A Comparative Study of Propellants Containing Al or ZrC/Graphite as Combustion Stabilizer

Tai-Kang Liu*, Song-Ping Luh*, Huey-Cherng Perng*, Hwa-Shu Chiu*

The combustion instability of the AP-based reduced smoke propellant was experimentally investigated with a T-burner and its smoke visibility was assessed with a smoke chamber. The only formulation variable investigated was Al or ZrC/Graphite which serves as the combustion stabilizer. It was found that ZrC/Graphite propellant produces relatively less smoke than Al propellant. Both ZrC/Graphite and Al were found to suppress combustion instability, although, depending on the formulation, ZrC may not be less than 1.5% in order to have good effect.

Experimental results were discussed further with the window bomb photographs, the physical properties of additives/combustion products, as well as the thermochemical considerations. In addition, the controversial literature findings on the effect of additive particle size on combustion instability were pointed out and briefly reviewed.

INTRODUCTION

During the past decade, many studies have been devoted to reduced smoke or minimum smoke propellants. Usually these propellants contain less than 2% of metal in formulation. For many years aluminum was employed for acoustic stabilization in such propellants. However, as the aluminum gaining specific impulse and density while also generating smoke in rocket exhaust, the use of aluminum becomes in a dilemma. The present study reports the experimental results of the AP-based composite propellant containing either Al or the "standard depressant" ZrC/graphite as stability additives. T-burner and smoke chamber were respectively employed to assess the propellant combustion instability and smoke visibility.

EXPERIMENTAL

Six formulations were prepared using Baker Perkins laboratory blender. Basically they were tailored to conform to the specific requirements while at the same time to isolate the effect of either Al or ZrC/graphite with respect to the baseline formulation. The formulations and the predicted delivered specific impulse are summarized in Table 1.

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*4th Research Department, Chung Shan Institute of Science and Tech.
Lungtan, Taiwan R. O. C.
TEL 02-3814014

A classical T-burner covering frequency range of 0.3-3 kHz was employed to study the longitudinal mode pressure coupling combustion instability of the propellant formulation. Implementation with the T-burner included a 701A Kistler pressure transducer, a series of signal conditioning electronics, and a PC/AT computer. Software was specially designed for the computer so that the oscillating pressure signal can be conveniently processed and the results graphically reported. The T-burner schematic is shown in Fig. 1. The test pressure was fixed at 20 atm since previous experiments have shown that the response at high frequency decreases with increasing pressure, thus the most severe stability condition for nonaluminum propellant is expected at low pressure and high frequency (1).

Al m^3 cubic smoke chamber was employed to study the propellant smoke visibility under different environmental (temperature and relative humidity) conditions. By measuring the light attenuation of the 0.95 mW He-Ne laser beam (λ-632.8 nm, calibrated with standarized neutral density filters for transmittance), the light transmittance through the smoke in specific wavelength was obtained. All experiments

Table 1 Propellant Formulation

	Group 1			Group 2		
	baseline effect Y138	Al effect Y139	ZrC/C Y140	baseline effect Y143	Al effect Y142	ZrC/C Y141
HTPB	13.25	13.25	13.25	13.64	13.64	13.64
PNBr	2.0	2.0	2.0	–	–	–
AP*	85.0	85.0	85.0	85.0	85.0	85.0
Al**	–	2.0	–	–	1.6	–
ZrC***/C	–	–	1.5/0.5	–	–	1.1/0.5
predicted delivered Isp(100atm)	236.2	239.4	232.7	238.7	240.6	235.5

*AP blend 400/225/90/20 micron for group 1, 225/20/5 micron for group 2
**median size 5.8 micron
***medain size 5.5 micron

