ARTICLE IN PRESS

Combustion and Flame xxx (2014) xxx-xxx

Contents lists available at ScienceDirect



Combustion and Flame

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

The effect of encapsulated nanosized catalysts on the combustion of composite solid propellants

Sarah Isert^a,*, Lori J. Groven^b, Robert P. Lucht^c, Steven F. Son^c

^a Maurice J. Zucrow Laboratories, School of Aeronautics and Astronautics, Purdue University, West Lafayette, IN 47906, USA
^b Department of Chemical and Biological Engineering, South Dakota School of Mines and Technology, Rapid City, SD 57701, USA
^c Maurice J. Zucrow Laboratories, School of Mechanical Engineering, Purdue University, West Lafayette, IN 47906, USA

ARTICLE INFO

Article history: Received 17 September 2014 Received in revised form 20 November 2014 Accepted 21 November 2014 Available online xxxx

Keywords: Composite propellant Nano-catalyst Planar laser-induced fluorescence Flame structure Burning rate

ABSTRACT

In solid rocket propellants, nano-sized catalysts are expected to be more effective than their micron-sized counterparts due to their higher surface area and increased contact with the oxidizer. However, propellant processing becomes more difficult and ultimate mechanical properties can be negatively impacted as catalyst size is reduced. One proposed solution to these issues is to encapsulate a nano-sized catalyst inside the oxidizer. We have previously created composite particles with nano-sized iron oxide catalyst encapsulated in fine ammonium perchlorate (AP). In this paper we explore the effect of the composite particles on the burning rate and flame structure of an AP-based composite propellant. The propellant containing the composite nano-iron oxide/AP particles was compared to a baseline propellant without catalyst, a propellant formulated with micron-sized catalyst, and a propellant with nano-sized catalyst mixed directly. All catalyzed propellants contained the same catalyst mass fraction. High-speed (5 kHz) OH planar laser-induced fluorescence (PLIF) and high-speed surface imaging were used to examine differences in flame structure and coarse crystal burning characteristics and determine how the encapsulated catalyst influences the global burning rate. It was found that the burning rate of the propellant with the encapsulated catalyst was 90% higher than that of the baseline propellant, 44% higher than the propellant with micron-scale catalyst, and 15% higher than that of the propellant with nano-sized catalyst added directly. Our results indicate that the high global burning rate of the propellant with the encapsulated catalyst is due to an accelerating effect on the fine AP/binder matrix burning rate, assumed to be caused by the intimate contact between the fine AP and catalyst. At elevated pressures (4.4-7.1 atm) flame structure and burning surface morphology change as the catalyst size and location is changed. The coarse crystals are observed to protrude more from the burning surface as the catalyst size decreases. The microscale flame structure is observed to transition from jet-like to lifted arches as the pressure and catalyst size increase.

© 2014 Published by Elsevier Inc. on behalf of The Combustion Institute.

Combustion and Flame

1. Introduction

Ammonium perchlorate (AP) is currently the most commonly used solid propellant oxidizer. Ballistic properties of ammonium perchlorate composite propellants (APCP) can be tailored by changing the AP particle size. Decreasing the AP particle diameter increases propellant burning rate; however, decreasing the particle diameter past a certain point no longer provides a burning rate increase and can cause processing problems and safety concerns during grain manufacture [1–3]. In addition to the dependence on particle size, burning rates and ballistic properties of APCP

* Corresponding author at: 500 Allison Rd., West Lafayette, IN 47907, USA. *E-mail address:* sarah.isert@gmail.com (S. Isert). may be modified by the use of various additives, including catalysts. Iron oxide (Fe_2O_3) is an attractive catalyst as it is relatively non-toxic, easy to manufacture, and is a good burning rate modifier at rocket pressures. The burning rate modifications provided by iron oxide are high, well-characterized, and reproducible [1,4–15].

Catalysis is a surface phenomenon. As such, the catalyst becomes more effective as the contact area with the reactant, in this case the oxidizer, increases [5]. Increasing the catalyst percentage can increase the number of catalysis sites and therefore burning rate, but after a certain catalyst percentage there is no increase in burning rate, and propellant performance decreases due to the larger percentage of an inert ingredient [16,17]. Catalyst efficacy also increases as the catalyst size decreases due to the larger number reaction sites available for the same catalyst mass percentage

http://dx.doi.org/10.1016/j.combustflame.2014.11.040

0010-2180/ \odot 2014 Published by Elsevier Inc. on behalf of The Combustion Institute.

Please cite this article in press as: S. Isert et al., Combust. Flame (2014), http://dx.doi.org/10.1016/j.combustflame.2014.11.040

[4,6–9]. The large total surface area of the fine catalyst particles, however, can lead to an increase in propellant mix viscosity. If the propellant mix viscosity becomes too high, propellant grain casting becomes unfeasible, as has been seen in propellant mixes with large amounts of fine AP [2,3,18].

Ideally the catalysts would be put in direct contact with the AP crystals instead of (as is conventional practice) added to the binder. There have been a few reports in the literature of methods that cause the catalyst or dopant to be in direct physical contact with the AP crystals [10,19–23]. Encapsulation of iron oxide has been achieved in our group by rapidly crystalizing the AP in the presence of nanoscale catalyst particles that act as nucleation sites [20]. Although the modified particles have been well characterized, they had not been formulated in a propellant and characterized for combustion modification prior to this study.

