Some Statistical Considerations in the Burning of Composite Solid Propellants

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Propagation of a deflagration wave through a nearly ordered composite propellant is considered. The intent is to see some effects of the particle packing statistics on the propagation rate. A highly oversimplified model of the deflagration physics is considered, and only the statistics are emphasized. It is found that, for propellants of usual packing density, the least-time path of burning to a given point depends primarily on the packing statistics in a line parallel to the burn rate vector; there is little effect of particles only a few particle diameters to the side of this line. In a simplified model that may be solved analytically, there are clearly seen particle-size, pressure, and packing density effects on burn rate which are due to the statistics alone and not the deflagration physics.

Introduction

THE heterogeneity of a composite solid propellant gives severe problems when it comes to modeling the combustion behavior. If one is positioned on a line parallel to the mean burn rate vector and follows the surface as it regresses, a time-dependent process is seen by the observer, involving alternate binder and oxidizer burnthrough (excluding metalized propellants) and other unsteady processes such as bubbling melts and melt flows. Alternatively, at any instant of time, if the observer looks over the surface, a spacewise heterogeneous process is involved. Precision in modeling, therefore, requires consideration of both time dependence and space heterogeneity.

Glick[1] and Glick and Condon[2] have tackled the space heterogeneity problem by application of a statistical method to the propellant structure. In their method, a combustion model is coupled with a statistical description of various fuel-oxidizer pair sizes to yield an average burn rate. The combustion model favored is the BDP[3] model, which is essentially a steady-state model and ignores the transients that would be found in an actual propellant burnthrough problem. There are also difficulties in the statistical description of the propellant surface. An early assumption in Ref. 1 is that for any particle of oxidizer the probability of a particular "size" of binder pocket being adjacent to the oxidizer is independent of the oxidizer size. This assumption is tantamount to assuming complete disorder to the propellant array. Furthermore, it is assumed that the existence of a particular oxidizer size at one point does not influence the allowable particle size at a neighboring point; this again is an assumption of complete disorder to the propellant structure.

These two difficulties, unsteadiness and a statistical description of the propellant surface, actually are coupled. The deflagration physics will, in fact, affect the surface configuration. It therefore would seem reasonable to actually track the deflagration front in time, using an appropriate deflagration model, to see how the surface details unfold with time. There are obvious difficulties with such an approach, however, because one still would need a combustion model and a statistical description of the propellant packing.

Concerning the propellant packing statistics, there is a real issue concerning the degree of disorder to the oxidizer particles imbedded in a binder matrix. For nonspherical particles with multimodal particle size distributions and wide cuts about a nominal particle size, the observation of Ref. 2 that a propellant is a quite disordered structure is probably correct, even if the propellant is near the maximum possible solids loading. On the other hand, it is possible to conceive of a nearly ordered propellant structure if single-particle-size, spherical particles are packed at nearly the maximum solids loading. This becomes a completely ordered structure in the limit that the oxidizer loading is a maximum and the particle-size cut is of zero variance about the mean particle size (a delta-function size distribution). Such a packing is shown in Fig. 1. The lattice points of the array form an oblique parallelpiped. A perfectly ordered structure also would result with spherical particles in a multimodal distribution at the maximum solids loading if the particles had narrow cuts about each size in the distribution. The propellant of Fig. 1 is, in principle, possible to manufacture, although it would not be a practical propellant. If one now backs off slightly from the maximum packing density, a nearly ordered structure would result with particles near but randomly displaced from their ordered lattice points. This is the kind of propellant to be considered in this paper.

The motivation for considering the nearly ordered structure is twofold. First, it is a simple structure through which to track an unsteady deflagration wave. Second, the statistics of the packing are sufficiently simple to investigate the question of what is the lateral extent of influence of one point upon the other. Stated otherwise, given the deflagration arrival at a point, how was the time of arrival affected by particles above, but transversely separated from, the vertical axis running through the particle of interest?

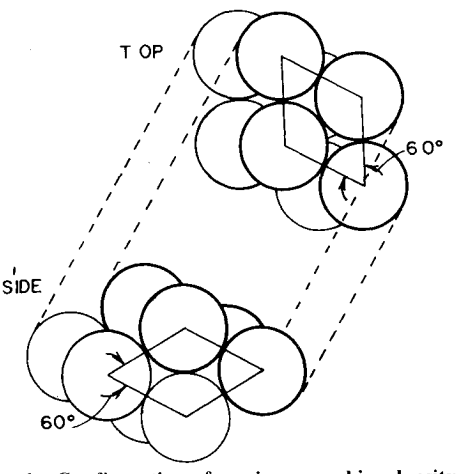

Fig. 1 Configuration of maximum packing density.

