Aerojet-General Corporation also found that fine (1 μ) Al_2O_3 would suppress unstable burning, and that the fine material was more effective than larger (3 and 6 μ) grades of the oxide.[4] Once again, calculations made using the Epstein and Carhart formula[2] showed some correlation with particle-size optimization effects in the Aerojet-General Corporation test configuration.

The importance of optimum particle size has been demonstrated with other materials. For example, carbon black has shown some effectiveness. However, the only material tested at the Rohm & Haas Company in the petrin-acrylate system had an average diameter of 0.03 μ, and was ineffective, undoubtedly because this size was too small for the motor geometry tested. Other sizes tended to inhibit polymerization.

Conclusions

A number of experiments showed that there are several mechanisms by which high-frequency oscillatory burning may be suppressed or its magnitude substantially decreased. The most significant finding is that the results of motor firings correlate well with photographs of the burning process of those additives that liberate large amounts of energy. It appears possible to make predictions of the effectiveness of these additives.

It has been shown that certain additives, e.g., Silon S, Fe_2O_3, and carbon black, can absorb sufficient sonic energy to eliminate high-frequency oscillations. However, the speculation that the nucleation of the Al_2O_3 formed from burning aluminum gives particles of the optimum size for damping by the particle-drag mechanism is not supported, because the region of the additive's burning appears to be the determining factor.

Increasing the heat required to vaporize a unit mass of propellant can decrease the magnitude of oscillation, and the effect of an additive with high thermal conductivity has been investigated, but a higher concentration is required before much of an effect will be realized from either of these approaches.

Because the extent of the attenuation of sound by a suspension is affected by motor geometry and the resulting characteristic frequencies, incorporation of an additive that suppresses oscillations by releasing energy appears to be a more practical method of eliminating combustion instability than one based solely on particle damping. Moreover, the photographic method similar to that employed here offers an excellent method of predicting the ability of a specific additive to suppress oscillations in a given propellant system.

Acknowledgments

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Early Investigations of Solid-Propellant Combustion Instability in Russia

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Nomenclature

A	= oscillation amplitude
c	= heat capacity
K_1	= coefficient of increment per unit burning area
K_2	= coefficient of oscillation absorption
P	= pressure
q	= heat of chemical reaction in reactionary layer of condensed phase
S	= initial combustion area
T_o	= starting temperature
T_s	= the temperature of condensed phase surface
t	= time
t_c	= relaxation time of condensed phase
t_g	= relaxation time of gaseous flame
u	= combustion rate
v	= combustion-product flow rate
x	= coordinate
α	= rate of decrease in burning area
β	= temperature coefficient of combustion rate, $1/T_0(\partial u/\partial T_0)_P$
ρ_g, ρ_c	= densities of gaseous and condensed phases, respectively

Introduction

DESIGNING and improving solid-propellant rockets, both before and during World War II, resulted in the discovery of phenomena that were inexplicable within the framework of views and knowledge available at that time. These phenomena included 1) anomalous pressure rise when long charges were employed, known today as erosive combustion; 2) spontaneous combustion damping, known as anomalous combustion or low-frequency motor rocket combustion instability; and 3) secondary peaks of pressure that arise during propellant burning in the combustion chamber, today known to arise from high-frequency instability. To explain these phenomena, it was necessary to create new modern theories for propellant combustion and internal ballistics of solid-fuel rockets. Contrary to the classical theory that focused primarily on steady burning, these new theories required the study of unsteady burning and its interaction with gasdynamic and heat processes in a rocket motor. Russian scientists contributed significantly to developing these theories. In particular, during World War II, Zel'dovich elaborated a theory of unsteady propellant burning,[1,2] as well as theories for erosive combustion[2,3] and low-frequency combustion instability,[2,4] which retain much of their validity today. Experimental research on erosive and anomalous combustion (i.e., low-frequency combustion instability), which confirmed the Zel'dovich theories, were conducted by Leipunskii[5] and other Russian scientists. This Note describes some of the early experimental combustion studies in Russia on the third class of the previously noted phenomena of modern internal ballistics of solid-propellant rockets, that is, high-frequency unsteady propellant burning.

