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REDUCED YIELD DETONATION CHARACTERISTICS IN LARGE FAILURE DIAMETER MATERIALS

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We have made detailed measurements of the approach to steady, self-supported propagating shock waves at greatly reduced yield in composite propellants. Propagation velocities are less than one half the theoretical value expected for full reaction at the sonic plane.

Previous experimental studies[1] have given evidence of similar behavior. Also, previous theoretical work[2] in an analytic form has shown the possibility of reduced yield detonations. We have developed a reaction model coupled with a hydrodynamic code that together provide a description of the coupling of the complex reaction behavior with shock propagation and expansion in energetic materials. The model results show clearly that if the dependence of reaction rate on pressure is of sufficiently low order and the mode of consumption is by "grain burning" the calculated detonation behavior closely parallels the observed non-ideal results.

We describe the experiments, the reaction model, and compare experimental and calculational results. We also extend the model to predict results in the unexplored regime of very large size charges.

INTRODUCTION

Our purposes in this paper are two-fold, first to report the rather remarkable reactive shock behavior of a typical propellant material and, second, to present analysis based on a reactive model of such material to display special features of their detonation behavior, Our model-based analysis predicts detonation behaviors that remain to be confirmed experimentally. The available experimental observations provide only a limited coverage of possible initiation and detonation behavior. We have extended the analysis to explore the full regime and have included illustrations of the dependence of the results on parameters in the model.

We will refer to the ammonium perchlorate (AP), aluminum(Al), polymeric binder propellant as 1.3 propellants. For convenience, other materials containing large amounts of AP with lesser amounts of high explosive such as RDX or HMX we will refer to as composite explosives even though some of these materials may be used as propellants and in a few cases may be officially classified as 1.3.

There have been relatively few studies devoted to detonation behavior in large charges of 1.3 propellants.[1,3,4] There was evidence of the propagation of detonation in all of these studies. However, as we will show, the approach to steady behavior is very prolonged so that the evidence may very well not be conclusive.

Considerable work[5-8] has been performed on smaller charges of 1.3 materials (charges whose diameter, d, is considerably smaller than the failure radius d_c). With the exception of the reports by Bai *et al.*[9] and Boteler and Lindfors,[10] results of shock loading tests have failed to reveal any significant reactive response during the time of shock transit. Our results are consistent with the preponderance of evidence.

Experimental work on composite explosives[11-14] containing large amounts of AP and work on pure granular AP (less than TMD) reveals behavior similar in nature but much less extreme than that which we observe and predict for 1.3 propellant. The composite explosives and granular AP offer the opportunity to observe detonation behavior at values of $d/d_c >> 1$ and to examine the relation of the reaction rate to the detonation behavior. We recover the measured[11,13,14] linear dependence of propagation speed with reciprocal diameter with specific parameter values.

Previous analyses by Leiper,[15] Guirguis,[2] and by Andersen and Chaiken[16] have presented numerical and analytic work to describe the non-ideal behavior in composite materials. Leiper and Guirguis have pointed out the possibility of "eigendetonations" (i.e. steady detonations with reduced yield and variable sonic plane conditions depending upon charge diameter). Leiper's description is based on a rather arbitrary multiple reaction step process while Guirguis' analysis is based primarily on analytic relationships between reaction rates and hydrodynamic flow. Both analyses are primarily limited to steady flow. Their analyses were applied to composite explosives where the effects are much less extreme than in the 1.3 propellant examined here. Anderson's analysis was applied to 1.3 materials in advance of the SOPHY test studies and focused on temperature dependence of initiation and burn rate. Their estimated failure diameter was approximately 10 times the value measured later. They did not discuss the sonic plane conditions or rate of approach to steady propagation. Westmoreland[17] and Tarver[18] applied models to composite explosives similar to the modeling analysis presented in this paper. Laminar burn rates were not explicitly incorporated and a form factor describing "hot spot" burning was used. Tarver[6] in a later paper applied a grain burning model to a 1.3 material. Their results and predictions were quite successful but were applied to a limited regime of composite behavior.