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Although the propellant of Fig. 1 is clearly not a practical propellant, it is intended for use to see some statistical effects that are surely present in real propellants. The model to be employed for the deflagration process is also highly oversimplified and is selected not for its deflagration physics credibility but its demonstration of statistical effects.

This work should be considered complementary to the work of Refs. 1 and 2; this is intended to answer several questions that cannot be answered at the present time by their procedure. Although the analogy is not precise, the work here may be considered a Lagrangian approach to the problem where the deflagration front is tracked in time. The approach of Refs. 1 and 2 may be likened to an Eulerian view where a time-stationary average surface is under observation.

Analysis

Lattice Statistics

The lattice structure of Fig. 1 is the starting point and represents the maximum packing density for spherical particles. If V_0 is any volume containing a large number of particles, the volume V of particulate material is given from elementary geometry as $V/V_0 = 0.6981$, which is the maximum packing density. Consider keeping the same lattice geometry, but shrinking each particle in size by an equal amount. Then each particle is in an ordered array with $V/V_0 < 0.6981$, and the particles are separated from each other by an equal amount at their points of closest approach. This perfectly ordered propellant actually would have a mild anistropic burning behavior. In any principal lattice plane and along a direction parallel to a principal lattice axis, everything looks the same. But, by making planar cuts in an arbitrary plane and moving along an arbitrary line, the configuration changes to an observer, as compared with observations on the principal planes and directions. As an assumption of the analysis, observation will only be made on a principal plane and along a principal direction.

Now consider Fig. 2a, where a typical three-particle element is viewed, and consider small random displacements of the particles from their ordered lattice points. The particle radius is r, the distance between ordered lattice points is d, the displacement of the center of any particle from its ordered position is the vector δ and the distance between two particles along the line of centers is x. The statistics to be employed consist of assigning a probability distribution to the δ's and then following the burn through the matrix.

To simplify things further, the "one-dimensional" approximation will be made that δ has only a vertical component δ and that all planes connecting particle centers which are parallel to the paper in Fig. 2 have the same set of δ's. Consequently, only events in the plane of the paper are of interest (there is no interaction between adjacent planes). One now has the situation of Fig. 1b. From elementary geometry, the distance $A-A'$, which is x for particles 1 and 2, is

$$x = \{(3d^2/4) + [(d/2) - \delta_2 - \delta_1]^2\}^{1/2} - 2r \tag{1}$$

An approximation to Eq. (1), which will be used and is valid if $(d-2r)/d \ll 1$, is

$$x = d - 2r + [(\delta_1 - \delta_2)/2] \equiv \Delta + [(\delta_1 - \delta_2)/2] \tag{2}$$

Equation (2) is valid for well-packed propellants if the δ's are also restricted so that $\delta_i/d \ll 1$.

There is, in principle, no restriction on the magnitude of δ. However, if one draws a sphere (circle) of influence about the ordered lattice center of $r+\delta_m$, where δ_m is the maximum expected δ, it is clear that these spheres will overlap if $\delta_m > (d-2r)/2$. Consequently, if $\delta_m > (d-2r)/2$, one particle will interfere with the allowable positions of an adjacent particle(s). This again would complicate the statistics, so it is required that $\delta_m \leq (d-2r)/2$. This may be viewed physically as a "mixedness" assumption. That is, if the propellant is well mixed, no large voids of particles would be expected and each particle would be near its ordered lattice point.

