NASA TECHNICAL NOTE

NASA TN D-2736

NASA TN D-2736

EFFECT OF OXIDIZER PARTICLE SIZE ON SOLID-PROPELLANT COMBUSTION STABILITY

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • APRIL 1965

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SUMMARY

Amplitudes of the longitudinal mode of oscillation were measured in side-vented cylindrical combustors loaded with a composite solid propellant. The binder consisted of a butadiene - carboxylic acid copolymor cross-linked with an epoxy resin, and the oxidizer was ammonium perchlorate. Mean oxidizer particle size was varied by changing the proportions of unground and ground perchlorate while keeping the total quantity constant at 81 percent by weight.

The experimental results indicate a sharp rise in oscillation amplitude at a well-defined mean oxidizer particle size: 40 to 45 percent ground perchlorate in a 6-inch-long combustor; 20 to 25 percent ground perchlorate in a 12-inch-long combustor. A similarity parameter calculated on the basis that burning velocity is determined by the rate of turbulent mixing is shown to be in reasonable agreement with the observed behavior.

A few experiments conducted with aluminized propellant indicate that the suppression effect associated with the aluminum addition is probably due to energy absorption at or near the surface.

INTRODUCTION

In recent years a number of theories have been advanced to describe the conditions under which a gaseous- or liquid-propellant rocket combustor will operate in a resonance mode (refs. 1 to 4). Very generally, these theories all indicate that a decrease in the characteristic time of the combustion process with respect to the wave time tends to destabilize the system; that is, an increase in the local burning rate has a destabilizing influence, other factors being constant. If these theories are applicable to solid-propellant systems, decreasing the oxidizer particle size should have a destabilizing effect. The experiments reported in reference 5 show in a qualitative manner that such an effect does exist. In addition, Horton and coworkers (refs. 6 and 7) have measured combustion instability (using acoustic admittance as a parameter) as a function of oscillation frequency for three different oxidizer size distributions. Their results qualitatively confirm the observations of reference 5.

An alternative approach developed by McClure and coworkers for solid-propellant combustion (refs. 8 and 9) is based on acoustic theory and treats the combustion zone as a boundary condition. In this case, the stability of the system is determined by the sign and the magnitude of the acoustic admittance at the boundary. In reference 10 the effect of oxidizer particle size on the acoustic admittance has been calculated by an extension of McClure's theory. It is found that a decrease in oxidizer particle size should have a stabilizing effect.

The experiment described herein was undertaken to test these two opposed predictions. Hollow cylindrical grains of composite propellant were burned in side-vented combustors of the design used by Price (ref. 11). Oxidizer particle size was varied by changing the proportion of ground and unground perchlorate. A few tests were also conducted with aluminized propellant to determine if the presence of aluminum would lead to greatly different behavior.

An analysis is presented that is based on the assumption that the rate of combustion of the composite propellant can be represented by the rate of turbulent mixing from a grid of point sources.

SYMBOLS

a,b	constants in burning-rate equation
c	velocity of sound
d	oxidizer average particle diameter
d_g	oxidizer average particle diameter of ground fraction
d_u	oxidizer average particle diameter of unground fraction
g	conversion constant
L	combustor length
M	molecular weight of combustion gas
P	mean combustion pressure
R	gas constant
r	linear regression rate of propellant
s	source spacing
T	temperature
$\mathcal{T}$	turbulence intensity
t	mixing time

t_w	period of oscillation
x	mixing length
α	mixing parameter, $\mathcal{T}\frac{x}{s}$
γ	specific-heat ratio
ρ	gas density
ρ_s	propellant density
φ	volume fraction of oxidizer in propellant
ψ	weight ratio of ground to unground oxidizer

Subscripts:

6	6-in. combustor
12	12-in. combustor

EXPERIMENTAL PROCEDURE

The combustor configuration, shown in figure 1, is very similar to that described in reference 11. Cylinder lengths of 6 and 12 inches were used with