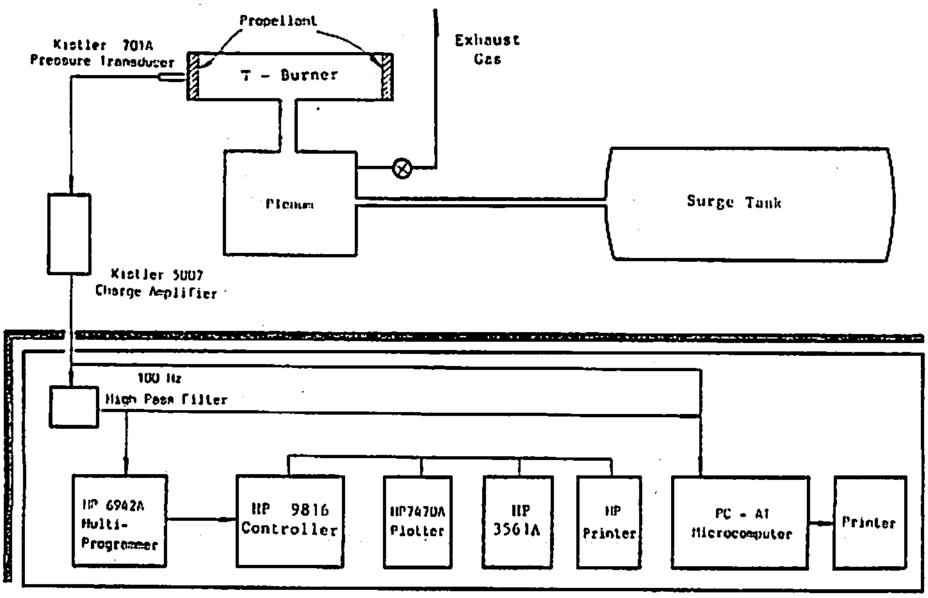

Fig. 1 T-burner schematic

were conducted using equal-weight propellant in order to give the same basis. The schematic of the test chamber is shown in Fig. 2.

RESULTS AND DISCUSSION

A. Combustion Instability

The use of metal particles for stability control is based on the known particulate damping theory which in turn is based on the energy dissipation by viscous drag. Although the effect of ZrC and graphite is evident in promoting combustion stability at high frequency and low pressure, the change in low frequencies is still not quite clear (2). Our T-burner experimental results for both ZrC/C and Al propellants are shown in Table 2. The oscillation frequency and the sum of linear stability growth and decay constant ($\alpha_g+\alpha_d$) are reported if oscillations were detected.

Comparing Y140 & Y141 in Table 2, it was found that the minimun amount of ZrC may not be less 1.5% in order to completely suppress combustion instability

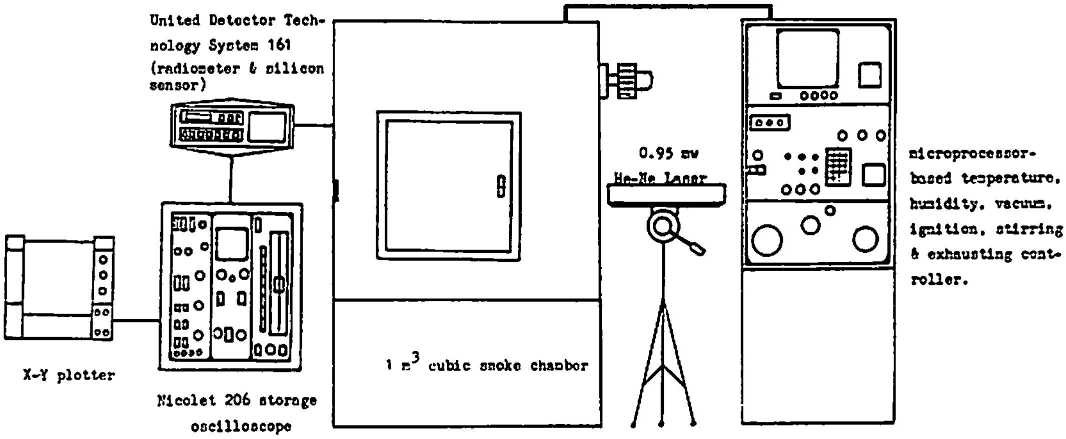

Fig. 2 Smoke chamber schematic

Table 2 T-Burner Test Result

formulation	Group 1			Group 2		
	Y138 baseline	Y139 2% Al	Y140 1.5% ZrC 0.5% C	Y143 baseline	Y142 1.6% Al	Y141 1.1% ZrC 0.5% C
Burner length, cm						
19	x	x	x	x	x	x
38	1280 Hz	x	x	1320 Hz	x	1320 Hz
	13.97			8.30		12.51
50	840 Hz	x	x	1000 Hz	x	x
	9.50			8.33		
59	740 Hz	x	x	780 Hz	x	800 Hz
	10.71			10.59		11.6
80	540 Hz	x	x	560 Hz	x	x
	9.08			12.68		
104	360 Hz	x	x	420 Hz	x	x
	8.76			10.88		

% denotes no oscillations being detected. Numerical values denote frequency and $\alpha_g+\alpha_d$ obtained from averaging of two experiments.

in the frequency range studied. In addition to the lesser amount of ZrC in Y141, the smaller AP particle size of Y141 compared with Y140 also contributes to the combustion instability. Regarding the frequency effect, all additives are effective in stability control down to the lowest frequency of around 400 Hz in our study. Besides, all Al formulations were found to exhibit stable combustion over the entire frequency range.