For illustrative purposes, the probability distribution for the δ's will be the uniform distribution, whereby the differential probability dP of finding δ between δ and $\delta + d\delta$ is

$$dP = d\delta/2\delta_m \tag{3}$$

Obviously the particle must be somewhere between $-\delta_m$ and $+\delta_m$, so that

$$P(-\delta_m \leq \delta \leq \delta_m) = \int_{-\delta_m}^{\delta_m} dP = 1 \tag{4}$$

Finally, two cases will be considered. The first, corresponding to Fig. 2b, will be periodic about two column

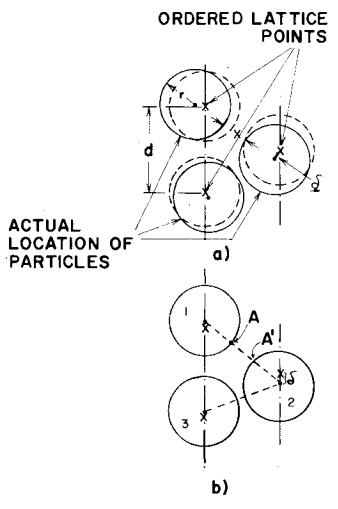

Fig. 2 Configuration of a nearly ordered array of spherical particles.

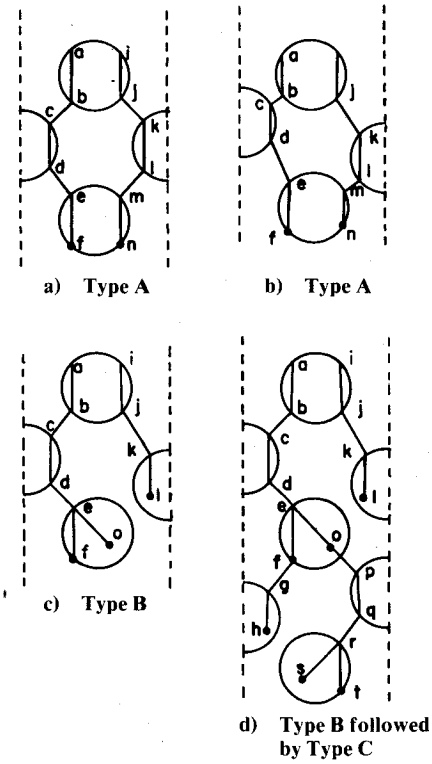

Fig. 3 Various situations in a three-column nearly ordered array of particles.

widths and will be symmetric about the left-hand centerline on Fig. 2b. This means that only the region between the two centerlines need be considered for burning calculations. The second case is shown in Fig. 3a. This case considers symmetry about the left-hand centerline and periodicity about three column widths. Thus, in case 1, only two columns can interact. In case 2, three columns can interact. In principle, one could continue this escalation to an infinite number of column widths which would be the limit of a truly heterogeneous, random propellant. It will be found that this is not necessary.

Mechanistic Model

It is assumed first that the oxidizer particles are monopropellants that have a planar deflagration rate of $\dot{r}_0$ at pressure p and cold temperature T. Second, it is assumed that the particles are ignited at the point of closest approach to a preceding adjacent particle. (The binder burns through at the thinest point, and ignition of the oxidizer does not occur until the binder burnthrough has occurred.) Third, the consumption of the oxidizer is by a spherical outgoing deflagration wave at rate $\dot{r}_0$ which emanates from the ignition point or points. Fourth, after the deflagration wave has reached the point of closest approach to an adjacent oxidizer particle, there is a binder burnthrough time calculated by

$$t_b = Kxe^{\alpha x} \tag{5}$$

This law will be discussed later. Finally, it is assumed that oxidizer ignition is instantaneous after binder burnthrough.

Researchers and practitioners in this field will find this model unacceptable from the standpoint of reality. It is not presented, however, for its realism in deflagration physics. It has the necessary elements to test the statistics of the oxidizer matrix and to yield information of the effect of the statistics of the net propagation rate.

Equation (5) has the property that $t_b \propto x$ for low α, but, for general α, t_b is a nonlinear function of x. For large x, the binder burnthrough time increases at an increasingly fast pace with the burnthrough distance. This behavior is expected to have its counterpart in reality. The self-deflagration wave in the oxidizer contains an advancing thermal wave in the solid phase which will precede the gas-phase reactions in arrival at the binder burnthrough point. This mechanism alone will provide energy for binder gasification. Following the gas-phase arrival, there is a convective flow of hot gases from the receding oxidizer which can provide more heat transfer to the binder. Moreover, a diffusion flame between the binder gases and oxidizer gases may form and will add further heat transfer for gasification. However, as the oxidizer surface recedes from the binder burnthrough point, there is a longer gas flow path, and a thicker heat-transfer "boundary layer" will exist at the burnthrough point. This will reduce the heat-transfer rate, making it more difficult to burn through the binder. Consequently, Eq. (5) embodies this expectation, although the exact analytical form probably is not correct.

