Impact initiated combustion of aluminum exposed to mechanical pre-activation

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Abstract. The impact initiation of as-received and mechanically activated aluminum powder compacts is investigated through uniaxial stress rod-on-anvil impact experiments. The compacts reveal light emission due to combustion reaction at velocities greater than 165 m/s. Mechanical pre-activation, such as that achieved via high-energy ball milling (HEBM) or high strain machining, strain hardens the starting materials, affecting their combustion initiation behavior. The starting materials are characterized by their lattice strain, size, and surface area to volume ratio. High speed imaging reveals that the threshold velocity (minimum velocity necessary for reaction initiation) changes as a function of the mechanical pre-activation.

1. Introduction

Intermetallic-forming energetic materials are highly desirable for several applications due to their energy release characteristics and their mechanical properties. The impact initiation of intermetallic reactions, such as Ni + Al, Ta + Al, and W + Al, is dependent on the mixing of the two reactants, the varying elastic/plastic properties of the reactant metals, the degree of mechanical pre-activation, and the reactant morphologies [1–3]. The understanding of the initiation of these intermetallic reactions is further complicated by the ability of aluminum to combust on its own.

The combustion of aluminum is assumed to proceed according to the well-known reaction [4],

$$Al + O_2 \rightarrow AlO + O, \tag{1}$$

which relies on the movement of atmospheric oxygen through the Al_2O_3 shell in order to interact with the aluminum. The properties and behavior of the Al_2O_3 shell and thus the reactivity of aluminum have been shown to be a function of the size of the particles [5], the temperature that the aluminum particles are exposed to [6], and the degree of mechanical pre-activation [7].

High energy ball milling (HEBM) of powders is often used to control the degree of mechanical pre-activation. Depending on processing parameters and the reactant constituents, HEBM has been observed to either increase or decrease the amount of energy necessary for reaction initiation under impact conditions [8–10]. Processing by HEBM encourages the mixing of reactants and exposes fresh reactant surfaces. However, it also causes strain hardening of the constituents. In the case of the Ni + Al system, reactivity has been shown to be enhanced after short milling times, due to the increased mixing of reactants and the exposure of fresh reactant surfaces. Yet, after long milling times, reactivity diminishes as a result of work hardening and the creation of localized interfacial reaction between the Ni and Al [9].

