Radiative Ignition of Double Base Propellants: II. Pre-ignition Events and Source Effects

L. DeLuca,* T. J. Ohlemiller,† L. H. Caveny,‡ and M. Summerfield§
Princeton University, Princeton, N. J.

In this second paper of a two-part study, emphasis is on the pressure dependent pre-ignition events in double base propellants and the influence of radiation source (arc image vs laser) on observed ignition behavior. The pressure-dependent (<21 atm) ignition domain of a PNC/MTN double base propellant is examined using controlled exposure lengths together with high speed movies and an infrared detector to monitor flame development. There is a brief flux-dependent period of transient flame development after gasification begins; this is followed by a relatively long, flux-dependent period of steady-state, radiation assisted burning before a self-sustaining condition is reached. The nature of this condition for self-sustainment is not yet well-defined. The ignition behavior seen with an arc furnace or a laser is generally quite close for both double base and composite propellants, if optical effects (reflection, penetration) are factored out. An exception is fast deradiation extinction, seen only with the laser; greater radiation penetration in the arc image wavelength region precludes this phenomenon.

I. Introduction

As pointed out in the companion paper,[1] thermal radiation is a convenient energy input for studying solid propellant ignition behavior. By varying such factors as radiant flux, ambient pressure, and propellant compositions, much can be learned about the processes underlying the macroscopic ignition response, e.g., which of these factors controls that behavior in differing ambient conditions and what is needed to predict ignitability quantitatively in new circumstances. At the same time, one must be aware that the unique nature of radiation and the characteristics of the devices used to produce it can affect the test results. The present study is a systematic survey of radiation ignition behavior of a variety of propellants, intended to illustrate both these positive and negative aspects.

In the companion paper,[1] a generalized ignition behavior "map" (log of irradiation time vs log of radiant flux) was presented. The boundaries on this map were discerned by go/no-go testing and by monitoring of IR radiation from the developing flame. For clarity, we summarize them again here in the order they are encountered as irradiation time increases: L_{1a} = first gas evolution; L_{1b} = first IR signal from the surface region indicating the beginning of exothermicity; L_{1c} = incipient flame indicated by a roughly 50-fold increase in IR signal; L_{1d} = self-sustained flame indicated by steady burning if flux is removed; L_2 = deradiation extinction boundary indicated by disappearance of a well-developed flame when radiation is removed too quickly.

The companion paper[1] concentrated on certain propellant formulation effects; it illustrated how these boundaries are shifted strongly by the optical properties of the propellants. This was shown to obscure the comparison of different types of propellants, but unmodified double base propellants clearly were more ignitable than the AP or HMX composites examined. The HMX composites had a much more pronounced separation of L_{1a} and L_{1d} boundaries (because of the pressure dependence of the latter) than did the *AP* composites examined. Catalysts added to double base propellants have no clear effect on their L_{1a} boundary but have a pronounced effect on L_{1d}.

The present paper is focused both on clarification of events occurring during ignition and on the effects that the radiation source has on the overall results. Both an arc image furnace and a laser ignition apparatus were used; they are described in Ref. 1. The compositions of the various propellants also are given in Ref. 1.

II. Observations of Flame Development in Double Base Propellants

The laser ignition apparatus is adapted best to the study of the processes leading up to the crossing of the L_{1d} (self-sustained ignition) boundary; it permits visual observation (via high speed movies) and IR emission monitoring (with a fast-response, gold-doped germanium detector described in Ref. 1). Such observations are difficult, if not impossible, in the arc image furnace because of scattered arc radiation.

A. Pressure Dependent L_{1d} Boundary

Before describing this study, however, it is of interest to compare the overall ignition behavior in the intermediate to low pressure range as seen with the arc image and the laser. Figure 1a is an ignition map for propellant 10 (catalyzed NC/TMETN double base), showing boundaries obtained in the two experimental setups. More detailed comparisons of results obtained with the laser and arc image will be made in Sec. III. For now, it is sufficient to note that the boundaries are qualitatively similar; separation of the L_{1a} (first gasification) and L_{1d} (self-sustained ignition) boundaries occurs in both setups at pressures below 21 atm, and the separation increases as pressure decreases. On the other hand, the absolute values of the separations depend on the apparatus.

This separation between L_{1a} and L_{1d} is found in various propellants but at differing conditions, particularly pressure. Similar behavior in other catalyzed double base propellants is seen in Fig. 4 of Ref. 1 and in Fig. 1b of this paper. The HMX composites in this study (Fig. 6b of Ref. 1) show this behavior

Presented as Paper 73-176 at the AIAA 11th Aerospace Sciences Meeting, Washington, D. C., Jan. 10-12, 1973; submitted May 19, 1975; revision received Jan. 5, 1976. This paper is based on research sponsored by the U.S. Army Research Office in Durham under Grant DA-ARO-D-31-124-72-G119 and monitored by the Ballistics Research Laboratories, Aberdeen Proving Ground, Md. The research was performed in the Aerospace and Mechanical Sciences Department of Princeton University.

Index category: Combustion Stability, Ignition, and Detonation.

*Graduate Student.

†Research Staff Member.

‡Senior Member of Professional Staff. Associate Fellow AIAA.

§Professor of Aerospace Propulsion. Fellow AIAA.

in the same pressure range as the catalyzed double base propellants (below 21 atm). The *AP* composites in this study (Fig. 6a in Ref. 1) do not show any pressure-dependent L_{Id} boundary down to 5 atm; it has been found by other investigators[2-4] at 1 atm. This behavior was not seen for noncatalyzed double base propellants in this study since their ignition response is dominated by other factors, to be discussed in Sec. III.

