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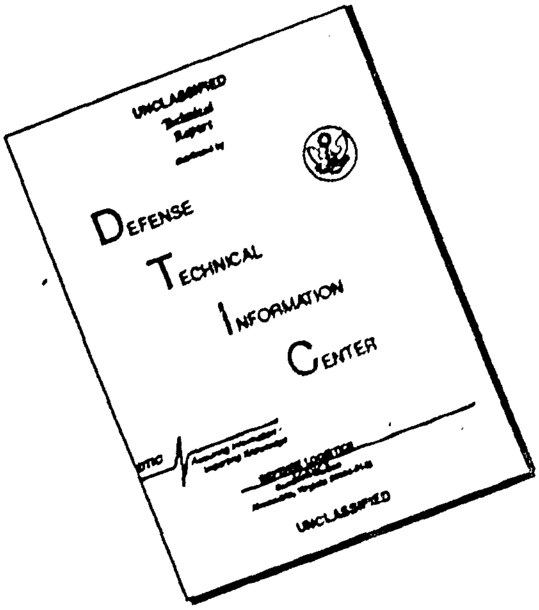

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THE MECHANISM OF DEFLAGRATION OF PURE AMMONIUM PERCHLORATE

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and Keith E. Rumbel

Atlantic Research Corporation
Alexandria, Virginia

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The Mechanism of Deflagration of Pure Ammonium Perchlorate[*]

Raymond Friedman, Joseph B. Levy, and Keith E. Rumbel

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I. SUMMARY:

Results of new deflagration experiments with ammonium perchlorate, both pure and catalyzed, have been presented. These include studies of rates of deflagration, pressure limits of deflagration, surface temperature, and effects of incident radiation on deflagration. These and previous results have been interpreted in terms of a tentative qualitative model of the deflagration process, summarized below.

The crystalline ammonium perchlorate sublimes to ammonia and perchloric acid vapors by an endothermic process. Heat is generated in a gas-phase oxidation-reduction process perhaps 10^{-5} cm (30 mean free paths) above the surface, at 100 atm. The final temperature is about 930°C and the surface temperature is one or two hundred degrees cooler. Heat is conducted back through this thin layer to supply the energy for preheating and vaporizing the crystal. The propagation rate is governed primarily by the rate of heat generation in the gas phase and the thermal conductivity of the gaseous layer just above the surface.

The low-pressure flammability limit is caused by radiative heat loss to the surroundings, while the previously reported high-pressure limit is caused by convective heat loss and may be eliminated by

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suitable shielding of the sample. Catalysts accelerate the burning rate by projecting from the surface into the thin gaseous reaction zone and producing local acceleration of reaction rate. Very low concentrations of catalyst, however, act primarily to reduce flammability by raising the lower pressure limit, because the catalyst increases the emissivity of the burning surface and hence the rate of radiant heat loss.

The foregoing model, while probable, is not rigorously established, and is presented at this time partly to stimulate further research.

II. INTRODUCTION:

Experiments (1) have shown that at certain ambient conditions a steady deflagration wave will propagate through pure ammonium perchlorate which has been pressed into an essentially void-free pellet. The rate of propagation for this process at typical rocket pressures is of the same order of magnitude as the burning rate of many ammonium perchlorate-oxidized solid propellants, so the ammonium perchlorate decomposition flame may well exert a controlling influence on propellant burning rates. Evidence of the interrelation between the decomposition flame and the propellant flame may be adduced from data showing variation of composite propellant burning rate with pressure, ammonium perchlorate particle size, and catalyst or inhibitor effects. Such discussion will be presented in another paper now in preparation. The present paper is concerned with deductions in regard to the mechanism of the ammonium perchlorate decomposition flame, which are based largely on new experimental findings reported below.

III. EXPERIMENTAL RESULTS:

Certain experimental findings in regard to the deflagration of pure and catalyzed ammonium perchlorate have been previously reported (1). Further results are described here.

A. Pressure Limits and Catalyst Effects

Figure 1 is the curve of deflagration rate vs pressure curve for pressed pellets of ammonium perchlorate 4 mm square and 38 mm long, burned at essentially constant pressure in a nitrogen atmosphere. The propagation was downward; no inhibitor was needed on the sides of the samples to prevent the formation of a conical burning surface.

The lower-pressure limit was previously reported as 45 atm. In the present tests a much more efficient ignition technique was used, which consisted of an energetic propellant mixture instead of a hot wire, and the lower limit was accordingly extended to 22 atm. The present lower-limit value is believed to be independent of ignition energy, since specimens ignited at the limit pressure with the present powerful ignitor would often burn partially before extinction. Experiments have also shown that the lower limit is insensitive to sample size and to the substitution of helium for nitrogen as the ambient atmosphere.

In previous tests an upper-pressure limit of about 250 atm was found. More recent tests with samples ignited on a deeply recessed surface and with standard-size asbestos-wrapped samples have shown that this effect was due to convective cooling, which becomes increasingly effective with increasing pressure. Figure 1, in contrast to previously reported data (1), shows that deflagration rate increases with increasing pressure at least to 340 atm, the limit of the apparatus. The high-pressure data points on the curve were taken with asbestos-wrapped specimens.

Some of the unusual effects of the addition of copper chromite powder (Harshaw Chemical Company, Cu-0202) on the lower pressure limit were previously described (1). Small additions raised the limit, while large additions lowered it. Further systematic experiments concerning the effect of additive concentration on pressure limits have now been made down to very low catalyst concentrations, the more efficient ignition technique being used, with results as shown in Figure 2. The copper chromite powder, the weight-average particle size of which was 8.5 microns, was mechanically mixed with ammonium perchlorate and the mixture was pressed into pellets at 50 tons per square inch. Additions of as little as one part in 20,000 are sufficient to raise the lower-limit pressure four-fold. The effect is maximized at three parts per thousand of catalyst. Above this level, both the upper and the lower pressure limits widen. A cross-plot is shown in Figure 3 of the deflagration rate vs additive concentration at 204 atm, a pressure at which burning was obtainable at all concentrations (cf. Figure 2). Deflagration rate is essentially independent of catalyst level up to one part per thousand and then rapidly increases at higher concentrations. It should be noted that three effects - increase of upper pressure limit, decrease of lower pressure limit, and increase of deflagration rate - all occur at about the same catalyst level.

At high catalyst levels (4 parts per 100 parts mixture) the deflagration rate maximizes and then decreases sharply with further additions. The catalyst perhaps may act as a diluent at such concentrations. The lower pressure limit, surprisingly, continues to decrease up to 40 parts of catalyst per 100 parts of mixture.

The data shown in Figure 2 were obtained with specimens of 4 mm square cross-section and downward propagation. The upper pressure limit, at least, is known to be strongly affected by sample size and geometry, so the results in Figure 2 are mainly of interest insofar as they indicate trends in combustion behavior, and quantitative conclusions should not be drawn solely from these limits.

B. Surface-Temperature Studies

Photographs have been taken of the reacting surface of deflagrating specimens of ammonium perchlorate by means of infra-red sensitive film. This radiation is obviously the sum of thermal emission from the surface and chemiluminescence (if any). Assignment of a value for the surface temperature from densitometric measurements of such photographs depends on the assumptions made for the ratio of chemiluminescence to thermal radiation and for the surface emissivity. If the minimum conceivable values are assumed for these two unknown quantities, one then may obtain an upper limit for the surface temperature. If this is found to be substantially below the measured final gas temperature, then a significant conclusion may be drawn.

