

# Effects of interfacial Al oxide layers: Control of reaction behavior in micrometer-scale Al/Ni multilayers



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## ABSTRACT

Exothermic reaction behaviors of compression-bonded Al–Ni multilayers are investigated through controlling the interfacial layers of the multilayers. The native interfacial Al oxide layers between the Al/Ni bilayers are intentionally preserved to induce a thermal explosion (TE) during slow heating with up to a few tens of K/min. In contrast, the thermal annealing (TA) occurs in the highly shear-deformed multilayers with many broken Ni layers because the ductile Al fills the gaps in the broken Ni layers to form reactive Al/Ni interfaces. Increased onset temperatures of 865–893 K and activation energies, 470 kJ/mol, are measured in the multilayers exhibiting the TE. The onset temperature is increased by 60–80 K over that of the multilayers exhibiting the TA. The influences of the interfacial native Al oxide layers that are preserved in order to delay the Al/Ni inter-diffusion on the exothermic reaction behavior of the compression-bonded multilayers are discussed.

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## 1. Introduction

Mechanically bonded reactive Al/Ni multilayers, which have various types of bonding mechanisms, such as ball milling, cold rolling, and compression bonding, have been of significant interest to many researchers [1–10] because they have numerous advantages including low costs and simple processes [2,3,5–9]. They are suitable for fabricating micrometer-scale multilayers. Moreover, the Al/Ni multilayers can be used with high temperature bonding, local heat sources, and igniters used to initiate secondary reactions that result from the restricted local heat [10–12].

In order to fabricate a homogeneous intermetallic compound (IMC) and to apply a local heat source with high temperature bonding, sufficient energy is required to combust the Al/Ni multilayers [7,13]. Therefore, mechanically bonded Al/Ni multilayers must be combusted using a laser [11] or flame [14]. However, these ignition methods induce damage in the neighboring layers [15,16]. Alternatively, Rabinovich et al. designed rapid heating rates, i.e., rates higher than 600 K/min, to induce a thermal explosion (TE) in an Al/Ni powder compact system [10]. However, the rapid heating was only achievable using a specially designed substrate heater, and it made the process control difficult [17–19]. In contrast, Al/Ni multilayers that are slowly heated at a few tens of K/min reacted in the thermal annealing (TA) mode [20–22]. In order to enable TE in Al/Ni multilayers via slow heating, the multilayers

must combust. Other researchers reported that the ignition of the multilayers in combustion synthesis is a function of the interfacial conditions, heating rates, thermal coupling, and layer thicknesses [23,24]. This indicates that the interfacial and layer thickness controls are critical conditions in designing a combustion system via slow heating.

A recent study by Kuk et al. demonstrated that the compression-bonded Al–Ni multilayers have highly uniform and continuous bilayers [7–9]. Moreover, the interfacial conditions between the Al and Ni layers are controllable using compression conditions [25], while other mechanically bonded Al/Ni multilayers cannot be altered due to the shear stress [1–6]. In a previous study, the TA occurred in the broken areas of the Ni in Al/Ni compression-bonded multilayers, and the interfacial oxide layer was impaired in the damaged positions of the Ni layers [7]. In contrast, the interfacial oxide layers between the Ni and Al layers can be preserved and the damaged positions of the Ni layers removed. Likewise, the interfacial oxide layers between the Ni and Al layers were controlled using compression-bonded Al/Ni multilayers. The onset temperatures of the TE were measured and the activation energies were calculated via the Kissinger and Horvitz analysis in order to design the effects of the interfacial oxide layers [26,27].

This research provides a new mechanism for inducing TE via slow heating. In addition, we used the concept of interrupting the reaction of the combustion elements to provide a method to control the combustion synthesis. This process can be used for homogeneous IMC generation, environmentally friendly Pb free bonding, high temperature bonding without thermal damage for electronic assemblies, emergency bonding for aerospace, local heat sources for munitions, propellant

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ignition, and as igniters for initiating secondary reactions at low temperature slow heating [28].