B. Photographs of the Developing Flame Zone

The process occurring between L_{Ia} and L_{Id} was called "flame development" in Ref. 1; this is a roughly descriptive title, but, as will be seen here, this process is complex and not well understood. A qualitative picture of events for a noncatalyzed double base propellant is shown in the first three photographs in Fig. 2a. (These are extracted from a high-speed movie, using a shadowgraph to bring out the gas flow patterns; a complete sequence can be found in Ref. 5.) Note that the pressure is sufficiently high enough (21 atm) so that this propellant will sustain ignition; the high pressure speeds flame development. The L_{Ia} boundary is crossed at 0.007 sec when gasification begins; this continues for a period of time, clearly becoming stronger by 0.016 sec. A complete flame, including the visible flame zone, first appears at 0.018 sec (not shown); this visible flame is seen as the bright area at the top of the frame at 0.034 sec. Certain precautions are necessary in interpreting these photographs. The visible flame zone is probably not at all important as a source of heat feedback to the propellant surface under these conditions;[6] it is too far away. The fizz zone, much closer to the surface and nonemitting, is the important gas phase reaction zone.[6] It seems reasonable, however, to take the appearance of the visible flame zone as a rough signal of strong development of the fizz zone, since the two are coupled in sequential order. (Note, however, that the visible flame is more subject to three-dimensional cooling.) Then one can conclude from Fig. 2a that a substantial amount of time elapses between onset of gasification and full development of the important gas phase reactions. Figure 2b will be discussed at the end of Sec. II.E.

C. IR Signal from the Developing Flame

Further evidence on this evolution of the gas phase reaction zone comes from the IR detector monitoring the emissions just above the surface. Results for propellant 6 (noncatalyzed double base) obtained within the laser apparatus are shown in Fig. 3a. A faint signal is that which first is discernible above the baseline; a strong signal is about 50 times larger than this. Comparison of the time of onset of the faint IR signal with first gasification (L_{Ia}), as seen in high-speed movies, shows that the two are essentially coincident. The strong signal cannot be given such an absolute meaning, except to say that, for a noncatalyzed double base propellant, it is near the maximum and, therefore, corresponds to a rather well-developed flame zone.

The faint signal line essentially coincides with the L_{Ia} (first gasification) boundary (compare with lowest line in Fig. 6a) for propellant 6. The faint signal line departs somewhat from L_{Ia} at high fluxes; this may be the result of localized gasification at image hot spots which are not as well smoothed by conduction at high fluxes. Such pinpoint gasification areas may not emit a sufficient IR signal to be picked up by the detector. This faint signal line is independent of pressure, as shown in Fig. 3b for propellant 10 (catalyzed double base); this is what one would expect if it corresponds to first gasification which is the result of condensed phase, pressure independent, irreversible reactions. It is also independent of the substitution of air for nitrogen, which indicates that the external oxidizer does not participate significantly in the initial gasification process.

The strong signal lines for the noncatalyzed propellants clearly depend on the state of the gas phase (Fig. 3a); in-

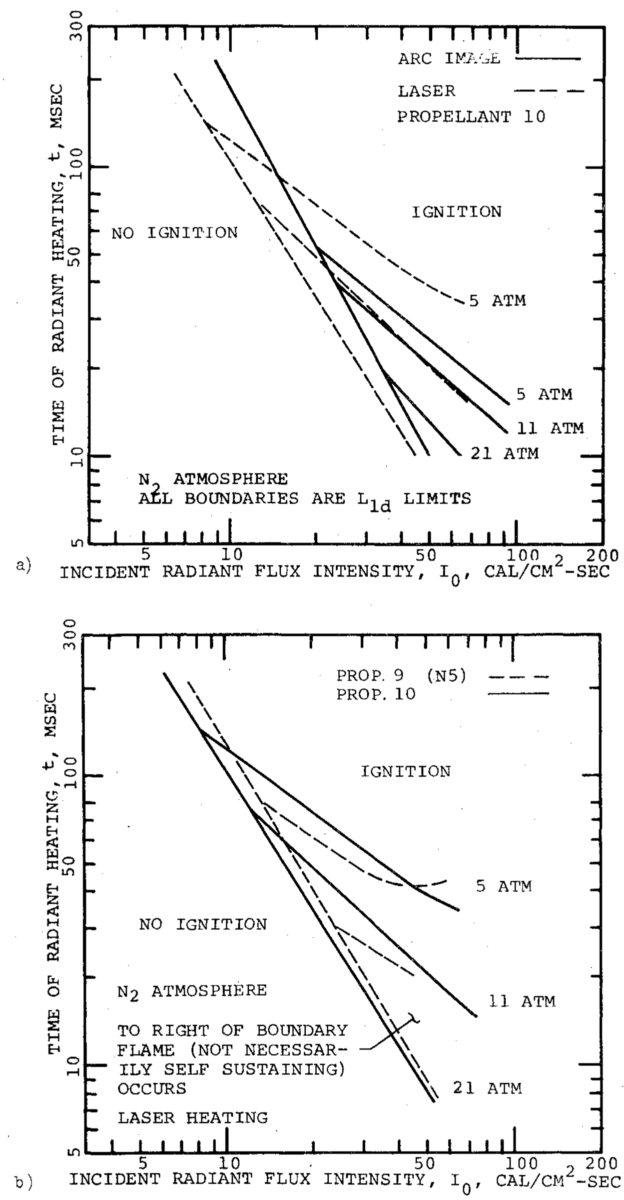

Fig. 1 a) Catalyzed DB propellant 10 tested in the arc image and laser ignition apparatus showing that pressure sensitivity is characteristic of the propellant not the ignition apparatus; and b) Catalyzed DB propellants 9 and 10 tested in the laser ignition apparatus showing that pressure dependence of ignition boundaries is a property of catalyzed DB propellants.

creased pressure or substitution of air for nitrogen shortens the delay between a faint and a strong IR signal by shifting the latter to earlier times. The roughly parallel nature of the faint and strong signal lines for propellants 6 and 10 in nitrogen suggests that the delay between the lines corresponds to a nearly constant step upward in surface temperature. Note that this step upward is accomplished in much less time at high fluxes than it is at low fluxes. The behavior of the strong IR signal lines in relation to the weak signal lines as flux varies is basically what one would expect of a transient ablation process; the time to approach a steady ablation process varies inversely with the external radiant flux.