The photographs were taken through the window of a test bomb at 500 psi, the burning surface making an angle of approximately 45 degrees with the optical axis, with photographic conditions as follows: exposure, 1/100 sec; ratio of image distance to lens diameter, 4.2; film, Kodak High-Speed Infra-Red; filter, Corning No. 7-69. The filter cuts off below 7100 Angstrom units, while the film sensitivity curve peaks at 8100 and has dropped a factor of ten at 9100 Angstrom units. The strands were 4 mm square, and the burning rate was such that the surface regressed about 30 microns in 1/100 second. Calibration was obtained by photographing an electrically heated rod of measured temperature and known emissivity through the same window. Further details are presented in Appendix A.

By densitometric measurements of photographs obtained by this procedure, an upper limit for the surface temperature was calculated. The emissivity was taken to be 0.2, a reasonable low-limit estimate, since measured total emissivities of a list of 13 nonmetallic solids at 800°C are all above this value (2). The lower limit for chemiluminescence is obviously zero. On this basis, and with the use of Planck's law, the upper-limit value for surface temperature was computed to be 616 ± 5°C.

This result is based on radiation in the spectral range 7100-9100 Angstrom units. Other experiments with different film and filters in the spectral range 6000-6750 Angstrom units gave an upper-limit temperature about 100 degrees higher. An even higher apparent temperature was obtained when the entire visible spectrum was utilized. This may mean that there is a chemiluminescent component in the visible part of the spectrum. Visual observation in a darkened room revealed a reddish glow from the spontaneously reacting surface and a much fainter pinkish glow from the product gases. Gaydon (3) reports an ammonia alpha-band due to NH_2 which emits prominently in the red for H_2/N_2O flames and which may be involved in this situation.

These considerations notwithstanding, the surface temperature must be below 816 ± 5°C, unless the emissivity is somehow below 0.2. The product-gas temperature at 500 psi has been measured with thermocouples as 932 ± 7°C (1). It is clearly indicated, therefore, that the temperature rises at least by 100°C because of exothermic gas-phase reactions.

This conclusion would be invalidated if substantial absorption of the emitted radiation by the product gases occurs. The absorptivity of water vapor, a prominent constituent, is well known (4,5). A calculation shows that absorption by water vapor in the spectral region under consideration is entirely negligible, for the present geometry. It does not seem likely that any other species to be expected in high concentration in the product gases could absorb strongly enough to cause difficulty.

Another possible objection is that the effective surface emissivity of the ammonium perchlorate may be abnormally low because of the steep temperature gradient just below the surface. On the other hand, the known local roughness of the reacting surface should tend to increase the effective emissivity.

In summary, the observations reported herein strongly suggest that the deflagration of ammonium perchlorate involves a substantial contribution from the gas phase.

A previously reported attempt to measure surface temperature of deflagrating ammonium perchlorate (6) was unsuccessful because it was not possible to fabricate sufficiently small thermocouples.

C. Effect of Added Radiation on Burning Rate

Ammonium perchlorate will not normally deflagrate below 22 atmospheres. However, a radiant-heating apparatus shown in Figure 4 was found to produce conditions permitting steady deflagration at atmospheric pressure.

The radiant-heat source is a 1000-watt projection lamp having a coiled tungsten filament with an effective area of 1 cm square, operated at a slight overvoltage. A pair of front-surface paraboloidal mirrors forms the optical system. The first mirror has a diameter of 11 inches and an f-number of 0.364, the second a diameter of 12.5 inches and an f-number of 0.48. The system has been calibrated with a radiant-flux meter which consists of a blackened cavity in an externally insulated copper block, the temperature rise of which may be followed. It has been possible to achieve an irradiance up to 20 cal/sq cm-sec with this system.

Ammonium perchlorate pellets of 4 mm square cross section and 38 mm long are fed horizontally into the focal point of the radiation apparatus from a tube water-jacketed to prevent radiant heating of the sides of the specimen. Pure ammonium perchlorate ignites in a few seconds and continues to deflagrate as long as an adequate radiant flux is maintained. Below about 10 cal/sq cm-sec, deflagration can neither be produced nor maintained, although very slow sublimation occurs. Figure 5 shows the variation of the radiation-induced deflagration rate with radiant intensity. The deflagration rate was taken as the feeding rate necessary to yield steady-state deflagration at the tube opening.

For pure ammonium perchlorate there is a critical radiation level (10 cal/sq cm-sec, as shown in Figure 5) below which only slow sublimation occurs and above which deflagration occurs. The deflagration rate is essentially linear with radiation flux, and extrapolates to a finite rate at zero radiant flux.

Addition of catalyst profoundly increases the deflagration rate and reduces the threshold flux required for ignition. The intercepts at zero radiant flux for the three curves with 0, 0.5 and 3.0

per cent Cu-0202 catalyst increase in the ratio 1:1.9:3.1, while the slopes increase in the ratio 1:4:13. The increase in the intercept values may be a measure of the chemical augmentation of deflagration rate by the catalyst, while the greater increase in slope with increasing catalyst concentration may include this effect and also the greater absorptivity of the surface for radiation when catalyst is present. No method has been devised for measuring the surface reflectivities under actual deflagration conditions, but some crude measurements of reflectivities at room temperature have yielded results as follows:

Specimen	Reflectivity
Pure NH_4ClO_4	0.63
NH_4ClO_4 + 0.5% Cu-0202	0.32
NH_4ClO_4 + 3.0% Cu-0202	0.21

Presumably the same trend of decrease in reflectivity with increasing catalyst level exists at deflagration conditions, so the higher dependence of the slopes of Figure 5 on the catalyst level may be in part attributable to this effect. It is also possible that a "worm-holing" effect is involved as the radiant energy penetrates the translucent perchlorate and is absorbed by the catalyst particles. A similar effect has been discussed for double-base propellants (7).

A number of radiation-induced deflagration experiments with other additives have been carried out, the results being generally similar to, but not as striking as, those with copper chromite. An experiment with a three per cent addition of magnesium oxide was noteworthy, as the radiation-induced combustion was associated with a fusion of the specimen. Some of the material burned and the rest ran down the side of the apparatus and resolidified. Addition of either calcium oxide or magnesium perchlorate to ammonium perchlorate led to a similar effect. It is postulated that under pre-deflagration conditions the magnesium oxide or calcium oxide is converted to the corresponding perchlorate, which forms a meltable eutectic with ammonium perchlorate.

DISCUSSION OF LOWER PRESSURE LIMIT:

Spalding (8) and Mayer (9) have independently shown that a nonadiabatic gaseous flame must possess a sharp flammability limit which is reached by progressive change of any independent variable which reduces the flame temperature, as long as the rate of heat loss does not vary as rapidly with a change of flame temperature as does the rate of heat generation. Their mechanism involves the following sequence: flame temperature is dropped slightly; reaction rate decreases; deflagration rate decreases and flame gets thicker; heat loss from reaction zone becomes a larger fraction of heat generation, and this further lowers flame temperature; the reaction rate decreases further, etc. The situation eventually either stabilizes at a reduced combustion intensity or the flame goes out, depending on the numerical parameters. It is proposed that this mechanism is broadly applicable to the lower pressure limit of ammonium perchlorate.

For deflagrating ammonium perchlorate, the rate of radiant heat loss from the hot solid-gas interface is essentially pressure-independent while the deflagration rate, and hence the over-all of heat generation decreases with decreasing pressure. Hence, reduction of pressure must produce a lowering of flame temperature, and at some critical point the flame is weak enough that extinction can occur by a Spalding-Mayer instability of the gas-phase flame.