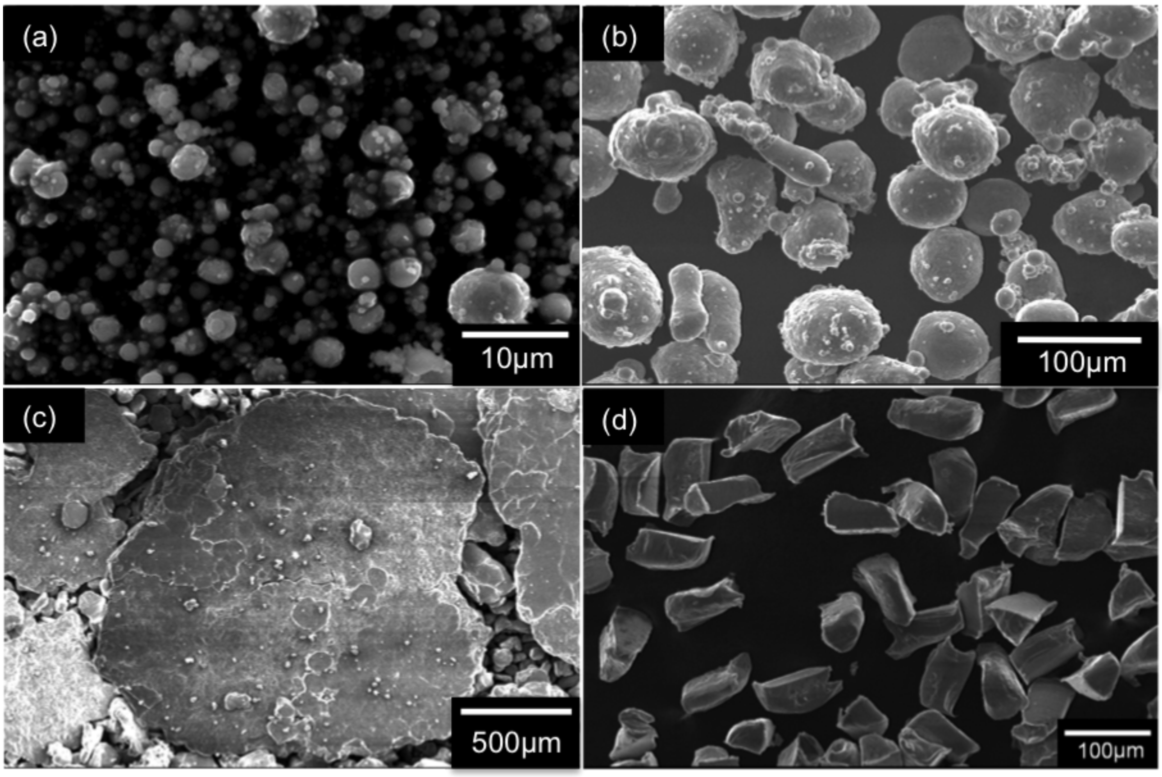

Figure 1. Micrographs of various aluminum powders. (a) H-2 Aluminum powder, (b) H-60 Aluminum powder, (c) H-60 milled for 10 minutes, (d) Highly strained chips of 6061 aluminum.

In order to better understand the variables dominating the initiation of intermetallic-forming reactions under impact conditions, the initiation of combustion in aluminum is assessed by comparison of mechanically activated aluminum powders to as-received aluminum powders. The compacts reveal light emission due to combustion reaction at velocities above a threshold velocity, which changes as a function of the mechanical pre-activation and initial particle size.

2. Experimental

The affect of mechanical pre-activation on the impact-initiated combustion of aluminum powder compacts was studied by comparison of as-received aluminum powders to aluminum powders that had been strain-hardened by either ball-milling or by high strain machining. Spherical, as-received powders of two different diameters were investigated, a 7.5 μm diameter powder (Valimet H-2, figure 1a), and a 105 μm diameter powder (Valimet H-60, figure 1b). A Saltykov analysis of the particle size distribution gave a normal particle size distribution for both as-received powder sets.

The third powder type (figure 1c) was prepared by loading 10 g of Valimet H-60 aluminum powder into an alumina cell and ball milling with a 12.6 mm diameter alumina sphere in a SPEX mill for 10 minutes. During the milling process, the spherical powders were observed to fracture, plastically deform, and cold weld. Resulting particles had a flattened, flake-like shape and gave a bimodal distribution of particle sizes that centered around diameters of 10 μm and 800 μm. The fourth powder type, highly strained aluminum 6061 chips (figure 1d) was prepared by Shankar et. al. [11, 12] using a plane-strain machining method. These chips contain ultrafine grains that strengthen the Al6061 and are approximately 50 μm $\times$ 50 μm $\times$ 30 μm in size[13].

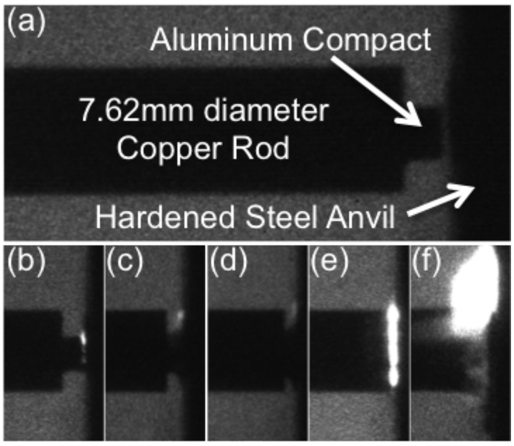

Figure 2. High-speed images of a HEBM aluminum powder compact affixed to a copper projectile impacting a hardened steel anvil at 302 m/s. (a) immediately prior to impact. (b-d) t = 0.05 to 2.5 μs following impact: light emission during compaction and deformation of pellet. (e) t = 6.5 μs following impact: light emission after complete deformation of the compact. (f) t = 9.5 μs following impact: sustained reaction of the HEBM pellet as the Cu projectile makes contact with the anvil.

SEM and XRD were performed on both the as-received and highly-strained aluminum powders. Line broadening observed in the XRD measurements was analyzed by the Williamson-Hall method to determine the retained lattice strain for each sample type.

All starting powders were quasi-statically pressed into a $\approx$ 95% dense cylindrical powder compact (2.0 $\pm$ 0.1 mm high $\times$ 3.19 mm diameter). The surface area to volume ratio S_v) of each powder type was measured by an intersection counting method [14]. The compacts were cross-sectioned, polished, etched with a Kellers reagent, and imaged by light microscopy (Leica DM IRM). Cycloids were super-imposed on the image, and the average number of intersections between the cycloid and the microstructural boundaries per unit of the cycloid length (P_L) were related to the surface area per unit volume, S_v via the relationship, $S_v = 2P_L$.