Discussion

Peaks in the pressure of a rocket combustion chamber, which appear some time after ignition, were detected in 1938 by Dernovoy.[6]

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These researchers (as well as others) determined the propellant parameters and burning conditions that were necessary to generate these pressure peaks and they studied how these conditions influenced peak characteristics. In these experiments, the chamber pressure (including pressure peaks) was recorded by rather low-frequency techniques that prevented detection of the high-frequency oscillations. It was found that the composition of propellant and its caloric content strongly influenced combustion stability, propellants with higher caloric content being more inclined to unstable combustion. It also followed from these experiments that the length-to-diameter ratio of the charge must be diminished, or pressure must be increased, to stabilize burning. It also was found that, at unstable burning, the average combustion rate in the channel exceeded the combustion rate of the outer propellant cylindrical surface. Various forms of round and periodic cracks appeared in a somewhat sinusoid manner along the axis of the channel.

Because these peaks in a pressure-time diagram appeared first when cooled propellant was burned (-10 to -70°C), their occurence was explained by charge cracking, as well as nonsimultaneous burnout of propellant components. Later, these peaks were also found at temperatures from 0 to $+50^\circ$C, where the propellant is not brittle and does not crack. As a result, it became evident that the main reasons for the appearance of these peaks are identical throughout the entire temperature range. Because these pressure peaks were not thought to result from acoustic phenomena at that time, they were explained by nonsimultaneous burnout of propellant components and thermal explosion of incomplete burning products.

Further experiments were conducted to investigate unstable burning in cases in which both relatively inert and chemically active distinct substances were present as admixtures. The development of this line of investigation was stimulated by the fact that, during World War II (about 1942), magnesium oxide (MgO) was included to chemically stabilize double-base propellant by absorbing nitrogen oxides evolving from the propellant. It soon was noticed that the presence of MgO eliminated the pressure peaks. Other thermally stable particles also were found to eliminate this kind of combustion instability, whether they were included in the propellant composition or formed as the result of burning (like Al_2O_3). At that time, their action was explained by the fact that a high-melting-point grid arose on a propellant burning surface, which accumulated the heat, as well as by a variety of other effects.

For the first time, the acoustic character of this unstable combustion behavior was described in the early works (1949–1953) of the American scientists Grad,[7] Smith and Sprenger,[8] and Wimpress.[9] These works were reviewed by Price.[10]

In Russia, the acoustic nature of unstable solid propellant burning in rocket chambers has been investigated since 1958 by the present author and his colleagues at the Moscow Institute of Chemical Physics. In the Russian experiments, the high-frequency oscillations (3–100 kHz) were measured by the waveguide transducer. In 1959, longitudinal and tangential oscillation modes were observed in the channel of a double-base propellant charge during unstable burning.[11,12] In 1960, these studies were expanded to examine unstable combustion more closely.[11] Specifically, tests were conducted to investigate how the charge geometry, pressure, heat of combustion, and initial propellant temperature influenced the amplitude and spectrum of acoustic oscillations, as well as the magnitudes of the pressure peaks (Fig. 1). It was shown theoretically and experimentally somewhat later[13] that at pressures above the critical levels for soft self-excitation of auto-oscillations lies a region of the rigid (nonlinear) excitation (Fig. 2).

Zel'dovitch's theory of unsteady propellant combustion[1,2] was expanded to include the case in which a surface layer of overall-exothermic reactions exists in the immediate vicinity of a burning surface and has an influence on combustion. This theory is based on the fact that t_g is substantially shorter than warmed-up t_c, because $t_g/t_c \approx \rho_g/\rho_c \ll 1$. Accordingly, processes in the gaseous phase are considered as being in a quasisteady state. The temperature of the condensed-phase surface has been taken to be constant, $T_s = \text{const}$. Theory[1] predicts that the combustion is stable when $\beta(T_s - T_0) < 1$, whereas for $\beta(T_s - T_0) \geq 1$ (i.e., at low temperatures), the combustion is unstable. It has been found[11] that if the chemical reaction occurs in the condensed phase the criterion for stable propellant combustion is $\beta(T_s - T_0) - q/C = 1$. The heat q produced in a condensed phase stabilizes combustion. The criterion that incorporates reactions in the condensed phase provides better agreement with the experimental data.

