Ignition and combustion of mechanically alloyed Al–Mg powders with customized particle sizes

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A B S T R A C T

Adding aluminum to propellants, pyrotechnics, and explosives is common to boost their energy density. A number of approaches have been investigated that could shorten aluminum ignition delay, increase combustion rate, and decrease the tendency of aluminum droplets to agglomerate. Previous work showed that particles of mechanically alloyed Al·Mg powders burn faster than similarly sized particles of pure aluminum. However, preparation of mechanically alloyed powders with particle sizes matching those of fine aluminum used in energetic formulations was not achieved. This work is focused on preparation of mechanically alloyed, composite Al·Mg powders in which both internal structures and particle size distributions are adjusted. Milling protocol is optimized to prepare equiaxial, micron-scale particles suitable for laboratory evaluations of their oxidation, ignition, and combustion characteristics. Oxidation of the prepared powders is studied using thermo-analytical measurements. Ignition is characterized experimentally using an electrically heated filament setup. Combustion is studied using a constant volume explosion setup for the powder cloud combustion, and a laser ignition setup for characterization of combustion rates and temperatures for individual particles. For the prepared materials, ignition and combustion characteristics are compared to those of pure Al. It is observed that the mechanically alloyed powders ignite at much lower temperatures than Al. Once ignited, the particles burn nearly as fast as Al, resulting in an overall improvement of the combustion performance.

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1. Introduction

Metal-based, binary, mechanically alloyed and nanocomposite materials offer advantageous performance as fuel additives in energetic formulations for propellants, explosives, and pyrotechnics [1–5]. Recent research is focused on aluminum-based materials with reactivity enhanced compared to pure Al powders. These materials have high combustion enthalpies (typical of Al), with tailored density, high energy density, and reduced ignition delays (i.e., alloys or composites of Al·Mg, Al·Ti, etc.) [6–11]. In addition, research was active to produce nano-sized powders of reactive metals [12–14], which achieve high burn rates due to highly developed reactive surface [15]. New materials prepared by arrested reactive milling [16] combine advantages of conventional, micron-sized composite or alloyed powders with reactivity of energetic nanomaterials [7,17–21]. In order for these materials to be practically useful, their particle size distribution should be adjusted to make them compatible with the existing protocols for preparation of energetic formulations. However, cold welding that often occurs due to the ductile nature of Al prevents synthesis of mechanically alloyed powders with useful size distributions. Process control agents (PCAs) have been used previously to avoid or minimize cold welding; similarly, smaller particle sizes can, in principle, be obtained by using smaller milling balls [22]. However, Al-based mechanically alloyed powders with attractive compositions prepared to date [9,23] were too coarse for practical applications. Long milling durations and increased PCA amounts can be used to further reduce particle size, but milling tools and PCAs contaminate the desired formulations [22].

This paper discusses the preparation and characterization of mechanically alloyed Al·Mg powders with customized particle size distributions. Al–Mg alloys have been explored for applications in energetic materials for a long time. Some of the first applications were in pyrotechnic formulations [24–26] while more recently, such alloys were successfully added to propellants [27]. It has been reported that Al–Mg alloys ignite at much lower temperatures than pure Al [28,29]. Combustion of alloys was also studied, with most detailed experimental observations on coarse particles with dimensions exceeding 100 μm [30–32]. It was reported that depending on composition, either Al or Mg determine the alloy combustion dynamics [33]; alternatively, a staged combustion behavior was described, in which Mg combustion preceded that of Al [34]. Despite significant attention to combustion

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characteristics of Al–Mg alloys [31,35,36], combustion of fine particles of such alloys has not been studied in detail, most likely because such fine particles are not readily available. Thus, enabling preparation of fine Al–Mg alloy powders by mechanical alloying, a very versatile materials manufacturing method, is of substantial interest. The issue is challenging primarily due to high ductility of Al, which tends to result in cold welding and coarse product particles. In previous research, elemental iodine was milled with aluminum resulting in particle size reduction [37]. In this work, iodine is used as an additional PCA enabling a better control over the particle shapes and sizes for an Al–Mg alloy. A 2-step ball milling technique is implemented with the first milling step used to achieve the desired structural and compositional refinement. Iodine is added and smaller milling balls are used for the second milling step aimed to reduce particle sizes. Combustion and ignition of the prepared materials is investigated and correlations between different oxidation, ignition, and combustion characteristics are studied.

2. Experimental

2.1. Material synthesis

Starting materials included elemental powders of Al (Atlantic Equipment Engineers, 99.8% pure, −325 mesh) and Mg (Alfa-Aesar, 99.8% pure, −325 mesh). Powders were blended and mechanically milled using a Retsch PM-400 MA planetary mill. Nominal powder composition was $Al_{0.47}Mg_{0.53}$, or 1:1 for Al:Mg by weight. The rotational speed was set to 350 rpm. The direction of rotation was set to reverse every 15 min.

The powders were milled in two steps. The first step was aimed to achieve compositional homogeneity between components as in conventional mechanical alloying. The second step was added to tailor the particle size distributions of the produced powders using additional PCA and altered milling conditions. The powder prepared in the two-step (2-step) milling was compared to a reference mechanically alloyed sample prepared in one step (1-step) with a duration equal to the total milling time of the 2-step procedure. Additional comparisons were made with the powder prepared using only the first step (1st step) of the 2-step procedure.