D. Gasification Rate Between L_{Ia} and L_{Id}

Various pieces of evidence indicate that the gasification between L_{Ia} and L_{Id} is not simply a passive ablation, driven only by the radiant flux; it is assisted also by the developing flame. The decreased delay between L_{Ia} and L_{Ic} (strong IR signal) with increasing pressure indicates that a gas phase

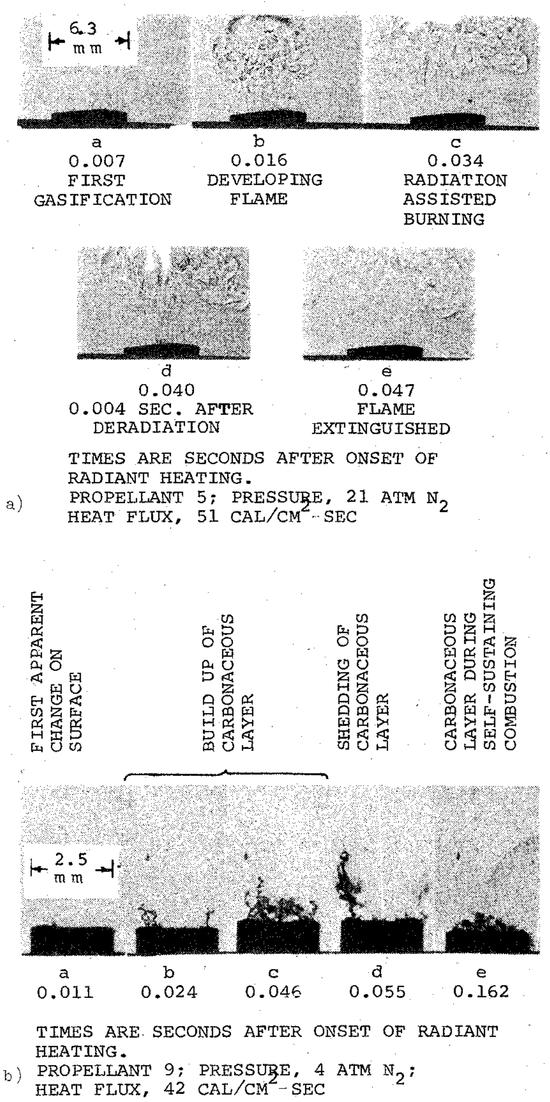

Fig. 2 a) High-speed shadowgraph movie illustrating flame development on noncatalyzed DB propellant; note that visible flame in c) and d) has a large standoff distance. Sample diameter is larger than the 2.5-mm size used elsewhere in this study; and b) High-speed shadowgraph movie showing carbonaceous layer formation on the surface of catalyzed DB propellant.

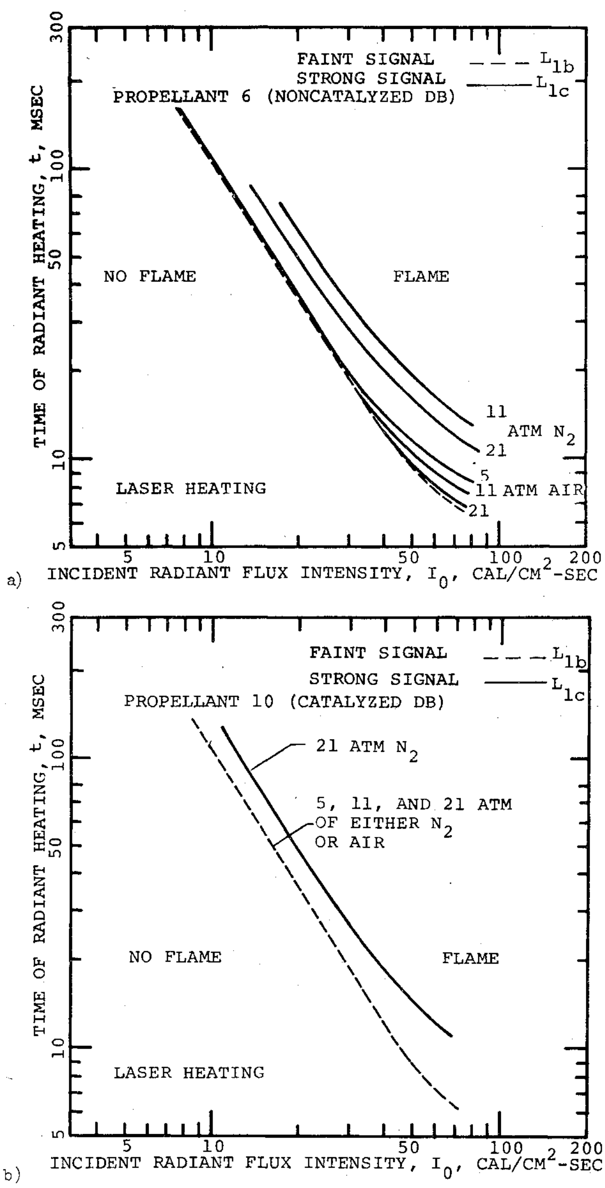

Fig. 3 a) Pressure dependence of strong surface region IR signal for noncatalyzed DB propellant 6. (Weak signal is independent of pressure and atmosphere.); and b) Pressure independence of initial surface region IR signal for catalyzed DB propellant 10.

reaction process is occurring (this also could be partly because of increased optical thickness of the gases above the surface as pressure increases). The decreased delay with air also points to an accelerated gas phase reaction development.

