

Comments on "Alumina Size Distributions from High-Pressure Composite Solid-Propellant Combustion"

E. W. PRICE,* J. E. CRUMP,† H. C. CHRISTENSEN‡
U.S. Naval Ordnance Test Station, China Lake, Calif.

AND

R. SEHGAL§
Jet Propulsion Laboratory, Pasadena, Calif.

A RECENT paper by Provinelli and Rosenstein¹ reported observations of the nature of condensed phase material collected on glass slides exposed near the surface of burning solid-rocket propellant samples. The paper relates the observations to a model of combustion zone processes presented in earlier reports^{2,3} and concludes that "reasonable agreement" is obtained. The body of research reported in Ref. 1-3 appears to be a major contribution to the subject of aluminum combustion in solid-propellant rocket motors, and it is probably safe to presume that this was the objective of the work.

It is the view of the present authors that the research in Ref. 1-3 has only limited relevance to rocket-motor behavior and is not in all respects internally consistent. This view is expressed here in order to minimize the impact of this work on the credibility of past and future work in this field. Although a detailed critique of the paper cannot be accommodated here, we will list the major criticisms and hope that they serve the stated purpose.

In the paper,¹ results are described from experiments in which samples of condensed phase materials were collected from combustion of pieces of solid propellant, the samples being collected by exposure of glass slides near the burning surface of the propellant. It is implied in the paper that the condensate collected on the slide is representative of final reaction products of normal propellant combustion. This conclusion is suspect for two reasons: 1) the sampling was done in close proximity to the burning surface, where aluminum combustion may not be complete and 2) the deposition of condensate on the slide is not likely to provide a representative sample of the droplets in the sampling region.

Examining first the question of completeness of combustion, the authors cite electron diffraction tests to support the claim of completeness. The result is suspect for several reasons, not the least of which is the observation earlier in the same program² of aluminum content from 25 to 50% in product samples. Although the techniques of the electron diffraction tests are not described in sufficient detail to evaluate, results could be misleading because the method is sensitive only to the surface composition of particles in the samples and would not necessarily establish whether the collected particles were completely burned. Thus the paper does not establish whether the collected samples are reaction products or partially burned droplets.

Regarding the impartiality of the sampling procedure, it seems unlikely that the fine Al_2O_3 condensate typical of aluminum combustion⁴⁻⁸ would deposit effectively on the collector slide because of the presence of convection of propellant gases. Thus there would be a tendency to collect primarily larger particles with higher mass-to-drag ratios. The sensitivity of the experiment to this sampling bias is suggested by the extreme difficulty encountered in retaining

the fine fraction of the sample during post-test depressurization of the test vessel.²

The present authors share the view of Povinelli and Rosenstein that the large particles obtained in the samples resulted from agglomeration of the original fine aluminum particles. However, we have observed⁵⁻⁹ that aluminum particles burn with a detached flame zone around the particles, in which large quantities of Al_2O_3 "smoke" are formed. Aluminum oxide is usually present also on the aluminum particle surface. The total mass of the (typically $1\ \mu$) smoke particles is of the same or larger order than the surface oxide. Although well supported by other work, this view is not consistent with the results of Ref. 1. We believe this is due to the deficiencies of the sampling procedures noted in the foregoing, deficiencies that lead to substantial exclusion of the smoke fraction and to uncertainty that the remaining fraction was typical of residual surface oxide.

