Combustion and Flame 192 (2018) 59–70

Contents lists available at ScienceDirect

Combustion and Flame

journal homepage: www.elsevier.com/locate/combustflame

Aluminized composite propellant combustion modeling with Heterogeneous Quasi-One dimensional (HeQu1-D) approach

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ARTICLE INFO

Article history:
Received 17 September 2017
Revised 30 October 2017
Accepted 24 January 2018

Keywords:
Aluminum
Catalyst
Composite propellants
Radiation
Modeling

ABSTRACT

The Heterogeneous Quasi One-dimensional (*HeQu1-D*) model for AP/HTPB composite propellant combustion is extended to aluminized propellants. Following the serial burning approach of *HeQu1-D*, a statistical particle path through an aluminized propellant is taken to consist of aluminized binder-matrix coated AP particles of various sizes. Extending the idea of homogenization of fine-AP particles, aluminum particles are assumed to be homogenized with the binder-matrix. Large Al particles (nominal size $\geq$ 15 µm) in the binder-matrix are assumed to get ejected into the gas phase and hence do not contribute to heat feedback to the burning surface. With these modifications, the model is shown to accurately predict the burn rate variation with pressure and initial temperature of propellants using Al of nominal sizes in the range of 15–50 µm (termed conventional aluminum, CAl). The experimentally observed reduction in propellant burn rate with substitution of AP by CAl is shown to be due to the increase in fuel richness and energy sink effect of melting of Al. Catalytic effects due to addition of burn rate modifiers (Fe_2O_3 in space shuttle booster propellant, for instance) are also accounted for by modifications to gas phase rate parameters. Increase in burn rate up to 25% with Al particles of a few µm (3–6 µm) in comparison with non-aluminized propellant is explained by recognizing that there will be non-negligible fraction of sub-micron Al due to the associated particle size distribution. This leads to heat release by sub-micron Al combustion close to the surface and an enhancement in the heat feedback, by a combination of convective and radiative mechanisms. Dramatic increase in burn rate with sub-micron Al (a factor of 4–5 as compared to CAl) is captured by invoking radiation from fine-Al/Al_2O_3 particles to propellant surface. Agglomeration of sub-micron Al particles is invoked to explain the saturation of burn rate enhancement with reduction in Al particle size and the effectiveness of sub-micron Al substitution in smaller fractions (bi-modal Al). Predictions for over fifteen different propellants with varying fractions of fine and coarse aluminum from earlier literature have been presented. Comparisons of the predictions from the model with experimental results from different sources is shown to be excellent-to-good for most cases. The hitherto unknown radiation dominated ablation ($\dot{r} > 50$ mm/s) of coarse AP particles and the effect of Al particle size on propellant temperature sensitivity are brought out. The MATLAB® code based on the model offers opportunity for design of AP-HTPB composite propellants with combination of fine and coarse aluminum with confidence.

1. Introduction

Aluminum fraction in composite propellants varies between 18% (launch vehicle class) to a few % (smokeless tactical missile class). Nominal aluminum particle sizes used in typical application propellants vary from 15 to 50 µm, with 25 µm being the commonly used size, termed conventional aluminum (CAl). Smaller Al sizes are mostly used only in laboratory research - for instance, the 5.65, 5 and 3 µm used in [1–3] respectively. Insight into the effect of substitution of AP with conventional (15–25 µm) and smaller size (a few µm) Al can be obtained by comparing the burn rates of the following propellants - one non-aluminized with 86% AP (Mix-I from [4]), other two aluminized with 68% AP and 18% Al (C1 - 25 µm Al and C2 - 5.65 µm Al from [1]) (all three have same AP distribution). Burn rates of these three propellants taken from [1,4] are shown in Fig. 1.

Burn rate of propellant C1 with 25 µm Al is lower than Mix-I by 23% at 70 atm and has a pressure index of 0.33 compared to 0.44

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https://doi.org/10.1016/j.combustflame.2018.01.042

Nomenclature

$\dot{q}_R$	radiative flux (W/m^2)
$\dot{r}$	linear burn rate (mm/s)
σ_p	temperature sensitivity (%/K)
d_0	diffusion distance (µm)
d_{AP}	AP particle diamter (µm)
d_{pm}	premixed cut-off diamter (µm)
F_i	mass fraction of component *i* in aluminized binder-matrix
f_i	mass fraction of component i
g_f	geometric factor
H_s	surface enthalpy change (kJ/kg)
K_r	gas phase reaction rate (s/m^2-atm)
l_i	line average fraction
T_0	initial temperature (K)
T_f	flame temperature (K)
T_s	surface temperature (K)
t_{bm}	binder thickness (µm)
T_{eff}	effective gas phase temperature (K)
x^*	flame standoff distance (µm)
ABM	aluminized binder-matrix
BRM	burn rate modifiers
CAl	conventional aluminum
UFAL	ultra fine aluminum