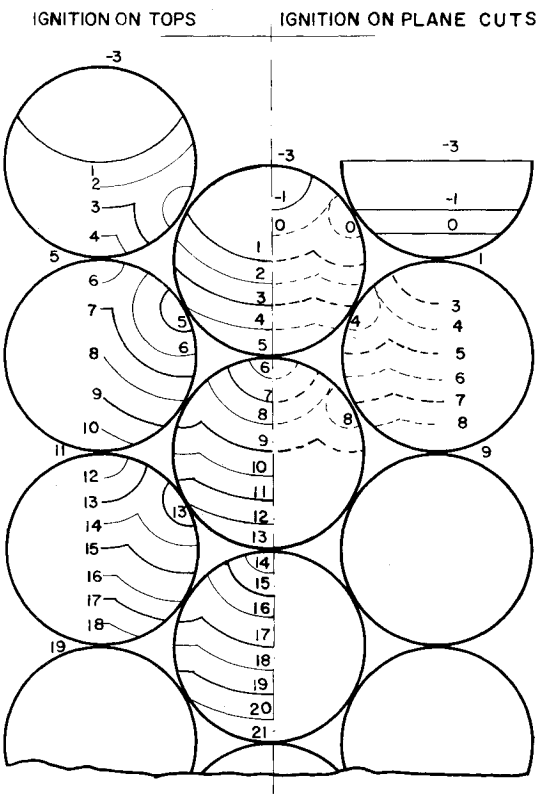

Fig. 4 Propagation through an ordered array of AP; labels are contours of constant times.

The binder could, in principle, burn through to a particle adjacent and to the side or below a given oxidizer particle. However, the only heat source to a binder directly below the particle of interest is by an advancing thermal wave. It was shown in Ref. 4 that, although the energy in this thermal wave is often sufficient to pyrolyze the binder, the rate is often much too slow. This is so because the pyrolysis rate drops rapidly because the surface temperature drops rapidly as the initial pyrolysis carries away the initial portion of the binder. Binder burnthrough points to the side of a particle have additional convective, hot, and, perhaps, reacting gases to aid the burnthrough process. Consequently, it will be assumed in the analysis that burnthrough is only allowed sideways and not directly downward. This necessitates consideration of at least a two-particle column width for the analysis.

Case 1: Two-Column Model

It is first necessary to demonstrate that the ignition details will not alter the results to be presented. Consider Fig. 4, which is a two-column case at the maximum packing density. In this case, burnthrough at the bottom of particles can also occur. Equal time contours have been drawn on Fig. 4 assuming ignition in two different situations at time $= -3$ units. On the left is ignition on the tops of two particles, and on the right ignition is assumed on a plane cut through the propellant. By graphical construction, it is seen that the ignition transient disappears after only two particles in the columns have been consumed. After that, a surface profile that is periodic in time emerges. This indicates that the ignition will have no lasting effect or induce any wild oscillatory behavior in the sequence, and it is sufficient to consider an arbitrary set of particles in the interior of the propellant for analysis.

The two-column model is as shown in Fig. 3a but with the dotted line on the right moved one column width to the left, to exclude the third column. The propagation path for a typcial element is ignition at point a, self-deflagration to b, binder burnthrough to point c, and then the process repeats. Considering a long column of length D containing N particles, the total time to burn through distance D is

$$t = t_{\mathrm{AP}_1} + t_{b_{12}} + t_{\mathrm{AP}_2} + t_{b_{23}} + t_{\mathrm{AP}_3} + \ldots t_{b_{N-1,N}}$$

Since the individual binder burnthrough times depend upon the δ_i,

$$t = t(\delta_1, \delta_2, \ldots \delta_N)$$

and the δ_i are random variables. The expected value of t is, using Eq. (3),

$$E(t) = \int_{-\delta_m}^{\delta_m} \ldots \int_{-\delta_m}^{\delta_m} \frac{\mathrm{d}\delta_1 \mathrm{d}\delta_2 \ldots \mathrm{d}\delta_N}{(2\delta_m)^N} t(\delta_1, \delta_2 \ldots \delta_N) =$$

$$= N \int_{-\delta_m}^{\delta_m} \int_{-\delta_m}^{\delta_m} (t_{\mathrm{AP}_1} + t_{b_{12}}) \frac{\mathrm{d}\delta_1 \mathrm{d}\delta_2}{4\delta_m^2} \tag{6}$$