Supporting evidence of three kinds may be cited to this view:

I. If this theory is correct, precooling of the sample below room temperature should reduce the flame temperature as effectively as the low-pressure radiant heat-loss mechanism, and precooled strands should be nonflammable even at high pressures. On cooling to minus 18°C, it was indeed found that ignition could not be obtained at any pressure up to 270 atm (1).

II. The theory implies that addition of external radiant energy should promote flammability. It has been found that stable atmospheric flammability is obtainable when a radiant flux of at least 10 cal/sq cm-sec is introduced. A portion of this flux is reflected and a portion is transmitted into the interior of the sample, subsequently becoming lost by conduction to the sides. The remainder, which is effective in heating the surface, is clearly comparable in order of magnitude with $1.36\ \epsilon(T_s/1000)^4$ cal/sq cm-sec, the rate of radiant loss from the surface, if T_s, the surface temperature (°K), is of the order of 1000-1100°K (ϵ is the surface emissivity.)

III. Addition of very small proportions of copper chromite, a black powder, very greatly affects the low-pressure limit (Figure 2), reducing flammability, while larger proportions promote flammability. Other dark additives behave similarly *. This is readily understandable if the primary effect of a small proportion of the black additive is to increase the emissivity of the surface, promoting heat loss, and seems inexplicable on any other basis.

In view of this evidence, a heat-loss mechanism for this limit seems to be established. The insensitivity of the limit to sample size and to nature of ambient gas (nitrogen vs helium) is further evidence that radiative heat loss is more important than convective loss.

DISCUSSION OF PROPAGATION MECHANISM:

The reaction

$$NH_4ClO_4\ (c) \longrightarrow HCl\ (g) + 1.5\ H_2O\ (g) + 0.5\ N_2 + 1.25\ O_2$$

is exothermic by 39.3 kcal at 25°C. This corresponds to an adiabatic flame temperature (at constant pressure) of 1136°C. This conclusion is modified slightly by the pressure-dependent equilibrium

$$2\ HCl\ (g) + 0.5\ O_2 \rightleftharpoons H_2O\ (g) + Cl_2$$

which is exothermic by 13.7 kcal at 25°C upon shifting to the right. At 100 atm the equilibrium ratio of Cl_2 to HCl is 0.11 and the corresponding calculated adiabatic flame temperature is 1167°C.

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* Addition of 0.25 per cent platinum black raised the lower limit from 22 atm to above 120 atm. Cf. also (1).

[illegible]ocouple measurements [illegible] product-gas analyses [illegible] that the deflagration reactions [illegible] necessarily proceed to equilibrium. Nevertheless propagation occurs and [illegible] the above-indicated energy is released in the decomposition [illegible] 100 atm. The first problem which must be considered is the establishment of the location at which this energy is liberated.

The two broad possibilities are condensed-phase and gas-phase exothermic reactions. Three types of experimental evidence, reviewed below, strongly suggest that the significant heat generation occurs in the gas phase:

I. It has been shown (10) that vacuum-heating of ammonium perchlorate produces sublimation with substantial recovery of recondensed ammonium perchlorate. The original decomposing crystal develops a porous structure which may be associated with a constant sublimation rate of the intermosaic matter (11). When the crystal is heated at 200-430°C under an inert gas instead of under vacuum, sublimation is reduced and gaseous oxidation-reduction products may be recovered. The rate of sublimation increases more rapidly with increasing temperature than the rate of decomposition. These facts may reasonably be interepreted by assuming a competition between two processes: (A) the crystal sublimes to NH_3 and $HClO_4$ vapors which diffuse away and recondense at a distant cold surface, and (B) an oxidation reduction reaction occurs. At the higher temperatures characteristic of deflagration (above 900°C) direct gas-phase reaction of NH_3 and $HClO_4$ would presumably be quite rapid. Thus, the above facts imply that gaseous oxidation-reduction reactions must occur at deflagration temperatures.

II. The rate of deflagration of pure ammonium perchlorate increases with increasing pressure, as [illegible] shows. This dependence upon pressure, as well as the existence of a lower-pressure limit of flammability, is evidence that gas-phase reactions are involved. The deflagration rate is of the order of one cm/sec at [illegible] atm. As was concluded in the discussion of the lower pressure limit, these small samples will not burn at atmospheric pressure because of [illegible]. However combustion is obtained when an incident radiant flux of [illegible] cal/sq cm sec is provided. This is at least comparable with the rate of radiant heat loss from the surface. If the deflagration were purely a surface and not a gas-phase phenomenon, one would expect the deflagration rate attained at atmospheric pressure with such an incident radiant flux to be of the order [illegible] the measured rate is 1/40 of this. It does not seem possible to account for this [illegible] difference in rate unless exothermic gas-phase reactions are involved. Finally, the dotted line in Figure 1 [illegible] slope of one, or a direct proportionality between burning rate and pressure. It is easily shown that this behavior corresponds to a second-order gas-phase reaction.

VII. The surface temperatures deduced from intensity of surface-emitted radiation [illegible] the surface temperature is below the final gas temperature.

These arguments, taken together, seem convincing enough to provide a good basis for a model [illegible] heat generation in the gas phase. The partial development of such a model has been described in Appendix B.

The next problem to be considered [illegible] whether the mechanism of the vaporization process during deflagration [illegible] properly be described as a thermodynamic or a kinetic [illegible] equilibrium exists between solid and vapor (if [illegible]), the rate of condensation being nearly as large as the rate of [illegible], and the net rate of volatilization being determined by the rate of supply of heat. The kinetic mechanism is based upon [illegible] as fast as it can at the prevailing surface temperature, and thus being rate-limiting. In the latter case, the rate would be determined by the surface temperature which in turn would be governed by a heat balance. In both cases, then, the endothermic [illegible] rate is ultimately limited by the rate of heat supply. If external radiant energy were added to the burning surface, the rate of sublimation would increase in either case, but the surface temperature would remain [illegible] in the first case and rise in the second.

not consider the secondary effect whereby the added heat raises the flame temperature, hence the gaseous reaction rate, and ultimately the steepness of the temperature gradient and the conducted heat flux to the surface.

Another question of importance is the size of the hypothesized gaseous reaction zone. An estimate of its size may be made as follows: At [illegible] atm, when the deflagration rate is [illegible] cm/sec, it is readily calculated that a heat flux of [illegible] cal/sq cm sec is required to supply a sensible heat of [illegible] kcal/mole (assuming [illegible] rise) and a heat of sublimation of 56 kcal/mole for ammonium perchlorate. If the thermal conductivity of the gas just above the surface is 0.00016 cal/sq cm-sec-°K (estimated), then the temperature gradient in the gas phase required to produce this heat flux is [illegible] × 10[illegible] °K/cm. Since the total temperature rise in the gas phase cannot be larger than a few hundred degrees, the flame thickness must be of the order of [illegible]. For radiation-assisted deflagration at one atmosphere, when the deflagration rate is 0.0[illegible] cm/sec, the gaseous zone as estimated by the above procedure would be [illegible]/0.025 or 40 times as thick, which is still only a few microns.

DISCUSSION OF CATALYST EFFECTS:

The primary function of a catalyst such as copper chromite is believed to be the augmentation of the gaseous reaction rate in the zone just above the surface. This effect is strikingly shown in Figure 3, and is also evident in isothermal decomposition-rate experiments at 200-300°C (1[illegible]).

However, the catalyst seems to possess a dual function associated with its ability to absorb and emit radiant energy, which is particularly important when the catalyst concentration is low or when a large external radiant flux is present. The trends shown in Figures 2 and 5 have already been discussed from this viewpoint.

