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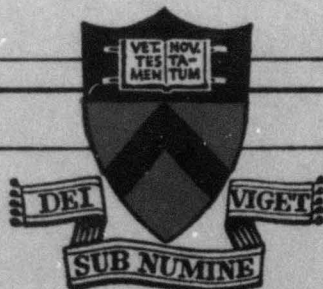
ROLE OF ALUMINUM IN SUPPRESSING INSTABILITY
IN SOLID PROPELLANT ROCKET MOTORS

Aerospace & Mechanical Sciences Report No. 840 ^{AMS-}

by

Martin Summerfield and Herman Krier

July 1968



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Transmitted by

Martin Summerfield

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Principal Investigator

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Technical Supervisor for this program is Dr. Bernard T. Wolfson, Propulsion Division, Directorate of Engineering Sciences, Air Force Office of Scientific Research.



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ABSTRACT

Combustion instability in large high-performance solid propellant rocket motors was a major difficulty standing in the way of practical development in the U.S., until the discovery in 1956, after much testing, that powdered aluminum added to the propellant suppresses instability. This discovery made it possible for the national solid rocket program to move forward rapidly. The exact mechanism of the action of aluminum has long been a mystery. Aluminum is almost unique in its effectiveness; magnesium is the only other additive known to be equally effective. A theory is advanced in this paper, based on the previously published KTSS theory of non-steady propellant burning, to the effect that it is the melting of the aluminum on the burning propellant surface, not its particular burning characteristics, that reduces the amplitude of the propellant burning rate fluctuations. The melting leads to the formation of a thin layer of molten aluminum that, through its thermal inertia, reduces the temperature response of the surface and therefore the burning rate response. The stabilizing mechanism is powerful: a very thin layer with only partial coverage of the surface produces a very large reduction in the acoustic admittance. Unfortunately, aluminum is objectionable (magnesium, too) because it produces smoke, flash, and ions in the exhaust jet. The paper concludes with an assessment, on the basis of the KTSS theory, of the possibility of ever finding an equally effective substitute and with some directions for investigation.

I. Speculations About The Role of Aluminum

Powdered aluminum metal is widely used as an effective propellant additive for the suppression of oscillatory combustion instability in solid propellant rocket motors, but the reason for its effectiveness has long been a matter for conjecture. In early reports^(1,2) it was hypothesized that the aluminum alters the structure of the combustion zone in some beneficial way so as to reduce the oscillatory "drive" of the flame, that is, so as to reduce the burning rate/pressure response ratio, but the actual mechanism eluded description. More recent studies⁽³⁻⁶⁾ have focused on a different hypothesis, that the stabilizing action of the aluminum is simply the result of an increase in the acoustic damping of the motor, achieved by the production of a dusty combustion gas containing fine particles of aluminum oxide in the central gas core.

The latter may be effective in some cases, especially at high frequencies of 5000 cps and more; however, experimental attempts to stabilize a rocket motor by adding not metallic aluminum but simply fine aluminum oxide to the propellant have met with only scant or doubtful success. Metallic aluminum in the propellant, on the other hand, functions effectively at the more troublesome lower frequencies (500-2000 cps), as shown by combustion oscillation tests in T-burners⁽¹¹⁾, where it reduces the measured response ratio markedly. So the suspicion lingers that the main effect of aluminum in stabilizing a motor is on the driving process and not so much on the damping process.

It is the purpose of this paper to present speculatively a specific mechanism that would support the first viewpoint, that the metal additive reduces the burning rate/pressure response ratio. It will be shown theoretically that, when the aluminum particles melt and agglomerate on the propellant surface (as they are seen to do), the resulting thin metal layer can provide a thermal inertia that can greatly reduce the temperature oscillations of the burning propellant surface in the presence of pressure oscillations. The effect is spectacular. It takes only a very thin layer, about 10-20 microns, to do the job.

This surface phenomenon, melting and agglomeration, is observed in all cases during combustion of aluminized propellant. After studying close-up motion picture sequences, Watermeier, et. al.⁽⁷⁾ stated, "In some instances, especially at high pressure and high initial aluminum

concentration, the propellant burning surface would appear to be almost covered with a blanket of molten aluminum." With such a condition in mind, it is logical to try to take this blanket into account in the formulation of a non-steady flame theory for aluminized propellant, to see how it affects the dynamic burning rate response.

This analysis is restricted, by the basic assumption of a quasi-steady gas phase, to the low frequency range, where the amplification, if any, is due to the thermal lag in the solid phase; amplification at higher frequency ($> 5000\text{cps}$) may be possible as a result of nonsteady processes in the gas phase, but no acceptable theory has yet been developed for that domain. Moreover, it is believed, on the basis of empirical evidence, that the most disturbing region of solid rocket motor instability is in the low frequency range, so the present analysis is appropriate to the problem.