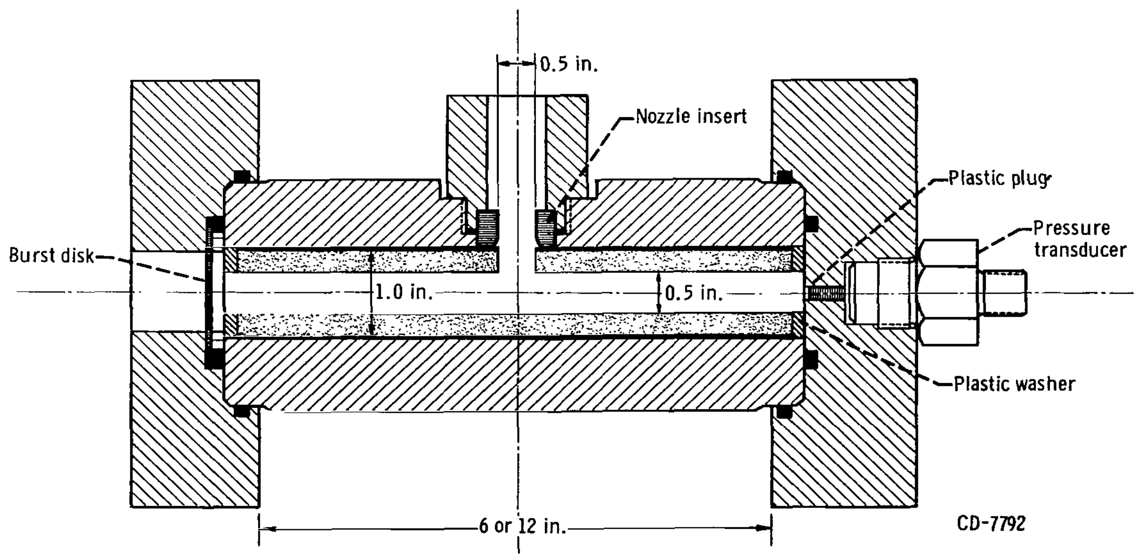


Figure 1. - Cross-sectional view of side-vented combustor.

a propellant charge having an inside diameter of 0.5 inch and an outside diameter of 1.0 inch, which was contained in a paper-base phenolic tube having a

1-inch inside diameter, a 1.0965±0.0005-inch outside diameter, and a 5.875-inch length. The 12-inch combustor was loaded with two such charges.

The strain-gage pressure transducer had a flat response to 10 kilocycles. Its output was recorded on magnetic tape at a speed of 60 inches per second. The pressure data were analyzed by playing the tape back at 15 inches per second and recording on a galvanometer. The mean pressure was derived by playing the tape back through a 40-cycle response element, and the oscillation frequency and the amplitude were derived by playing the tape back through a 2000-cycle response element.

The propellant binder was a butadiene - carboxylic acid copolymer cross-linked with an epoxy resin. The ammonium perchlorate, which contained no anticaking agent, was stored, ground, and weighed at approximately 20-percent relative humidity. Particle size distributions are shown in figure 2 for the ground and the unground perchlorate and for the aluminum powder used in the several formulations. Average particle diameters were 9.1, 67.7, and 6.7 microns for the ground perchlorate, the unground perchlorate, and the aluminum powder, respectively.

