



Exploring the reaction mechanism in self-propagating Al/Ni multilayers by adding inert material



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ABSTRACT

We present a new class of reactive materials termed “inert-mediated reactive multilayers” (IMRMs) that use inert material to decouple the effects of chemistry and maximum temperature in a reactive multilayer. Important considerations in the selection of composition and thickness for reactive and inert material in an IMRM are detailed. We then present the results from a specific set of IMRM samples that we fabricated using 23-nm-bilayer 1:1 Al:Ni reactive sections and 2:3 Cu:Ni inert sections. In these samples we observe a systematic reduction of heats of reaction, maximum reaction temperatures, and reaction propagation velocities as the volume fraction of reactive material is reduced. At the same time, metrics sensitive to the reaction mechanism and products indicate that there is little if any cross-contamination between inert and reactive material in any of the samples. This indicates that the IMRM samples all undergo the same net reaction ($\text{Al/Ni} \rightarrow \text{AlNi}$) but at a range of different flame temperatures (roughly 1950 K to 1300 K). Using existing theoretical models for the relationship between flame temperature and propagation velocity, we analyze the experimental data to obtain the activation energy for the mixing process and find that this value varies significantly as the maximum reaction temperature changes. At high reaction temperatures we observe a very low activation energy (26 kJ/mol) which suggests diffusion of Ni in molten Al is the rate controlling mixing mechanism in agreement with the conclusions of other studies focused on un-mediated Al/Ni reactive multilayers. However, as the reaction temperature decreases the activation energy appears to shift to much larger values implying a change in the reaction mechanism. We postulate that this shift indicates that solid products are able to form earlier in the reaction at these temperatures, impeding atomic diffusion and intermixing.

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1. Introduction

Interfacial reactions can, in principle, occur anywhere two different miscible materials are in contact. They are especially relevant in nanoscale multilayers, since these structures exhibit unusually high ratios of interfacial area to volume. In the particular case that the interfacial reaction is highly exothermic, these are termed “reactive multilayers” [1] and a phenomenon known as a self-propagating reaction can be observed where the heat released by local mixing is sufficient to heat the adjacent material and produce a self-sustaining reaction front that moves through the sample with a characteristic velocity. Practically, reactive multilayers are of interest as heat sources for bonding [2,3], thermal batteries [4], and ignitors [5,6]. Scientifically, self-propagating reactions present a unique opportunity to investigate the processes that control atomic mixing and phase formation at the extremes of

concentration gradient and heating rate. Such scientific investigations feed back into practical applications by enabling improved predictions of performance over a range of operating conditions and by identifying ways that performance can be tuned for specific applications.

One of the most-studied reactive multilayer systems is Al/Ni. Al and Ni are readily available, relatively inexpensive, and have a fairly large enthalpy of reaction (1381 J/g [7]). Al/Ni multilayers have been fabricated using a range of physical vapor deposition techniques as well as by mechanical processing, and have been investigated using differential scanning calorimetry (DSC) [8,9], time-resolved x-ray diffraction [10,11], and time-resolved transmission electron microscopy [12], as well as in many theoretical studies [13–18]. The references here are only a small sampling of the available literature – the system is generally very well characterized.

Despite this extensive body of work, several questions remain. Perhaps the most interesting is, “What physical processes control the reaction rate in self-propagating Al/Ni multilayers?” A second, related question might be, “What role, if any, do intermetallic phases play in determining the reaction rate?” In order to answer

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these questions directly we would need to be able to resolve atomic-scale processes occurring at the reacting interface on extremely short timescales (< 100 ns). Existing instruments are capable of satisfying either the spatial or temporal resolution demands, but so far no instrument can meet both requirements simultaneously.

Instead, the best available experimental results regarding this question have been obtained by quenching self-propagating reactions. This relaxes the time-resolution constraint, allowing the use of conventional transmission electron microscopy to examine the interfaces of the partially reacted sample in high-resolution. Based on quenched microstructures it has generally been proposed that mixing into an Al-rich molten phase is the most likely rate-controlling process during self-propagating Al/Ni reactions [17–19]. However, this conclusion cannot be confirmed without experimental observation in the self-propagating state.

Another way to circumvent experimental limitations is to use molecular dynamics (MD) simulations. Simulations allow observation of the reacting interface in granular detail and at arbitrary temperatures. MD simulations of the Al/Ni reaction have been performed in a number of different thermodynamic ensembles, most notably the isobaric-adiabatic (NPH) and isobaric-isothermal (NPT). Studies using the NPH ensemble are most analogous to real experiments. In these [13–15], researchers observe a rapid temperature rise and the melting of Al relatively early in the reaction, similar to what is observed in time-resolved experiments [10–12]. Isothermal (NPT) simulations are less realistic but easier to analyze. Xu et al. conducted isothermal simulations of the Al/Ni interfacial reaction at multiple temperatures ranging from 2000 K (near the adiabatic flame temperature of 1:1 Al/Ni) down to 1300 K, and observed that different mechanisms controlled the mixing rate depending on the simulation temperature [16]. Detailed explorations at even lower temperatures (1000 K) have also been performed [17,18]. These results are suggestive but again call for experimental validation, especially given that the MD potentials used [20–22] are known to be incapable of reproducing the Al-rich Al/Ni intermetallic phases.