The aluminum powder compacts were affixed to the lapped end of a copper projectile (38.1 mm high $\times$ 7.62 mm diameter) via 24 hour Hysol epoxy and impacted against a hardened steel anvil using a 7.62 mm gas gun. The impact event was imaged via a high-speed framing camera (IMACON 200) in order to determine the initiation of a combustion reaction during the crush-up process. The observation of light in several consecutive frames captured by the high-speed framing camera was used as indicative of an impact-induced combustion reaction (figure 2). Each powder compact was tested at a range of velocities, from 140 m/s to 320 m/s, to determine the threshold velocity, defined as the minimum velocity necessary for reaction to initiate under each condition. All experiments were performed under ambient environmental conditions.

Table 1. Summary of powder compact properties.

Sample Type	Size (μm)	Surface area to volume ratio (mm^{-1})	Lattice strain (η)	Threshold velocity for reaction (m/s)
Valimet H-2	7.5	1885.7	1.00×10^{-4}	235
Valimet H-60	105	105.7	-1.30×10^{-3}	195
HEBM Valimet H-60	10 to 800	163.4	1.20×10^{-3}	165
Highly Strained Al 6061	50×50×30	130.8	6.00×10^{-3}	235

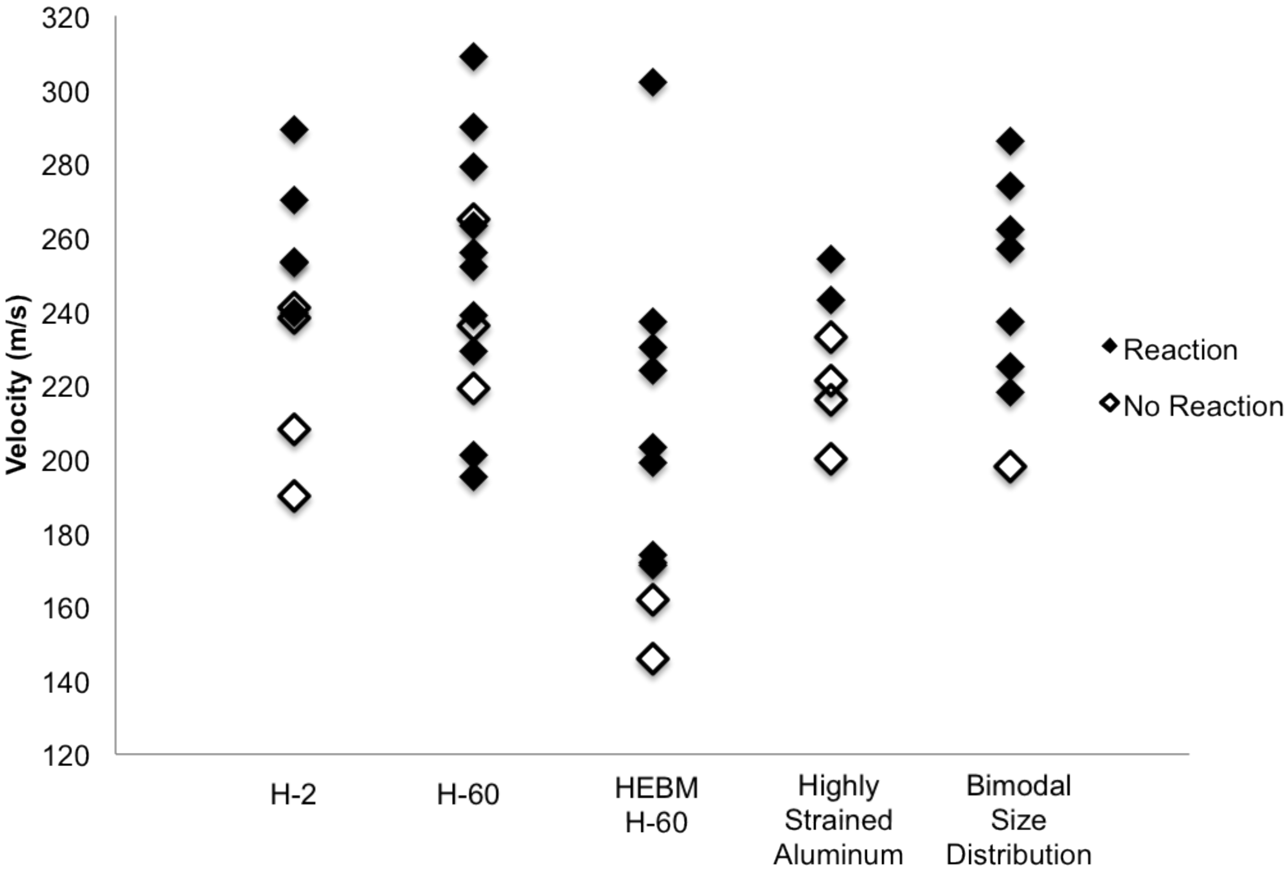

Figure 3. Reactivity of each sample type with respect to velocity of impact.