The response function of the two nonmetallized propellant is shown in Fig. 3. They were calculated from experimental data using the equation

$$R_b=\frac{\bar{P}}{4\rho_p\bar{r}\bar{a}}\left(\frac{\alpha_g+\alpha_d}{f}\right)$$

where $\bar{a}$ is sound speed which can be estimated with $\bar{a}=2fL$, L denotes the burner length. P, ρ_p, r and f denote respectively pressure, propellant density, burning rate and oscillation frequency. It is clear that the response decreases with increasing frequency in the experimental range studied. Thus the nonmetallized propellants are more susceptible to combustion instability in the low frequency range.

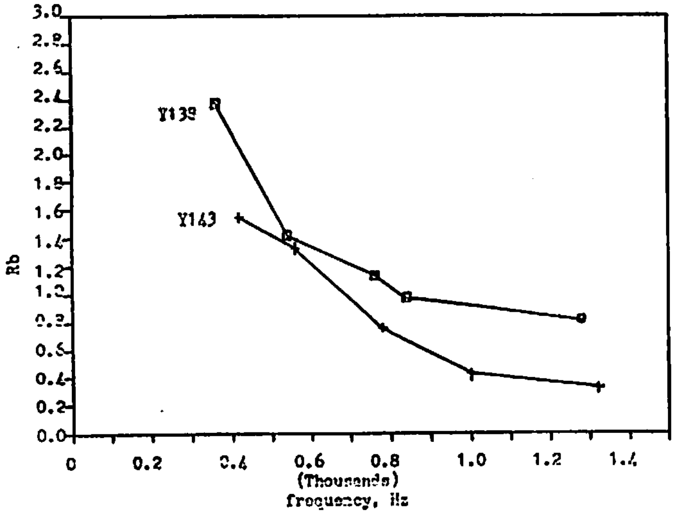

Fig. 3 Response function of non-metallized propellant

Table 3 GSD Testing Summary of Combustion Stabilizers

	incandescent particles		
candidate stabilizer	population at propellant surface	location of first appearance	population above propellant surface
Al, 5 micron	dense	on surface	dense
ZrC, 3 micron	very dense	on surface	dense

Window bomb movie was adopted by Rudy and Bain (3) to rank the propellants relative to the apparent ignition location of the stability additive. They found that those additives that reach ignition at or near the propellant surface are most effective. Their findings are summarized by Braithwaite et al. (4) in Table 3 for the selected case of our interest. Our window bomb photographs in Fig. 4 showing the similar results. Moreover, much more incandescant particles above propellant surface were observed for ZrC propellant than for Al popellant. Consider that ZrC has a much higher m.p. than Al or Al_2O_3 (Table 5), it is more likely that ZrC does not completely burn at the propellant surface but rather in gas phase without too much changing in size during burning. It is believed that the latter process contributed to the "damping" effect which lead to the reduction of the response. Since the weight percent of ZrC is lesser than Al in the individual formulation group (Table 1), the capability of ZrC in stability control is even more evident.

Regarding the effect of particle size of stability additives on viscous particle damping, there exists some conflicting findings during the last ten years. King (5) believes that ZrC does not change particle size during burning so that it is important to choose the right particle size distribution of ZrC to "tune" the combustion so that some specific dampings can be obtained. Beckstead et al. (6) found that changing particle size of burning Al particles can have significant effect on particle damping. The calculaed damping based on model that accounts for the changing particle size of Al changed by a factor of two over the typical range of conditions used in the T-burner. Derr et al. (10) found that pressure oscillation in motor arises from nonmatching of experimental particle size distribution of additives with the theoretical ones for optimum damping. They are the believers who emphasize importance of the size effect on particle damping, which can be predicted by particle damping theory. Evans and Smith's experiments (7) indicated that SiC