Our approach differs considerably from the previous work in several respects. The current numerical model is more general in that it treats initiation and the approach to steady propagation or to failure. It is based on an attempt to provide a physical description of the reaction process following shock stimulus. As we will describe, our modeling analysis has explicitly incorporated measured burn rate dependence, separate aluminum burning, and inert shock measurements together with calculated equation of state parameters in order to produce realistic predictions. More important, the assumptions in the model and the assignment of parameters can be separately subjected to critical examination. Thus it is our hope that we have laid the groundwork for future studies.

MODEL OF IGNITION AND GROWTH OF REACTION

The model we use to describe the growth of energetic reaction in propellant is based on the original model described by Lee and Tarver,[19] and modified by us to incorporate the idea that the oxidation of the aluminum metal has a clear and distinct rate of reaction that differs in its pressure dependence from the rate of decomposition of the AP oxidizer and the oxidation of the polymeric binder HTPB. As implemented, the model uses JWL equations of state to represent (a) the unreacted material, (b) the products of burning binder in AP decomposition products, and (c) the products of burning aluminum in (b). Analysis of the composition of the latter two using CHEETAH[20], a thermochemical equilibrium program, shows that over a wide pressure range, oxygen from H_2O and from CO_2 is used to burn the aluminum metal, but CO is not further reduced.

In our implementation of the model, there are two parameters that track the extent of the two reactions, λ_1 and λ_2. The extent of reaction 2 depends on the progress of reaction 1, according to our view that aluminum can burn only when water and carbon dioxide are present. The mixture rule for the equations of state is that pressure and temperature are equilibrated. The mass fraction of reactants is given by $1-\lambda_1$, the mass fraction of the intermediate products is $\lambda_1-\lambda_2$, and the mass fraction of final products is λ_2. We impose the additional constraint that the intermediate product may not have a negative mass fraction.

The equation of state parameters for the unreacted propellant were fit to Hugoniot data taken for this and similar propellants.[5,21] The JWL parameters for the intermediate products were fit to CHEETAH calculations of the energy released on the adiabat from a theoretical ideal detonation of AP, HTPB, and non-reacting aluminum metal. The JWL parameters for the final products are taken from a renormalization of the CHEETAH calculation that reduces the available energy from the final products by 20%. The historical precedent for doing so is that the predictions of thermochemical calculations for aluminized propellants have always exceeded the measured values even in cases where the aluminum is consumed. The argument relies on comparison of calculational and experimental results of cylinder tests on 1.1 propellant of varying diameters. We give the parameter values for the equations of state in the Appendix.

REACTION RATE EXPRESSIONS

The decomposition of AP and the oxidation of binder material is treated as a single reaction, which can be expressed schematically as

$$AP + Binder + Metal \Rightarrow AP/Binder\ products + Metal \quad (1)$$

The rate of this reaction is specified by

$$\frac{d\lambda_1}{dt} = I(1-\lambda_1)^{a_i}(\mu - c)^{\eta} + G_1(1-\lambda_1)^{a_g}\lambda_1^{y}r'(p) \qquad (2)$$

The first term represents the initiation of propellant by a shock wave that produces an excess compression, μ. The excess compression is simply related to the density ρ by $\mu = \rho/\rho_0 - 1$. Here ρ_0 is the reference density. The limit c represents a pressure cut-off, so that lower pres-

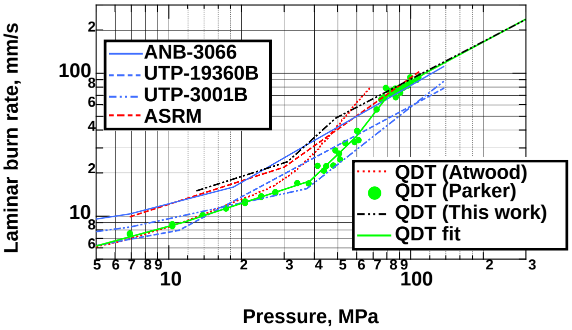

Figure 1. Closed bomb and strand burner measurements of the laminar burn rates of several similar propellants.

