



Effect of packing density on flame propagation of nickel-coated aluminum particles



Dilip Srinivas Sundaram, Vigor Yang*

School of Aerospace Engineering, Georgia Institute of Technology, Atlanta, GA 30032, USA

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ABSTRACT

The combustion-wave propagation of nickel-coated aluminum particles is studied theoretically for packing densities in the range of 10–100% of the theoretical maximum density. Emphasis is placed on the effect of packing density on the burning properties. The energy conservation equation is solved numerically and the burning rate is determined by tracking the position of the flame front. Atomic diffusion coefficients and reaction rate of isolated nickel-coated aluminum particles are input parameters to the model. The burning behaviors and combustion wave structures are dictated by the heat transfer from the flame zone to the unburned region. Five different models for the effective thermal conductivity of the mixture are employed. The impact of radiation heat transfer is also assessed. As a specific example, the case with a particle size of $79\ \mu\text{m}$ is considered in detail. The burning rate remains nearly constant ($<1\ \text{cm/s}$) up to a packing density of 60%, and then increases sharply toward the maximum value of $11.55\ \text{cm/s}$ at a density of 100%. The Maxwell–Eucken–Bruggeman model of thermal conductivity offers the most accurate predictions of the burning rate for all loading densities.

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1. Introduction

Aluminum particles with diameters greater than $100\ \mu\text{m}$ ignite only upon melting of the amorphous aluminum oxide (Al_2O_3) layer at $2350\ \text{K}$ [1]. The particle ignition characteristics can be significantly enhanced by substituting a nickel coating for the oxide layer [1]. For example, the ignition temperature of $\sim 2.5\ \text{mm}$ nickel-coated aluminum particles is $\sim 1600\ \text{K}$, which is substantially lower than the melting point of aluminum oxide [2]. This phenomenon is attributed to the presence of exothermic reactions between nickel and aluminum atoms [2,3]. The flame speed of micron-sized particle dust clouds can be quadrupled by employing nickel-clad aluminum particles [4]. At nano scales, nickel coating may be used to enhance the active aluminum content of the particle [5]. As a result, nickel-coated aluminum particles have been studied for propulsion and energy-conversion applications [1–5].

Combustion synthesis is an attractive method for manufacturing nickel aluminides ($\text{Ni}_x\text{Al}_{1-x}$), which are advanced structural materials for a variety of engineering systems [6,7]. The adiabatic reaction temperature of a nickel–aluminum system is as high as $1910\ \text{K}$ [8], considerably greater than those achieved in conventional methods. The high temperatures drive out volatile impuri-

ties, producing relatively pure materials. The process can be operated in two modes. In the propagation mode [9,10], the powder is ignited at one end and a self-sustaining combustion wave propagates through the unburned mixture. The thermal explosion mode, which is applicable for systems with low exothermicity, involves heating the entire sample to the ignition temperature [11,12]. The reactant pellet is typically obtained by compacting the powder in a die using a hydraulic press [13].

Recently, the cold spray method has been employed to synthesize fully dense energetic materials [13]. The particles were entrained in a carrier gas and accelerated through a de Laval nozzle and directed to a substrate. The primary advantage of this method is the low process temperature, which helps to minimize reactions during deposition. Unlike a cold pressing technique, the product density is relatively uniform in both radial and axial directions [13]. The starting material may consist of nickel and aluminum particles or nickel-coated aluminum particles [12,13]. Understanding their burning properties is thus of paramount importance for material synthesis applications.

The physicochemical behaviors of nickel–aluminum systems has been extensively studied by means of experiments [2,10,11–14] and molecular dynamics (MD) simulations [3,15–17]. The materials of interest include multi-layered foils [11,15], core–shell structured particles [3,12–14,16], and consolidated blends of nickel and aluminum particles [10,12,13]. The reactivity of Ni–Al systems

* Corresponding author. Fax: +1 404 894 2760.

E-mail address: vigor.yang@aerospace.gatech.edu (V. Yang).