More definitive still are the data shown in Fig. 4. This is a record of the thickness of propellant gasified as a function of total exposure time to the radiation; the propellant is the same as that in Fig. 3b, and the flux is fixed at one level. The results in Fig. 4 provide a record of the gasification history seen as one moves along a vertical line (at 37 cal/cm^2-sec) in Fig. 1. The L_{Ia} (first gasification) boundary is crossed at about 13 msec. The rate of gasification thereafter depends on the ambient pressure; it is substantially higher than 11 atm than at 5 atm. In both cases there is a brief transient growth of the gasification rate and then it reaches a steady-state rate dependent on pressure.

Table 1 puts these steady gasification rates into a more informative context. Note first that they agree well with rates obtained in another radiation-assisted burning experiment using the arc image furnace rather than the laser.[7] That experiment was a steady burning experiment in which the arc radiation supplemented the heat feedback from a fully developed flame. The implication of the agreement between

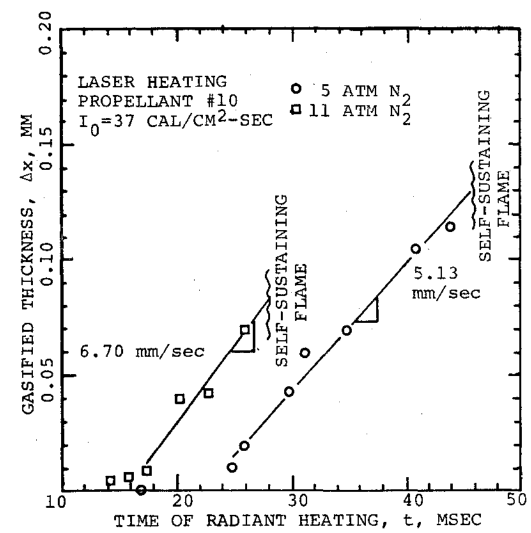

Fig. 4 Pre-ignition surface regression of a catalyzed DB propellant subjected to laser radiation showing occurrence of stationary gasification process prior to the self-sustained flame.

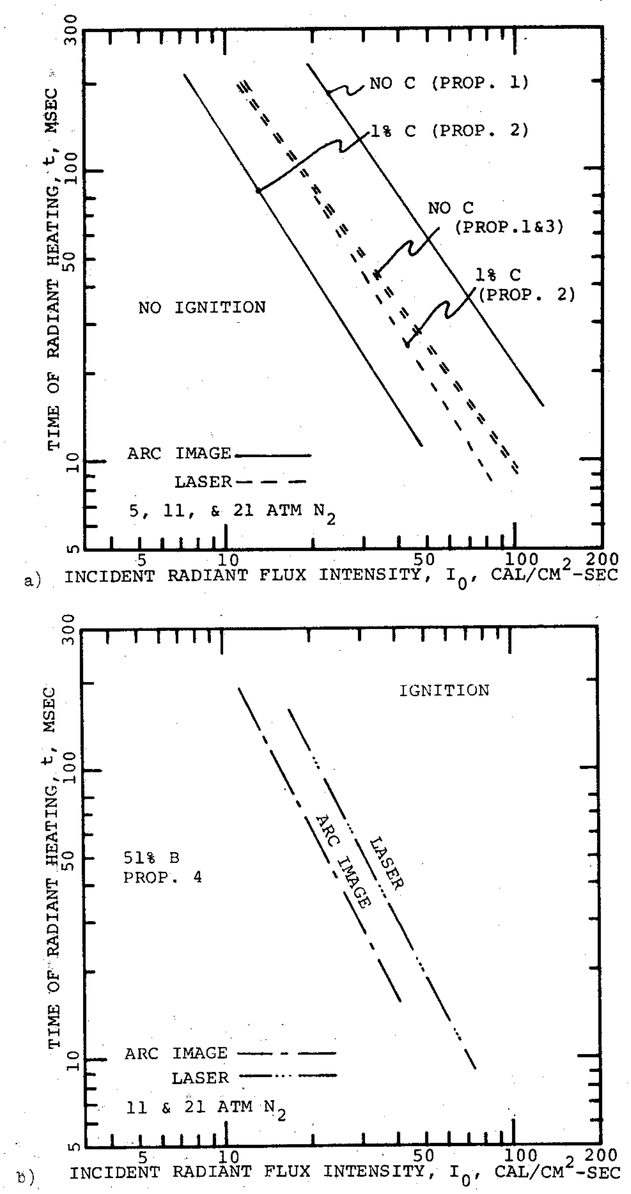

Fig. 5 a) Ignition of AP composite propellants 1, 2, and 3 demonstrating dependence on radiation source and independence of pressure (contrast with Fig. 1). (Absorbed laser radiation is affected only slightly by carbon powder added to the propellant.); and b) Effect of radiation source on a nearly opaque highly metallized propellant.

