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Ignition mechanism of nickel-coated aluminum particles

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Abstract

The use of nickel-coated aluminum particles (10– $100~\mu m$) as a fuel component in propellants is expected to improve the performance characteristics of solid rocket motors. They can also be used for combustion synthesis of nickel aluminides. The ignition mechanism of such particles, however, is not well understood. In this study, to provide sufficient spatial and time resolution, single ~ 2.5 -mm Ni-coated Al particles were laser-heated in argon and carbon dioxide atmospheres. The ignition process was investigated using a high-speed digital video camera, thermocouple measurements, and analysis of particles quenched at different preignition stages. It was shown that the particle ignition temperature is $\sim 1325~\text{C}$ and does not depend on atmosphere (Ar or CO_2) or Ni content. The established ignition mechanism includes exothermic intermetallic reactions and phase transformations of different Ni–Al compounds. Specifically, the melting of NiAl $_3$ at 854 $^{\circ}\text{C}$ plays a critical role in particle ignition.

Keywords: Heterogeneous combustion; Metals; Aluminum; Ignition; Propellants; Combustion synthesis

1. Introduction

The elucidation of the ignition mechanism for Nicoated Al particles is of interest for propulsion applications. It has been proposed [1] to replace the Al powder in aluminized solid rocket propellants with Ni-coated Al particles to improve the combustion characteristics of Al. Such particles can be produced by a variety of methods (e.g., electroplating, electroless plating, and chemical vapor deposition), which provide uniform coatings and are relatively inexpensive.

The use of Al powder as a fuel additive in solid rocket propellants improves performance by increasing the specific impulse and eliminating combustion instabilities. There exist, however, some drawbacks to the use of Al powder in propellants. Aluminum particles ignite at $\sim\!2000\,^\circ\text{C}$ due to the existence of a thin alumina (Al₂O₃) surface layer that prevents the oxidation of Al at lower temperatures. There is also a tendency for Al particles to agglomerate during propellant burning, which leads to incomplete combustion of Al and slag formation inside the solid rocket motor [2].

Nickel-coated Al particles show promise for overcoming such drawbacks. Prior studies [1,3] demonstrated that coating Al with Ni reduced particle agglomeration during propellant combustion. In addition, experiments [4] on the combustion of air/metal particle clouds have shown that the flame propagation velocity for Ni-coated Al particles is significantly higher than that for Al particles. It is thus expected that the utilization of Ni-coated Al particles will increase the efficiency of aluminized rocket propellants.

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In addition, Ni-coated Al particles can potentially be used to synthesize nickel aluminides, which are lightweight materials with high melting points, good strength characteristics, and excellent corrosion and oxidation resistance. These features make such materials attractive for use in aircraft turbines and coating applications. Nickel aluminides are produced by various methods, including combustion synthesis, which can be conducted as a batch process using compacts of either Al and Ni powder mixtures [5–10] or Ni-coated Al particles [10,11]. Also, experiments in microgravity (conducted in NASA's drop tower, in research aircraft, and later onboard Mir space station) demonstrated propagating combustion fronts in clouds of Ni-coated Al particles in inert atmosphere [12]. These observations suggest that nickel aluminide powders could also be synthesized by a continuous combustion process where Ni-coated Al particles are suspended in an inert gas flow.

For both the propulsion and materials synthesis applications, a better understanding of the ignition mechanism for single Ni-coated Al particles is critical. Note that an earlier mechanism postulated that particles ignite in oxidizing atmospheres due to thermal stresses, which cause the shell to crack and peel away from the surface, allowing oxidation of the bare Al core [4]. This mechanism does not account for intermetallic reactions between Ni and Al and is clearly inapplicable to ignition in inert atmospheres.

Useful information on the interactions between Ni and Al was obtained in prior studies of the combustion synthesis of nickel aluminides from compacted mixtures of Ni and Al powders [5-10]. It was shown that the ignition process involved the melt-spreading of liquid Al over solid Ni to form intermediate nickel aluminides prior to the synthesis of the final product (NiAl or Ni₃Al). Studies [10,11] on the combustion of compacted Ni-coated Al particles indicate a similar ignition mechanism, whereby the solid Ni shell cracks due to thermal expansion of the liquid Al core, allowing Al to melt-spread and react with the Ni surfaces. Reaction inside the particles to form NiAl₃ was also detected, which indicates that, while the meltspreading of Al plays a major role in the ignition of Ni-coated Al particle compacts, single particles may ignite in a different manner.

To provide insight into the ignition and combustion of single Ni-coated Al particles, experiments were conducted involving laser ignition of electrodynamically levitated particles (diameter 30–100 μ m) in different gas environments (air, O₂, CO₂, Ar) [13,14]. In addition, single Ni-coated Al particles (100–125 μ m) were injected into a vertical, preheated tube furnace to determine *critical ignition temperatures* [14]. The results indicated that the Ni coating dramatically decreases both the ignition delay time

and the critical ignition temperature (by $\sim 1000\,^{\circ}\text{C}$) of Al particles. It was hypothesized that exothermic intermetallic reactions between liquid Al and solid Ni play a major role in the ignition mechanism. The experimental technique, however, lacked the spatial and temporal resolution required to reveal ignition details

In the present work, laser ignition of ~ 2.5 -mm Ni-coated Al particles was performed to establish the sufficient resolution needed for investigating the ignition mechanisms and determining particle temperatures and microstructure during reaction. The substantial ignition delay for the larger particles allows the ignition processes to be effectively studied at different reaction stages using high-speed digital video recording, thermocouple data acquisition, and particle quenching. To determine the influence of atmosphere on ignition, experiments were conducted in both argon (Ar) and carbon dioxide (CO₂) environments. The importance of studying ignition in CO2 atmosphere is twofold. First, CO₂ and H₂O are the primary gases that oxidize Al during the burning of aluminized solid rocket propellants [2,15]. Second, the high ignition temperature of Al prevents its consideration for metal/CO₂ propulsion on Mars [16–18]; thus Ni-coated Al particles are promising candidates for this application.

2. Experimental

The particles used for this research were 99% pure Al spheres (diameter 2.38 mm, Alpha Aesar) coated with Ni by Federal Technology Group (Cleveland, OH) using a cyclic electroplating method [19]. The coating process includes the removal of surface alumina, thus allowing the direct deposition of Ni onto Al. In this work, particles with three different coating thicknesses were studied: 7.6 µm (6 wt% Ni), 47 μm (29 wt% Ni), and 147 μm (58 wt% Ni). Fig. 1 shows images of unreacted, cross-sectioned samples obtained using scanning electron microscopy (JEOL JSM-840). Regarding the wide range of Ni content investigated in this study, it is beneficial to minimize Ni fraction for propulsion (i.e., 6 wt% Ni), while stoichiometric ratios are preferred for materials synthesis applications (i.e., 58 wt% Ni).

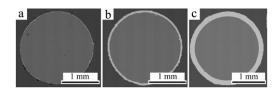


Fig. 1. SEM images of Ni-coated Al particles: (a) 6 wt% Ni, (b) 29 wt% Ni, and (c) 58 wt% Ni.

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