(A note on terminology: In some writings, "low frequency" is a term applied to oscillations of the order of 100 cps or lower, and corresponds to oscillations of chamber pressure without wave motions in the gas core (like a Helmholtz resonator), and "intermediate frequency" is applied to frequencies of several hundred to a few thousand cps, where the lowest resonant wave motions occur in the chamber (like an organ pipe). Our term "low", however, refers to the source of the amplification, that is, to oscillations arising from solid phase behavior; it so happens that it encompasses both the "low" and "intermediate" frequency ranges of the usual rocket motors.)

(Before entering into the present analysis, it is interesting to note that a different interpretation of the non-steady solid propellant flame zone has been adopted in the series of papers written recently by Novikov and Ryazantsev in Moscow.⁽¹³⁾ The series started with some earlier theoretical ideas of Zel'dovich. As a consequence of the different model, they have been led to a different interpretation of the source of combustion instability. We do not agree with that physical model; our reasons are expressed best in the KTSS paper^{(8).})

II. Theoretical Reduction of Acoustic Admittance

The combustion model described in Ref. 8, which we shall denote here as the KTSS Model, will be altered to include the thin molten layer of aluminum. Figure 1 shows schematically the regions to be analyzed. The thickness, ℓ , is probably less than 100 microns and may well be only of the order of 10-20 microns; also, for this analysis, the area coverage will be taken to be complete. We have no way of predicting this thickness, but we note that both the thickness and the fraction of surface area covered must be in some way the result of a dynamic balance between the rate at which aluminum powder is brought to the surface and the rate of ejection of molten aluminum into the gas phase. It will depend, therefore, on the particle size of the aluminum, on the weight fraction of the aluminum in the propellant, on the linear burning rate (i.e. on the pressure), on the structure of the burning surface (smooth or rough, solid or molten, therefore on fuel type and oxidizer particle size), and on the gas velocity past the surface (rate of removal by shear).

It is assumed that the gas phase flame structure is essentially unaltered by the aluminum layer on the surface or by the ejected droplets of molten aluminum, and that the fuel and AP pyrolysis products bubble through the melt to sustain the normal granular diffusion flame. (9,10) A one-dimensional analysis is retained.

The interfacial heat balance condition at 2 is

$$-\lambda \left. \frac{\partial T}{\partial x} \right|_{Al \text{ @ } 2} = \dot{q}_g = \frac{\Phi(\tau(t))}{\tau} \quad (1)$$

where $\dot{q}_g$ = instantaneous heat flux from the gas phase.
 $\Phi(\tau)$ depends on the kinetic rate of reaction in the gas phase, as explained in Ref. 8.

At position 1,

$$-\lambda \left. \frac{\partial T}{\partial x} \right|_{s.p.} = -\lambda \left. \frac{\partial T}{\partial x} \right|_{Al \text{ @ } 1} + \tau \rho \tilde{Q}_s \quad (2)$$

To describe the temperature field inside the aluminum layer accurately, the full non-steady heat conduction equation in a moving medium would be required. However, as a result of the large thermal conductivity of the molten aluminum, the temperature profile can be taken as nearly flat, although oscillating in time. Then the partial differential equation for the temperature of the aluminum becomes a lumped-temperature ordinary differential equation, and the temperature of the aluminum layer (taken uniform) is then the same as that of the propellant surface. (It can be shown that this approximation is valid if the burning rate is of the order of 1 cm/sec and the frequency of oscillation is greater than about 10 cps.)

$$T_{al} \approx T_s \quad (3a)$$

$$(c \cdot l \cdot \rho)_{al} \frac{dT}{dt} \Big|_{al} = \frac{\Phi[p(t)]}{r} + \lambda \frac{\partial T}{\partial x} \Big|_{s.p} + \rho r \tilde{Q}_s \quad (3b)$$

$\tilde{Q}_s$ = is the overall heat release per unit mass at the propellant surface due to exothermic decomposition, modified to take into account the endothermic melting of the contained aluminum.

The above equation (3b) is then rewritten (in non-dimensional form) as the interior heat flux boundary condition at the propellant surface:

$$\frac{\partial \theta_s}{\partial x} = \frac{-P^{2n}(P_m^n - H)}{R} - HR + C \frac{\partial \theta_s}{\partial \tau} \quad (4)$$

where

$$C = \frac{(c \cdot l \cdot \rho)_{al} \cdot r_o}{\lambda_{s.p.}} \quad (5)$$

The notation used in (4) above is that used in the KTSS theory (8), and appropriate definitions are given again for convenience in the Nomenclature.