sures do not cause initiation. This was set to correspond to an impact stress of approximately 150 bar. Tests by Merrill on a similar propellant showed no reaction from low-velocity impacts. The value of *c* used results in no reaction at the impact velocity tested there. The parameter *I* was reported by Tarver[6] in his simulations of impact induced reactions at high stress levels. He also reports the use of a small value for the parameter a_i.

The second term represents the growth of reaction. The laminar burn rate, r′, of QDT propellant for the Titan IV SRMU and other similar propellants have been measured by us and by others.[22-24] The various measurement techniques and laboratories result in somewhat differing burn rates. (See Fig. 1) We chose to use the data of ours and Parker[23] The piecewise power-law fit is given in the Appendix. In our high-pressure measurements, the value of the power was constant and equal to the lowered value at the highest pressure of Parker. The 1 atmosphere burn rate is consistent with that measured by Merrill.[25] The aluminum metal, in the form of 29 micron diameter powder, is assumed to burn in the products of AP and binder. Pokhil *et al.*[26] report the time for the combustion of aluminum in an oxidizing gas atmosphere to be approximately 2 millisec for a 30 micron sphere. The dependence of burn-up time on gas pressure is approximately -0.3,[27] so that the rate of combustion would be proportional to the 0.3 power of pressure. Hermsen[27] reports a factor of 3 faster burn-up times for the same pressure and diameter, as a consequence of the assumed erosive burning present in the turbulent flow of the exhaust gases in rocket motors. Both reports show the dependence on oxygen concentration to be the 0.9 power. As a consequence, we have chosen the equation for the metal combustion to be

$$\frac{d\lambda_2}{dt} = G_2(R\lambda_1 - \lambda_2)^{0.9}(1-\lambda_2)^{0.667}p^{0.3} \tag{3}$$

The stoichiometric ratio, R, is taken to be 1.42, based on CHEETAH calculations of the product species throughout the pressure range 0.09 to 0.7 GPa. This is the ratio of oxygen concentration considering only H_2O and CO_2, to the oxygen needed to burn the aluminum completely to Al_2O_3 The two-thirds power on the form factor is appropriate for spheres burning from their surface. We fit the coefficient $G_2 = 0.00953$ for pressure in Mbar, time in microsec to match Pokhil's data interpolated for 29 micron spheres. For our model, we defined the time to burn as the time taken to burn between 10% and 90% of the aluminum present.

EXPERIMENTS

The growth of reaction parameter G_1 (Eq. 2) and the form-factor, are fit by comparing simulations of Critical Diameter (CD) tests with measured results. Critical diameter tests of several similar 1.3 composite propellants were performed at Edwards AFB. Cylinders with diameters between 0.4 and 1.5 m were tested. In those tests, the cylinders were 2.5 to 4 diameters long. The booster charge was as-poured density (1.22 g/cc) AP. The booster was the same diameter as the propellant and between 0.5 and 2 diameters long. Experimental results for various similar propellants include propagation velocity at locations more than 2 diameters downstream of the AP/propellant interface. The propagation velocity midway between two pins is measured in the experiments (and in the simulations) by recording the arrival time of the shock front at various axial locations on fixed radial lines. The arrival times are differentiated to establish the propagation velocity. We found it convenient to treat both simulations and experiments alike by averaging two adjacent velocity values and reporting that velocity at the midpoint as described by Banas.[3] For equally-spaced pins, this procedure is equivalent to differentiating by skipping a pin, and recording the velocity at the skipped pin location. For all propellants from 0.4 to 0.9 m diameter, the propagation velocity measured at two diameters downstream was less than 2.6 km/s, which corresponds to a pressure less than 1 GPa.