The apparent extreme thinness of the gaseous reaction zone which has already been discussed has a bearing on the mechanism by which catalyst particles are effective in augmenting the deflagration rate.

It seems reasonable that the primary effect of the catalyst particles must be to increase the rate of the rate-controlling gaseous reaction. If the reaction zone were substantially thicker than the catalyst particle diameter, the catalytic effect would occur as the gas-borne catalyst particles are carried through the zone. However, the catalyst particles, of the order of 10^{-3} cm diameter, are roughly 100 times as thick as the reaction zone at 100 atm, and hence would be effective while still partially embedded in the solid ammonium perchlorate. This would have two consequences. In the first place, the catalyst would move through the reaction zone at the solid velocity rather than the much higher gas velocity, and thus the catalyst would have a longer time to act. In the second place, the augmented heat-release rate around each catalyst particle would cause local pockets to be burned into the perchlorate which later flatten out, the over-all higher deflagration rate being associated with this process. This deduction suggests that needle-like catalyst particles oriented normal to the burning surface should be highly effective.

The "worm-holing" mechanism by which catalysts may act when a sufficient radiant flux is present should also be discussed. In this mechanism, a catalyst particle below the surface is heated by radiant energy. It is certainly possible that such effects may be important for propellants with very high flame temperatures and corresponding intense radiation emission. However, several arguments may be put forth which suggest that there is no need to invoke this effect in explaining presently described experiments:

1. When catalyzed ammonium perchlorate deflagrates at elevated pressure without externally added radiation, the products are not very hot (930°C), so the radiant flux emitted would be relatively low. Yet the deflagration rate, of the order of five cm/sec, is comparable to that of energetic propellants in which ammonium perchlorate is the oxidizer, and which have far higher flame temperatures and correspondingly more intense radiation fluxes. Hence, radiation cannot be too important as a rate-controlling process in elevated-pressure deflagration.

II. The burning rates of ammonium perchlorate-based propellants are at least roughly the same in large rockets and in small strand burners, although the radiation environment is grossly different.

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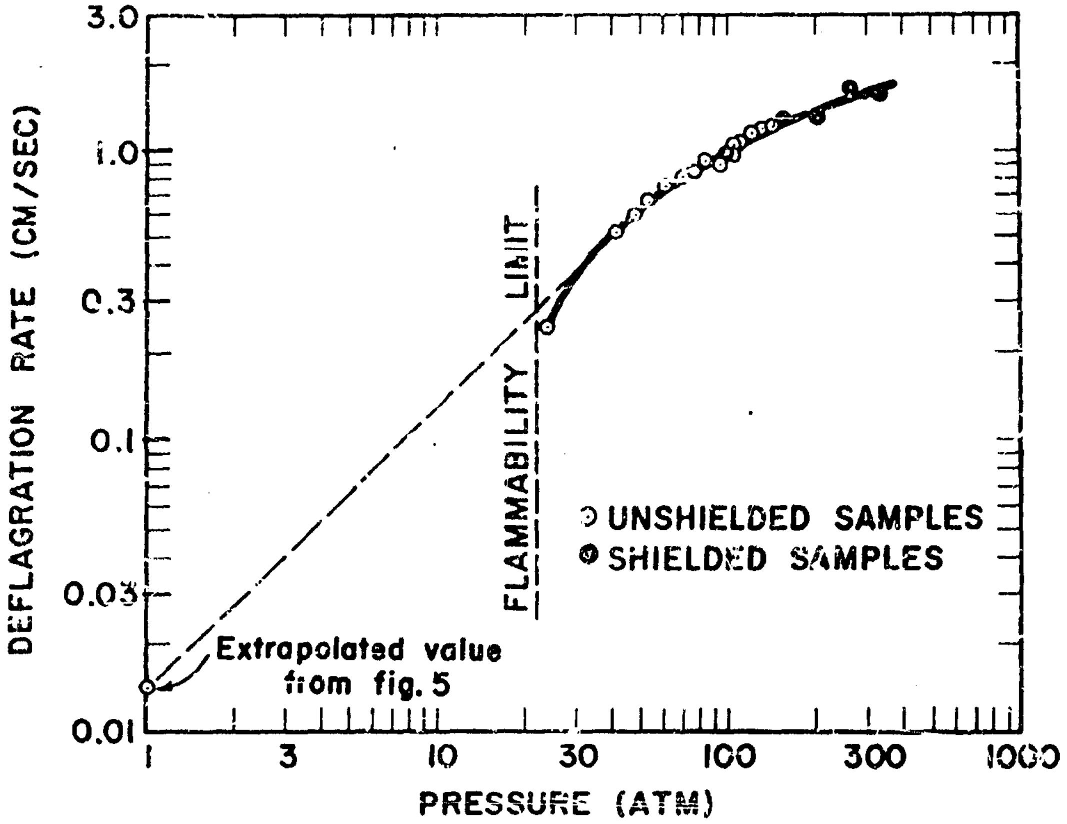


FIG. 1 VARIATION OF DEFLAGRATION RATE OF PURE AMMONIUM PERCHLORATE WITH PRESSURE, AT 25°C INITIAL TEMPERATURE

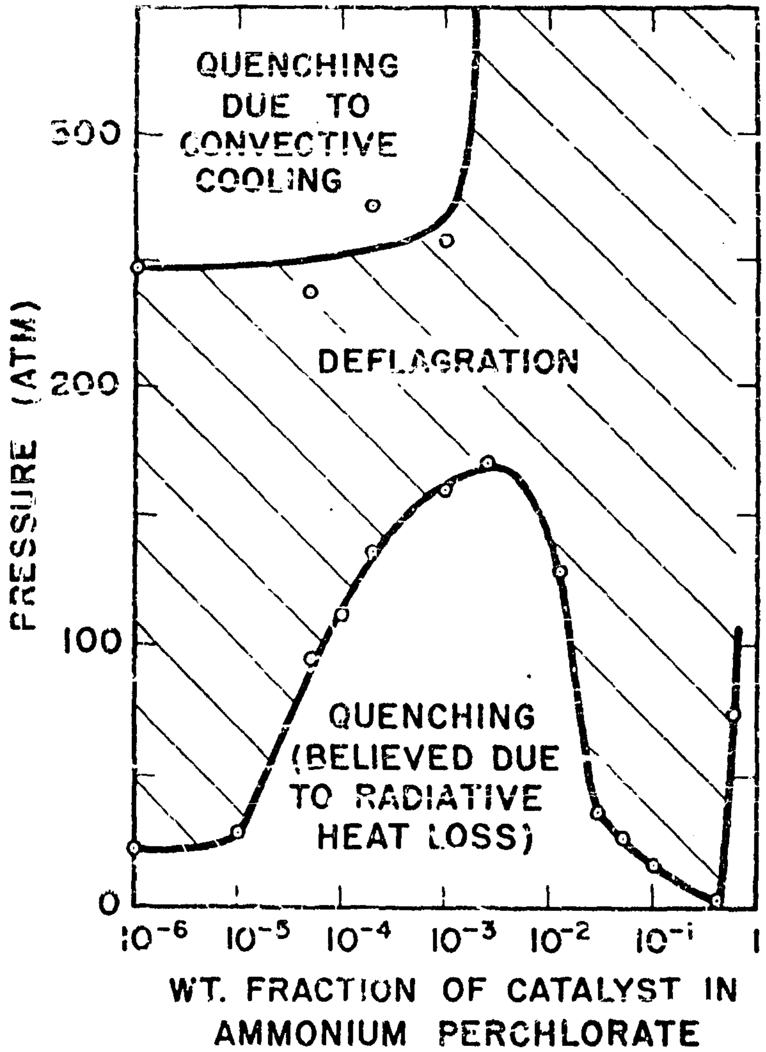


FIG. 2 EFFECT OF COPPER CHROMITE CONCENTRATION ON PRESSURE LIMITS OF DEFLAGRATION OF AMMONIUM PERCHLORATE INITIALLY AT 25° C.

