



Exploring mechanisms for agglomerate reduction in composite solid propellants with polyethylene inclusion modified aluminum



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ABSTRACT

In composite solid propellants, shortening particle residence time at the burning surface and inducing particle microexplosions could decrease aluminum agglomeration, thus reducing two-phase flow losses in a rocket motor. We explore this by using aluminum particles modified with low-density polyethylene (LDPE) inclusion to drive intraparticle outgassing, which could break apart composite particles, yielding smaller and faster burning fragments during composite solid propellant combustion. We find that use of these particles in a propellant results in more prompt particle ignition, and surface residence time is decreased. For composite propellant burning at 6.9 MPa, mean coarse agglomerate diameter is decreased from 75.8 μm (spherical aluminum) to 29.0 μm (Al/LDPE 90/10 wt.% particles). Thermal analysis with DSC/TGA shows that 10 wt.% LDPE inclusion in aluminum (1.5% of propellant weight) results in enhanced oxidation characteristics that are more similar to nanoaluminum than neat spherical aluminum. Thermochemical equilibrium calculations indicate LDPE inclusion decreases specific impulse by 1.0% from 262.7 to 260.0 s, but it is expected that in a motor, LDPE inclusion could produce a net increase in specific impulse due to a substantially reduced agglomerate size. This work shows that reduced agglomeration is possible using gas generating inclusion materials that are only weakly reactive with aluminum.

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1. Introduction

Increasing the specific impulse performance of solid rocket motors by reduction of two-phase flow losses remains an elusive goal. Two-phase flow losses, which occur from kinetic and thermal non-equilibrium between condensed phase particles and surrounding product gas are prevalent for aluminized formulations. In a typical aluminized composite solid propellant, two-phase flow losses can be significant, resulting in a ~3–5% reduction in specific impulse [1,2]. The dissimilarity between propellant burning surface temperature (~500–700 °C) [3] and aluminum particle ignition temperature in a propellant (reported by different authors to vary in the range of 1400–2100 °C) [4–6] is a particular cause for formation of large agglomerates. During propellant combustion, aluminum particles emerge from solid propellant and aggregate at the propellant burning surface. Aluminum aggregates can

remain at the burning surface until they reach their ignition temperature, spending a significant amount of time at temperatures above the aluminum melting point. This results in aluminum particle sintering or coalescence into larger aggregates or agglomerates [7–9].

Prior efforts to reduce two-phase flow losses have attempted to reduce product agglomeration by using smaller aluminum particles. Use of smaller, nanoscale aluminum (nano-Al) particles in composite propellant results in shorter burning surface residence time due to faster particle heating and ignition at a lower temperature than micrometer-scale aluminum [10]. Replacement of micrometer-scale aluminum with nanoaluminum can decrease coarse agglomerate size to 2 μm [9,11,12]. However, the high oxide content of nanoaluminum particles (~10–50 wt.%), and poor aging characteristics can reduce propellant specific impulse [13]. Additionally, nanoaluminum's high specific surface area (~10–50 m²/g) can result in unprocessable viscosities [14] and poor mechanical strength [15], causing motor failure. Consequently, nanoaluminum is not currently used in fielded propellant formulations.

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Additional efforts to reduce agglomeration have focused on overcoming the drawbacks of nanoaluminum by using micrometer-scale particles that undergo low temperature, exothermic reactions to accelerate their ignition and thus reduce burning surface residence time. To this end, metallic [16–19] and polymeric [18,20] particle coatings have been employed to achieve limited agglomeration improvement through exothermic reaction with underlying aluminum cores. Nickel coatings react in the condensed phase to first form nickel aluminides, and in propellant combustion, this was reported to reduce coarse agglomerate size by ~20%, but agglomerates (~122 μm) were still about five times larger than the initial aluminum particle diameter (~22 μm) [17]. Fluoropolymer coatings react with underlying aluminum and aluminum oxide during heating to form an aluminum fluoride coating that upon sublimation at ~1300 °C might aid separation of aggregated aluminum particles at the burning surface. Use of fluorinated coatings (polymethylfluoroacrylate or fluorocarbon chlorosilanes) [18,20] reduces coarse agglomerate size, however agglomerates are still one and two orders of magnitude larger than initial aluminum particle sizes. In general, since particle coatings act to prevent agglomeration of one aluminum particle with another, coarse agglomerate sizes are limited by the initial aluminum particle size.

Coarse agglomerates smaller than initial particle sizes are theoretically possible if breakup of aluminum at the burning surface can be achieved. Toward this goal, partial replacement of aluminum with mechanically activated Ni/Al reactive particles in a hydroxyl-terminated polybutadiene (HTPB) and ammonium perchlorate (AP) composite solid propellant decreases coarse agglomerate size from 235 to 90 μm (1 atm pressure) [21], but due to the high quantity of nickel present in particles (~70 wt.%), theoretical specific impulse is reduced by ~25 s. Higher theoretical performance is possible through use of poly(tetrafluoroethylene) (PTFE) inclusions within micrometer-scale aluminum particles, which result in only a 2–7 s decrease in theoretical specific impulse [22]. Low level (10–30 wt.%) PTFE inclusion modification was found to expedite aluminum ignition and produce coarse agglomerates smaller in diameter than the initial aluminum. Upon ignition at the propellant burning surface, it is hypothesized that exothermic, gas-producing reactions of aluminum with PTFE within these particles can cause them to breakup into smaller, faster burning particles. In a propellant, replacement of spherical aluminum with similarly sized Al/PTFE (70/30 wt.%) composite particles can reduce the coarse product agglomerate diameter by ~66% [22]. Though PTFE inclusion is effective at reducing agglomeration, the mechanism responsible for agglomerate size reduction is not entirely clear. It is possible heat generation from exothermic aluminum-PTFE reaction shortens aluminum particle residence time at the burning surface, decreasing chances of particle coalescence. Alternately, it is possible that gas produced from both PTFE decomposition and from aluminum fluoride sublimation causes the particles to break apart.