The 1.5 m diameter test took place at Edwards AFB on November 18, 1996. The results of the 1.5 m SOPHY test[1] were similar, except for the near-booster response, which was noticeably more reactive. The TNT booster used in SOPHY resulted in considerably faster propagation in the first diameter. Because of this similarity, we also calculated the 1.8m SOPHY test, which was still more reactive (faster propagation downstream). Despite the differences in the SOPHY ANB-3226 propellant formulation, which was 69% AP, 15% Al, 16% PBAN, and the QDT propellant (69, 19, 12% HTPB), the similar behavior in the 1.5 m test led us to believe that a single model would suffice. We found that using the grain-burning form factor,

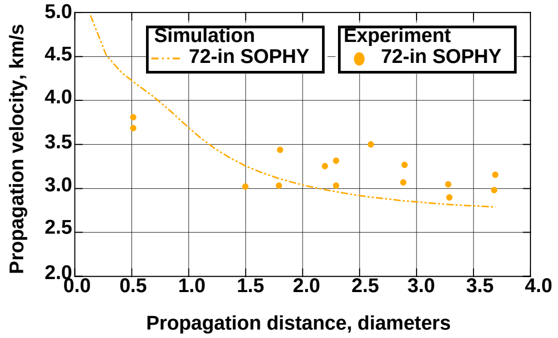

Figure 2. Experimental results and simulation of the 1.8 m SOPHY CD test.

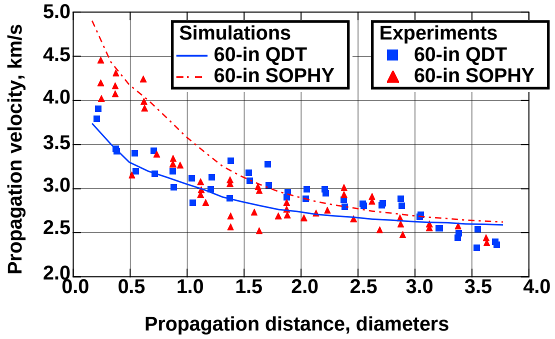

Figure 3. Experimental data and simulations of the 1.5 m SOPHY and PIRAT tests

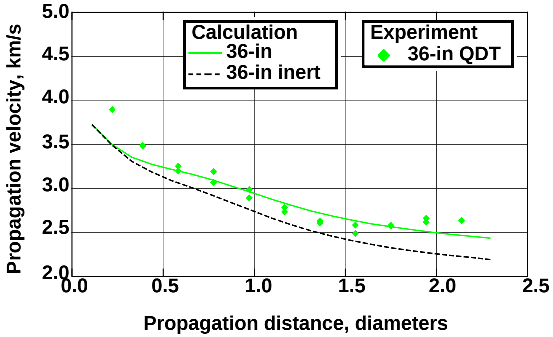

Figure 4. Experimental results and simulations for the 0.9 m CD test with reaction (solid) and without any reaction (dash)

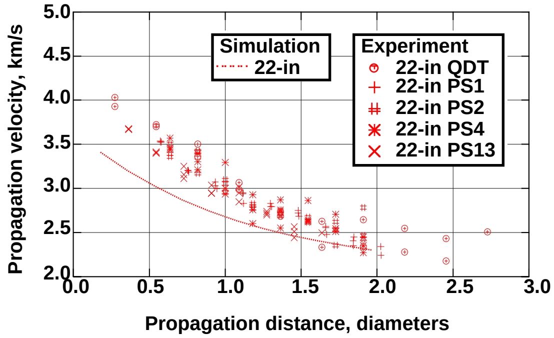

Figure 5. Experimental results and simulations for the 0.5 m DSWA and PIRAT CD tests

$$FF = (1-\lambda)^{0.667}\lambda^{y} \qquad (4)$$

and either the pairs of values (y, G1) (0.1, 4.5) or (0.33, 11.75), we could match the two SOPHY tests and all test results 1.5 m diameter and smaller. In the following section, we discuss the consequences of other choices for the value of y. We show the computed and experimental results for CD tests in various sizes (Figs 2-5). Air shock overpressure measurements, made during these tests, are consistent with explosive yields of 50% for the smallest diameters, and 100% for the largest diameters. These explosive yields are much greater than the shock front conditions in the propellant would indicate. These are reduced yield (or failing) detonations.