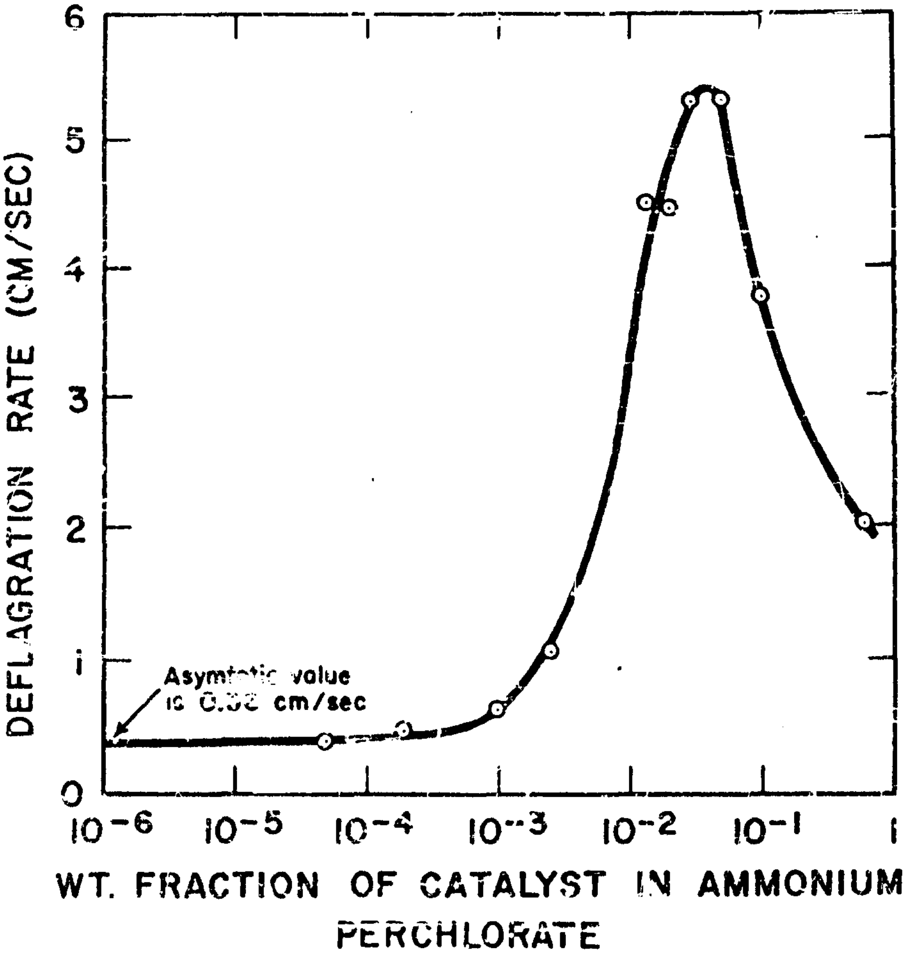


FIG. 3 EFFECT OF COPPER CHROMITE CONCENTRATION ON AMMONIUM PERCHLORATE DEFLAGRATION RATE AT 204 ATM AND 25°C AMBIENT CONDITIONS