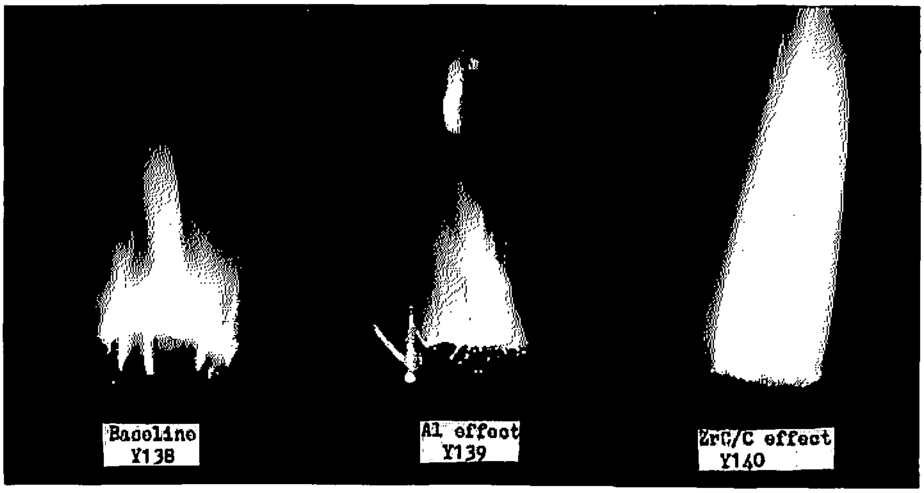

Fig. 4 A comparison of combustion phenomena (P = 0.5 atm)

Table 4 Afterburning Potential and Total Condensibles Based on the Chamber Pressure of 100 kg/cm²

formulation	nozzle exit temp. K	nozzle exit mass percentage				
		CO	H_2	H_2O	HCl	total condensibles
Y138	1254	12.00	1.20	23.51	26.35	49.86
Y139	1340	13.77	1.25	22.38	25.83	48.21
Y140	1227	12.63	1.28	22.14	25.83	47.97
Y143	1361	9.31	0.79	25.86	26.84	52.70
Y142	1418	11.17	0.90	24.28	26.35	50.83
Y141	1315	10.59	0.93	24.21	26.35	50.56

stabilizer does not change particle size after combustion. Contrary to above studies, they concluded that the size of stabilizer may not be too precise but that the mass fraction appears to be of primary importance. Besides, Zimmerman and Ditore (1) also found that changing particle size of ZrC or Al_2O_3 had no significant effect on the response function over the experimental range studied. These conflicting results may not be resolved at the present, but should remain to be a research subject in the future.

B. Smoke Visibility

Assessment of propellant plume has been one of the important topics in the reduced smoke propellant research. Although different complex facilities were developed to simulate the real flight conditions, the conventional smoke chamber was found capable of yielding the same relative propellant ranking as the more advanced versions (8). Our experimental results using similar facilities are shown in Fig. 5 under different enviromental conditions. A dilution ratio of 60 : 1 was calculated for the present study using 17.6 gram sample of each formulation. Regarding smoke visibility, it was found that both groups of propellants show consistent ranking, i.e., Al propellant generating the highest smoke concentration followed by ZrC/C propellant. Baseline propellant always produces the lowest concentration. Changing the amount of additives also changes the relative smoke visibility proportionately.

Thermochemical prediction of the secondary smoke related combustion products of different propellants is shown in Table 4. Afterburning arises from burning of CO and H_2 fuel species to form secondary smoke. Since the percentage of afterburning (in 50 to

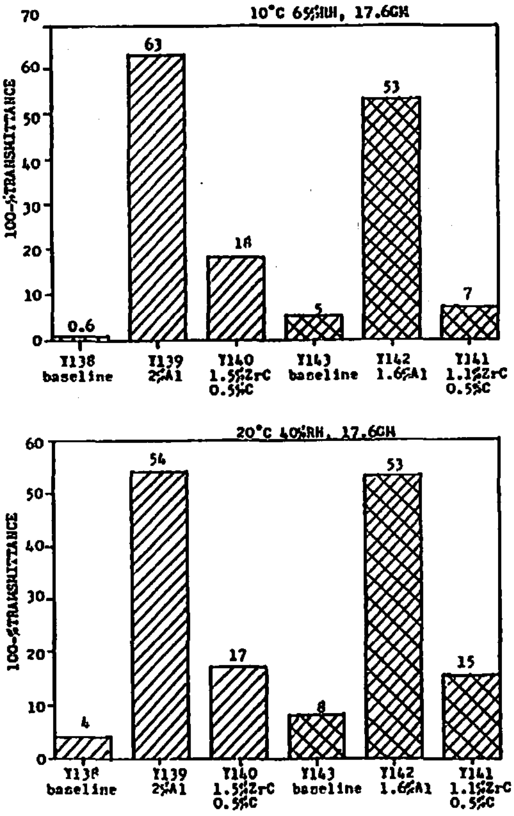

Fig. 5 A comparison of smoke visibility under different environmental conditions.