MODEL ANALYSIS, RESULTS AND DISCUSSION

To illustrate the behavior of 1.3 propellant as described by our model, we performed a set of computer experiments of under-initiated cylinders. In these computer experiments, a 4-diameter long cylinder of propellant is stimulated by impacting one end with a steel disk that has the same diameter and is a quarter-diameter thick. By under stimulating, we could observe the growth of reaction to finally achieve a steady peak pressure that was higher than the initial shock value. In this way, distinguishing a steady reaction from one that decays was unambiguous. In all cases reported, an input pressure of approximately half of the steady propagating shock was sufficient to reach steady state in four diameters. All steady values reported here had propagated 3 to 4 diameters. The result for steady propagation velocity as a function of reciprocal diameter is shown in Fig 6. For the choice (y, G_1) = (0.33, 11.75) we replicate the results of the 1.5 and 1.8 m CD tests. The result for under-stimulation was that diameters of 2 m and smaller did not propagate. The reaction started but then died out. A 2.5 m diameter calculation showed steady propagation. The approximately linear variation of propagation velocity with reciprocal diameter has been reported[11,13,14] for a number of composite explosives containing large amounts of AP.

The original choice for the values of (y, G_1) as (0.1, 4.5), also recovers the measured propagation velocity of the 1.5 and 1.8 m CD tests. The behavior of propagation speed with reciprocal diameter is even more extreme, and is well fitted by an exponential decay with reciprocal diameter. (See Fig. 6) For this case, a critical diameter cannot be identified. Even small sizes propagate a small amount of reaction at scarcely larger than the sound speed in unreacting propellant. The shape of the form factor is shown in Fig 7 for various choices of y, (Eq. 4). If a "hotspot" model (y, G_1) = (0.667, 56.225) is chosen, it is possible to match the results of the 1.5 m test, but the calculated result of the 1.8 m test is a reduced yield detonation propagating at 5 km/s.

Based on the above limited evidence we have selected y = 0.33. The physical description corresponding to this form factor is as follows. Starting from widely spaced localized ignition sites, "flames" spread subsonically in thin sheets to enclose rather large (10-30 mm) regions of propellant. The grain burning regime is attained after only a small fraction (.03) is consumed. This implies a flame thickness of approximately 1 mm. This picture is based on our model and has not been directly confirmed.

INITIATION

We sought to characterize the initiation behavior of this propellant by calculating the "Pop" plot of run-distance to detonation as a function of pressure for thick-plate stimulus. We found that this could not be done in an unambiguous way. We examined a 5 m diameter cylinder, which is 2.5 times the critical diameter. In the usual experiment, steady detonation is achieved in the plane-strain region, less than one radius propagation. In our calculation (Fig. 8) steady detonation does not occur within one radius, even for an initial pressure of 0.15 Mbar. The peak pressure for full detonation at infinite diameter would be over 0.3 Mbar (Fig. 9). After achieving a maximum at one radius propagation, side rarefactions reduce the peak pressure. At 3 to 4 diameters downstream, the steady peak pressure of 0.15 Mbar (Fig. 9) is achieved.

REACTION ZONE

Another remarkable feature revealed in our analysis is the extremely low value of energy release forward of the sonic plane. The sonic plane separates the region behind the shock front. Behind that plane, disturbances cannot communicate with the shock front, so energy released in the reaction cannot contribute to the strength of the shock. It is the Chapman-Jouget (CJ) plane of detonation theory. We show the distance between the shock front and the sonic plane as a function of reciprocal diameter in Fig. 10. These distances are much larger than commonly obtain in explosives. For our nominal model, the distance is approximately 1 meter.