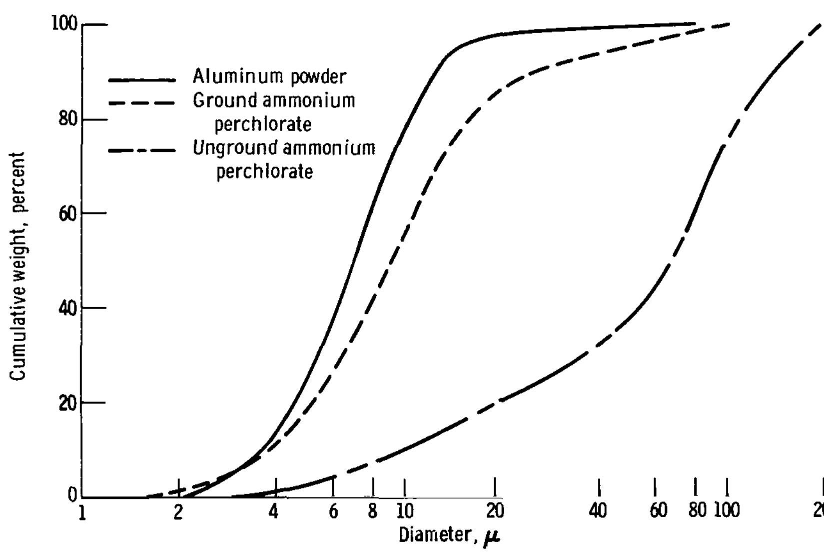
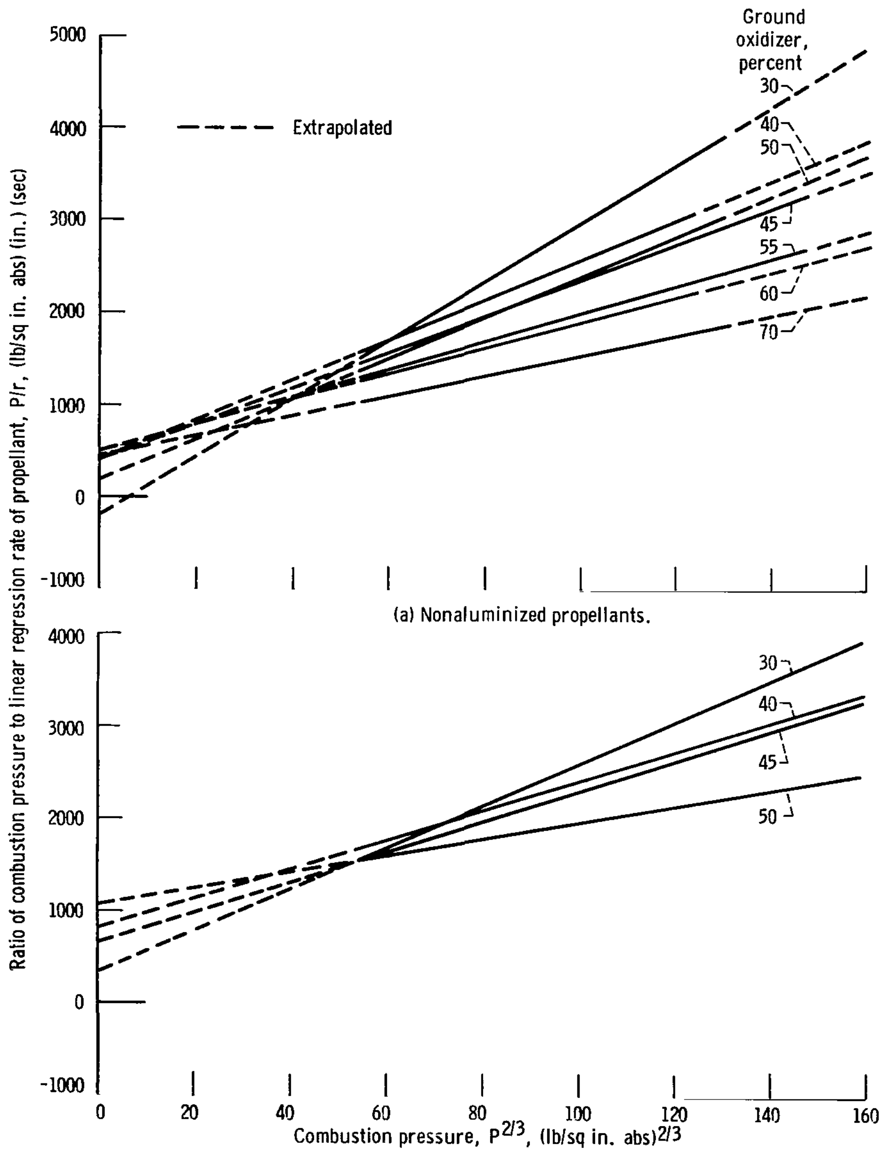


Figure 2. - Particle size distributions of ammonium perchlorate and aluminum used in propellants.

The composition of the nonaluminized propellant was 18.8 percent binder, 0.2 percent magnesium oxide, and 81 percent ammonium perchlorate. The aluminized propellant consisted of 18.4 percent binder, 0.2 percent magnesium oxide, 79.4 percent ammonium perchlorate, and 2.0 percent aluminum. The proportions of ground and unground perchlorate were varied to obtain a range of average oxidizer particle sizes. In all formulations the weight ratio of perchlorate to binder was held constant at 4.3.