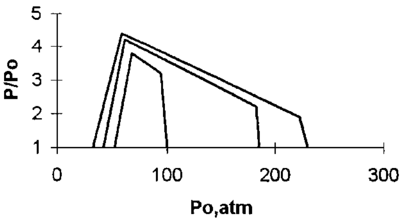

Fig. 1 Heights of peaks of pressure (P/P_0) as the function of the pressure P_0 immediately preceding the peak. Here $\Delta P = P_m - P_0$, where P_m is the maximal pressure in the peak. Charges consist of the double-base propellant A; Exterior diameter = 40 mm; channel diameter = 8 mm; charge lengths = 150 mm (outer curve), 112 mm (middle curve), 75 mm (inner curve).

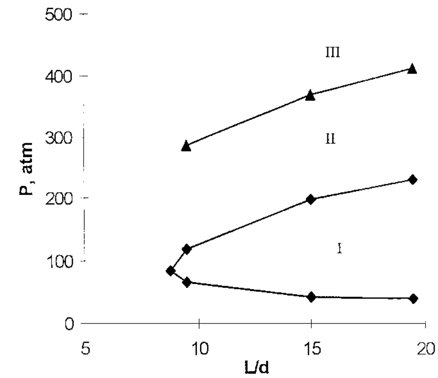

Fig. 2 Regions of unstable combustion of the propellant A: soft self-excitation (I), rigid excitation (II), absolute stability (III); P pressure, L/d = ratio of the charge length (L) to the initial channel diameter (d).

Propellant combustion in the nonsteady flow of burning products, i.e., nonsteady erosive combustion, has been examined with the assumption that the relaxation time of the gas boundary layer is less than the relaxation time of the propellant condensed phase. To this end, the transformation used in theory,[1] namely,

$$U(P, T_0) \rightarrow U\left[P, \left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)_s, T_s\right]$$

has been generalized to the case in which products that flow along a burning surface influence the rate of combustion:

$$U(P, V, T_0) \rightarrow U\left[P, \left(\frac{\mathrm{d}T}{\mathrm{d}x}\right)_s, T_s\right]$$

The erosion coefficient was demonstrated to be a function of frequency; thus, it exceeds the steady-state value for fast-changing gas flow rates.

On the basis of the unsteady combustion theory,[1] the expression for acoustic conductivity of the propellant combustion surface was evaluated as the function of combustion parameters and combustion conditions.[11] It was shown that the dimensionless acoustic conductivity χ of a burning surface is on the order of 10^{-3}–10^{-2} and depends weakly on oscillation frequency.[11] The small magnitude of χ compared to unity is a crucial factor in understanding the essence of unstable propellant combustion and in exerting control over this process. For this reason, the sound radiation from charge channel and nozzle and the sound absorption in combustion products (in the presence of condensed particles) compete with sound amplification in the combustion zone to determine the overall stability.

Applying known concepts from the acoustics of aerosols to unstable propellant burning provides a quantitative method for predicting the stabilizing action of condensed particles produced by the combustion.[11] The damping coefficient is a function of oscillation frequency, size, and density of particles. Damping estimates, which agree with experimental measurements, revealed that the condensed particles can effectively dampen unstable combustion. These concepts of the acoustic nature of unstable combustion explain peculiarities of combustion in the flow stagnation area. Acoustic waves are additionally strengthened in the zone where propellant burns out. Outside the stagnation area the burnout zone is destroyed because of quasisteady-state flow of hot products of combustion.

The most important experimental problem was to definite the sound amplification coefficient of the burning surface. In 1960, a method for measuring acoustic conductivity of a burning-propellant surface was developed.[11] This method, referred to as the method of threshold oscillations, allowed one to determine the amplification coefficient at a threshold of the auto-oscillations. This threshold is achieved when total sound amplification is equal to the sum of acoustic losses. In 1961, we measured acoustic conductivity of a surface of propellant combustion by this method, presumably for the first time.[11] With the assumption that, for longitudinal modes, the acoustic energy was lost mainly by sound radiation from the charge channel, and using a theoretical coefficient of that sound radiation, we determined the acoustic conductivity for a double-base propellant. This coefficient was of the order of 10^{-3} at 70 atm pressure.[11] Later measurements using this same propellant were made at a higher frequency (50 kHz) and at pressures from 50 to 100 atm using this method of threshold conditions. The resulting acoustic conductivity also turned out to be of the order of 10^{-3}. In the latter case, auto-oscillations perpendicular to the surface arose in a gap between two parallel plates of propellant. The main acoustic losses were in the propellant plates because the combustion products contained no condensed particles. These losses were calculated from the known equations of acoustics, using predetermined acoustic propellant parameters.[14]