The low value of the propagation velocity relative to the infinite diameter result suggests that little of the energy of reaction is released at the sonic plane. We show the result for the fraction of the energy released (including both reactions) at the sonic plane in Fig. 11. For our nominal model at 2.5 m diameter, the reactive shock propagates with less than 15% of the energy released at the sonic plane. For the extreme model (y, G_1) = (0.1, 4.5), less than 3% of the energy is released at the sonic plane at 2 m diameter. It is important to remember, however, that substantially all of the energy is eventually released. However, it is not released in such a way that it can affect shock propagation. We show profiles (Fig. 12) on the center-line of the 5 m diameter cylinder using the nominal model, about 3.7 diameters downstream from the initiation. The shock pressure and the fraction of energy released are shown as a function of axial distance. There are no features that would make the CJ plane identifiable. This is simply the location of the place where the sum of local sound velocity and material (particle) velocity equal the steady propagation speed of the reactive shock. The reactions continue to release energy behind the sonic front, which contributes to radial expansion of the propellant cylinder and to air blast, but not to the propagation speed of the reactive shock. Experimental pressure measurements would report the von Neumann spike pressure. Determination of the "CJ" or sonic plane pressure requires further analysis.

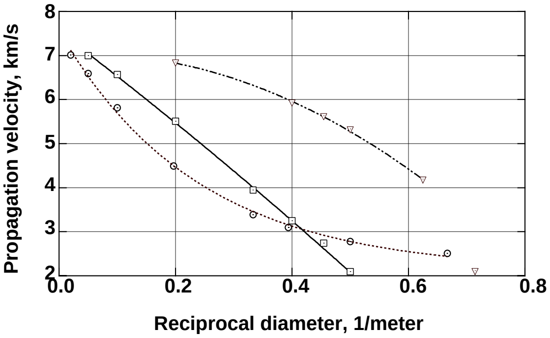

Figure 6. Calculated steady propagation velocity as a function of reciprocal charge diameter for y=0.1 (dot), 0.33 (solid), and 0.667 (dash-dot)

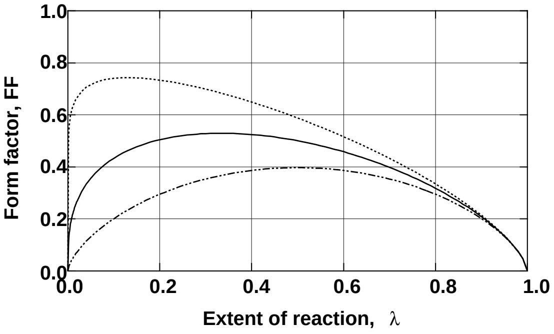

Figure 7. Form factor evaluated for different values of the y power 0.1 (dot), 0.33 (solid) and 0.667 (dash-dot)

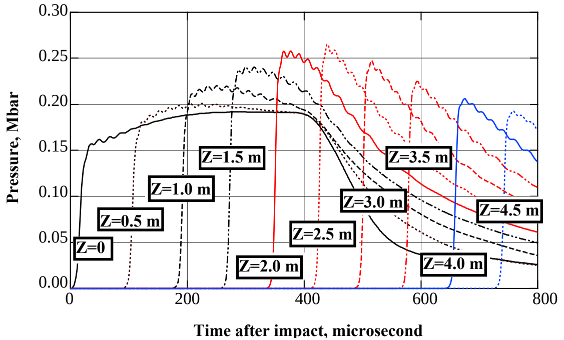

Figure 8. Calculated pressure histories on the centerline of a 5m diameter cylinder. The plane-strain one-dimensional region extends to 2.5 m.

