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Ignition of aluminum-rich Al–Ti mechanical alloys in air

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Abstract

Ignition of metastable Al–Ti mechanical alloys with titanium concentrations from 10 to 25 at% was investigated experimentally. A thin layer of powder was coated on an electrically heated filament. The ignition instant was identified from the powder's radiation measured in real time. Simultaneously, filament temperatures were measured using a high-speed infrared pyrometer to determine the ignition temperature. The experiments were conducted at different filament heating rates in the range of 3×10^3–2×10^4 K/s to determine the ignition kinetics. The ignition temperatures and kinetics were compared to the respective characteristics of the phase changes and oxidation steps observed for the same mechanical alloys using thermal analysis. It was shown that at the heating rates exceeding 10^3 K/s, the exothermic formation of a metastable $L1_2$ phase of Al_3Ti occurring during heating of the Al–Ti mechanical alloys triggers their ignition. This conclusion was confirmed by additional ignition experiments in which annealed mechanical alloys already containing this transition Al_3Ti phase did not ignite in the same temperature range as fresh mechanical alloys. The ignition kinetics identified for Al–Ti mechanical alloys based on thermal analysis and on ignition experiments enables one to predict ignition temperatures as a function of both composition and heating rate. Specifically, extrapolation is possible to higher heating rates typical for aerosol flames.

Keywords: Metal powder ignition; Reaction kinetics; Heated filament; Phase changes

1. Introduction

Powders of metastable aluminum-based alloys have been proposed as potential solid fuels in order to increase burn rates of aluminized propellants and other energetic formulations [1–3]. While the overall combustion enthalpy would not be affected significantly, the phase changes occurring on the metastable relaxation of such alloys were expected to accelerate the reaction kinetics [1,4]. Feasibility tests have indeed shown higher burn rates for flames of aerosolized powders of several aluminum-based alloys (e.g., Al–Mg, Al–Ti, Al–Li, and Al–Zr) in air [1–3]. Such alloys typically include amorphous metallic phases or a supersaturated solid solution of an alloying element in aluminum. In addition, equilibrium intermetallic phases can be present. The exothermic phase changes occurring in such systems result in the decomposition of supersaturated solid solutions and formation of intermetallic phases. The enthalpy of such phase changes is much less than that of oxidation. However, it was hypothesized that if timed

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properly, this additional heat release could significantly accelerate ignition and as a result increase the global combustion rate.

Among several aluminum-based alloys considered in previous preliminary work [5], particle combustion rates for aluminum-rich Al–Ti mechanical alloys were found to exceed most significantly the combustion rates of aluminum particles of similar size. Therefore, a more detailed study of this material system was warranted. Specifically, it was of interest to establish which of the phase transformations occurring in Al–Ti mechanical alloys upon heating would affect their ignition. A study of the phase changes in samples heated in argon was conducted using differential scanning calorimetry [6,7]. The exothermic formation of metastable and equilibrium Al_3Ti phases was observed to occur in several steps. Exothermic subsolidus transformations resulting in the formation of the metastable $L1_2$ and $D0_{23}$ phases occurred in the range of 500–800 K. The stable $D0_{22}$ phase of Al_3Ti formed in the temperature range of 990–1040 K above the eutectic melting of Al (∼935 K). A detailed description of the transitions leading to the formation of the stable $D0_{22}$ form of Al_3Ti is given in Ref. [6]. It was also clear that the oxidation mechanism of an Al–Ti mechanical alloy would differ from that of pure aluminum. Different alumina polymorphs, oxides of titanium, or ternary oxides form resulting in either slower or faster oxidation as compared to pure aluminum. To characterize the oxidation processes for such alloys, a thermogravimetric study combined with differential thermal analysis was carried out in which the mechanical alloy samples were heated in an oxygen flow [8,9]. For Al-rich alloys, the oxidation proceeded stepwise. The thermal kinetics of the oxidation steps was not observed to correlate with the thermal kinetics of the exothermic phase changes in which Al_3Ti formed. Thus, when mechanically alloyed Al–Ti powders are heated in an oxidizing environment, their ignition is preceded or accompanied by three weakly exothermic transitions leading to the formation of increasingly stable Al_3Ti phases, while oxidation occurs concurrently in several distinct steps.

This paper is aimed at determining whether it is possible to identify the process governing ignition of the aluminum-rich Al–Ti mechanical alloys. The ignition in practical systems usually occurs at very high particle heating rates (e.g., 10^3–10^6 K/s) that exceed by far the typical heating rates of thermal analysis (<100 K/min). Therefore, the approach of this study was to study the kinetics of ignition of the Al–Ti mechanical alloys directly and compare the results to the kinetics of the phase transitions and oxidation steps identified earlier by thermal analysis.

2. Materials

Aluminum–titanium mechanical alloys containing 10, 15, 20, and 25 at% titanium were used in the experiments. The starting materials were elemental powders of Al (Alfa Aesar, 99.8%, −40 + 325 mesh) and Ti (Alfa Aesar, 99%, −325 mesh). The powders where mechanically alloyed for 15 h using an 8000D series shaker mill by SPEX CertiPrep. The mechanical alloying was carried out within zirconia vials. The amount of powder used in the alloying cycle was 5 g and the total weight of milling media (3/8 inch diameter ZrO_2 balls) was 50 g, giving a ball to powder mass ratio of 10. Before alloying, 2 wt% of a process control agent (stearic acid) was added.

The size distributions of the obtained powders were measured using a Beckman Coulter LS 230 laser diffraction particle size analyzer. The size distribution parameters, including the volume mean particle sizes, median particle sizes, and standard deviations obtained for different powders, are shown in Table 1. Particle-size distributions for different mechanical alloy compositions were qualitatively similar. Gener-

Table 1
Characterization of particle sizes of synthesized Al–Ti mechanical alloys using low-angle laser light scattering

Alloy	Volume mean diameter (μm)	Median diameter (μm)	Standard deviation (μm)
$Al_{0.90}Ti_{0.10}$	28.08	22.06	19.96
$Al_{0.85}Ti_{0.15}$	27.99	22.85	19.04
$Al_{0.80}Ti_{0.20}$	22.70	16.00	17.86
$Al_{0.75}Ti_{0.25}$	19.42	13.83	16.19

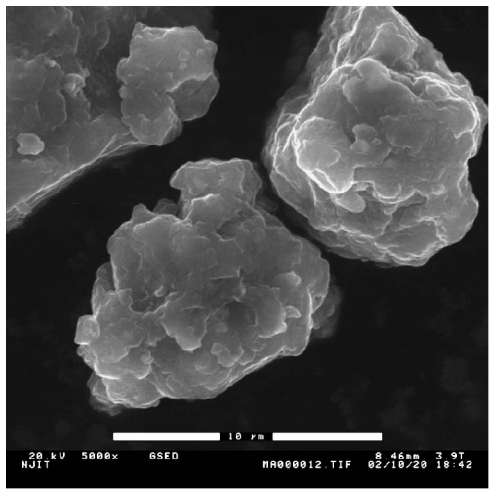

Fig. 1. SEM image of $Al_{0.90}Ti_{0.10}$ mechanical alloy particles.