Efforts are made in Ref. 1 to derive meaning from photographic studies of the combustion, particularly regarding motion of aluminum about the propellant surface during burning (the supporting agglomeration model² calls for random motion and collisions of particles on the surface, but does not discuss the mechanism by which such motion is caused). No sample photographs or statistically significant amount of original data on particle motion are presented, nor is the measuring and averaging procedure described. It was noted that data on motion of particles were obtainable only at low pressures because of the lack of resolution due to "soot." It was not indicated whether external illumination was used to reveal particles not yet heated "to self-luminosity," and no information is provided regarding the size of the particles whose motion was measured. In the experience of the present authors, the resolution of such photography would preclude observation of the motion of most of the unagglomerated ($6\ \mu$) aluminum, and even larger droplets would be only poorly resolved until they became self-luminous. Thus it seems unlikely that the particle velocity measurements reported could be typical of preagglomeration velocity of random motion, or even of motion of typical agglomerates, except near onset of ignition. In any case the absence of data at pressures above 50 psi precludes any general conclusion regarding the mechanism of agglomeration and combustion. The authors apparently did not consider the possibility that agglomeration could occur without lateral motion simply by accumulation of aluminum at sites on the surface where the binder is being pyrolyzed. This simpler mechanism is more consistent with the observations of the present authors,⁹ including those made by photography at pressures up to 1000 psi, with surface illumination by a high-intensity xenon arc; including also observations of the effect of oxidizer particle size on agglomerate size seen in the combustion; and including observations of extensive accumulation of unagglomerated aluminum on the surface of quenched propellant samples. Thus we conclude that the observations of particle motion used as a basis for comparing the agglomeration model and the particle size distribution are not sufficiently well documented to warrant acceptance, and we question that they are representative of the motion of the particles considered in the model. Further, there is reason to doubt the relevance of "collision theory" to agglomeration, and there seems to be no need to resort to it since agglomeration can occur by accumulation without mobility of the particles during residence on the propellant surface.

A further criticism concerns the relevance of the theory (reported in Ref. 2 and used in Ref. 1). The model pictures the propellant as a cubic crystal lattice array of spherical oxidizer particles of equal size, with appreciable space between adjoining particles. Combustion of the aluminum (contained in the binder in the space between oxidizer particles) is viewed as proceeding as rapidly as oxidizer gas can diffuse into the fuel region. Agglomeration is presumed to be possible by a collision process if the particle is not con-

Received April 12, 1965.

* Head, Aerothermochemistry Group, Research Department. Associate Fellow Member AIAA.

† Supervisory Physicist, Research Department. Member AIAA.

‡ Consultant on Combustion and Propulsion. Member AIAA.

§ Engineering Specialist. Associate Fellow Member AIAA.

sumed or removed from the surface before collision. A more realistic model would necessarily recognize that the propellant consists of a dense three dimensionally packed array of non-uniform oxidizer particles with most of the aluminum contained in "cusp-shaped" binder pockets bounded by surrounding oxidizer particles. In these pockets the aluminum is protected on the binder surface from oxidation by transpiration of binder products, and further protected by the presence of an oxide coating that prevents ignition until very high temperatures are reached. Thus the theoretical model is deficient in its picture of the geometry of the diffusional situation, and even more deficient in its picture of wandering, burning aluminum droplets looking for partners. It seems much more likely that agglomeration is concurrent with ignition of accumulated, oxide coated particles, and that agglomerate size is governed by the microscopic heterogeneity of the propellant. In any case, there is ample evidence in the literature that the aluminum particles will not oxidize at propellant surface temperature at a rate governed by oxidizer diffusion as presumed in the model. We do not contend that this "argument" disproves the model proposed in Ref. 1 and 2, but rather raises a number of issues that would have to be resolved before the model merited acceptance. We feel the issues cannot be resolved in favor of the model, and will seek to establish this point when recent work is fully documented.

In summary, we believe that the results reported in Ref. 1 have doubtful relevance to combustion in rocket motors, are derived from a sampling procedure that does not warrant the conclusions reached, and are correlated by a theoretical model that is so unrealistic as to lack known qualitative attributes of the combustion process. The "reasonable agreement" between experiment and theory is considered to be little more than rationalization.