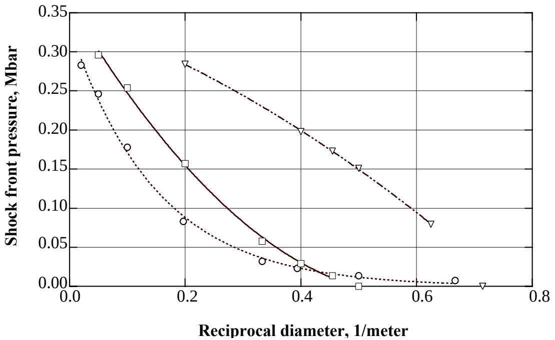

Figure 9. Calculated shock front pressure for y = 0.1 (dot), 0.33 (solid) and 0.667 (dash-dot)

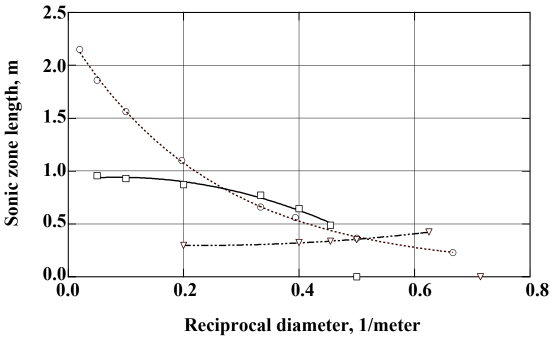

Figure 10. Calculated distance between shock front and sonic plane on the axis of symmetry for y=0.1 (dot), 0.33 (solid), and 0.667 (dash)

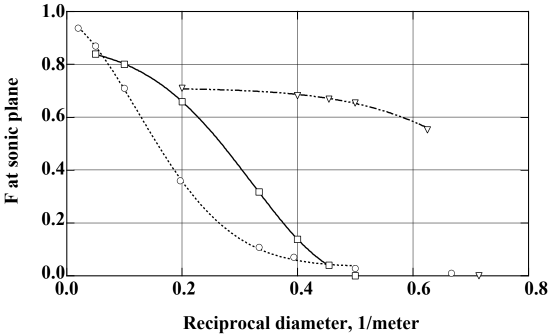

Figure 11. Fraction of energy released, F, at the sonic plane on the centerline of the propellant cylinder for y=0.1 (dot), 0.33 (solid), and 0.667 (dash-dot)

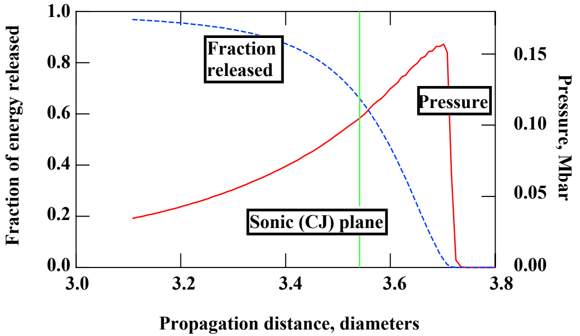

Figure 12. Calculated centerline profiles of pressure and fractional energy released for a 5m diameter cylinder, y=0.33

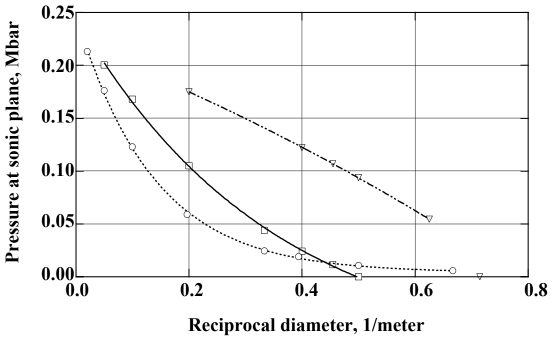

Figure 13. Calculated pressure at the sonic plane for y = 0.1 (dot), 0.33 (solid) and 0.667 (dash-dot)