The propellant ingredients were combined in a sigma-bladed mixer, transferred to a deaerator, and then forced, while warm, from the deaerator into the phenolic tubes under vacuum. Each tube was held vertically in a support that sealed the bottom end and guided a 0.500-inch-diameter core rod down the axis of the filled tube. Six assemblies were prepared at a time and cured for 16 hours at $85^{\circ}\pm2^{\circ}$ C in a forced-draft oven. After the core rods were pulled out, excess propellant was trimmed off, and the absence of significant defects in the grain was ascertained from X-ray photographs.



(b) Propellants containing 2 percent aluminum.

Figure 3. - Burning rates of propellants (plotted according to ref. 11).

The outside diameter of each grain was measured to the nearest 0.001 inch so that the larger ones could later be inserted into the larger motors to give a consistently tight fit. Each grain was wrapped in a plastic bag and stored at -109° F for at least 16 hours before being loaded into the motor case.

The chilled grains were coated on the outer surface with silicone grease before being rammed into the similarly lubricated combustor cases. After warming to room temperature, a 1/2-inch-diameter opening was cut radially through the side of the grain at the nozzle location. Plastic washers were attached at each end of the grain to prevent both spalling and burning of the end surfaces. Approximately 1.5 milliliters of ignition powder consisting of equal parts of polytetrafluoroethylene and magnesium powder were distributed along the length of the charge. A 6-inch length of nichrome wire (4.1 ohms/ft) was embedded in the igniter powder, and the leads were brought out through the nozzle. A 10-volt alternating-current input was sufficient to ensure reproducible ignitions.

Linear regression rates (strand burning rates) for each composition were measured in a Crawford-type bomb prior to the combustor tests. The rate data were correlated by the method of Summerfield (ref. 12). The faired lines based on a least squares fit are shown in figure 3. From the regression rates, the nozzle sizes were selected to obtain about the same average mean pressure (1500 lb/sq in. abs) for each composition over the range where instability occurred. Actual values ranged from about 1300 to 1800 pounds per square inch absolute.

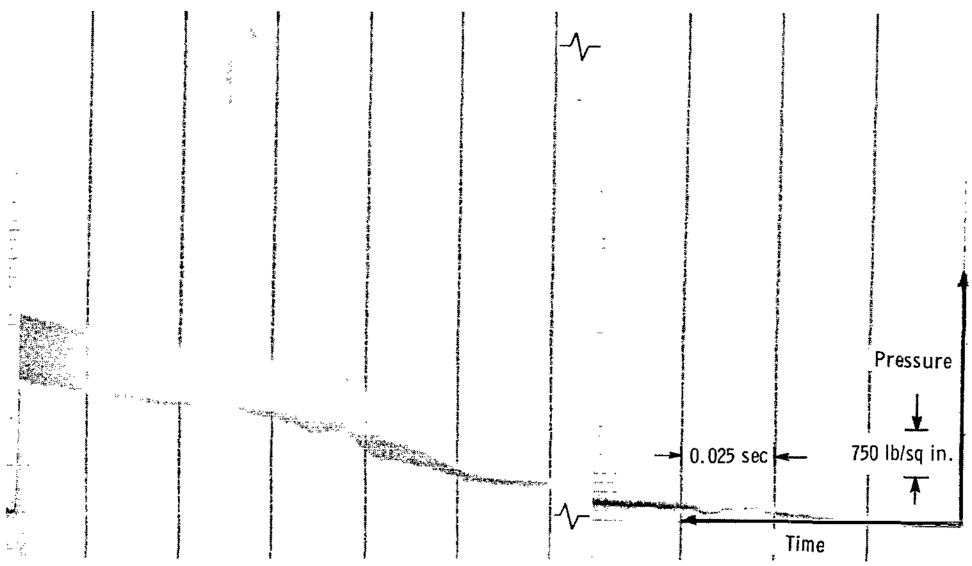


Figure 4. - Typical pressure trace showing commencement of oscillatory combustion.

RESULTS

All the runs exhibited similar characteristics. After ignition, the combustion pressure increased almost linearly with time, as expected from the charge geometry. When the mean pressure reached a value in the range 600 to 1000 pounds per square inch, instability started spontaneously and the amplitude increased rapidly. Simultaneously, the mean pressure increased to a higher level. Following this transient, the oscillation amplitude reached a steady value and the mean pressure resumed its regular increase until combustion was terminated by bursting of the safety disk. A typical pressure trace is shown in figure 4. In this reproduction, the oscillograph paper speed was reduced in order to keep the figure size manageable. The individual cycles are not evident.