Nomenclature

c	burning-time constant
C_p	specific heat
D	pellet diameter, diffusion coefficient
d_p	particle diameter
E_A	activation energy
h	heat transfer coefficient
k	imaginary part of refractive index
K_0	frequency factor
L	pellet length
l_0	optical thickness
n	real part of refractive index
Q_a	absorption efficiency
Q_{conv}	convective heat transfer rate
Q_{gen}	heat generation rate
Q_r	heat of reaction
Q_{rad}	radiative heat transfer rate
Q_s	scattering efficiency
r	core radius
R	particle radius, universal gas constant
T	temperature
t	time
t_b	burning time
x	spatial coordinate

Greek

α	volume fraction
ρ	density
λ	thermal conductivity, wavelength
ε	emissivity
σ	Stefan–Boltzmann constant
$\dot{\omega}$	reaction rate
ϕ	volume fraction
β	extinction coefficient
η	fraction of burned reactants

Subscripts

a	ambient
f	fluid, flame
ign	ignition
m	mixture
P	preheat zone
p	particle
R	reaction zone
r	radiation
u	unburned
1	Maxwell–Eucken structure

is facilitated by melting of aluminum [2,11,12], although reactions have also been observed to occur at temperatures substantially lower than the melting point of aluminum (933 K) [18,19]. The predominant reaction pathway is the dissolution (or diffusion) of nickel atoms in liquid aluminum, formation of various inter-metallic phases at the interface, and precipitation of NiAl, a highly exothermic process [2,11]. Table 1 shows the heat of formation of several Ni–Al compounds [6]. NiAl has the highest heat of formation on a per mol-atom basis. Ignition may be caused by melting of eutectic mixture (Al–NiAl₃) [12], intermediate product (e.g. NiAl₃ or Ni₂Al₃) [12] or formation of NiAl [2]. The melting temperatures of different Ni–Al alloys are also given in Table 1. For nickel-coated aluminum particles, mechanical fracture of nickel shell may further assist the ignition process [12,16]. It is important to recognize that the ignition behavior of isolated particles could be different from that of powder compacts, due to the effects of heat losses and collective phenomenon [2]. The reactivity can be enhanced by both thermal and mechanical stimuli [14].

The flame propagation characteristics of Ni–Al multi-layered foils [20–22] and particulate systems [10,13,23,24] have been widely studied, while those of nickel-coated aluminum particles are yet to be understood. Dean et al. [13], recently, measured the burning rates of pressed and cold sprayed nickel–aluminum samples over a density range of 55–100% of the theoretical maximum density (TMD). For pellets containing nickel and aluminum particles, the burning rate decreases with increasing pellet density. An opposite trend was, however, observed for nickel-coated

aluminum particles. Samples with densities lower than 55% of the theoretical maximum density (TMD) were not considered, since they were mechanically unstable. It is apparent that the effect of packing density on burning properties of nickel-coated aluminum particles needs to be further explored. The present work aims to address this issue for densities in the range of 10–100% TMD. Five different models for the effective thermal conductivity of the mixture are employed. The model that accurately captures the measured variations of the burning rate with packing density is then determined.

2. Theoretical framework

The theoretical analysis treats one-dimensional and planar flame propagation of a uniformly packed pellet of nickel-coated aluminum particles in an argon environment. In reality, non-uniform packing of particles often takes place and results in density

Table 1
Heat of formation and melting point of Ni–Al inter-metallic compounds [2,6].

Species	Heat of formation, kJ/mol	Melting point, K
NiAl ₃	–150.624	1127
Ni ₂ Al ₃	–282.42	1406
NiAl	–118.4	1910
Ni ₃ Al	–153.134	1668

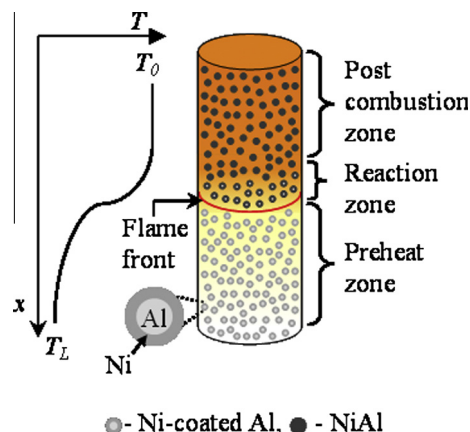


Fig. 1. Schematic of the physical model of flame propagation of nickel-coated aluminum particles.

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