of Mix-I. It is clear that the substitution of AP by CAl reduces the burn rate and index. It is important to note that the AP/HTPB ratio of Mix-I (86/14 = 6.1) is closer to stoichiometry (88/12 = 7.33) compared to that of C1 (68/14 = 4.9). This increase in fuel richness of C1 combined with the energy sink effect due to melting of Al particles at the surface of the propellant is later used to predict its burn rate behavior. On the other hand propellant C2 from [1] with 5.65 µm Al in place of CAl burns at a rate comparable to Mix-I (non-aluminized) at all pressures, in spite of the fuel richness and Al energy sink effect brought out earlier. This is argued to be due to heat release from the sub-micron fraction in 5.65 µm Al used in C2. Ishihara et al. [2] obtained experimental results by systematically substituting 5 µm Al in bi-modal AP/HTPB propellant with 82% total solids (A-0, 5, 10, 20 in Fig. 1). The burn rate data, though limited, is consistent with that from [1] - substitution of 5 µm Al for AP has marginal effect on burn rate. Therefore the effects of fuel richness due to decrease in AP fraction and the Al melting related surface energy sink effects seem to be counteracted by Al particle related processes in the gas phase. Ishihara et al. [2] have shown that the radiation received by propellant A-20 (with 20% 5 µm Al) is about 20% of the gas phase flux at 35 atm - this is consistent with the observation about sub-micron fraction oxidation close to the surface for 5 µm Al. For Al nominal sizes of 15 µm and above since the sub-micron fraction is negligible the compensating mechanisms are absent leading to decrease in burn rate compared to the non-aluminized counterpart. Thus Al particles of nominal size about 5 µm and less cannot be classified as CAl. Results from extensive experimental studies by Price and group (see for instance [5,6]) are also consistent with these observations - with use of 3 µm aluminum leading to burn rate increase by 35% at 69 atm compared to propellants with 15 and 30 µm aluminum. The corresponding non-aluminized compositions (89% AP) were not considered in [6]. But predictions for the non-aluminized compositions were obtained using the HeQu1-D model and the results show that the burn rates of compositions with CAl (15 and 30 µm Al) are 20% lower and that of the compositions with 3 µm Al is 15% higher than the non-aluminized cases, consistent with the observations made so far. Pressure index also increases with

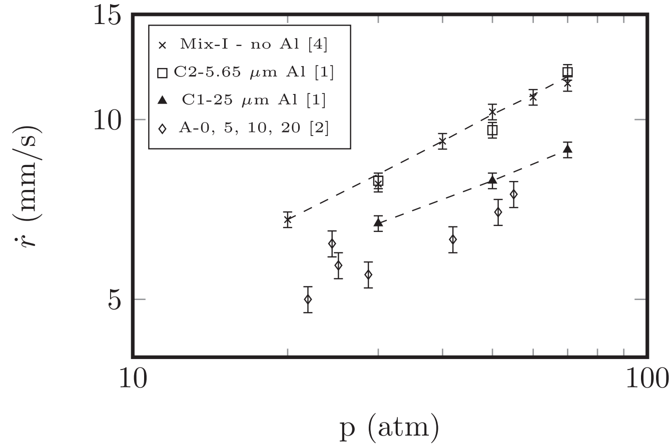

Fig. 1. Effect of substitution and particle size of Al for AP refs [1,2,4]; error bar is 3% for data from [1,4] and 5% for [2].

replacement of 30 µm Al with 3 µm Al from about 0.3 to 0.48 consistent with earlier observation related to propellants from [1,4]. Note that differences in burn rate index in general are difficult to quantify compared to burn rate itself.

Recent interest in high burn rate propellants ($\sim$ 40 mm/s) has led to the use of ultra-fine aluminum (UFAl) which belongs to sub-micron sizes. Studies due to Dokhan et al. [6] on propellants of 89% total solids with 18% aluminum indicate that use of UFAl can increase the burn rate of propellant dramatically - a factor of 5 compared to propellants with CAl. Even 20% substitution of CAl with UFAl is shown to lead to three times increase in the burn rate as compared to CAl propellants indicating a shift in the combustion mechanism as compared to propellants with CAl.

These observations on the effect of aluminum particle size can be rationalized and predicted using a modeling framework by incorporating the following two points - (1) ignition of Al is a strong function of particle size and oxide coating (purity) and (2) Al added to any propellant has an associated size distribution (usually log-normal as found for space shuttle Al [7]). Arguments are presented later to show that, almost all of conventional Al particles (nominal 15 µm and above) ignite sufficiently far away from the propellant surface to have any significant influence on the convective and radiative fluxes to the propellant surface leading to a burn rate decrease compared to the non-aluminized propellant. As the nominal size is decreased, the fraction of sub-micron Al oxidizing sufficiently close to the surface increases (due to the associated particle size distribution) and contribution to the heat flux to the surface increases leading burn rate increase compared to CAl propellants - propellants with 3, 5 and 5.65 µm Al from [1,2,6] are cases in point. At sub-micron nominal sizes all of the aluminum will oxidize very close to the surface causing dramatic increase in burn rate - as shown later, almost all the heat flux with UFAl comes from radiation due to the inverse dependence of radiative flux on Al particle size. Rupture of oxide coating due to volume expansion caused by melting of aluminum core in sub-micron sized particles is a possible ignition mechanism of UFAl particles (see [8,9]). This mechanism is clearly dependent on the surface-to-volume ratio and hence can become dominant at sub-micron scales. Similar considerations are applicable to high surface area flaky aluminum with thickness of few tens of nanometers which are shown to enhance burn rates compared to CAl by a factor of about 2.5 in [1]. By incorporating these additional features, the recently developed '*Heterogeneous Quasi One-dimensional (HeQu1-D) model*' [10] for non-aluminized propellants is extended to account for aluminum effects. Catalytic effects due to addition of burn rate modifiers (Fe_2O_3, for instance) are also accounted for by