## 2. Experiments

Ni was sputtered from Ni target (purity 99.99 wt.%) on 5.8  $\mu\text{m}$ -thick Al foils (purity 99.5 wt.%) using a DC magnetron sputter. Before the Ni was sputtered via a sputter system from A-Tech System Ltd., Korea on the Al foils, the Al foils were cleaned using RF argon plasma. The deposition began at a vacuum of less than  $2 \times 10^{-6}$  Torr ( $2.7 \times 10^{-4}$  Pa) and it was maintained in an Ar atmosphere of  $5 \times 10^{-3}$  Torr (0.7 Pa) during the sputtering. The Ni deposition thickness was set to 2.8  $\mu\text{m}$  for a target  $\text{Al}_3\text{Ni}_2$  phase after the exothermic reactions. The single layer of Ni-sputtered Al foil was cleaned again using RF argon plasma for the surface treatment. After the treatment, the Ni-sputtered Al foil was punched into circular disks with diameters of 10 mm and these disks were stacked to form a lamellar structure.

Each disk structure was stacked with 60 disks with a total layer thickness of  $460 \pm 20$   $\mu\text{m}$ . The stacked layers were pressurized at 875 MPa using an Instron 4400R (Instron Corp., USA) in order to mechanically bond the Al/Ni multilayers. The Al/Ni multilayers were placed in the compression mold to induce pure compression bonding during the compression. The Al/Ni multilayers were compression-bonded with a boron nitride (BN) lubricant at the surfaces of the upper and lower punches to reduce the frictional stress. The Al layer in the as-compressed multilayer was reduced to a thickness of 5.2  $\mu\text{m}$  and the Ni layers were reduced to thicknesses of 2.5  $\mu\text{m}$  in the multilayers pressed without lubrication.

The fabricated multilayers were heated from room temperature to 1000 K to induce a sufficient reaction with heating rates of 5, 10, 20, and 40 K/min in flowing Ar using a differential scanning calorimetry (DSC) 404C in order to analyze the activation energy of the compression-bonded multilayers. The as-compressed specimens were investigated using scanning electron microscopy (SEM), Philips XL30SFG, transmission electron microscopy (TEM), Tecnai TF30 ST, and energy dispersive X-ray spectroscopy (EDS) of the TEM, at the interfaces between the Al foils and the deposited Ni to compare the multilayers pressed with the BN lubricant and those pressed without lubrication. Microstructures of the DSC-tested specimens were measured via SEM and the specimens were pulverized to measure an X-ray diffractometer (XRD), Rigaku Ultima 4 in order to identify the resulting phases after TE during the DSC tests. The radiation, wavelength and power of the XRD were Cu  $K\alpha$ , 0.15418 nm, and 40 kV 300 mA, respectively. The outer 1 mm edge of each specimen was removed to eliminate the edge effects during the slow heating. The average overall length of the interfaces with the Al oxide layers and those of the interfaces without the oxide layers between the Al and Ni layers were measured from tens of cross-sectional SEM images of the multilayers pressed with the BN lubricant.

## 3. Results and discussion

### 3.1. Before reaction

The microstructural features of the multilayers were investigated using the cross-sectional SEM images (Fig. 1). Fig. 1(a) presents the cross-sectional SEM image after the compression bonding using a compression mold without lubrication. Unbroken Ni is feasible using thin film multilayers and molds. The mold minimized the formation of broken Ni layers because the frictions between the specimen and the punches reduced lateral shear deformation of the multilayers. Larger horizontal planes between the top and bottom of the mold reduced the shear stress during compression. Moreover, the shear strain was restricted due to the frictional stress between the molds and specimens. Ji and Wang determined that there are three zones in the specimens during high compression bonding with interfacial friction [25]. First, at