The data are shown in figures 5 and 6, where average peak-to-peak pressure amplitude, normalized by the mean pressure over the range where oscillations occurred, is plotted as a function of weight percent of ground material in the oxidizer. Each data point represents an average value of amplitude derived from six firings. The observed oscillation frequencies were 3600±200 cps in the 6-inch-long combustor and 1800±100 cps in the 12-inch-long combustor. These frequencies correspond to the first longitudinal mode of oscillation.

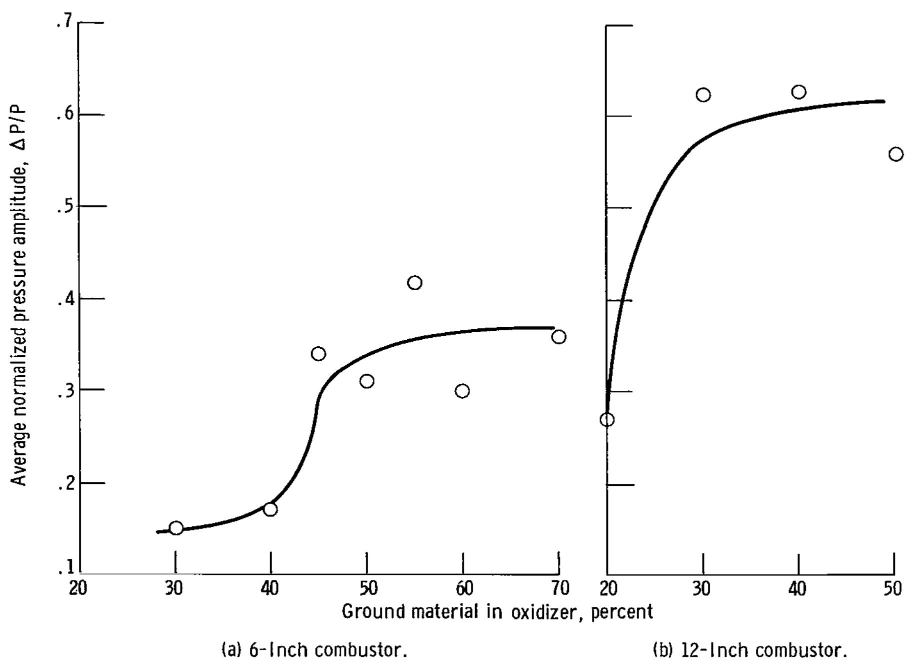


Figure 5. - Pressure oscillation amplitude in combustor as function of ground material in oxidizer.

In both the 6- and 12-inch combustors, the oscillation amplitude was nearly twofold greater at the highest percent ground oxidizer than at the lowest percent (figs. 5(a) and (b)). This agrees qualitatively with the results

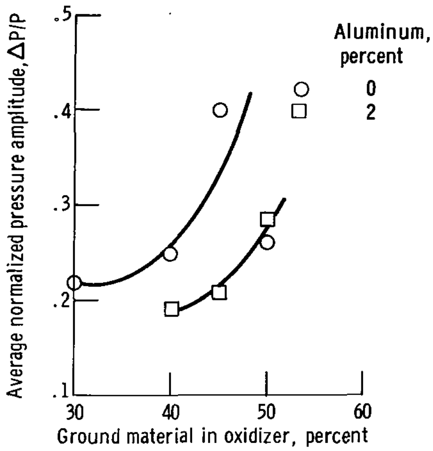


Figure 6. - Pressure oscillation amplitude as affected by addition of aluminum. 6-Inch combustor.

in referencss 6 and 7, and contradicts the prediction of reference 10 as discussed in the INTRODUCTION.

For the 6-inch combustor, an abrupt increase in oscillation amplitude was observed as the percent ground material was increased from 40 to 45 percent (fig. 5(a)). A similar increase was observed in the range 20 to 30 percent ground material for the 12-inch combustor (fig. 5(b)).

The addition of aluminum (fig. 6) appeared to shift the locus of the amplitude change to a higher percent of ground oxidizer. The abnormally low amplitude observed for 50-percent ground material with no aluminum is inexplicable at this time.