For small pressure disturbances the temperature in the solid and the burning rate will be perturbed from their mean values, so that:

$$\begin{aligned} P &= 1 + P' & \theta &= \bar{\theta} + \theta' \\ R &= 1 + R' & \frac{d\theta}{d\tau} &= \frac{d\theta'}{d\tau} \end{aligned} \quad (6)$$

For oscillatory perturbations, we assume

$$\begin{aligned} \theta' &= \tilde{\theta}(x) e^{i\omega\tau} \\ \frac{d\theta'}{d\tau} &= i\omega\theta' \end{aligned} \quad (7)$$

Then, as shown in the KTSS theory, the linearized heat equation for the solid

$$\frac{\partial \theta'}{\partial \tau} - \frac{\partial \theta'}{\partial X} - \frac{\partial^2 \theta'}{\partial X^2} + m e^{-X} \theta'_s = 0 \quad (8)$$

must be solved for θ'_s , but now with the modified boundary condition at $X = 0$ (i.e. with the aluminum thickness parameter, C):

$$\frac{\partial \theta'_s}{\partial X} + (A - i\omega C) \theta'_s = B P' \quad (9)$$

where

$$\begin{aligned} A &= (2H - 1) m \\ B &= (2H - \frac{1}{m} - 2) n \\ P' &= (\Delta P) e^{i\omega\tau} \end{aligned}$$

The method of solution of (8), with the boundary condition (9) and the boundary condition that $\theta' \rightarrow 0$ as $X \rightarrow \infty$, is quite similar to that described in Ref. 8 and will not be repeated here.

The intrinsic stability boundary discussed in the KTSS theory now occurs when the pyrolysis exponent m and the surface heat release H fit the modified characteristic equation:

$$2CA^3 + (1-3C)A^2 + [C(1-4m-mC)-1]A = m(2+C^2m-C-2C^2) \quad (10)$$

The modification lies in the presence of C . Solution of (10) gives $m = m_1[A]$ (or $m = m_2[H]$) on the boundary and is shown in Figure 2. The meaning of this boundary is that it divides the (m, H) plane into two regions, one corresponding to propellants that burn stably in the presence of small perturbations of pressure, the other corresponding to propellants that can be excited to wild burning rate behavior by a small pressure perturbation. (The latter are dynamic-ally unstable, although they do satisfy the equations for steady-state burning.)

It is to be noted that the region of stable burning of the propellant is considerably enlarged by the presence of the thermal inertia at the surface, for typical values of the aluminum thickness parameter, C . For example, with no aluminum melt ($C = 0$), the surface temperature perturbation grows without bound for surface heat release $H = 0.83$ and pyrolysis exponent $m = 6$. With aluminum thickness $C = 1$ (about 10 to 20 microns) the instability does not occur for $m = 6$ until H reaches the high value of 0.90. Physically, since H is a measure of the heat release at the surface, this means that the propellant formulation can accept a very high percentage of ammonium perchlorate or other exothermic constituent before it begins to display intrinsic instability. In fact, such high percentages are usually impractical, since they cannot be processed by acceptable techniques. That means that, as far as practical formulations go, $C = 1$ stabilizes them.

In passing, it should be noted that it is necessary to avoid a propellant formulation that even comes near to the intrinsic instability line in Fig. 2. As shown in Ref. 8, when a propellant with this tendency to intrinsic instability

is coupled to a rocket chamber that has a resonant frequency in the sensitive range, combustion instability can occur even if H and m are on the safe side of the line. Nevertheless, the thermal inertia of the aluminum layer is still a stabilizing effect.

The real part of the acoustic admittance for an aluminized propellant completely covered with the melt is shown in Figure 3 as a function of the non-dimensional frequency, $\bar{\omega}$ for $C = \frac{1}{2}$ and $C = 1$. The calculation was similar to Eq. (27) of Ref. 8. If the effect of the mean outward flow from the surface is neglected--a small correction usually--the propellant combustion zone can amplify an incident pressure wave when $\text{Re}[Y] > 0$. Figure 3 shows that this amplification region along the frequency abscissa is markedly narrowed when C is nonzero. In fact, for $C > 2$, and for the particular combustion parameters listed, there will be no frequency region at all where amplification may occur.

For only partial coverage of the burning surface by the aluminum melt, the real part of Y was calculated with the one-dimensional assumption that the mass flux perturbation, $\dot{m}$, is given by:

$$\dot{m}' = (1 - \epsilon) \dot{m}'_{\text{uncov.}} + \epsilon \dot{m}'_{\text{cov.}} \quad (11)$$

ϵ is the fraction of surface covered with the melt. Figure 4 gives $\text{Re}[Y]$ vs. $\bar{\omega}$ when $\epsilon = 0.5$. It is seen that even a half-coverage of the surface is very effective.

It was mentioned above, right after Equation (3), that the melting of the aluminum as it comes to the burning surface has a second beneficial stabilizing effect on the dynamic burning rate response. The endothermic melting of the aluminum occurs at nearly the same place as the exothermic decomposition of the propellant, i.e. on the surface, and so the effect of the melting is to depress the value of H , thus shifting the propellant toward the stable region of Fig. 2. The magnitude of this reduction is proportional to the concentration of aluminum in the propellant; we calculate a reduction of about $\frac{1}{2}$ percentage point in H for each percent of added aluminum. This stabilizing effect was discussed in Ref. 8, when it was thought that the reduction

in H was the main effect of aluminum addition; we had not thought of the thermal inertia effect at that time. Now we see that it is the thermal inertia that may be the more potent of the two effects.