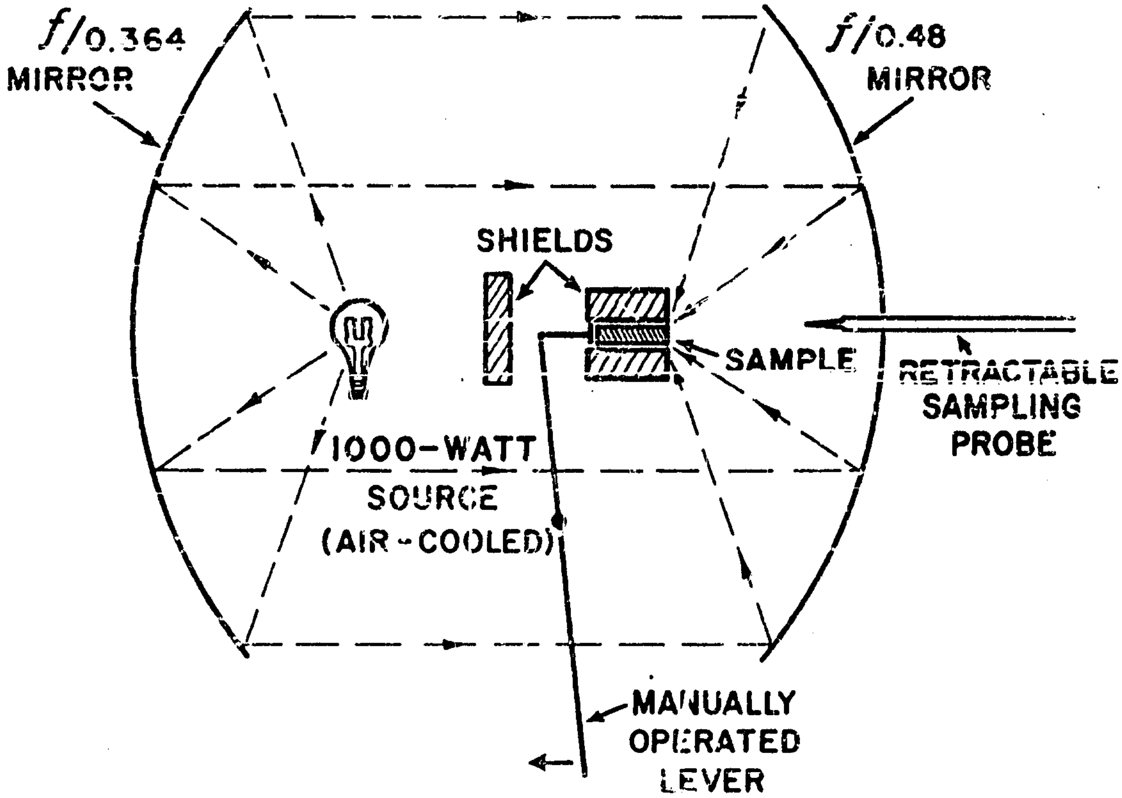


FIG. 4 APPARATUS FOR STUDYING EFFECT OF IRRADIATION ON DEFLAGRATION

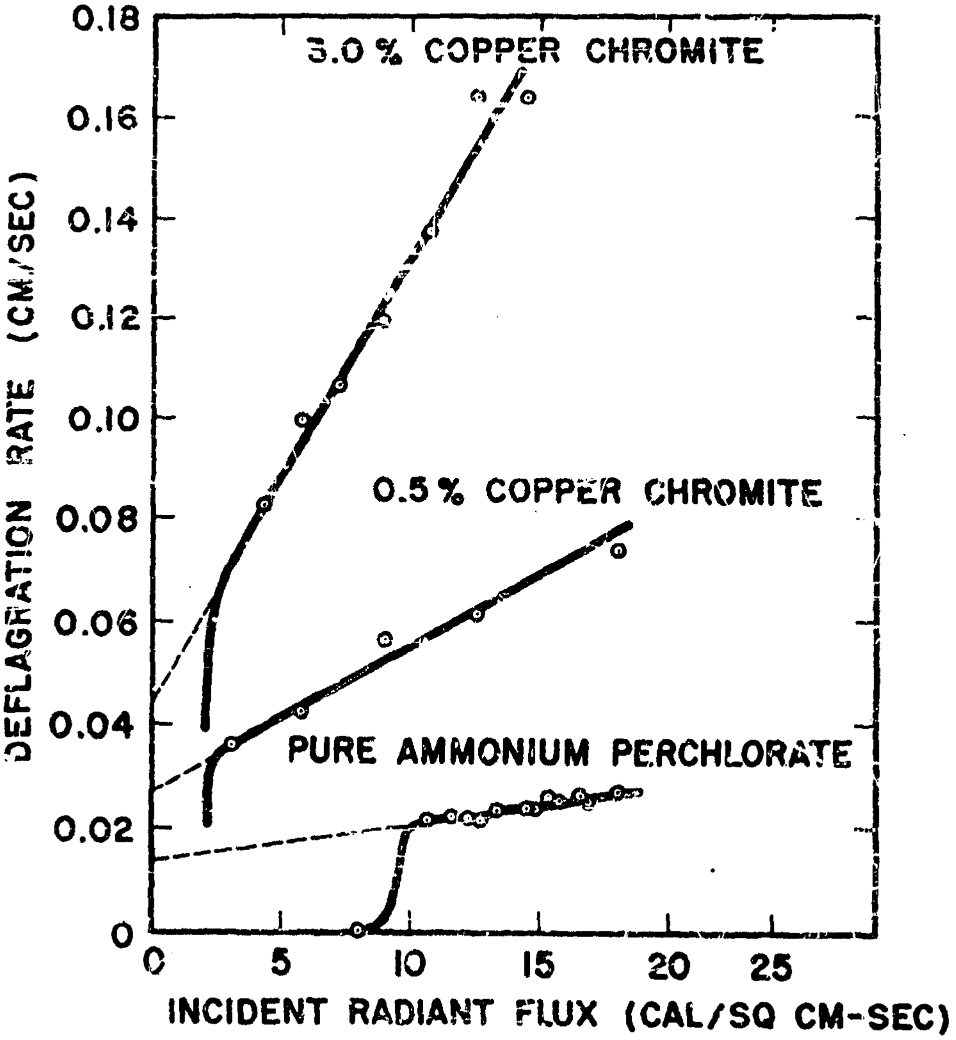


FIG. 5 EFFECT OF INCIDENT RADIANT FLUX INTENSITY ON DEFLAGRATION RATE OF PURE AND CATALYZED AMMONIUM PERCHLORATE AT ONE ATMOSPHERE

APPENDIX A

Details of Surface-Temperature Measurements and Calculations

I. Film Densitometry

On September 13, 1957, a series of seven snapshots of a reference body at various temperatures and two snapshots of burning strands were taken and developed together. Densitometric measurements of the reference body are tabulated:

T (°C)	Density (arbitrary units)
650	0.62
700	1.40
750	2.15
800	2.34
850	2.47
900	2.55
950	2.70

(The reference body is a stainless steel tube heated by electrical resistance, the outer surface being coated with Bureau of Standards Ceramic Glaze A-418. The temperature was measured with a chromel-alumel couple within the tube. A check on the validity of this measurement was obtained by applying a material of known melting point (816°C) to the outside of the tube.)

The two strand photographs taken together with the above calibration were found to have densities of 1.65 and 1.80, corresponding to apparent temperatures of 715°C and 725°C. (These would be true temperatures only if the emissivities were the same for the reference body and the strand surface.) All photographic conditions were the same for the calibration and the deflagration photographs except that in one case there was air at atmospheric pressure in the optical path and in the other case, nitrogen at 500 psi and combustion products.

In a second series of photographs on September 17, 1957, consisting of three calibrations and two deflagration photographs, calibration values were:

T (°C)	Density (arbitrary units)
700	1.20
750	1.50
800	1.90

(These values are seen to differ slightly from the earlier calibration.) The two strand densities were each 1.60, corresponding to apparent temperatures of 730 and 730°C.

II. Filter and Film Characteristics

Kodak supplies the following data for spectral sensitivity of their high-speed infrared film:

Wavelength (microns)	Log sensitivity
0.65	+ 0.60
0.70	+ 0.75
0.75	+ 0.80
0.80	+ 0.95
0.85	+ 0.90
0.90	+ 0.10
0.93	- 1.00
0.97	- 2.00

Thus the sensitivity is roughly a thousand times as great at 0.80 microns as at 0.97 microns.

The Corning 7-69 filter which was used transmitted as follows, according to Corning:

Wavelength (microns)	Transmittance (%)
0.20 - 0.70	0
0.71	0 +
0.72	6
0.73	25

0.74	45
0.75	65
0.80	80
0.85	80
0.90	71
0.95	57
1.00	35
1.05	15
1.10	5

III. Calculation of Upper-Limit Temperature from Apparent Temperature

Since the emissivity of the strand surface is unknown, one can try to estimate upper and lower limits of emissivity by comparison with other materials. Only the lower limit of emissivity may be estimated, however, since the upper limit in the spectral region of interest may be affected by chemiluminescence in an unpredictable way.

McAdams (2) lists the following total emissivities for nonmetals at 800°C (1470°F):

Nickel oxide	0.66	
Aluminum trioxide	0.27	
Cuprous oxide	0.66	
Magnesium oxide	0.22	
Iron oxide	0.87	
Thorium dioxide	0.22	
Silica	0.45	
Zirconium silicate	0.55	
Magnesite brick	0.38	(1832°F)
Carborundum	0.92	(1850°F)
Building brick	0.45	(1832°F)
Firebrick	0.75	(1832°F)
Carbon	0.53	(1900°F)

No value is below 0.2, so this is taken as the lower limit. It is assumed that these values for total emissivity are applicable to spectral emissivity in the vicinity of 0.8 microns.

The total emissivity of the ceramic coating on the reference body has been measured by De Corso and Coit (ASME Trans., 77, 1189 (1955)), as 0.89 at 725°C. The assumption will be made that this is a grey body.

The following expression, which follows at once from Planck's law, was used to calculate the temperature of the perchlorate surface from the foregoing data:

$$\epsilon\lambda_x / \left[\exp\ (1.439/\lambda T_x) - 1 \right] = \epsilon\lambda_R / \left[\exp\ (1.439\lambda T_R) - 1 \right]$$

Here, $\epsilon\lambda$ is spectral emissivity, λ is wave-length (cm), T is temperature (°K) and subscripts x and R refer to unknown and reference bodies. As discussed, $\epsilon\lambda_x$ is taken as 0.2 and $\epsilon\lambda_R$ as 0.89. T_R is taken as 998°K (725°C), the average of the above-described four data points. The wave-length λ is taken as 8×10^{-5} cm (0.8 micron) on the basis of the filter and film cut-off characteristics, shown above. (A calculation shows that a ten per cent error in choice of wavelength would cause only a one per cent error in temperature.)

With the above values, T_x comes out to be 816°C.

Measurements and calculations for other films and filters were handled similarly and will not be detailed here, since no quantitative conclusions were based on them.

APPENDIX B

A Mathematical Model for a Deflagrating Homogeneous Solid Propellant, Including an External Radiant Flux

The Model: It is assumed that a one-dimensional homogeneous solid (which might be ammonium perchlorate, as discussed in this report) sublimes at a surface, the vapor reacting exothermically above the surface with a resulting feedback of heat to the surface. Steady-state regression of the surface results.

This model would also be applicable to a liquid monopropellant which vaporizes and reacts exothermically in the vapor phase just above the surface.