Table 5 Properties of Additives. Associated with Combustion Instability and Smoke Visibility

additives	density g / cm³	m.p. ℃	size range micron
Al	2.35 (1000 C)	600	6−9
Al_2O_3	3.4−3.9	2007	predominantly submicron
ZrC	6.73	3540	5−10

100% range) has little effect on prediction of the secondary smoke of the AP-based reduced smoke propellants (9), however, transmittance variation due to its effect are expected to be small. Besides, the near invariness of HCl and H_2O condensibles concentration is also indicated in Table 4. These results, along with the prediction (8) of condensation boundaries for reduced smoke propellant at sea level for our conditions, suggest that the primary smoke, namely Al_2O_3 or ZrC, contributes to the variation of the smoke. Results shown in Fig. 5 indicate that replacing Al with ZrC reduces the visibility by 3 to 8 times.

Observations in Fig. 5 may be explained from the classical optical theory that the size of particulate matter in solid propellant should be chosen to avoid the maximun scattering for visible light in exhaust, or, alternatively, the material should be chosen with higher density which, for a given mass fraction for acoustic damping, minimizes the particles per unit volume of exhaust gas and hence low smoke. The properties of additives in the present study are shown in Table 5. It is clear that the higher m.p. of ZrC may keep the particle size rather unchanged during combusttion. ZrC reacts heterogeneously in solid phase without melting during combustion. Al, on the other hand, reacts in phase and produces Al_2O_3 particles mostly in submicron range in the exhaust, thus produces a high value of number of particles per unit volume and hence high smoke. Besides, the Al/Al_2O_3 has a lower density than ZrC which even aggravates the smoke generation. Our observations on relative smoke are in good agreements with the study of Evans and Smith (7). They found a 4 times reduction of smoke/light attenuation when 1.4% Al was replaced by 1.4% SiC in the propellant. Consider that SiC has a density of 3.22 g / cm³ which is lower than 6.73 g / cm³ of ZrC, the much better result of smoke/light attenuation (up to max. of 8 times) in our study could be realized.

SUMMARY

The role of ZrC/C and Al was experimentally investigated with respect to their behavior to suppress combustion instability and smoke visibility. It was found that ZrC/C formulation is better than Al formulation in reducing smoke visibility. This was attributed to the different physical and optical properties between these two additives and their combustion products. Formulaions without metallic additives were found to exhibit combustion instability, showing the significance of particulate damping effect during combustion. Nonmetallic formulations are much more susceptible to instability problem in low frequency range. Regarding the ZrC mass fraction, It was found that, depending on formulation, the amount of ZrC may not be less than 1.5% in order to suppress the combustion instability. More incandescent particles above propellant surface were observed for ZrC

propellant than Al propellant in window bomb experiments, substantiating its role particle damping. In addition, controversial literature results on the effect of additive particle size on combustion instability were pointed out, thus leaves the subject rather inconclusive at the present time.

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燃焼安定剤としてアルミニウムあるいは炭化ジルコニウム/グラファイトを含む推進薬の比較研究

Tai−Kang Liu*, Song−Ping Luh*,
Huey−Cherng Perng*, Hwa−Shu Chiu*

不安定燃焼を抑制し，かつ噴出ガスの無煙化を確保するためにアルミニウム(Al)と炭化ジルコニウム/グラファイト(Zr/C)を添加した推進薬について実験的研究を行った。Zr/CはAlよりも無煙化に有効であり，この結果はこれら2種類の添加物の物理的及び光学的特性の違いから起因していることがわかった。金属添加物を含まない推進薬組成では不安定燃焼が発生するが，微粒子によって容易に減衰させることができる。また同時に，不安定燃焼は低周波領域では微粒子の添加により敏感である。Zr/Cの質量分率が1.5%以下では不安定燃焼抑制の効果が少ない。窓付き燃焼器での観察によると，Zr/C推進薬はAl推進薬よりも燃焼表面で白熱光を発した微粒子が見られ，減衰の効果が大きいことを示している。これらの結果は，過去に報告されている結果と異なり，本論文では実験的事実のみを報告するだけに留どめる。

(*中山科学研究院　第4研究所　中華民国，台湾省桃園龍潭郵政一號)