When these two effects are put together into a single calculation of acoustic admittance for an AP propellant, we get the result shown in Fig. 5. The particular values of area covered and thickness of melt are just assumptions, but they seem reasonable in the light of the motion pictures we have studied (7). The stabilizing effect is striking.

We conclude speculatively that aluminum suppresses instability because it melts on the surface of the burning propellant and not because it burns.

III. History of the Combustion Instability Problem

It is a matter of historical interest that reports of the occurrence of unstable burning in solid propellant rockets began to appear in the U. S. about 1941-2, just about when pressure sensors and recording instrumentation with high resolution capability began to become available in rocket development laboratories. However, the laboratories that reported combustion instabilities in the early '40's were only those concerned with nitrocellulose-nitroglycerin propellants; the laboratories developing perchlorate-base composite rocket propellants never encountered instability. Thus, the NC/NG rocket designers began as a routine matter to incorporate resonance rods, grain perforations, and other schemes in the motors to damp out instability; the perchlorate rockets on the other hand, contained no such devices.

However, those early perchlorate rockets were made with potassium perchlorate, not ammonium perchlorate. Although AP would have been desirable as a replacement just to get rid of the annoying KCl smoke (more than one-third of the ejected mass by weight), it was not yet available industrially. Then, about 1945-46, when industrial sources of AP emerged and the switch-over from KP to AP took place, combustion instability began to enter the perchlorate rocket industry just as in the NC/NG double-base industry. (This early history can be put together from various published sources, such as the pages noted in the four books cited in Ref. 12, but it is well known to the senior author who participated in some way in all of it, since 1940.)

Our interpretation of this bit of history from the standpoint of combustion mechanism is as follows, in the light of what we have learned in developing the KTSS theory. Dynamic instability of a rocket engine in the 100-2000 cps frequency range depends on the existence of a strong exothermic reaction at or close to the pyrolyzing solid surface. A nitrocellulose-base propellant, especially when energized by the addition of a large percentage of nitroglycerine, has this property to a strong degree, and so such rockets show marked tendencies toward instability. An AP-base propellant has the same characteristic (the heat release in this case comes from the exothermic decomposition of the AP), and so it has a similar tendency toward instability. However a KP-base propellant does not. So the early perchlorate rockets were stable. (13)

The KTSS theory shows that during incipient oscillatory burning of an AP or an NC/NG propellant the associated

oscillating heat release close to the surface causes a relatively large oscillation of surface temperature; this in turn enhances the amplitude of the initial burning rate oscillation, and by repeating itself this cycle leads to instability. The role of added aluminum, as indicated above, is to reduce the amplitude of the surface temperature oscillation and thus break the cause-and-effect cycle responsible for instability.

Prior to 1956 the large solid propellant rocket engine program in the U. S. was in trouble. It was plagued with serious instabilities, especially since the high performance type engines then intended for such demanding programs as Minuteman and Polaris required the use of high specific impulse propellant formulations. These formulations are characterized by a large value of the surface heat release parameter H , at least 0.75, which the KTSS theory shows (Ref. 8) makes them very sensitive to pressure disturbances. Mechanical damping devices (resonance rods, etc.), used with success in the 1940's in small artillery rockets and anti-aircraft rockets, proved impractical for the large Minuteman and Polaris engines.

In 1956 Mr. H. M. Shuey of the Rohm and Haas Company discovered that the addition of powdered aluminum, employed for many years in some propellants and explosives simply as a fuel additive to enhance the specific impulse or the flame temperature, had the remarkable effect of suppressing combustion instability. (For this discovery, he received the AIAA Research Award in 1964, after the technique was declassified.) Its success was dramatic and it is widely used in rockets today.

It was thought at the time that it was the peculiar burning characteristics of aluminum that did the trick, that the burning of aluminum had some beneficial effect on the gaseous flame structure. However, the only thing that high speed close-up movies ever disclosed was just the opposite, that the aluminum barely started to burn near the surface, that unburned molten aluminum was continually swept away by the combustion products into the gas core of the rocket. (Even there, the burning is only partially complete; one of the persistent problems of rocket combustor development is to achieve complete combustion of the aluminum simply to avoid loss of specific impulse.)

It was in the search to identify the burning mode of the aluminum and to discern its effect on the flame structure, that is, to find out how it works, that Watermeier⁽⁷⁾ and others, using high-speed close-up photography, noticed

the heavy melt on the propellant surface. This observation remained for many years simply a curiosity, as far as we know, until we suddenly got the idea recently that this melting is, in fact, the essential mode of action of the aluminum. The idea of the thermal inertia was a logical corollary of the KTSS theory.