When the gas-phase exothermic reaction for such a process cannot be described quantitatively from *a priori* knowledge, its rate may be described formally in some fashion with adjustable parameters. We choose to do this in the simplest conceivable way so as to permit straight-forward analytical solutions to the resulting differential equations describing the model. Let the reaction rate be zero below an ignition temperature T_i, which is greater than (or in a special case equal to) the surface temperature T_s. Above T_i the reaction is assumed to proceed at a constant rate q (cal/cu cm-sec) until the reactants are consumed.

With this model, reaction rate is independent of reactant concentration, so that molecular diffusion in the reaction zone need not be considered. Heat is conducted from the reaction zone, through the gaseous preheating zone, and to the surface. The heat flux across the interface is $\dot{m}Q$, where $\dot{m}$ is the burning rate (gm/sq cm-sec) and Q is the sum of the heat necessary to raise the solid from ambient temperature T_a to surface temperature T_s and the heat of sublimation. As an optional feature, an external radiant flux q_R (cal sq cm-sec) may be assumed to pass without absorption through the gases and be absorbed at the interface; a negative value for q_R would correspond to radiant heat loss from the surface to the surroundings, the gas immediately above the surface again being transparent. The sketch in Figure B1 summarizes the model.

The mathematical behavior of the model will first be developed for q_R equal to zero, and then for either positive or negative values of q_R.

Mathematical Relationships (No Radiation): We assume that the gas thermal conductivity λ and isobaric heat capacity C_p are independent of temperature. The coordinate system is chosen to move with the solid-gas interface, so that $T(x)$ is time-invariant. Accordingly, the differential equation expressing conservation of thermal energy in the reaction zone is

$$\lambda \frac{d^2T}{dx^2} - \dot{m} C_p \frac{dT}{dx} + q = 0 \qquad \text{(B1)}$$

The boundary conditions on the downstream side of the reaction zone are

$$T = T_f \text{ and } \frac{dT}{dx} = 0 \text{ when } x = x_f \qquad \text{(B2)}$$

Since λ is constant, equation (B1) is readily integrated with boundary condition (B2) to yield

$$T = T_f - \frac{q\lambda}{\dot{m}^2 C_p^2}\left[e^{\dot{m} C_p (x - x_f)/\lambda} + \frac{\dot{m} C_p}{\lambda}(x_f - x) - 1\right] \qquad \text{(B3)}$$

and

$$\frac{dT}{dx} = \frac{q}{\dot{m} C_p}\left[1 - e^{\dot{m} C_p (x - x_f)/\lambda}\right] \qquad \text{(B4)}$$

In the gaseous preheat zone (between T_s and T_i) the equation for conservation of energy is

$$\lambda \frac{d^2T}{dx^2} = \dot{m} C_p \frac{dT}{dx} \qquad \text{(B5)}$$

The boundary conditions on the upstream side of this zone are

$$T = T_s \text{ and } \lambda \frac{dT}{dx} = \dot{m} Q \text{ at } x = x_s \qquad \text{(B6)}$$

Integration of (B5) with boundary conditions (B6) yields

$$\frac{dT}{dx} = \left[Q + c_p(T - T_s) \right] \dot{m}/\lambda \tag{B7}$$

At the ignition plane $x = x_i$, where the gaseous preheat zone described by equation (B7) and the reaction zone described by equation (B4) meet, dT/dx must be continuous if a steady state exists. Hence, (B4) and (B7) may be equated at this plane, yielding

$$\left[Q + c_p(T_i - T_s) \right] \dot{m}^2 c_p/\lambda q = 1 - e^{-\dot{m} c_p(x_f - x_i)/\lambda} \tag{B8}$$

It is possible to simplify this equation by eliminating the quantity $x_f - x_i$ as follows. The over-all energy balance of the flame may be written

$$q(x_f - x_i) = \dot{m} \left[Q + c_p(T_f - T_s) \right] \tag{B9}$$

The reaction-zone thickness $x_f - x_i$ may be eliminated between equations (B8) and (B9). This introduces a new parameter T_f, of course, but this is readily evaluated in any specific case from thermodynamic considerations. The resulting equation can be expressed most compactly in dimensionless form:

$$\alpha(1 + \beta) = 1 - e^{-\alpha(1 + \gamma)} \tag{B10}$$

where

$\alpha = \dot{m}^2 c_p \, Q/\lambda q$

$\beta = c_p(T_i - T_s)/Q$

$\gamma = c_p(T_f - T_s)/Q$

Equation (B10) permits calculation of the burning rate $\dot{m}$ when the other parameters are known. Since equation (B10) is transcendental, it is not possible to express $\dot{m}$ explicitly as a function of the other quantities. However, an inspection of the dimensionless groups shows at once that

$$\dot{m} \sim (\lambda q)^{1/2} \tag{B11}$$

An exact quantitative description of the relations between α, β and γ is given in the following table, which might readily be extended by further computations:

$(1 + \gamma)/(1 + \beta)$	$\alpha(1 + \beta)$
1.00	0
1.05	0.1
1.11	0.2
1.19	0.3
1.28	0.4
1.39	0.5
1.53	0.6
1.72	0.7
2.01	0.8
2.56	0.9
∞	1.0

For the special case where γ is small compared with unity, equation (B10) may be expanded in good approximation according to

$$e^{-x} \doteq 1 - x + \frac{x^2}{2}$$

This yields the asympototic expression

$$\alpha \doteq 2(\gamma - \beta)/(1 + \gamma)^2 \quad \text{for } \gamma \ll 1 \tag{B12}$$

Mathematical Relationships (Radiation Included): An external radiant flux q_R (cal/sq cm-sec) is assumed to be incident on the solid-gas interface and to be entirely absorbed, without reflection or re-radiation. A negative value for q_R corresponds to radiant heat loss from the interface to infinity. Equations to describe the foregoing model with this additional feature will be developed.

Equations (B1), (B2), (B3), (B4), and (B5) are still valid. However, T_f will obviously be a function of q_R, according to the relation

$$c_p(T_f - T_s) = c_p(T_f - T_s)_{q_R = 0} + q_R/\dot{m} \tag{B13}$$

This is based on the assumption that Q is independent of q_R.

Instead of equation (B6), the boundary conditions at x_s become

$$T = T_s \text{ and } \lambda \frac{dT}{dx} + q_R = \dot{m}\, Q \tag{B14}$$

Integration of (B5) with boundary conditions (B14) gives

$$\frac{dT}{dx} = \frac{\dot{m}}{\lambda}\left[C_p(T - T_s) + Q - \frac{q_R}{\dot{m}}\right] \tag{B15}$$

On equating (B4) and (B15), one obtains

$$1 - e^{-\dot{m}\, C_p(x_f - x_i)/\lambda} = \left[Q + C_p(T_i - T_s) - \frac{q_R}{\dot{m}}\right] \dot{m}^2 C_p/q\lambda \tag{B16}$$

The over-all energy balance now is

$$q(x_f - x_i) + q_R = \dot{m}\left[Q + C_p(T_f - T_s)\right] \tag{B17}$$

One can eliminate T_f between (B13) and (B17), and then eliminate $(x_f - x_i)$ between the resulting equation and (B16), finally to obtain

$$\alpha(1 + \beta - \sigma) = 1 - e^{-\alpha(1 + \gamma)} \tag{B18}$$

where $\gamma = C_p(T_f - T_s)_{q_R=0}/Q$

and $\sigma = q_R/\dot{m}\, Q$

Equation (B18) is seen to be identical to equation (B10) when β in (B10) is replaced by β-σ, and the previously tabulated solutions are valid on this basis. It should be noted that α contains q, which one would expect to be a function of $(T_i + T_f)/2$, while T_f in turn is a function of q_R, given by equation (B13). It is not appropriate here to speculate on the functional relation between reaction rate q and temperature, but obviously any desired function may be introduced.