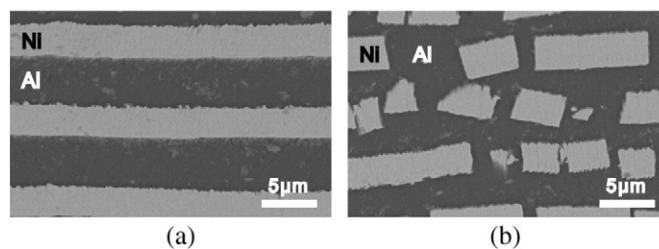


Fig. 1. Cross-sectional SEM images with the mechanically bonded interface between Al and Ni for the multilayers pressed (a) without lubrication and (b) with the BN lubricant.

the “strain forbidden zone”, no strain occurs at the top and bottom of the multilayers due to strong friction between the punches and the surface of the samples. In contrast, intensive strain occurs at the “intensive strain zone” which is at the outside of the “strain forbidden zone”. The “intrusion zone” is a stress forbidden area at the side wall of the specimens, and the shear deformation from the “intensive strain zone” is not propagated. In ultra-thin multilayers, there is neither the “intensive strain zone” nor the “intrusion zone”, and only the “strain forbidden zone” remains due to the friction during compressive stress.

In contrast, the BN lubricants reduced the frictional stress, and then the shear stress was accelerated vertically to the compressive stress, which resulted in increasing the “intensive strain zone” [25]. While the Al layers were ductile when stretched, the Ni layers were not sufficiently ductile to stretch but rather they were broken by the shear stress. Thus, the ductile Al filled the broken Ni layers to form reactive Al/Ni interfaces in the multilayers pressed with the BN lubricant (Fig. 1(b)). Fig. 2(a) and (b) presents the cross-sectional TEM images of the interfaces of Ni-sputtered Al foils in the multilayers pressed without lubrication and those pressed with the BN lubricant, respectively. Native amorphous Al oxide layers with thicknesses of approximately 5 nm were observed on the interfaces between the Al and Ni layers. In contrast, reactive Al/Ni interfaces were formed without in-between oxide layers at the interfaces between the broken Ni and these gaps were filled by Al in the multilayers pressed with the BN lubricant (Fig. 2(c)). No oxide layer was observed via EDS line scanning of the interfaces between Al and Ni. The native Al oxide layers were impaired due to the Al layers leaning at the interface between the Al and Ni layers as a result of the shear stress during the compression with the BN lubricant. It was easy for the TA process to occur via slow heating at the interfaces between the Al and Ni layers where the Al oxide layer was damaged [7]. In contrast, the multilayers without the lubrication preserved the Al oxide multilayers.

### 3.2. Reaction behavior

The DSC experiments were conducted on the multilayers pressed with the BN lubricant and without lubrication in order to determine the interface effects for the reaction behavior via slow heating. Fig. 3 presents the DSC traces on the multilayers during the slow heating at a rate of 5–40 K/min. The TA was generated from the multilayers pressed with the BN lubricant as a result of the inter-diffusion of the Al and Ni (Fig. 3(a)). Multi-peaks were found at the occurring TA multilayers, which mean the heat was released before the reaction of the main peak, which is a tendency of  $\text{Al}_3\text{Ni}_2$  [7]. The onset temperature (see Fig. 3(b) and (c)), the tendency of onset peaks of  $\text{Al}_3\text{Ni}$ , and peak temperature were similar to the previous research during the TA [7]. Despite the sample having the atomic ratio of  $\text{Al}_3\text{Ni}_2$ , the cross sectional SEM image of the multilayers pressed with BN lubricant demonstrated that  $\text{Al}_3\text{Ni}$  remained due to the partially Al rich areas after slow heating at a rate of 20 K/min (see Fig. 3(d)). In contrast, TE was generated in the multilayers pressed without lubrication due to the preservation of the native Al oxide layers between the Al and Ni layers (Fig. 3(b)). The DSC profiles of the TE, except the specimen at 20 K/min, appeared

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