IV. The Search for a Better Additive

For all its remarkable potency in suppressing instability, aluminum as an additive is a nuisance. It contributes an annoying smoke of alumina and aluminum in the jet. It enhances the visible jet flash and adds to the radiative heating of the aft end of the rocket structure. It contributes an easily ionizable component in the jet that interferes with microwave communications. We would like to find a substitute that would be less objectionable. The theory in this paper can be used as a guide to find a substitute, assuming that we look for an additive that will act in the same way to provide thermal inertia at the surface.

The following are therefore the desired characteristics for the additive we seek, in addition to the avoidance of smoke, flash, and ions: (1) The additive should be a solid with a melting point not higher than about 500-600 °C, in order to melt on the surface, to provide the desired blanket before being swept away as light particles in the gas phase. (2) The additive should have a high thermal conductivity, so that the molten layer thus formed will act as a thermal inertia without interfering with the heat feedback essential for steady burning. (3) It should produce an abundance of very small particles (solid product) in the gas core to contribute to the acoustic damping at higher frequencies. (4) The additive should serve as a fuel, in order to contribute to the specific impulse, but it should not burn on the propellant surface; it should burn up rapidly, however, in the gas core before being ejected from the rocket. (5) It should be physically and chemically compatible with the other components of the propellant and with the propellant processing method. (6) It should be cheap, available, and easy to handle.

Powdered magnesium is obviously a candidate to substitute for aluminum. It meets all of the above six requirements. And indeed, it works very well as an instability suppressant in rocket engines. Unfortunately, it is just as bad for smoke, flash, and microwave interference. Even more unfortunately, no other substance is known (and we have searched!) that can satisfy the above six requirements nearly as well as aluminum or magnesium, and indeed to our knowledge, no other equally effective additive has ever been found. It appears that what we want does not exist.

If an effective substitute is ever to be found, it will have to work by some other mechanism. But just what that other mechanism might be, we do not know. It is just conceivable, for example, that an inhibitor

might be found that would slow down the exothermic gas phase redox reaction in the AP decomposition so as to shift the heat release of the AP away from the surface out to the main flame 50 to 200 microns away (10); this would reduce H and stabilize the response. Continuing research on non-steady burning may some day give us a clue to an alternate mechanism; we might then know how to search for a substitute instability suppressant.

An alternative approach, of course, would be to replace AP with an oxidizer that is less exothermic in its decomposition, (small H), but there are not many practical choices, and the few that are known produce a much lower specific impulse or too much smoke or both. A chemical search for an instability-resistant oxidizer that would need no aluminum would be worthwhile.

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(b) Sutton, G. P., "Rocket Propulsion Elements", Second Edition, Wiley, N. Y., 1956, Fig. 9-3 on p. 310. (This illustration shows the complete design of a pre-1950 rocket made with ordinary potassium perchlorate propellant (unaluminized), and it is clear that it has no mechanical devices to suppress instability.)

(c) Barrere, M., Jaumotte, A., Fraijs de Veubeke, B., and Vandenkerckhove, J., "Rocket Propulsion", Elsevier, N. Y., 1960, p. 351. (It is stated that rockets with potassium perchlorate reportedly never displayed combustion instability, but that NC/NG rockets almost always exhibited instability (when no aluminum was added.)

(d) Zucrow, M. J., "Aircraft and Missile Propulsion", Vol. II, Wiley, N. Y., 1958, p. 524. (The author states that just prior to writing his book the instability problem in the U.S. was solved; he could not say how, but years later it was revealed that the method was the addition of aluminum.)
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NOMENCLATURE

List of symbols (Ref. 8)

c = specific heat, $\text{cal/g}^\circ\text{K}$

C = thermal inertia term, defined by Eq. (5)

$H = Q_s/c(\bar{T}_s - T_\infty)$, dimensionless heat release parameter

l = thickness of aluminum melt layer

m = power of propellant pyrolysis law

$\dot{m}$ = mass flux, $\text{g/cm}^2\text{sec}$.

n = pressure index in steady burning rate relation

$P = \dot{p}/\dot{p}_0$, dimensionless burning rate

$X = x/(\alpha/\dot{m}_0)$, dimensionless distance into the solid propellant

$\alpha = \lambda/\rho c$ thermal diffusivity of the solid propellant, cm^2/sec .

γ = ratio of specific heat of burned gas

ϵ = fraction of burning surface covered with melt

$\Theta = (T - T_\infty)/(\bar{T}_s - T_\infty)$, dimensionless temperature

λ = thermal conductivity, $\text{cal/cm sec.}^\circ\text{K}$

$\xi = (\bar{T}_s - T_\infty)/\bar{T}_s$

ρ = density, g/cm^3

$\tau = t/(\alpha/\dot{m}_0^2)$, dimensionless time

$\bar{\omega} = \omega(\alpha/\dot{m}_0^2)$, dimensionless frequency

r = burning rate, cm/sec .