Based on the technique for measuring acoustic conductivity of propellant in T-burner by Horton and Price,[15] the variable-surface techniques to measure this quantity was proposed in Russia. Later, this method was described in Ref. 14. The T-burner method[15] uses planar parallel propellant plates, the area of which does not change with time. In this case, it is necessary to measure growth rates of the oscillation amplitude after ignition and after the sample has burned out, to estimate acoustic conductivity of burning-propellant surface. The Russian experiments use propellant samples with a time-varying combustion area, namely cylinders that can burn only on the cylindrical surfaces (increasing area with time) or wedge-shaped plates attached to the T-chamber end-wall (decreasing area with time). In these cases, the oscillation amplitude A changes like this:

$$\frac{\mathrm{d}A}{\mathrm{d}t} = [SK_1(1 + \alpha t) - K_2]A$$

The increment of the oscillation amplitude increase, $(SK_1 - K_2) + \frac{1}{2}S\alpha K_1 t$, comprises a constant and a linear term with time. The linear term is proportional only to the incremental coefficient of burning surface. Measuring the magnitude of the linear term permits the coefficient of sound increment per unit area, which is related to the acoustic conductivity, to be measured.

This technique was implemented in two variants: in a T-burner and in a double Helmholtz resonator.[14,16] Both methods were used to measure the acoustic conductivity of double-base propellants N (890 cal/g) and A (1100 cal/g) at various frequencies and pressures. The results are shown in Fig. 3. Using the Helmholtz resonator allows one to make measurements at lower frequencies than those in the similar-sized T-burners. In 1962, the auto-oscillations in the channel of a propellant charge were recorded using film shot through a window in the rocket chamber.[17] There are "stars" on film shots with an even number of slots (from 2 to 12) that are nodal diameters of standing tangential oscillations, as well as nodal circles of radial oscillations and whirlwinds (Fig. 4).

An investigation was made of the interaction between oscillations in the gaseous phase and oscillations in condensed phase. For this purpose, longitudinal and tangential components of oscillatory rate were measured in the channel of a burning charge.[18] At nearly the same position, tension oscillations in a body of propellant were determined using a piezosensor. The maximum amplitude of auto-oscillations in the gas phase was shown to arise under conditions in which the ratio of the amplitude of the solid-phase oscillations to the amplitude of the gas-phase oscillations is minimal.[18]

Since 1960–61, experimental research on unstable combustion of propellant by measuring acoustic oscillations also were carried out in Russia by other research groups, but their results are not reflected in the given paper.

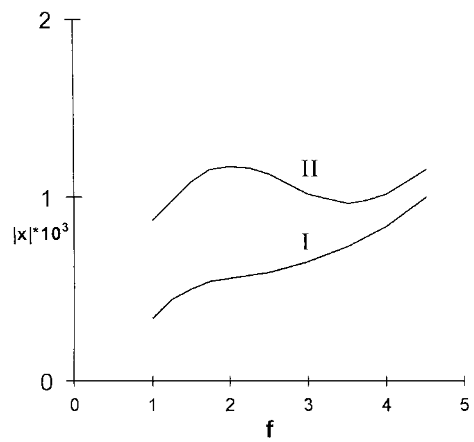

Fig. 3 Acoustic conductivity of the burning surface of the double-base propellant N χ is the dimensionless acoustic conductivity at pressure 25 atm (curve I) and at pressure 55 atm (curve II).