3. Results

Particle size, surface area to volume ratio, lattice strain, and degree of mechanical pre-activation were analyzed to aid in the description of the dominant mechanism of the impact initiated combustion of aluminum (table 1). The smallest, H-2, particles had the highest S_v, by an order of magnitude, of all samples tested. The remaining samples, H-60, HEBM H-60, and highly strained aluminum 6061 all had a S_v between 100 and 160 mm^{-1}. The lattice strain in the as-received spherical particles was negligible ($\leq 10^{-4}$). The HEBM H-60 particles had a lattice strain of 1.20E-3 and highly strained aluminum 6061 had lattice strain of 6.00×10^{-3}.

The reactivity of each powder compact type was plotted against the velocity measured during impact (figure 3). The smaller, 7.5 μm diameter particles, H-2, had a higher reaction threshold velocity (235 m/s) than the larger, 105 μm diameter particles, H-60 (195 m/s). Upon pre-straining by HEBM, the H-60 particles exhibited a drop in reaction threshold to 165 m/s. By comparison, the other highly strained aluminum particles, the plane-strained, equiaxed, ultra-fine grained aluminum chips had a significantly higher reaction initiation threshold velocity of 235 m/s. In order to examine the effects of particle size distribution alone, a fifth powder mixture was prepared, composed of a 1:3 by volume ratio of H-60 and H-2 aluminum. This mixture gave a reaction threshold velocity of 210 m/s.

4. Discussion

The threshold velocity for reaction for this set of experiments displayed correlations with both the degree of mechanical pre-activation and the starting particle size (table 1). Mechanical pre-activation of the aluminum powders both strain hardens the starting powder and exposes fresh reactant surfaces. The presence of strain hardening was confirmed by measuring the lattice strain using the Williamson-Hall method.

The lattice strain for highly strained aluminum 6061 was significantly higher than the lattice strain of any other sample tested but resulted in decreased reactivity. On the other hand, the other set of samples exposed to mechanical pre-activation, the HEBM Valimet H-60, showed enhanced reactivity. Strain hardening by mechanical pre-activation can have a pronounced effect on the particle-particle interactions, which dictates the energy input necessary for the compaction and deformation process within the powder compact during impact. Reactivity in the highly strained aluminum 6061 was most likely decreased because the harder, stronger particles required more energy be expended for compaction and deformation of these higher strength particles, leaving less energy available for reaction initiation. For the HEBM H-60 particles, high energy ball milling for a short period of time caused some strain hardening as observed by the lattice strain parameter but also resulted in plastic deformation in the aluminum powders. The plastic deformation achieved by HEBM might have aided in the compaction and deformation process during impact, leaving more energy available for reaction initiation. Additionally, mechanical pre-activation can result in creation of fresh reactant surfaces, which, in the case of aluminum particles, leaves a thinner, more easily penetrable Al_2O_3 coating, which serves to enhance reactivity.

With respect to particle size, the threshold velocity for reaction of the as-received aluminum powders was observed to decrease with increasing size. The bimodal particle size distribution, composed of a 1:3 by volume ratio of 105 μm to 7.5 μm particles showed decreased reactivity. Both smaller particles and the bimodal particle size distribution result in a powder compact that is more dense, with smaller, more uniform pore spacing, than a compact composed of mostly larger particles. Compacts containing larger particles have larger, more randomly distributed pores and are therefore subjected to increased plastic deformation and heating during the compaction and deformation process during impact. Although more energy is necessary to plastically deform the particles, the enhanced heating serves to augment the reaction in the larger particles.

5. Conclusion

The degree of mechanical pre-activation and particle size both influence the reactivity of aluminum under impact conditions. Powder compacts composed of the larger, 105 μm, particles were more easily initiated than the smaller, 7.5 μm particles. The impact initiated reactivity of the 105 μm particles became even further enhanced after those particles underwent processing by high energy ball milling. Yet, particles undergoing a rigorous high strain machining process were the least reactive, due to effects of work hardening. Studies that investigate a larger range of particles sizes and of HEBM processing times are needed to gain a clearer understanding of the mechanisms involved in initiating reaction in aluminum under impact conditions.

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