Although the location of the break in the amplitude curves was reproducible, the levels of amplitude were not reproducible from batch to batch. Therefore, the aluminized propellant data are compared in figure 6 with data for a batch of nonaluminized propellant prepared at the same time. For this reason, the curves in figure 5(a) and 6 for nonaluminized propellant are not the same.

DISCUSSION

When the data for the nonaluminized propellant are considered, the fact that the change in amplitude occurs at a smaller percentage of ground oxidizer for an increased wave time suggests that the onset of instability may be associated with the attainment of a given ratio of combustion time to wave time, in agreement with the theories of references 1 to 4. If this is actually the case, it is an important finding for it implies that these theories have sufficient generality to be applied to both liquid- and solid-propellant systems when proper account is taken of the differences in boundary conditions.

Following this lead, it is possible to explain the observed behavior by adopting a simplified model of turbulent mixing from a grid of point sources (ref. 13) that has been checked experimentally for hydrogen-oxygen combustion (ref. 14). Here it is assumed that the oxidizer particles can be considered to have a uniform size and spacing based on the mean diameter and the volumetric loading. The mixing parameter of reference 13 is $\alpha \equiv \mathscr{F}\left(\frac{x}{s}\right)$, which is constant for any given degree of mixing.

For the mixing length, the following expression is obtained on the basis of mass flow continuity:

$$x = \frac{r\rho_s t}{\rho} \tag{1}$$

For the spacing, the following expression (ref. 15) is used:

$$s = d\left[\left(\frac{\pi}{6\varphi}\right)^{1/3} - 1\right] \tag{2}$$

When equations (1) and (2) are used, the mixing parameter becomes:

$$\alpha = \frac{\mathscr{T} r \rho_s RTt}{MPd\left[\left(\frac{\pi}{6\varphi}\right)^{1/3} - 1\right]} \tag{3}$$

where the gas density has been evaluated by the ideal gas law.

For the longitudinal mode of oscillation, the wave time is given by

$$t_w = \frac{2L}{c} = \frac{2L}{\sqrt{\frac{\gamma gRT}{M}}} \tag{4}$$

A combination of equations (3) and (4) yields the ratio of mixing time to wave time:

$$\frac{t}{t_w} = \frac{\alpha}{2} \frac{Pd\left[\left(\frac{\pi}{6\varphi}\right)^{1/3} - 1\right]}{L\mathscr{T} r \rho_s} \sqrt{\frac{\gamma gM}{RT}} \tag{5}$$

If it is assumed that the rate of combustion is limited by the rate of turbulent mixing, equation (5) becomes the correct similarity parameter for describing the onset of instability.

Applying the burning-rate expression proposed in reference 12 yields

$$\frac{t}{t_w} = \frac{\alpha}{2} \frac{d\left[\left(\frac{\pi}{6\varphi}\right)^{1/3} - 1\right] \sqrt{\frac{\gamma gM}{RT}}}{L\mathscr{T} \rho_s} \left(a + bP^{2/3}\right) \tag{6}$$

where a and b are functions of d. The value of d may be obtained by computing the volume-to-surface ratio of the oxidizer. If it is assumed that the mixture consists of two fractions, each composed of spherical particles having the average diameter of that fraction, the expression for average oxidizer particle size is

$$d = \frac{\psi + 1}{\frac{\psi}{d_g} + \frac{1}{d_u}}$$

In figure 7, d is plotted as a function of the percent ground material in the oxidizer. For the nonaluminized propellants used in this study (fig. 3(a)),

$$a = 1264 - 144.8\times10^4\ d$$

$$b = 4.13\times10^4\ d - 8.135$$

where the numerical constants are based on values of P in pounds per square inch absolute, r in inches per second, and d in inches. Substitution of these values in equation (6) yields the final expression for the stability parameter:

$$\frac{t}{t_w} = \frac{\alpha}{2} \frac{d\left[\left(\frac{\pi}{6\varphi}\right)^{1/3} - 1\right]}{L\mathscr{F}\rho_s} \sqrt{\frac{\gamma g M}{RT}} \left[(1264 - 144.8\times 10^4 d) + (4.13\times 10^4 d - 8.135)P^{2/3}\right] \tag{7}$$