$R = r/\dot{m}_0$, dimensionless burning rate

Subscripts

g = gas

s = surface

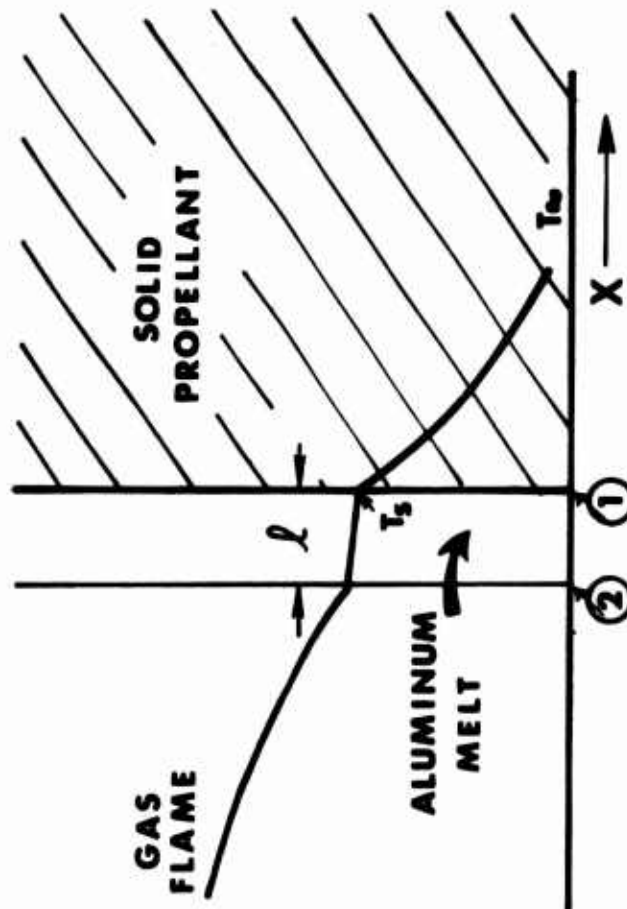
$s.p.$ = solid propellant

Al = aluminum

o = mean value during small oscillation

JP23 R 9182 68

COMBUSTION MODEL FOR ALUMINIZED
SOLID PROPELLANT BURNING



EFFECT OF THICKNESS OF MOLTEN ALUMINUM LAYER
ON STABILITY BOUNDARY (KTSS MODEL)