For large N, $D = N\ d/2$. Moreover, the burn rate is $\dot{r} = D/E(t)$, so that $\dot{r} = d/2\tilde{t}$, where, from Eq. (6),

$$\tilde{t} = \int_{-\delta_m}^{\delta_m} \int_{-\delta_m}^{\delta_m} \frac{1}{4\delta_m^2} (t_{\mathrm{AP}_1} + t_{b_{12}}) \mathrm{d}\delta_1 \mathrm{d}\delta_2$$

The distance between a and b on Fig. 3a is also slightly affected by the δ values, but, under the previous approximation of close packing, it is only weakly dependent upon the δ values. Approximately, $t_{\mathrm{AP}_1} = r/\dot{r}_0$, and $\tilde{t}$ becomes

$$\tilde{t} = \frac{r}{\dot{r}_0} + \frac{1}{4\delta_m^2} \int_{-\delta_m}^{\delta_m} \int_{-\delta_m}^{\delta_m} t_{b_{12}}(x) \mathrm{d}\delta_1 \mathrm{d}\delta_2 \equiv \frac{r}{\dot{r}_0} + \bar{t}_b \tag{7}$$

The expression for t_{AP_1} is where the assumption of the close-packed hexagonal array enters. The final relation needed is $t_{b_{12}}(x)$. Using Eq. (5) for the burnthrough law, a rather important observation may be made. If $t_{b_{12}} \propto x$, then $\tilde{t}$ becomes the time taken for the AP to burn plus the time required for cookthrough to take place through the average binder thickness. That is, the statistics add nothing to the problem. Only when $t_{b_{12}}$ is a nonlinear function of x does the statistical treatment give interesting results. The reason for this behavior, of course, is that if $t_{b_{12}} \propto \dot{x}$, the burnthrough rate is a constant and the slowness of cookthrough of thick layers is canceled by cookthrough of an equal number of thin layers. When the function is nonlinear, however, the statistics will weight the slower burnthrough layers in a heavier manner. Placing Eq. (5) in Eq. (7) and carrying out the integration and forming the overall burn rate expression, there results

$$\frac{\dot{r}}{\dot{r}_0} = \frac{1}{(2r/d) + (\bar{t}_b/d) R \dot{r}_0} \tag{8a}$$

$$\bar{t}_b = K \Delta e^{\alpha \Delta} \tag{8b}$$

$$R = 2(\cosh \alpha \delta_m - 1) / (\alpha \delta_m)^2 \tag{8c}$$

In Eqs. (8), $\bar{t}_b$ is the time to burnthrough of the average binder thickness. $R \geq 1$, depending upon the product of $\alpha\delta_m$; this is the nonlinearity effect of the binder burnthrough law. If $\bar{t}_b$ is short enough (if the binder on average burns at a faster linear rate than the oxidizer), $\dot{r}/\dot{r}_0$ may be slightly greater than unity, according to this model, since $2r/d \leq 1$, depending solely on the oxidizer loading level.

Comparing with experiment, for example, with the polysulfide-AP work of Bastress,[5] it is known that the burn rate ratio of Eqs. (8) will increase with a decrease in AP particle size, decrease in pressure (the propellant has a lower exponent than does pure AP), and an increase in oxidizer loading. In Eqs. (8), the first term in the denominator of the burn rate expression depends solely on the oxidizer loading level, whereas the second term depends upon all parameters. The factor $\bar{t}_b\ R/d$ will decrease with AP particle size at a rate depending on the magnitude of $\alpha\Delta$ and $\alpha\delta_m$; consequently, the particle-size effect is qualitatively predicted. If the binder burnthrough law is pressure-independent, as it is expected to nearly be, the pressure behavior is also qualitatively predicted properly, since $\dot{r}_0$ increases with pressure in the second term of the denominator. Furthermore, the oxidizer loading effect is also properly predicted because $\bar{t}_b\ R/d$ will decrease with an increase in oxidizer loading much faster than $2r/d$ will decrease.