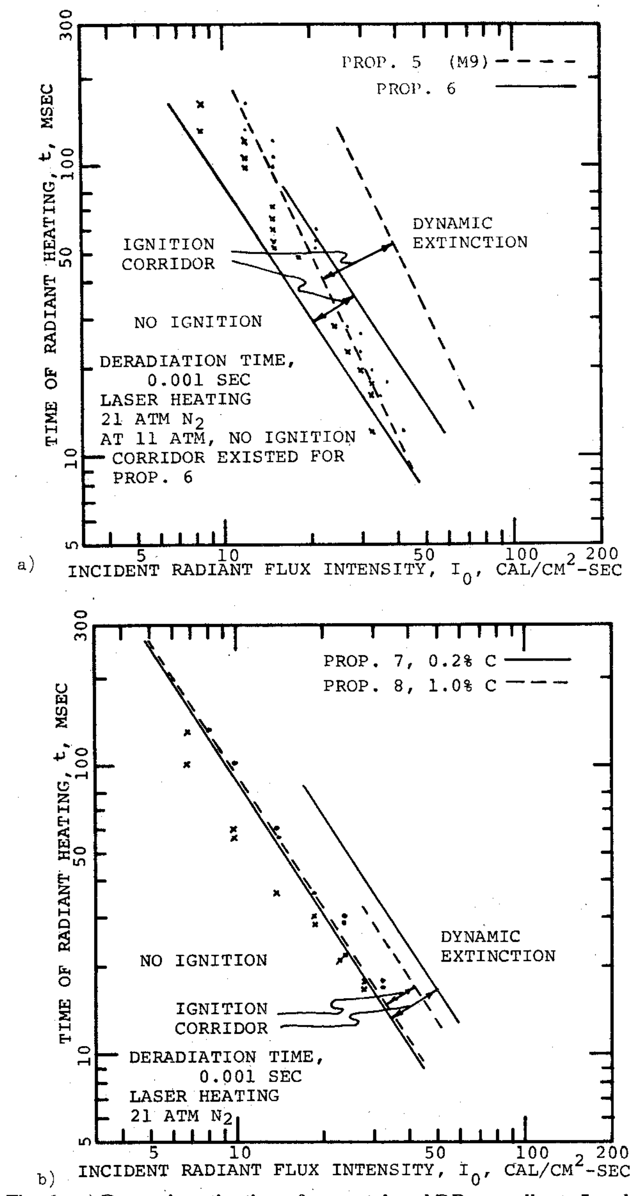

Fig. 6 a) Dynamic extinction of noncatalyzed DB propellants 5 and 6 tested in the laser ignition apparatus. (No such boundaries noted in arc image tests.); and b) Addition of carbon has only slight effect on dynamic extinction of noncatalyzed DB propellants 7 and 8.

the two is that the flame is equally well-developed during the steady-state portion of the gasification history.

Comparison of the present data with the standard strand burner rate measurements (no external radiation) indicates that the steady rates found in the present experiment are always higher. The ratio of laser-assisted rate to strand burner rate may be indicative of the relative importance of flame feedback and external radiation in yielding the observed laser gasification rate. For propellant 10 (catalyzed) at 5 atm the ratio is 3, and at 11 atm it is 2; the higher pressure has strengthened the flame and weakened the effect of the laser radiation. Comparison of this same ratio at 5 atm for propellant 10 and propellant 6 (noncatalyzed) gives 3 for the former and 3.25 for the latter. The external radiation has a somewhat greater effect on the noncatalyzed propellant, indicating that it has a weaker flame feedback at the same pressure; this is consistent with other studies of the effects of catalysts in double base propellants.[6]

E. Possible Explanations of the Delay Between L_{1a} and L_{1d}

The foregoing results, taken together, are somewhat paradoxical. They indicate that a fully developed, radiation assisted flame exists above the propellant surface after a relatively brief transient gasification and flame development process. Yet a much longer period of gasification is needed before the propellant can sustain ignition when the radiation is removed. For example, in Fig. 4, at 5 atm the propellant has reaches a steady state about 10 msec after it starts gasifying, but it must continue to gasify at this steady rate for another 20 msec before L_{1d} (self-sustaining ignition) is reached. Similar results were reported by Price[2] for JPN propellant (nitrocellulose/nitroglycerin) in an arc image furnace and by Kondrikov[8] for N-5 (catalyzed nitrocellulose/nitroglycerin) in the Princeton laser apparatus.

Price suggests that the pressure-dependent behavior separating L_{1a} and L_{1d} in double base propellants is a combination of flame development in the gas phase coupled with weakened heat release in the condensed phase; the latter is a hypothesized result of driving normal condensed phase reactants into the gas phase when external radiation forces an excessive degradation rate in the solid. Some of Kondrikov's results support this hypothesis qualitatively. These mechanisms are plausible as an explanation of the transient portion of the gasification history, but they seem to offer no reason for the required long period of steady gasification after the flame has developed.

Kondrikov suggests that this further delay is required to build up a radiation-attenuating layer of combustion products (see also Librovich[9] and Baer and Ryan[10]). In this model (derived from Zeldovich[11]) the flux must be reduced to a level no greater than that fed back to the solid by the normal steady flame at that pressure. Strictly speaking, then, this calls for total attenuation of the radiation before self-sustainment is possible. The evidence for attenuation of 10.6-μ laser radiation by double base propellant combustion products is ambiguous.[5,8] Mihlfeith et al.[12] measured attenuations of between 30 and 80% during arc image ignition of AP composite propellants. Regardless of this, it is difficult to see how attentuation of the laser radiation can balance exactly changes in flame structure (relaxation of Price's distended surface reaction zone) to yield a steady rate of gasification as seen in Fig. 4.

The exact nature of the processes occurring between L_{Ia} and L_{Id} for double base propellants (and also for the *AP* and HMX composites discussed in Ref. 1) is not yet well enough defined. It is not possible to say exactly what condition is fulfilled as L_{Id} is crossed, and self-sustained ignition becomes possible. Further experimental work is needed on the question of radiation attenuation and, perhaps, the three-dimensional aspects of ignition in a real experimental apparatus. These and possibly other factors may account for the substantial differences in the pressure-dependent L_{Ia}-L_{Id} separation seen in Fig. 1 for the laser and the arc image.