The pressure at the sonic plane, which we may call "CJ" pressure, although it is not the pressure supported by a fully reacted shock, is shown as a function of reciprocal diameter in Fig. 13. Note that the "CJ" pressure and the energy release do not reach theoretical values even at infinite diameter conditions although the reaction zone length approaches its asymptotic value. Given the rate dependence we chose for aluminum burning, the aluminum reaction rate is simply not rapid enough to reach completion in the passage of the reaction zone (about one meter) at the propagation speed 7.5 km/s. This is similar to the case with aluminum particles in high explosives. For this 1.3 propellant a large fraction of the aluminum is predicted to be consumed forward of the sonic plane, because the reaction zone is enormous. The much smaller distance between the shock front and the sonic plane for aluminized explosives, despite the higher temperature of the intermediate products, implies that for many formulations, a smaller mass fraction of aluminum may be burned in that zone.

CONCLUSIONS

The response of composite energetic materials, in particular, the 1.3 propellants commonly used in large booster motors, is at great variance with what has been observed in high explosives. In our modeling analysis, detonation behavior is strongly dependent on the form factor for the burn surface area and on the pressure dependence for the burn rate. The combination of low values for S/V, grain burning, and a nearly linear dependence on pressure results in a remarkably slow approach to steady flow, very low reduced yield detonation speeds, and very small fraction burned ahead of the "CJ' plane for charges near critical diameter.

The major energy release in this regime (near critical diameter) occurs downsteam of the "CJ" plane. This is totally at variance with the observed behavior of high explosives and much more extreme than somewhat similar behavior in composite explosives.

The confirmation or refutation of our extended results will require very large experiments or ingenious subscale experiments, perhaps using similar materials with much smaller failure diameters. We pose this as a challenge to detonation researchers.

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APPENDIX: PROPELLANT PARAMETERS

The equation of state used for the reactants, intermediate products, and final products all have the same JWL form

$$p = A\exp(-R_1\rho_0/\rho) + B\exp(-R_2\rho_0/\rho) + C_v\omega T\rho/\rho_0,$$

where the reference density, ρ_0, is 1.80 g/cc.

Table 1. Equation of state parameters

Parameter	Reactant	Intermed	Final
A, Mbar	205	16.059	8.0098
B, Mbar	-0.107876	0.09449	0.16183
R_1	10	5.84	5.3
R_2	2	1.3	1.4
ω	0.8	0.222	0.26
C_v, Mbar-cc/cc_0/K	$2.22\ 10^{-5}$	$1.61\ 10^{-5}$	$1.845\ 10^{-5}$
Q, Mbar-cc/cc_0	0	0.07	0.04

Note. The total Q released for both reactions is 0.11, the sum of the two energy values given

The values of the ignition and growth parameters appear in Table 2. In addition to the nominal values of y and G_1 given there, we used the two other pairs of (y, G_1) values (0.1, 4.5) and (0.667, 56.225)

Table 2. Ignition and growth parameters

Parameter	Value
I	1.1
a_i	0.222
c	0.002
n	4
G_1	11.75
a_g	0.667
y	0.33
G_2	0.00953
R	1.42

Note 1. The ignition term is set to 0 when the extent of reaction exceeds 0.04

Note 2. The pressure dependence of the laminar burn rate is specified by log-log interpolation of the following (burn-rate, pressure) pairs, where the burn rate is in cm/microsec, and the pressure is Mbar:
($1.016\ 10^{-7}$, $1.000\ 10^{-6}$), ($9.068\ 10^{-7}$, $1.151\ 10^{-4}$)
($1.763\ 10^{-6}$, $3.842\ 10^{-4}$), ($3.599\ 10^{-6}$, $5.957\ 10^{-4}$)
($6.988\ 10^{-6}$, $8.077\ 10^{-4}$), ($1.016\ 10^{-5}$, $1.164\ 10^{-3}$),
($4.441\ 10^{-3}$, 1)

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