It should be mentioned that there is a serious limitation when compared with experiment. In the experiments cited, $\dot{r}/\dot{r}_0$ was significantly greater than unity, except at sufficiently large particle size. The current model cannot produce such results unless the pure AP rate is augmented, perhaps by consideration of an alternate heat source from oxidizer-binder reactions. On the other hand, there are several AP-binder systems that do exhibit burn rates less than that of pure AP over a significant pressure, particle-size, and oxidizer loading range.[6]

The major point is that some of the trends apparent in experiment are contained in the statistical treatment. The deflagration model is so naive that it was not expected to show the full picture with regard to various variables.

Case 2: Three-Column Model

Returning to Fig. 3, consider the three-column model. The question now is whether or not there is significant interaction of the third column with the left two columns. In Fig. 3a, a perfectly ordered array is shown. Evidently the propagation path *abcdef* takes exactly the same time as path *ijklmn*. In this case, the same burn rate is obtained by considering either path, and the third column adds nothing to the problem. Consider next Fig. 3b, which is a somewhat disordered array. Here $x_{bc} = x_{lm}$ and $x_{de} = x_{jk}$. Again path *abcdef* takes exactly the same time as path *ijklmn*, and there is no interaction between the third column and the left-hand two columns. This kind of event will be called one of "type A."

Figure 3c, on the other hand, shows a different type of event. Here, if the nonlinearity in the binder burnthrough law is strong enough, $t_{jk} \gg t_{bc}$ or t_{de}. Consequently, path *abcdef* or *o* is faster than path *ijkl*. The bottom particle has been consumed solely by the left-hand path, and the right-hand path is stopped effectively at point *l*. This event will be called one of "type B," and the identical event with the left and right columns interchanged will be called one of "type C."

Neither type B or C events, if followed by the same type or one of type A, will alter a burn rate calculation as compared with a two-column calculation. All that is happening is that one column is being dragged along by the other. Since the burn rate is defined by the deepest penetration divided by the time, the left-hand column in the case of type B or A interaction is the sole determinant of the burn rate. Even a type B followed by type A followed by type C event does not increase the burn rate statistically. This is so because, immediately after the type A events, everything starts out "fresh" with the center column particle.

Consider now, however, Fig. 3d, where two opposite unusual events follow each other; in Fig. 3d, this is shown as type B followed by type C. Here, everything would start fresh at points *f* and *o*, but now t_{fg} is so long that the left-hand columns, t_{fg} would have had to be considered, and it would have decreased the expected burn rate. Here, however, path *opqrs* allows that long time interval to be bypassed. This is a clear interaction of all three columns which augments the burn rate. The question is, how frequent are such events? First of all, the event of Fig. 3d cannot happen unless the binder burnthrough law is nonlinear. Since the distance from *a* to *e* and *i* to *m* is always the same, the total time to burnthrough the binders would be the same if the law were linear; all events would be those of type A. Consequently, the nonlinearity is essential if the statistics are to give interesting effects. Second, the answer may be provided analytically, as in the two-column model, but the problem becomes so algebraically complex that it was done by computer.

A three-column array was set up on the computer with all δ_i's chosen by a standard FORTRAN random number generator. Again the uniform distribution of Eq. (3) should be reproduced if enough samples were taken. A baseline case where $\bar{t}_b = 0.5\ r/\dot{r}_0$ was chosen. An AP-binder propellant was assumed at 75% wt of AP with a binder specific gravity of 1.3. This yields $d/2r = 1.015$. If the mean binder burnthrough time were equal to the binder burnthrough time over the mean distance $\bar{t}_b$, this would yield $\dot{r}/\dot{r}_0 = 0.677$. The nonlinearity in

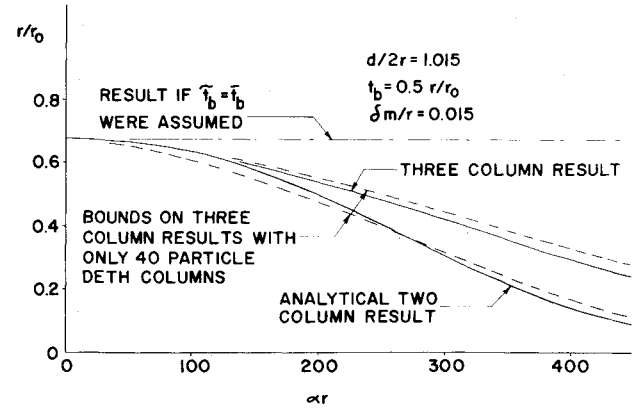

Fig. 5 Burn rate for the two- and three-column models as a function of the binder burnthrough nonlinearity parameter.