An apparently carbonaceous layer (Fig. 2b) is observed when catalyzed double base propellants are ignited at low pressure (~4 atm). This never is observed for *AP* composite or noncatalyzed double base propellants; also, this is less pronounced when catalyzed double base propellants are ignited at high pressure (21 atm). Formation, growth, and emission of carbonaceous filaments on the surface occurring prior to the time of self-sustaining combustion, appear to indicate a strong solid phase activity promoted by the catalysts. In all observed cases, the apparently carbonaceous layer is a necessary precursor for self-sustaining combustion.

III. Effects of Radiation Source on Observed Ignition Behavior

A. Effects on the L_I Boundary

As indicated in Ref. 1, neither the laser nor the arc image apparatus provides an ideal energy input. This ideal would be a uniform flux extending over an area of the sample that is much larger than both the thermal wave in the solid and the flame thickness in the gas. Furthermore, this flux would be absorbed totally at the surface and would induce exactly the same chemical response in the solid and gas phases as one gets in real applications of the material being tested.

Both sets of experimental apparatus deviate from the ideal for essentially the same reasons but to differing degrees for any particular reason. Both provide somewhat nonuniform fluxes over the irradiated area. With the arc this nonuniformity is a monotonic drop from a central peak; with the laser it is more gridlike as a result of a complex interference pattern. (More details can be found in Ref. 1). In both setups, the irradiated area is much larger than the thermal wave in the solid, but it may become comparable to the flame thickness in the gas if the pressure is low. (This depends on the propellant, but 1 atm is probably borderline for many.) In both, the ambient gas is cold. This is not the case in many propellant applications and the cold gas undoubtedly lengthens the pressure dependent separation between L_{Ia} and L_{Id}, as discussed in the previous section. This effect should be about the same for both setups since the sample sizes (and therefore the three-dimensional cooling effects) are the same. (This will be less true at high fluxes where the differing flux patterns on the same size sample face will tend to yield different gasification patterns and cooling effects in the gas.)

The two radiation sources fall in differing spectral regions: the xenon arc, visible and near infrared; the laser, one far IR wavelength, 10.6 μ. All propellants of interest have nonzero reflectivities and noninfinite extinction coefficients in both these spectral regions, but these nonidealities are generally worse for the arc radiation. The effects of these differing optical properties on the apparent ignition behavior of the *AP* composites described in Ref. 1 are shown in Fig. 5. The solid lines are L_I limits (first gasification and self-sustained ignition coincide) obtained in the arc apparatus; the dashed lines were obtained with the laser. As indicated in Ref. 1, a shift to shorter ignition delays, with no change in slope, indicates a change in reflectivity; a shift toward shorter delays, with an increase in slope, indicates an increase in extinction coefficient, with a possible simultaneous reduction in reflectivity. The available data on the optical properties of the propellants used in this study are summarized in Ref. 16.

Figure 5a shows that the apparent ignition behavior of the *AP* composite propellants depends on the radiation source. With the laser, propellants 1-3 give coincident L_I lines. (Note, propellants 1-3 have burning rates differing by 10% or less over the pressure range of interest.) Thus, one would conclude from this that carbon addition or changes in *AP* particle size do not affect ignitability. Whereas this is true for the two propellants shown here, one should be aware that, over a wider range of variables, effects of both carbon addition and *AP* particle size can be expected and they interact. In this heterogeneous propellant, the carbon goes only into the binder and will affect the propellant extinction coefficient only when it is less than that of *AP* crystals. Here, the limiting extinction coefficient appears to be that of the *AP* since it does not respond to carbon.

The arc image data do in fact reveal a different situation for the same propellants, because the *AP* extinction coefficient is less limiting in the range examined. The L_I line for propellant 1 appears to indicate that it is much less ignitable than propellant 2, which is identical to propellant 1 except for 1% carbon addition. Correction of the arc data for propellant 1 to account for its 29% reflectivity will shift its line to about the same position as the laser lines. The remaining shift needed to make it coincident with the line for propellant 2 presumably is obtained by the increase in extinction coefficient, though the change in slope (see Table 2 in Ref. 1) is less than one would expect from the indicated change in extinction coefficient.

Though the addition of carbon lessens the arc image optical effects on ignitability, it does not eliminate the difference between the data from laser and arc image; the L_I lines for propellant 2 do not coincide. Since it is unlikely that the difference in radiation wavelengths is causing chemical effects,[12,13] the fault probably lies in the persistence, for both radiation sources, of substantial but different in-depth penetration. The slopes in Table 2 of Ref. 1 indicate that this is so, since they are substantially less than -2. The slightly higher slope for the arc image data is consistent with the lesser ignition delays for propellant 2 in the arc apparatus.

Propellant 4 in Fig. 5b is loaded sufficiently with boron as to be essentially opaque; the slope for both arc and laser is -2. The L_I lines do not coincide, however. The difference may simply be reflection; all metals have high infrared reflectivities and, thus, would shift the laser data in the direction seen. Unfortunately the necessary propellant reflectance data are not available. Some small part of the difference also may be because of the differing flux distributions in the two sets of apparatus. This normally would be noticeable at high fluxes where lateral conduction cannot smear out these effects. Since the two curves are essentially parallel, this cannot be a major factor here.