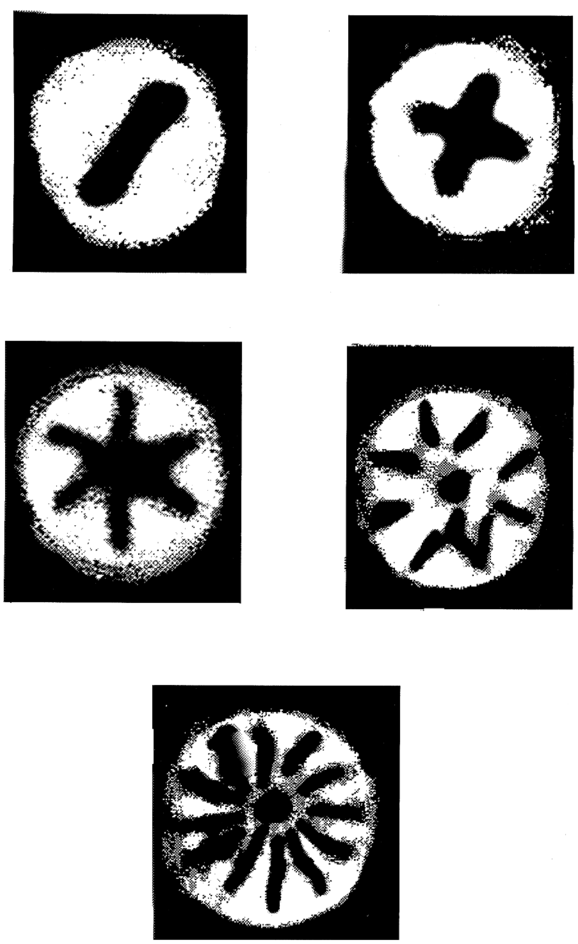

Fig. 4 Film of unstable combustion in the channel of the propellant *A* charge. One can see eight-angle star, i.e., four nodal diameters of the tangential mode of auto-oscillations.

Theoretical research on this phenomenon in Russia are reflected in the review by Novozhilov.[19]

Conclusion

This note describes some of the early combustion studies in Russia on high-frequency unsteady propellant burning. Actually, they began in 1938 when peaks in the pressure of a rocket combustion chamber were detected: a short time later, the limits for their existence depending on propellant parameters and burning conditions were investigated. It was also shown that thermostable particles eliminated these peaks. However, at that period, the reasons for these phenomena were not established. Acoustic character of this combustion instability was described by American scientists.

During 1958–1962 at the Moscow Institute of Chemical Physics, experimental and theoretical investigations were performed on various aspects of high-frequency (acoustic) combustion instability. In particular, the theory of interaction of acoustic waves with the propellant combustion surface was developed, and the coefficient of the sound reflection from a burning surface was measured experimentally.

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Underwater Incineration of Heterogeneous Propellants

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Introduction

DISPOSAL methods for processing demilitarization of ammunition are being addressed in many countries throughout the world. Processes for recycling certain materials, such as brass from cartridges, are technically simple, well developed, and financially very attractive. Processing other components, such as rocket propellants, however, remains an unsolved problem. Initial studies in developing an ecological and economically friendly method for processing heterogeneous solid rocket propellants are described. The basic approach is to incinerate the propellant in a neutralizing solution that transforms the ecologically undesirable combustion products into substances that commonly appear in nature. This approach offers the potential advantage of simultaneously eliminating gaseous hydrogen chloride and trapping the aerosols of aluminum oxide.

Method Selection and Description

A number of possible methods for destroying common explosives exist. Selecting the most effective method for a particular explosive depends on many factors. These factors include the quantity of propellant to be processed or recycled; the original production price of ingredients, such as ammonium perchlorate; the obtained price for products of the recycling process; the cost of facilities for the disintegration and extraction process or burning and fume-scrubbing process and so on. Some possible methods are outlined below.

1) *Direct combustion or detonation:* This method is very simple, but because of the production of harmful substances, it is unacceptable for ecological reasons.

2) *Physical–chemical processing:* Mechanical grinding of the propellant followed by solvent separation of AP and subsequent recycling for reuse. The major concern with this method is the processing of the insoluble remnants consisting of binding agent, additives (primarily aluminum), and nonextractable oxidant. Disposing of these materials presents the same problems and hazards as disposing of the original propellant. For example, experiments conducted in the Czech Republic show that the reaction of aluminum with cutting water can be very dangerous and can cause a disaster.

3) *Recycling materials as explosives:* Reuse of the original substances or their components in production of explosives for industrial purposes. This method does not eliminate collection of the poisonous substances and is therefore unacceptable.

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