To summarize, we are essentially taking T_f and q to vary with q_R, while T_i, T_s, Q, λ, and C_p are assumed independent of q_R. This choice is purely arbitrary; the other quantities could obviously be expressed as functions of q_R if one wished, at the cost of increasing complexity.

A qualitative understanding of the effect of the radiation parameter σ may be obtained from Figure B2, which is a plot of α vs (β − σ), for parametric values of γ from zero to infinity. It is seen that the burning-velocity parameter α increases with increasing σ and decreases to zero for sufficiently large negative σ, corresponding to extinguishment. The mathematical condition for extinguishment is

$$\gamma \leq \beta - \sigma \qquad \text{(B19)}$$

Physically, this means that the radiant heat loss (negative σ) is sufficient to reduce T_f below T_i.

Approximate relation (B12), valid for small γ, now becomes

$$\alpha \doteq 2(\sigma + \gamma - \beta)/(1 + \gamma)^2 \qquad \gamma \ll 1 \text{ and } \sigma \ll 1 \qquad \text{(B20)}$$

It follows that

$$\left(\frac{\partial \alpha}{\partial \sigma}\right)_{\gamma, \beta} \doteq \frac{2}{(1 + \gamma)^2} \doteq 2 \qquad \gamma \ll 1 \text{ and } \sigma \ll 1 \qquad \text{(B21)}$$

Approximate relation (B21) is interesting in that it might be tested experimentally if the variation of q with q_R were known.

Nomenclature

$\dot{m}$ = burning rate (gm/sq cm-sec)
C_p = specific heat of gas at constant pressure (cal/gm-°C)
Q = heat of sublimation plus sensible heat needed to raise solid from ambient to surface temperature (cal/gm)
q_R = radiant flux to or from surface (cal/sq cm-sec)
q = rate of gaseous exothermic reaction (cal/cu cm-sec)
λ = thermal conductivity of gas (cal/cm-sec-°C)
x = distance
T_s = surface temperature (°C)
T_i = ignition temperature (°C)
T_f = final flame temperature (°C)
α, β, σ, γ = dimensionless groups

Figure B1. Model of Solid-propellant Flame.

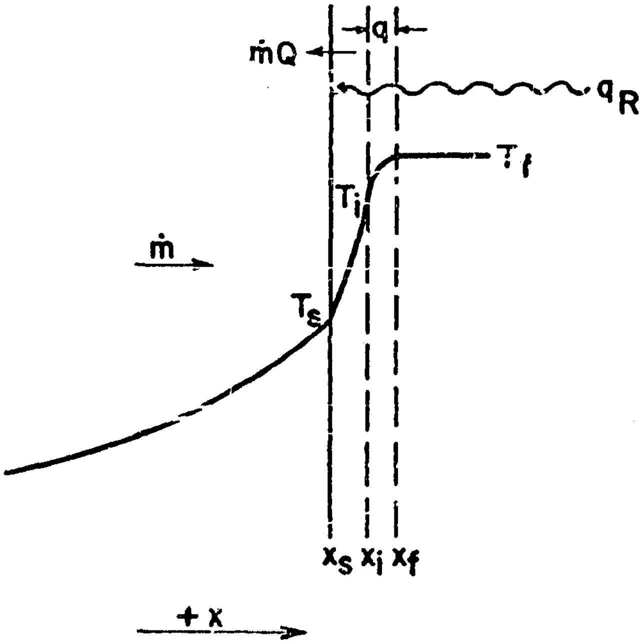
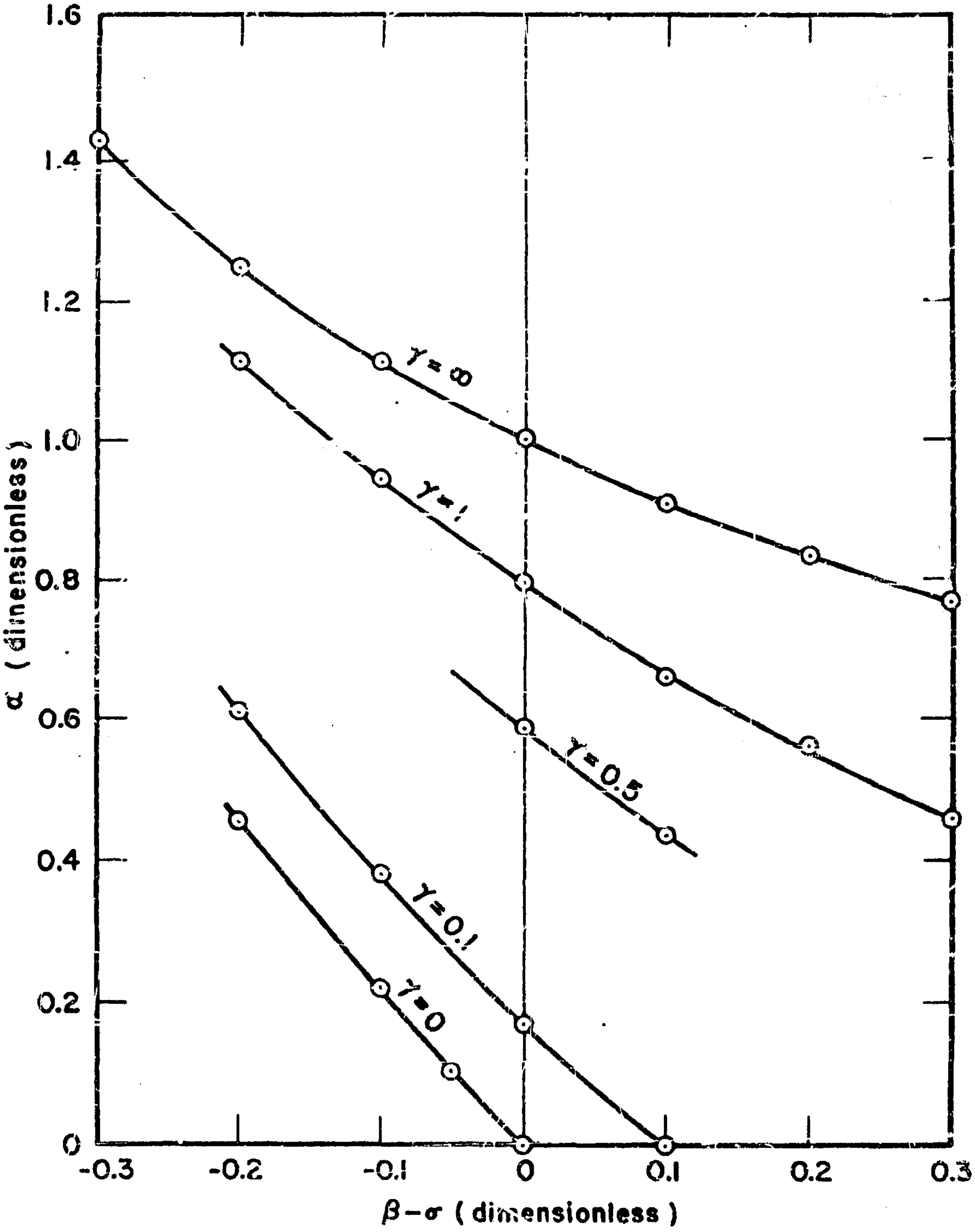


Figure B2. Numerical solutions of equation (B18).



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