For the 1st step milling, elemental powders of Al and Mg were loaded in 0.5-l steel milling vials with 9.5-mm diameter hardened steel milling balls. Vials were filled in argon with a 30-g powder load and 300 g of milling balls (charge ratio of 10). 50 ml of hexane (C_6H_{14}) was added to each milling vial as a PCA. The milling duration was 120 min. This completed the 1st step milling. For further processing, vials were opened in argon and iodine (I_2, chips, Sigma Aldrich, 99% pure) was added as 4 wt.% of the initial powder load to serve as a new PCA. After adding iodine, the vials were closed and milled for 5 min. to clean the 9.5-mm milling balls, which were then removed and replaced with the same mass of 3-mm diameter steel milling balls. The balls were replaced inside an Ar-filled glovebox. The second, softer milling step was then performed to produce mechanically alloyed powder with the desired finer particle sizes. The duration of the second milling step was 60 min. The total milling time was, therefore, 185 min. This milling time was also used to prepare reference samples milled in a 1-step process. Other milling conditions corresponded to those used in the first step of the 2-step milling, while iodine (4 wt.%) was included with the starting blend of Al and Mg powders.

The prepared powders were removed from the milling vials in the Ar-filled glovebox. The powder was left in the glovebox overnight, where it was exposed to an environment with low oxygen concentration (∼2%). This exposure passivated the powder surface so that the powder could then be recovered and handled in open air.

2.2. Material characterization

Scanning electron microscopy (SEM) was used to study powder morphology using a Phenom Tabletop Microscope by FEI Technologies Inc. Backscattered electron images were taken to inspect particle shapes and sizes. Particle size distributions of the prepared composites were measured with low-angle laser light scattering using a Beckman-Coulter LS230 Enhanced Particle Analyzer. Powder suspensions for analysis were prepared in ethylene glycol ($C_2H_6O_2$) and sonicated to minimize particle agglomeration. Phase compositions of the samples were analyzed using X-ray Diffraction (XRD) on a Phillips X'pert MRD powder diffractometer operated at 45 kV and 40 mA, using Cu Kα radiation (λ = 1.5438 Å). The patterns were collected between 10° and 85° at a rate of 0.625°/min.

Thermal stability and temperature-dependent phase transformations were studied using differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) using a Netzsch Simultaneous Thermal Analyzer STA409 PG with a DSC sample carrier and corundum sample crucibles. Oxidation experiments were performed in a mixed 1:1 ratio of oxygen (50 ml/min, Matheson, 99.98% purity) and ultra-high purity argon (50 ml/min, Matheson, 99.99%) at various heating rates between 5 and 40 K/min. Decomposition experiments were performed in argon only and at a heating rate of 5 K/min. Ignition of the prepared materials was analyzed using a heated filament ignition apparatus described in detail elsewhere [38,39]. A thin layer of powder was coated onto a nickel–chromium alloy wire (manufacturer-specified working temperature range up to 1953 K). The wire was heated by a DC current and its temperature was monitored optically using an infrared pyrometer focused on its uncoated surface adjacent to the powder coating. Concurrently, light emission from the powder coating was measured using a photodiode sensor, in which a sharp onset of light emission was identified as the ignition instant. The temperature measured by the pyrometer at that instant was assumed to be the ignition temperature. Such experiments were conducted in air for a range of heating rates between 10^3 and 10^5 K/s. Ignition instant was also identified from videos recorded using a high-speed camera (MotionPro500 by Redlake) at 500 fps.

Combustion was studied using a constant volume explosion (CVE) setup for powder aerosol combustion, and a laser ignition setup for characterization of combustion rates and temperatures for individual particles. The details of the CVE experiments were described elsewhere [40–42]. The aerosolized powders were ignited from the center of a nearly spherical 9.2-L vessel. The vessel was initially evacuated, after which the powder was introduced with an air blast by opening a solenoid valve connecting the vessel with a chamber filled with compressed air. After the air blast, ignition was triggered following a delay of 300 ms, provided to reduce turbulence. The targeted initial pressure in the explosion vessel prior to ignition was 1 atm. Combustion pressure traces were recorded using both static and dynamic transducers. The measured pressures traces are normalized by the initial pressure in the vessel (P/P_0). The rates of pressure rise, $d(P/P_0)/dt$, are also reported for the prepared composite and reference pure Al powder. (P/P_0) and $d(P/P_0)/dt$ are proportional to the flame temperature and combustion reaction rate, respectively [43].

For laser-ignited single particle combustion, the experimental technique is described in Refs. [44–46]. A vertically rising particle jet with low number density was generated using a vibratory powder feeder [47]. Particles ignited when crossing a CO_2 laser beam. Luminous streaks produced by the ignited particles were photographed. Four photomultiplier tubes (PMT, Hamamatsu H3164-10) were used to record the light emission from burning particles. The PMTs were equipped with interference filters (486, 532, 568, and 589 nm). Emission signals filtered at 532 and 589 nm were used for optical pyrometry. The 486 nm filter was