Figure 1 shows ignition data for a catalyzed double base propellant (propellant 10) obtained with the laser and with the arc image. The pressure dependent L_{Id} boundaries were

Table 1 Measured rates of pre-ignition gasification, showing the occurrence of a stationary, radiation sustained deflagration wave[a]

Propellant and ambient pressure	Gasification rate as measured in laser apparatus at $I_0 = 37$ cal/cm^2-sec	Burning rate as measured in a strand burner (no external radiation assistance)	Burning rate as measured in arc image apparatus[7] at $I_0 = 37$ cal/cm^2-sec
Catalyzed NC plastisol (propellant #10) at 5 atm N_2	5.13 mm/sec	1.7 mm/sec	4.8 mm/sec[b]
Catalyzed NC plastisol (propellant #10) at 11 atm N_2	6.70 mm/sec	3.3 mm/sec	6.4 mm/sec
Noncatalyzed NC plastisol (propellant #6) at 11 atm N_2	5.83 mm/sec	1.8 mm/sec	Not available

[a] Price, et al.[2] measured a pre-ignition gasification rate of 4.57 mm/sec for a JPN propellant subjected to $I_0 = 100$ cal/cm^2-sec at 1.09 atm N_2.
[b] Extrapolated value.

discussed in the previous section; we focus here on the L_{1a} (first gasification) boundary. Again the lines differ significantly for the two radiation sources. Table 2 in Ref. 1 indicates that the propellant is much more opaque at the laser wavelength; reflection is minimal for both wavelength ranges. The greater opacity at the laser wavelength is consistent with the shorter ignition delays found in the laser, but it is not consistent with the fact that the slope (−1.60) is less than that found with the arc (−1.77). (On the basis of its extinction coefficient, this propellant would be expected to have a slope very near −2.) The reason for this anomaly is not known. Apparently, in some cases there may exist factors other than differences in optical properties and flux distribution which can yield radiation source-dependent behavior. The unexplained difference here is not of an alarming magnitude, however.

B. Dynamic Extinction Effects

Dynamic extinction of a burning propellant is a phenomenon that can be induced by sudden removal of a supportive external heat flux just as well as by a sudden drop in ambient pressure. The underlying causes are fundamentally similar. The propellant initially burns at a high rate as a result of a high heat input to the surface. This high heat input may be because of either high pressure accelerating the gas phase flame or a moderate pressure flame supplemented by external radiation. Both situations yield a thin thermal wave in the solid. When the high pressure or supplementary flux is removed in a time comparable to the thermal relaxation time of the solid, the persisting steep gradient in the condensed phase may drag the surface temperature down to a point of no return, and extinction follows. This was first reported and modeled for the sudden radiation removal situation in Ref. 5. (Related work was reported earlier by Mihlfeith.[14]) The propellant was M-9 (nitrocellulose/nitroglycerin), and the radiation source was the same laser as was used in the present study.

Figures 6a and 6b show that this was not a result unique to M-9 propellant; the dynamic extinction boundary (L_2) is shown for propellants 5 (M-9), 6-8. The compositions of these last three are in Table 1 of Ref. 1; they are all NC/TMETN propellants with varied carbon content. Note that in all cases self-sustained ignition is found only in a corridor between the L_1 and L_2 boundaries. The ignition corridor vanishes at 11 atm or below.

It should be noted that there is a complication that enters in here than can obscure the L_2 boundary. If the propellant flame spreads during the irradiation to a portion of the sample that has a much less (or even zero) flux (e.g., the sides of the sample), that portion of the flame will not experience the same "shock" upon sudden radiation termination, and it will not be extinguished. (This has been observed in high-speed movies.) In a go/no-go test, the sample will appear to ignite normally and be self-sustaining, since all of it is consumed eventually. This masking of the L_2 boundary was seen for propellants 5-8 when the irradiation time was long (e.g., ~ 100 msec for propellant 6 at 21 atm), so that there was adequate time for lateral flame spreading. It is not shown in Fig. 6a and 6b because it confuses the basic point.

Note that this dynamic extinction phenomenon is different from the phenomena encountered in the gap between L_{1a} (first gasification) and L_{1d} (self-sustained ignition) at moderate or low pressures as discussed in Sec. II. There, self-sustained ignition could not be obtained until long after the flame had reached a steady state (with radiation); here, ignition can be obtained only before the flame has reached a steady state (with radiation). The model developed in Ref. 5 shows that the ignition corridor (L_1 to L_2) in the present case is possibly only because the preheated layer developed during heat-up broadens the thermal wave (lessening the flux into the solid) and stabilizes the combustion zone throughout the radiation termination event. When this layer is burned off, this stabilizing factor is lost and termination of the radiation causes extinction.

The question now arises as to why these same propellants, when tested in the arc image furnace, do not exhibit the same dynamic extinction phenomena. (Arc image data for propellants 6-8 are shown in Fig. 4 of Ref. 1; recall that the same sample size and same shutters were used there.) This behavior also was never reported by any previous investigators using the arc image furnace for a wide spectrum of propellants. The probable reason is implicit in the previous discussion. Anything which thickens the thermal wave in the solid lessens the flux into the surface and thereby lessens the degree to which the surface temperature is pulled down during radiation termination. Radiation transparency can have exactly this effect if the absorption depth is comparable to the thermal wave thickness in the solid. Table 2 of Ref. 1 indicates that propellants 6-8 are much more opaque in the infrared (10.6μ) than in the visible/near infrared. Even propellant 8, which has the highest extinction coefficient for arc radiation, will have a radiation penetration depth of the order of 100μ. This is comparable to the thermal wave thickness in a 100 msec ignition and substantially greater than the thermal wave for a 10 msec ignition. Added to this effect (which alone could probably preclude dynamic extinction with the arc) is the slower radiation termination time with the arc, even though the shutters are the same; the larger beam size in the arc makes the termination time about 2 msec instead of 1 msec. This is shown in Ref. 5 to broaden the self-sustained ignition region because it is a lesser shock.

With other investigators using arc furnaces, these effects may have been sufficient to preclude dynamic extinction, but yet another factor may have guaranteed its elimination as well. This is the lateral flame spread effect mentioned earlier. If one uses a sample that is large enough in diameter so that the front surface sees a continually decreasing flux along any radius from the central peak flux, these annular lower flux regions can ignite during irradiation. Since these lower flux regions are less over-driven by radiation (and also have thicker thermal waves), they experience less shock upon radiation termination and may not extinguish. In a go/no-go test the sample is consumed fully.