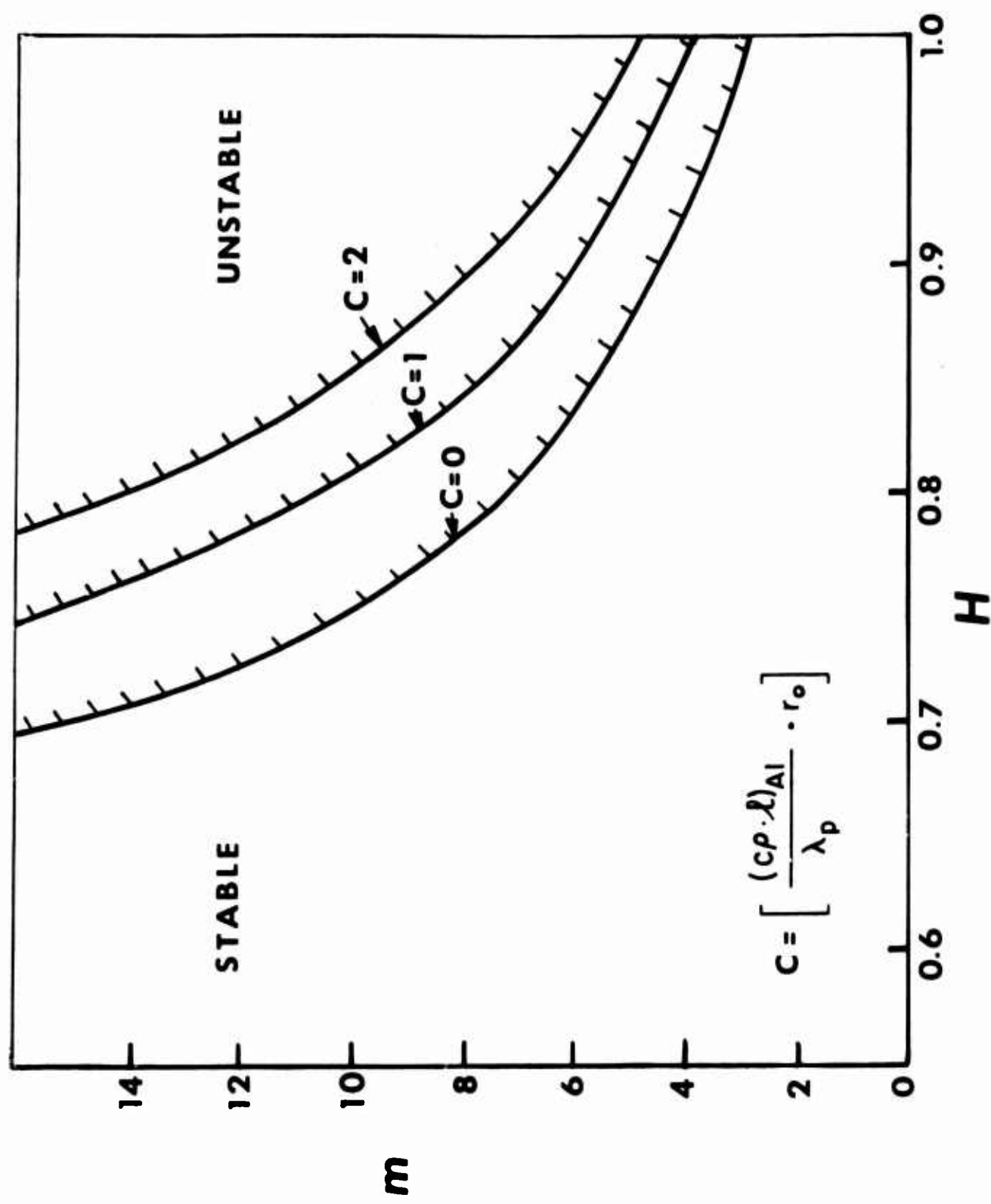
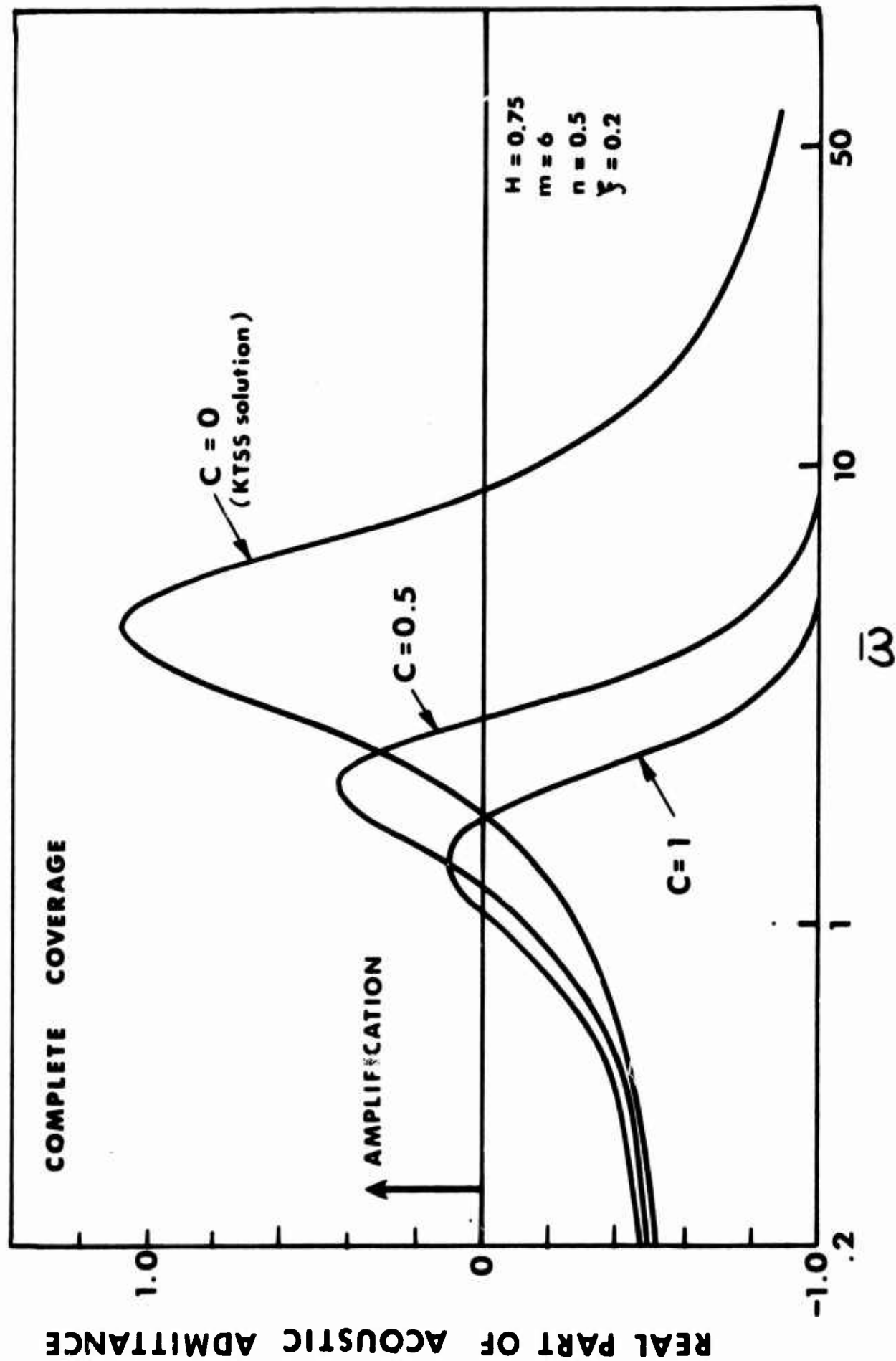