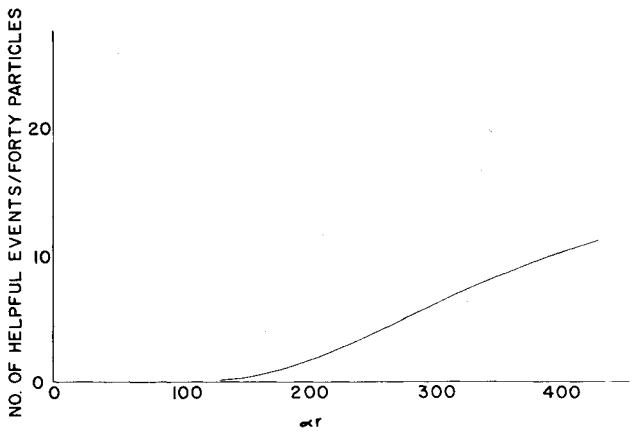

Fig. 6 Illustration of number of "helpful" events as a function of the binder burnthrough nonlinearity parameter.

the binder burnthrough law will, in general, reduce this value through Eqs. (8), but the three-column model will mitigate this drop.

The results are shown in Fig. 5. Shown is the analytical two-column result from Eqs. (8). This is a lower bound. An upper bound is the curve (constant) that would result if $\tilde{t}_b = \bar{t}_b$ were assumed. The three-column result lies in between and was obtained with a 120-particle column depth. With fewer particles, some severe scatter can occur, and approximate bounds on the results obtained with only a 40-particle depth column are shown. The scatter becomes worse at high α (strong nonlinearity) because any deviation from perfection in the random number generator strongly weights the long binder burnthrough events. This is also the reason why the exact (120-particle) curve lies close to the upper bound of the 40-particle curve; deviation from perfect sampling heavily weights the long-time events.

The breakaway of the three-column result from the two-column result begins precisely with the onset of events shown in Fig. 3d. Figure 6 makes a count of the number of "helpful" events vs α. Even at high α, the number of helpful events is relatively small compared with the number of particles in the column, but the effects on rate are striking because an extremely long-time burnthrough has been avoided by the helpful event.

The three-column result makes a 100% augmentation in the two-column result for $r\alpha \approx 400$. The augmentation is shown to about 25% at $r\alpha \approx 275$, where the burn rate is about half the zero-α value. This suggests that the two-column analytical result may be adequate for real propellants, since burn rate depressions from the monopropellant oxidizer rate often are not severe. That is, the effective α is probably not very large. The major conclusion, therefore, is that a two-column model probably is not too bad; there is little influence of neighboring columns on the two-column situation. Another way of saying this is that the horizontal correlation length scale is only of the order of a couple of particle widths. From a practical standpoint, this says that, if the transient problem of combustion through a column that is about as wide as two of the largest oxidizer particles can be solved, an important breakthrough in burn rate modeling will have been achieved.

Considering more than three columns would elevate the result in Fig. 5 toward the $\tilde{t}_b = \bar{t}_b$ line, but not very much at moderate α. There is a point of diminishing returns which is reached rapidly as the column width is expanded. The probability of a compound helpful event from extra columns becomes too small.

As δ_m is reduced toward zero, all curves collapse to the $\tilde{t}_b = \bar{t}_b$ line. If $\bar{t}_b$ is depressed by depressing K in Eq. (5), the curves all tend toward $\dot{r}/\dot{r}_0 = 1.0$. The other effects of oxidizer loading, particle size, and oxidizer size are the same with the three-column model as with the two-column case.

Conclusions

1) Using a simplified deflagration model and a nearly ordered array of spherical oxidizer particles in a binder matrix, a simple model predicts known effects of oxidizer size, oxidizer loading, and pressure level through the statistics of the particle packing.

2) A nonlinearity in the binder burnthrough time vs distance law is required to obtain interesting results from the statistical treatment.

3) The transverse correlation length scale for burning through the matrix is relatively small. Hence, combustion modeling of a column of propellant only a couple of particle diameters wide would be a useful undertaking. This is probably the most important conclusion of the work.

Acknowledgments

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