Returning to the laser ignition experiments, one finds that only noncatalyzed double base propellants exhibit this dynamic extinction in the range of parameters examined. Mathematical modeling of this phenomenon[5,15] has revealed that the susceptibility to extinguishment increases as the ratio of surface heat release to flame heat feedback increases. Experimental studies[6] of the effects of lead and copper catalysts on double base propellants have shown that they increase the flame feedback without affecting the surface heat release. This would stabilize these propellants and make them less susceptible to dynamic extinction. *AP* composites have most of their heat release in a gas phase *AP* flame followed by a diffusion flame. This preponderance of flame feedback similarly should stabilize them. Finally, it should be noted that any propellant which generates radiation absorbing products during ignition is, in effect, tending gently to terminate the radiation; this effect may be operative to varying extents with all propellants. It requires further research.

IV. Conclusions

In the pressure dependent ignition domain, double base propellants (and possibly also composites) exhibit a brief period of transient flame development followed by a prolonged steady-state gasification. Finally, some condition is fulfilled which allows the propellant flame to survive when the supporting radiation is removed. Current explanations of what this condition is are inadequate and further research on this point is indicated.

In comparing radiative ignition data from arc image and laser apparatus, one must bear in mind that both suffer, in varying degrees, experimental nonidealities. When these are factored out as much as is possible, the ignitability results with both types of radiative heating are comparable. An exception occurs in the case of noncatalyzed double base propellants which practically cannot be made as opaque to arc radiation as they are to laser radiation. This difference is the probable cause of the lack of dynamic extinction with the arc; the increased thermal wave thickness helps stabilize the propellant during the deradiation disturbance.

References

[1] DeLuca, L., Caveny, L. H., Ohlemiller, T. J., and Summerfield, M., "Radiative Ignition of Double Base Propellants, I. Some Formulation Effects *AIAA Journal* (this issue).

[2] Price, E. W., Bradley, H. H., Jr., Hightower, J. D., and Fleming, R. O., Jr., "Ignition of Solid Propellants," AIAA Paper 65-120, 1964.

[3] Beyer, R. B. and Fishman, N., "Solid Propellant Ignition Studies with High Flux Radiant Energy as a Thermal Source," *Solid Propellant Rocket Research, Progress in Astronautics and Rocketry*, Vol. 1, 1960, American Rocket Society, pp. 674-692.

[4] Shannon, L. J., "Composite Solid Propellant Ignition Mechanisms," Rept. AFOSR 67-1765, Sept. 1967, Air Force Office of Scientific Research, United Technology Center.

[5] Ohlemiller, T. J., Caveny, L. H., DeLuca, L., and Summerfield, M., "Dynamic Effects on Ignitability Limits of Solid Propellants Subjected to Radiative Heating," *Fourteenth Symposium (International) on Combustion*, The Combustion Institute, 1973, pp. 1297-1307.

[6] Kubota, N., Ohlemiller, T. J., Caveny, L. H., and Summerfield, M., "The Mechanism of Super-Rate Burning of Catalyzed Double Base Propellants," Rept. No. 1087, March 1973, Aerospace and Mechanical Sciences Dept., Princeton Univ., Princeton, N. J. (AD 763 786); also "Site and Mode of Action of Plationizers in Double Base Propellants," *AIAA Journal*, Vol. 12, Dec. 1974, pp. 1709-1714.

[7] Caveny, L. H., Ohlemiller, T. J. and Summerfield, M., "Influence of Thermal Radiation on Solid Propellant Burning Rate," *AIAA Journal*, Vol. 13, Feb. 1975, pp. 202-205.

[8] Kondrikov, B. N., Ohlemiller, T. J., and Summerfield, M., "Ignition and Gasification of a Double-Base Propellant Induced by CO_2 Laser Radiation." *Thirteenth International Symposium on Combustion*, Paper No. 129, Aug. 23-29, 1970, Univ. of Utah, Salt Lake City, Utah.

[9] Librovich, V. B., "Ignition of Powders and Explosives," *Zhurnal Prikladnoi Mekhaniki i Technicheskoi Fiziki*, Vol. 6, 1963, p. 74.

[10] Baer, A. D. and Ryan, N. W., "An Approximate but Complete Model for the Ignition Response of Solid Propellants," *AIAA Journal*, Vol. 6, May 1968, pp. 872-877.

[11] Zeldovich, Ya.B., *Doklady Adademiia, Nauk,* USSR, Vol. 2, 1963, p. 150.

[12] Mihlfieth, C. M., Baer, A. D., and Ryan, N. W., "The Response of a Burning Solid Propellant Surface to Thermal Radiation," TR-71-2664 (AD736047), Aug. 1971, Air Force Office of Scientific Research, Univ. of Utah, Salt Lake City, Utah.

[13] Bastress, E. K., Allan, D. S., and Richardson, D. L., "Solid Propellant Ignition Studies," Tech. Doc. Rept. No. RPL-TDR-64-65, Contract AF04 (611)-9065, Oct. 1964; also Bastress, E. K., "Test Methods for Solid Propellant Rocket Igniter Development," *Proceedings of Second ICPRG Combustion Conference*, 1966, pp. 551-561.

[14] Mihlfeith, C. M., Baer, A. D., and Ryan, N. W., "Propellant Combustion Instability as Measured by Combustion Recoil," *AIAA Journal*, Vol. 10, Oct. 1972, pp. 1280-1285.

[15] DeLuca, L. and Summerfield, M., "Nonlinear Stability Analysis of Unsteady Combustion Phenomena Induced by Radiation," (in preparation).

[16] Caveny, L. H., Summerfield, M. and May, I. W., "Propellant Optical Properties and Ignition Characteristics as Modified by Particulate Carbon," presented at AIAA 13th Aerospace Sciences Meeting, Jan. 1975; also *AIAA Journal*, to be submitted.