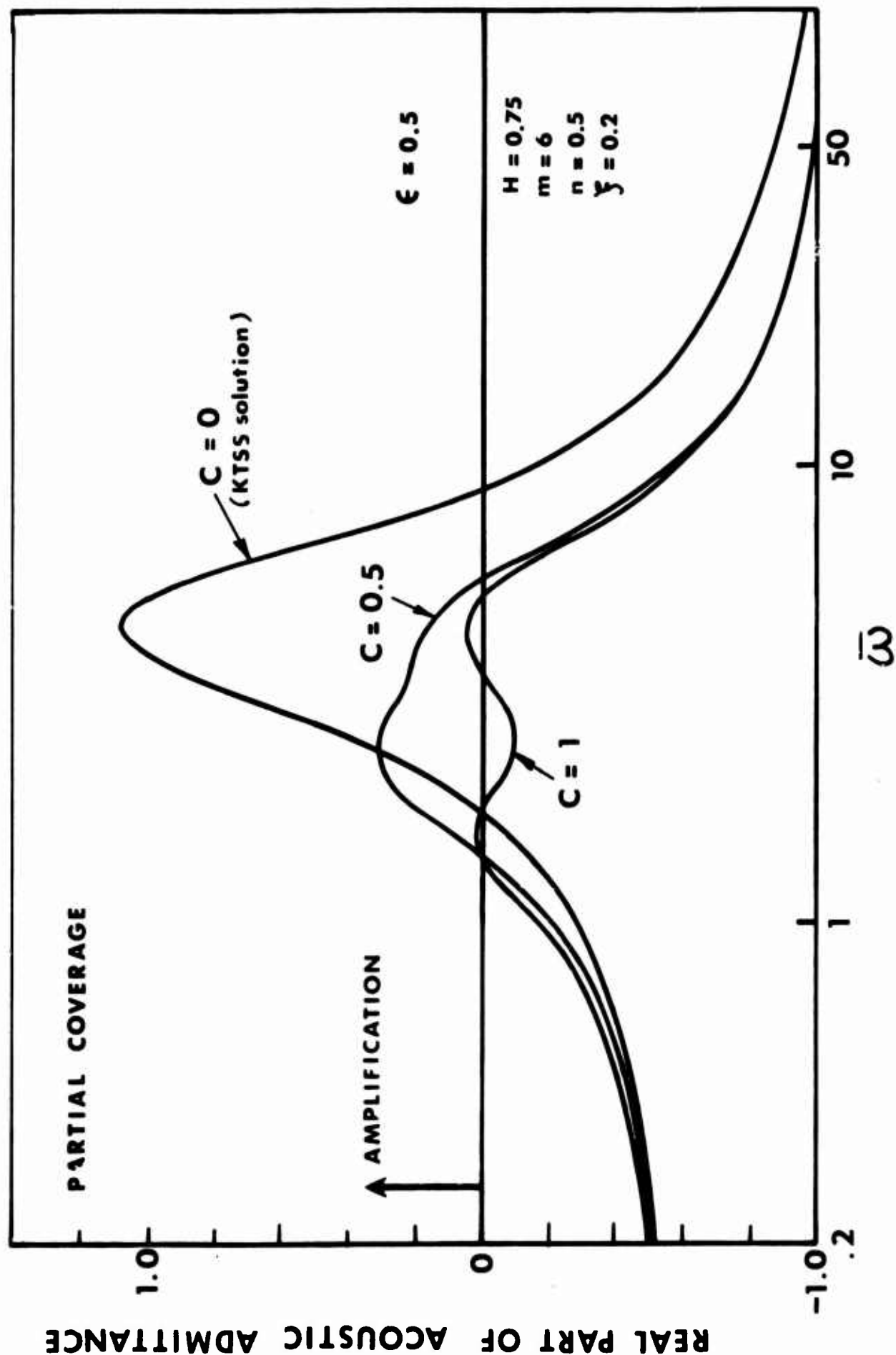
FIGURE 2

SP 23 E 4104 68

THERMAL INERTIA EFFECT OF THICKNESS OF MOLTEN ALUMINUM LAYER ON ACOUSTIC ADMITTANCE



THERMAL INERTIA EFFECT OF THICKNESS OF MOLTEN ALUMINUM LAYER ON ACOUSTIC ADMITTANCE



ESTIMATED EFFECT OF 10% ALUMINUM ADDITION AP COMPOSITE PROPELLANT