If it is assumed that turbulence intensity is independent of the particle size of the oxidizer, it is possible to compute the ratio of the similarity parameters for the experimental conditions under which the sudden change in amplitude occurs. For the 12-inch combustor, the change occurred in the range 20 to 30 percent ground material ($d = 10.2\times 10^{-4}$ in.), and the average pressure at which instability occurred was 1800 pounds per square inch absolute. For the 6-inch combustor, the change occurred in the range 40 to 45 percent ground material ($d = 7.1\times 10^{-4}$ in.), and the average pressure was 1300 pounds per square inch absolute. It is assumed, of course, that φ, ρ_s, and the gas properties are invariant. With these values, the ratio of similarity parameters given by equation (7) is

$$\frac{(t/t_w)_6}{(t/t_w)_{12}} = 0.8$$

The ratio, of course, should be unity for complete similarity. The deviation may be due to the simplifications that have been made in the analysis and evaluation.

The observed effect of oxidizer particle size on combustion stability is the reverse of that predicted in reference 10, where the theory of references 8 and 9 was extended to include the effect of oxidizer particle size on the real part of the acoustic admittance of the burning surface. This circumstance leads to the belief that a meaningful theory of combustion instability must include an expression for the characteristic time of the combustion process rather than treating the latter as a boundary condition.

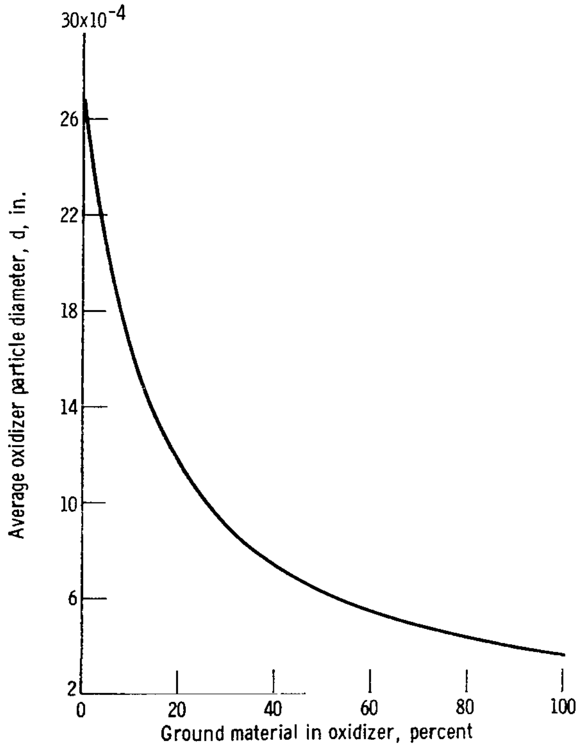


Figure 7. - Average oxidizer particle diameter computed from ratio of volume to surface area.

Although the data for aluminized propellant (fig. 6) are not extensive, they do indicate that aluminum produces an effect that is equivalent to an increase in mixing time. The net effect is equivalent to the damping observed by Horton (ref. 16). While the drag of bluff bodies, such as aluminum or aluminum oxide, in the combustion gases above the surface should produce damping, the increased mixing rate due to vortex formation should counteract this effect. It is more likely, therefore, that thermal energy absorbed by the additive at or near the surface is responsible for the observed behavior, in agreement with the data of reference 17 on the extinction of solid-propellant burning.

The melting and agglomeration of aluminum on the surface (refs. 15 and 18) should make it a more effective damping agent than aluminum oxide. This is in agreement with reference 16. If this view is correct, it might be profitable to investigate the effects of thermal capacity, heat of fusion, and liquid range of additives on their damping efficiencies.

SUMMARY OF RESULTS

The effect of oxidizer particle size on resonance amplitude was studied by burning cylindrical grains of polybutadiene - carboxylic acid - ammonium perchlorate propellant in a side-vented combustor. As the proportion of ground oxidizer was increased, a sudden increase in amplitude occurred. The proportion of ground oxidizer corresponding to this change varied inversely with the length of the grain.

When a simplified model of turbulent mixing was employed, it was shown that the change in amplitude level corresponds to a fixed value of the ratio of mixing time to wave time as predicted by several theories of liquid-propellant combustion instability. The addition of aluminum produces an effect equivalent to an increase in mixing time.

Lewis Research Center
National Aeronautics and Space Administration
Cleveland, Ohio, March 27, 1964

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