Here, we seek an experimental assessment of the rate-controlling processes in self-propagating Al/Ni reactive multilayers that does not require quenching or atomic-scale resolution of the reacting interface. In general, one of the easiest ways to experimentally characterize a reaction mechanism is to observe how the reaction rate varies as a function of temperature. This variation can be summarized in the activation energy, a macroscopic parameter that is often strongly correlated with the underlying microscopic processes at work. The primary challenge in applying this principle to Al/Ni reactive multilayers is that in a traditional multilayer the reaction temperature cannot be altered in a controlled fashion. In this work, we introduce a new class of reactive materials intended to address this need. Dubbed “inert-mediated reactive multilayers” (IMRMs), these materials incorporate an inert material to suppress the heating rate and maximum temperature without altering the chemistry of the reaction. The adiabatic flame temperature of an IMRM can be tuned continuously over a wide range (in this work we demonstrate temperature reductions as large as ~ 650 K) simply by varying the ratio of reactive material to inert material in the system. By using flame speed as a macroscopic indicator of the underlying interfacial reaction kinetics, this new class of materials can provide needed experimental support for conclusions derived from molecular dynamics and quenching studies.

In the following, we begin by laying out the basic design principles for IMRMs in general and for Al/Ni in particular. We then present the results obtained, assess them in light of the design requirements, and apply an analytical model for flame speed in reactive laminates to give scientific significance to the trends observed. We conclude by using these results to construct a picture of poten-

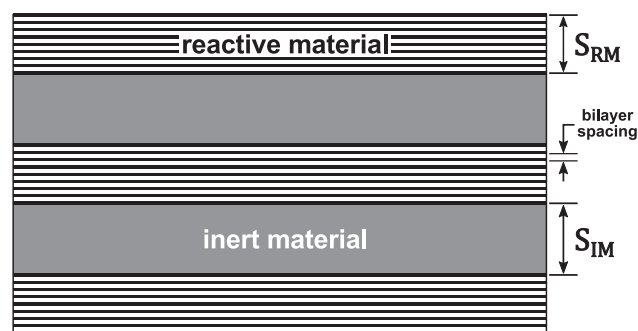


Fig. 1. A cross-sectional schematic of the generic design for an inert-mediated reactive multilayer (IMRM) showing alternating sections of reactive material (dark/light banded) and inert material (gray) with corresponding section thicknesses S_{RM} and S_{IM} . The multilayer bilayer spacing is also labeled.

tial reaction mechanisms over the temperature range 1250–1950 K, supported by previous experimental and theoretical results.

2. Design of inert-mediated reactive multilayers

A schematic diagram of the IMRM design is shown in Fig. 1. It consists of alternating sections of reactive material and inert material with characteristic section thicknesses S_{RM} and S_{IM} . Multilayer sections possess a second level of periodicity characterized by the “bilayer spacing”. A bilayer is the characteristic repeat unit of a two-component multilayer and consists of one layer of each material. The bilayer spacing is the sum of these layer thicknesses. In designing the IMRMs for this work, the principle design considerations were (1) what materials to use in each section, (2) how those materials should be arranged within the section, and (3) how thick the sections should be.

2.1. Section composition

The sections below outline the general considerations to be made when selecting materials for the reactive and inert sections, and how those decisions were made for the samples fabricated in this work.

2.1.1. Reactive material

The main requirements for the reactive component of an IMRM are a composition that is of scientific interest and a small bilayer spacing that will yield a high propagation velocity. The latter requirement is motivated by the expectation that quickly propagating sections will be able to accommodate the largest temperature suppression before quenching. The Al/Ni system is extensively well characterized and presents outstanding scientific questions of interest. Consulting a chart of flame speed versus bilayer spacing for 1:1 Al:Ni (e.g., [23]), a bilayer spacing of 23 nm yields a velocity very close to the maximum (~ 13 m/s). Consequently, the reactive component in this work was Al/Ni reactive multilayers with a 23 nm bilayer and a 1:1 molar ratio (approximately 3Al:2Ni by volume).

2.1.2. Inert material

The ideal inert material undergoes no chemical reactions upon heating that would add or subtract from the energy output of the sample, and does not chemically mix with the material in the reactive sections so as to alter the products formed. It also has high volumetric heat capacity and high thermal conductivity to maximize the temperature reduction per volume and promote thermal uniformity across the sample. These requirements suggest a metallic inert material, which would have the added benefit of being

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