References

- ¹ Povinelli, L. A. and Rosenstein, R. A., "Alumina size distributions from high-pressure composite solid-propellant combustion," AIAA J. 2, 1754-1760 (1964).
- ² Povinelli, L. A. "Effect of oxidizer particle size on additive agglomeration," NASA TN D-1438 (1962).
- ³ Povinelli, L. A. and Ciebuch, C. C., "Surface phenomena in solid propellant combustion," *Bulletin 18th Meeting, JANAF-ARPA-NASA Solid Propellants Group* (Chemical Propulsion Information Agency, Applied Physics Laboratory, Johns Hopkins University, Silver Spring, Md., 1962), Vol. II, pp. 387-400.
- ⁴ Davis, A., "Solid propellants: the combustion of particles of metal ingredients," *Combust. Flame* 7, 359-367 (December 1963).
- ⁵ Drew, C. M., Gordon, A. S., and Knipe, R. H., "Study of quenched aluminum particle combustion," *AIAA Progress in Astronautics and Aeronautics: Heterogeneous Combustion*, edited by H. G. Wolfhard, I. Glassman, and L. Green, Jr. (Academic Press Inc., New York, 1964), Vol. 15, pp. 17-39.
- ⁶ Brzustowski, T. A. and Glassman, I., "Vapor-phase diffusion flames in the combustion of magnesium and aluminum: I. Analytical developments, and II. Experimental observations in oxygen atmospheres," *AIAA Progress in Astronautics and Aeronautics: Heterogeneous Combustion*, edited by H. G. Wolfhard, I. Glassman, and L. Green, Jr. (Academic Press Inc., New York, 1964), Vol. 15, pp. 75-158.
- ⁷ Sehgal, R., "An experimental investigation of a gas-particle system," *Jet Propulsion Lab. TR 32-238* (1962).
- ⁸ Brown, B. and McArty, K. P., "Particle size of condensed oxides from combustion of metallized solid propellants," *Eighth Symposium (International) on Combustion* (Williams & Wilkins Co., Baltimore, Md., 1962), p. 814.
- ⁹ Crump, J. E., "Photographic survey of aluminum combustion in solid propellants," *Proceedings of the 1st Combustion Instability Conference* (Chemical Propulsion Information Agency, Applied Physics Laboratory, Johns Hopkins University, Silver Spring, Md., 1965), CPIA Publ. 68, p. 367.

Comment on "Buckling of a Cylindrical Shell Loaded by a Pre-Tensioned Filament Winding"

T. C. FAN*

The Rand Corporation, Santa Monica, Calif.

Nomenclature

D	= plate flexural stiffness, $Et^3/12(1 - \mu^2)$
E	= Young's modulus
f	= function
k	= elastic spring constant
k_y	= buckling coefficient
k_{yp}	= $f(\beta, Z)$ as defined in Ref. 1
K	= spring stiffness parameter
L	= length of cylindrical shell
m	= number of longitudinal waves
n	= number of circumferential waves
r	= radius of cylindrical shell
t	= thickness of cylindrical shell
Z	= curvature parameter
β	= $nL/\pi r$
μ	= Poisson's ratio

MIKULAS and Stein¹ are to be congratulated for their interesting note on the subject of shell stability with pre-tensioned filament winding. The result of this note undoubtedly is useful for the optimization study of an integrated system in the conceptual design. However, there are three points not yet discussed in the same note.

1) The minimization of the negative root of the characteristic equation [Ref. 1, Eq. (14)] may also determine the critical stress resultant. This operation can be performed by letting

$$\frac{\partial k_y}{\partial \beta} = \frac{\partial}{\partial \beta} \left\{ \frac{k_{yp} - [k_{yp}^2 + (4k_{yp}K/\beta^2)]^{1/2}}{2} \right\} = 0 \quad (1)$$

It seems that Eq. (1) may lead to a different result. It can be shown that [Eq. (14)]¹

$$\frac{k_y \beta^2}{k_{yp} \beta^2 + K} = \frac{(1 + \beta^2)^2}{\beta^2} + \frac{12Z^2}{\pi^4} \frac{1}{(1 + \beta^2)^2 \beta^2}$$

A quadratic in k_y can be reduced to Eq. (B5)²

$$k_y = \frac{(m^2 + \beta^2)^2}{\beta^2} + \frac{12Z^2 m^4}{\pi^4 \beta^2 (m^2 + \beta^2)^2}$$

which is linear in k_y if and only if there is no spring stiffness K vanishing in Eq. (14)¹ and m is taken to be unity in Eq. (B5).² Similarly their solutions can be reconciled if treated as below. If both roots of Eq. (14)¹ are considered, then Eq. (15)¹ becomes

$$k_y = \{k_{yp} \pm [k_{yp}^2 + (4k_{yp}K/\beta^2)]^{1/2}\} / 2$$

Then if K vanishes,

$$k_y = \begin{cases} 0 \\ k_{yp} = \frac{(1 + \beta^2)^2}{\beta^2} + \frac{12Z^2}{\pi^4} \frac{1}{\beta^2(1 + \beta^2)^2} = \end{cases}$$

Eq. (B5)² for $m = 1$

It indicates that only the positive root has significance for $K = 0$.

Therefore, for $K \neq 0$, Eq. (14)¹ is not identical to Eq. (B5)²; it may be necessary to establish further the uniqueness

Received May 7, 1965.

* Aero-Astronautics Department. Member AIAA.