., <u>Conduction of Heat in Solids</u>, 2nd. Ed., (Oxford University Press, London, 1959) p. 75

- 14. Sutton, D., and Wellings, P. C., "The Ignition of Solid Propellants by Radiant Energy", RPE-TR-66/4, Rocket Propulsion Establishment, April, 1966
- Jensen, G. E., and Cose, D. A., "Studies in Ignition and Flame Propagation of Solid Propellants", United Technology Center Report #UTC 2117.FR, June, 1966.

teritorial construction and the second se

3

Nomenclature

- T temperature
- t time
- K thermal diffusivity
- v convective velocity
- Q heat of reaction
- Z frequency factor
- C heat capacity
- E activation energy
- R gas constant
- ρ density
- thermal conductivity

 C_{ox} , C_{F} - concentrations of oxidizer and fuel, respectively

- D molecular diffusivity
- I_x radiation absorption function
- β fraction of photochemical products that form fuel vapor
- p combined conversion factor and fractional efficienc, of photochemical reaction (wavelength dependent)
- α absorptivity
- r reflectivity

 ΔT^* - difference between ambient and auto-ignition temperatures

subscript s - solid

Subscript g - gas





March 1999

1

: JPB R 4052 67

-24-



BUBBLING INDUCED IN A PURE PBAN POLYMER SAMPLE BY A LOW RADIATIVE HEAT FLUX FIGURE 3

大学の時代的にない。ためもの時代のないがあった

and the state of t



FIG. 4 COMPARISON OF ARC IMAGE IGNITION WITH OTHER METHODS OF IGNITION

-27 -

COMPARATIVE FEATURES (Part II)	FFECT OF AB- SURFACE SENSIFIVE DETECTABILITY OF DRBING AGENTS EFFECTS PHYSICAL FACTORS IGNITION SURFACE	egligible if Possible Gas Temp. and On-Catalytic Hetero. Reac. Mass Flow Rate	egligible if Possible and Diameter	egligible if Possible Interverse Regulant Tem	rob. Enhance Bubbling, Gas Eject. & Wavelength of Radiation, Ignition Somewhat Obscured gnitability Possible Hetero. Reac. Gas Comp., & Temp. (if by Radiation Gas Phase Mechanism)	e Bubbling, Gas Eject. & Wavelength of Radiation, Possible Hetero. Reac. Gas Comp., & Temp. (if Gas Phase Mechanism)	Possible Gas Heter J. Reac. and	egligible if Possible Gas Temp., Composition Ignition Unobscured on-Catalytic Hetero. Reac. and Mass Flow Rate	egligible if Possible Particle Temperature Ignition Unobscured on-Catalytic Hetero. Reac. and Diameter	egligible if Possible Power Input to Wire Ignition Unobscured on-Catalytic Hetero. Reac
	EFFECT OF AB- SORBING AGEN'IS				Prob. Enhance Bu Ignitability Po		Negligible if Po Non-Catalytic He	Negligible if Po Non-Catalytic He	Negligible if Po Non-Catalytic Ho	<u> </u>
1 <u></u>	IGN IT ION METHOD	Pyrogen (Hot Gas)	Pyrotechnic (Hot Part.)	Liquid	Arc Image Furnace	Laser	Shock Tube (End Wall)	Shock Tunne 1	Hot Solid or Liquid Particles	Heated Wire

Cont.

[eoitoerd

-28-

-28-



-29-



-30-

-00



. -31-

10

JPB'Z 9056



instan first and a second and the structure of the second s

JP. B & 4057

Fig. 8a Theoretical effect of radiant flux level on ignition energy









LO 0900 2.8 d?

Saverth out of the start her see stranger with the





- 36 -

1

UNCLASSIFIED

Security Classification

DOCUMENT CONFROL DATA - R & D

Security classification of title, body of abstract and indexing anne atlon must be entered when the overall report is classified)						
Princeton University	28. REPORT SECURITY CLASSIFICATION UNCLASS IF IED					
Department of Aerospace and Mechanical Sciences Princeton, New Jersey 08540	25. GROUP					

3. REPORT TITLE

A CRITICAL ANALYSIS OF ARC IMAGE IGNITION OF SOLID PROPELLANTS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates) Scientific Interim

5. AUTHOR(S) (First name, middle initial, last name)

M Summerfield and T J Ohlemiller

78. TOTAL NO OF PAGES 36	76. NO. OF REFS
98. ORIGINATOR'S REPORT NU	IMBER(\$)
AMS Report No. 789)
9b. OTHER REPORT NO(S) (Any this report)	other numbers that may be assigned
AFOSR 67-1534	
	36 90. ORIGINATOR'S REPORT NU AMS Report No. 789 90. OTHER REPORT NO(S) (Any this report)

1. Distribution of this document is unlimited.

11 SUPPLEMENTARY NOTES	AF Office of Scientific Research (SREP)
	1400 Wilson Boulevard
	Arlington, Virginia 22209

13 ABSTRACT

The arc image furnace is used widely for evaluating solid propellant ignition characteristics, but the applicability of such data to rocket motors is questionable. In arc image testing, radiation replaces the normal conductive and convective heating modes that exist in rocket motor ignition; radiation interacts with the propellant in a basically different way. Thus, for example, because radiation is absorbed in depth, the ignition delay is sensitive to propellant opacity. In addition, radiation can cause subsurface disruption of the propellan. A physical model is proposed in which incident radiation penetrates the propellant and is absorbed, causing thermal and photochemical decomposition and a temperature increase. Oxidative reactions start either in the gaseous boundary layer or on the surface and, by thermochemical action, lead to a flame. The system of differential equations for this complete model is complicated and has not been solved. However, an equivalent model of simpler character is solved, and an equation is produced for the ignition time delay as a function of radiation intensity pressure, and physico-chemical properties. The qualitative behavior of experim. tel radiative ignition data reported by Beyer and Fishman and by Bastress is explained by this theory.

DD FORM 1473

UNCLASS IF IED Security Classification

UNCLASS IF IED

and the strength

Security Classification

l de la constante de	KEY WORDS	LIN		LINK B		LINK C		
·			ROLE	WT	ROLE	₩T	ROLE	w۲
					ļĮ	l		
Arc Image Fùrnace								
						l		
Solid Propellant Igr	nition							
Radiative Ignition								
Ignition Theory								
Solid Fuel Ignition					ļļ	1		
	,							
						I		
							, . 	
							: · 1	
							1 · ·	
							! • ·	
							:	
							 i	
							•	
							İ İ	
				:				
والمسترجعا والمناقل ومنبعة التشنين فبالفاطيني والماكر والمتكر بالمتر					L			
		B-2	UNCLAS					