The objectives of this study are to determine how global burning rate changes with the catalyst size and location, particularly between a propellant with nano-sized catalyst mixed directly and a propellant with the catalyst encapsulated in the fine AP. This will be done using high-speed OH planar laser-induced fluorescence (PLIF) and surface imaging.

2. Methods

Four propellants were studied: a baseline propellant with no catalyst, a propellant with micron-sized iron oxide catalyst, a propellant with nano-sized iron oxide catalyst mixed into the binder, and a propellant with nano-sized iron oxide catalyst encapsulated in the fine AP. Table 1 gives the propellant formulations.

The propellants had an 80% solids loading and a 1:1 coarse-tofine AP particle size ratio. The coarse AP was on average 400 μ m in diameter (Firefox Enterprises) and the non-composite fine AP was on average 20 μ m in diameter (Alliant Techsystems). The size distributions of these materials have been reported elsewhere [24]. Two catalyst sizes were used: nominally 53 μ m (Firefox Enterprises) and 3 nm (Mach I Inc.). Particle size distributions are given in Refs. [6,25], respectively. The catalyst percentage was driven by the amount of iron oxide captured in the composite particles and was kept constant at 0.21% for all catalyzed propellants. The composite particles are described in Section 2.1.

The same binder formulation was used for all propellants and consisted of R45-M hydroxyl-terminated polybutadiene (HTPB) prepolymer (Firefox Enterprises), isodecyl pelargonate (IDP) plasticizer (RCS Rocket Motor Components), Desmodur E744 curative (Bayer Corporation), and Tepanol HX-878 bonding agent (3 M). The propellants were hand-mixed and cast into plastic molds 6.35 mm in diameter and 100 mm in length. They were cured at room temperature for at least 7 days prior to use. The small batch sizes did not allow for viscosity measurements.

A Crawford strand burner was used to measure global burning rates, and 5 kHz OH planar laser-induced fluorescence (PLIF) was used to examine microscale flame structures and coarse crystal burning characteristics. High-speed imaging was also used at 1 atm to visualize the burning propellant surface. These methods will described in further detail in Section 2.2.

2.1. Encapsulation method

Nano-sized iron oxide particles were encapsulated in fine AP using the fast-crash solvent–antisolvent technique described by Reese et al. [20]. Briefly, the procedure is as follows. Acetone was chilled in an ice bath to 4 °C. Chemical grade AP (Sigma–Aldrich) was added to the acetone and stirred until dissolved. Nano-sized iron oxide (3 nm Nanocat© Superfine Iron Oxide, Mach I Inc.) was added to the AP/acetone solution and the resulting colloid was bath sonicated to promote dispersion of the iron oxide through the solution. After sonication the ethyl acetate antisolvent was added to crash the AP onto the iron oxide and the mixture allowed to sit for 10 min. The precipitated composite particles were vacuum filtered, bath sonicated with hexane to remove surface particles, and dried in a vacuum oven. The resulting mean composite particle diameter ($D_{4,3}$) was 25 µm and the size distribution is reported elsewhere [20].

After enough material was accumulated for a propellant batch and mixed together, inductively coupled plasma-atomic emission spectrometry at Galbraith Laboratories (Knoxville, TN) was used to determine the iron content in the composite crystals. The iron content of the composite crystals was found to be 0.375%, which translates to an iron oxide content by mass of 0.57%. The catalyst percent in the propellant formulation was 0.21%. This catalyst mass fraction was kept constant for all the catalyzed propellants in this study.

2.2. Experimental methods

Three diagnostic techniques were applied: strand burns, surface imaging at 1 atm, and 5 kHz OH PLIF. These will be discussed below.

2.2.1. Strand burns and surface imaging

Strand burns to determine global burning rate (overall burning rate of the propellant) were performed at four pressures: 1, 5.8, 50, and 149 atm (0.1, 0.6, 5.1, and 15.1 MPa). At least three strands were burned for each propellant at each pressure. Propellants were burned in air at 1 atm. The strand burns at elevated pressures were carried out in a Crawford strand burner using nitrogen as the inert gas. Images were acquired at 1000 fps using either a Phantom 7.3 or a Phantom Miro eX4 high-speed camera.

The Phantom 7.3 high-speed camera was also used for strand burns and visual imaging of the propellants at 1 atm. For visual imaging, an Infinity K2 Long-distance Microscope Lens was used to examine the burning surface. Images were acquired at 1000 fps. A high-intensity light source was used to illuminate the propellant surface.

| Table 1 | Та | ble | 1 |
|---------|----|-----|---|
|---------|----|-----|---|

Propellant formulations.

| Propellant | Coarse AP (%) | Fine AP (%) | Composite particles (%) | Micron iron oxide (%) | Nano iron oxide (% |
|-----------------------------|-------------------------------|-------------------|-------------------------|-----------------------|--------------------|
| 1 | 40.00 | 40.00 | _ | _ | - |
| 2 | 39.89 | 39.89 | - | 0.21 | - |
| 3 | 39.89 | 39.89 | - | - | 0.21 |
| 4 | 40.00 | - | 40.00 | - | - |
| All propellants ha R45-M | ad 20% binder with the follow | ving composition: | | | 14.58 |
| | | | | | 2.92% |
| IDP | | | | | |
| E744 Curative | | | | | 2.30% |
| Tepanol HX-878 | | | | | 0.20% |

Download English Version:

https://daneshyari.com/en/article/10264287

Download Persian Version:

https://daneshyari.com/article/10264287

Daneshyari.com