The individual significance of these two mechanisms in agglomerate size reduction and aluminum combustion enhancement might be determined through developing and assessing inclusion materials that result in (1) substantial gas generation accompanied by little heat release or (2) heat release without gas production. Efforts addressing the latter [21] have shown heat release alone decreases agglomeration but can also significantly reduce theoretical specific impulse. However, the sole benefits of gas production remain unknown. The question remains for inclusion modified composite particles: Can inclusion outgassing alone fragment aluminum and reduce agglomerate size?

The objective of this work is to determine if and how the mechanism of inclusion outgassing can lead to agglomeration reduction within a composite solid propellant. To achieve this goal, the effects of gas production are isolated from heat generation

through the choice of low density polyethylene (LDPE) as a weakly exothermic, high gas production inclusion. Low-level (10 wt.%) inclusion modified aluminum particles are manufactured and used in a composite solid propellant to determine the effect on aluminum ignition and agglomeration. Results are compared with the previous work using 10 wt.% PTFE inclusion to understand the individual contribution of gas production. The effects of particle geometry and size on propellant density, theoretical propellant performance, burning rate, metal particle ignition, agglomeration, and metal combustion efficiency are all measured in order to determine how inclusion outgassing can alter propellant agglomeration.

2. Experimental

Thermochemical equilibrium calculations were performed on propellant, Al/polymer, and neat polymers using the Cheetah 6.0 equilibrium code [23] using the JCZS product library and JCZ3 gas equation of state. Equilibrium calculations performed on neat polymers were conducted at 0.101 MPa. For Al/polymer (90/10 wt.%) mixtures, the adiabatic flame temperature and amount of gas produced were computed at 6.9 MPa pressure. For propellant calculations, aluminum was replaced with up to 10 wt.% LDPE inclusion (1.5% propellant weight) in a composite propellant containing 71 wt.% ammonium perchlorate (AP), 14 wt.% hydroxy-terminated polybutadiene (HTPB), and 15 wt.% of either aluminum or Al/LDPE. A chamber pressure of 6.9 MPa and ideal expansion to equilibrium products at 0.101 MPa was assumed.

Five-gram batches of Al/LDPE composite particles were produced from fuel-rich mixtures of 90 wt.% aluminum (<45 μm , 99.5%, Atlantic Equipment Engineers) and 10 wt.% LDPE (500 μm , Alfa Aesar) via mechanical activation in a SPEX 8000D mill. Neat powders were continuously milled for four hours in an argon-filled (99.999%, Airgas) 50 mL steel vial (SPEX SamplePrep 8007) using 10 mm hardened steel media (Grade 1000, Frantz Manufacturing) and a charge ratio of 10. Milled, materials were processed in an argon environment and stored in hexane prior to incorporation in propellant.

Prior to use in a propellant formulation, Al/LDPE composite powders were dry sieved to between 25 and 75 μm , and their size, morphology, and thermal behavior were determined. The size distributions of the sieved MA composite powder were verified using a Malvern Mastersizer 2000 with Hydro 2000 μP dispersion unit and isopropyl alcohol as the medium. These size measurements were compared to measured distributions of similar spherical, flake, and Al/PTFE composite particles that were tested in propellants in previous work [22]. The thermal behavior of 1.5–3 mg samples of sieved powders as well as nanoaluminum (Novacentrix 80 nm) was determined by simultaneous DSC/TGA (TA Instruments Q600) over a temperature range of 100–800 °C using a 20 °C/min heating rate and 100 mL/min flow of 20/80 vol.% O₂/Ar gas.

Propellant was manufactured using procedures described elsewhere [22]. Briefly, propellant consisted of 14 wt.% of a HTPB binder (cured with isophorone diisocyanate, 8.3 wt.%), 71 wt.% AP (80 wt.% coarse 200 μm and 20 wt.% fine 20 μm , ATK), and 15 wt.% of sieved Al/LDPE 90/10 wt.% composite particles. The propellant was mixed in a 30 g batch for 20 min in a 250 mL container (McMaster-Carr 42905T25) using a LabRam (Resodyn) vibratory mixer at 90% intensity. Propellant was deaerated for 15 min at <35 mbar prior to being packed into 5.8 mm diameter, ~6 cm long cylindrical strand molds and cured in air at 60 °C for approximately seven days. After curing, the density of the propellant was calculated using Archimedes principle based on the mass of an approximately one-gram piece of propellant measured both unsubmerged and submerged in a bath of anhydrous isopropyl alcohol. The

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