Combined Properties of Al/Ni Reactive Multilayer Films During High-Rate Heating Using a Pulse of Current

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Abstract. We use pulses of current to investigate the combined properties of Al/Ni multilayer films under high-rate heating by measuring current-voltage waveforms and the flyer velocity. Explosion occurs earlier in the Al/Ni multilayer foil than in the Al and Ni films, suggesting that the addition of Al/Ni affects the electrical exploding process, which is not controlled solely by resistance. The experiments indicate that the differences between the physical properties of the Al and Ni films is a factor to causing higher plasma electron temperature and absorb more electrical energy than the single Al or Ni film. The flyer velocity of the Al/Ni multilayer film is much lower than that of the Al film due to the high adiabatic index of Ni.

1. Introduction
Composite materials contain two or more materials with different physical or chemical properties. When combined, materials exhibit different characteristics than the individual components, such as their strength, electron transport, and phase transitions [1]. Multilayer films are composite materials consisting of alternating layers of two or more different materials, which can be easily deposited and integrated into electronic devices to exploit their microelectromechanical properties and have been applied in many fields [2]. Reactive multilayer films comprise two components and releases the chemical energy stored by the two individual reactant components. These heterogeneous solids can be stimulated by an external source to promptly release the stored chemical energy in a sudden emission of light and heat [3-5]. Al/Ni is a reactive multilayer film that has a wide range of potential applications in the fields of ignition, welding, propulsion, and safe and arming devices. The reaction properties of the Al/Ni multilayer films have been investigated based on thermal explosion and a self-sustained propagating reaction using differential scanning calorimetry [6], time-resolved transmission electron microscopy [7], time-resolved x-ray diffraction [8], and in many theoretical studies [9]. All of the above-mentioned investigations focused on the onset temperature, reaction heat, reaction velocity, threshold for ignition, etc., under low-rate heating. C. J. Morris et al [10-12] reported experiments on heated microscale Ni/Al multilayer films at high heating rates of $10^{11}$-$10^{12}$ K/s by using a capacitor discharging unit (CDU). They found that uniformly heating Ni/Al multilayer films can lead to a rapid, thermally explosive type of reaction characterized by a high flyer velocity. However, the combined properties of Al/Ni multilayer films under high-rate heating by using a pulse of current are complex and involve resistance, phase transitions, the heat of the reaction, etc. Thus, the combined properties of Al/Ni multilayer films are worth investigating by designing samples with different reaction heats and resistances.

In this work, three kinds of samples were designed related to differences in heat reaction, resistance,
and reaction rate. The combined properties of the Al/Ni multilayer films were systematically investigated by varying the structure of the Al and Ni multilayers. The explosion performance of the Al/Ni multilayer films was exhibited in terms of the discharging current-voltage, exploding time, absorbed electrical energy, phase transition time, and flyer velocity.

2. Experimental Section

2.1. Fabrication of Multilayer Films

It has been reported that the energy release rate, chemical energy stored in the multilayer films, and room-temperature resistivity was correlated with bilayer thickness. The stored energy in a multilayer decreases as decreasing bilayer thickness due to pre-reaction intermixing layer at the Al-Ni interfaces during layer deposition. The intermixing layer is also influenced on room-temperature resistivity. Meanwhile, reducing layer thickness can improve energy release rates because of extremely short distance among reactants, and relatively large intermixed region. So, we fabricated five types of films, including two single-metal films and three multilayer films, as shown in Figure 1. The total thickness of the films was 4 μm. Sample types A and B were traditional metal foils without additional energetic materials. The type C sample theoretically exhibits an energy release rate several orders of magnitude higher than the type D, and E samples. Meanwhile, the chemical energy of type C is much lower than type D, and E due to the pre-reaction intermixed thickness. The type D and E samples were the same room-temperature resistivity and chemical energy.

Each bilayer consisted of an aluminium layer and a nickel layer in a 3:2 thickness ratio to maintain the overall 1:1 atomic composition. The samples were deposited by magnetron sputtering from Ni, and Al targets powered at 90 W. The base pressure prior to the sputtering was less than 5×10⁻⁴ Pa, and the sputter gas was argon at a flow velocity of 30 sccm. Multilayer films were deposited with a bilayer spacing of 500 nm, and the bilayer was repeatedly deposited to yield a total thickness of 4 μm. Each film began and ended with a nickel layer due to high oxidation property of the Al film in air for the multilayer samples. The deposition parameters of different metals are listed in Table 1.

![Figure 1. Sample types designed for this paper](image)

**Table 1. Deposition parameters of metals**

| Metal    | Deposition pressure Pa | Deposition power W | Deposition rate nm/s | Flow rate of Ar sccm |
|----------|------------------------|--------------------|----------------------|----------------------|
| Nickel   | 0.4                    | 60                 | 8                    | 30                   |
| Aluminium| 0.6                    | 90                 | 20                   | 30                   |
2.2. Fabrication of Multilayer Foils
High-rate heating was performed by passing an electrical current through a small region with a high-current pulse duration of ~100 ns. Therefore, the patterned multilayer foil was fabricated by a traditional wet process. The ceramic substrate was cleaned with acetone, alcohol and deionized water in an ultrasonic bath for 5 min. Then, the cleaned substrate was placed in an oven for 30 min at 100 °C. Next, different samples were deposited by magnetron sputtering. A positive photoresist (AZ5214E) was spin-coated onto the multilayer films at 5000 rpm for 60 s and pre-baked in an oven for 90 s at 100 °C. Afterward, the layer was partially exposed to ultraviolet (UV) radiation with the intensity of 16 mJ/cm² through a mask. The UV-exposed region of the multilayer film was dissolved by a developer (NaOH solution). Subsequently, the entire film was heated to 120 °C to prevent the patterned region from etching. Finally, the regions of the multilayer films exposed to UV were removed by the Al etchant at 30 °C. Multilayer foils were designed as squares with a length and width of 0.6 mm.

2.3. Fabrication of the Electrical Transduction Unit
Figure 2 shows the schematic image of the electrical transduction unit. The multilayer foil, flyer and barrel were manufactured separately. The electrical transduction unit was integrated by screws or welding. When a specific value of high current is applied to the transmission line conductors, the foil bursts and creates plasma under the Kapton flyer. The energy stored in the plasma is efficiently directed towards the flyer, which is sheared off by the barrel and accelerates through the barrel at a high velocity. The dimensions of the electrical transduction unit are summarized in Table 2.

![Figure 2. Schematic image of electrical transduction unit](image)

Table 2. Dimensions of electrical transduction unit

| Foil width (mm) | Foil length (mm) | Foil thickness (μm) | Kapton thickness (μm) |
|----------------|------------------|---------------------|-----------------------|
| 0.6            | 0.6              | 4                   | 20                    |

2.4. Characterization Methods
The microstructure of the samples was characterized using transmission electron microscopy (TEM). The type C sample was processed by a focused ion beam (FIB) prior to the TEM observation. The multilayer composition was determined using an energy dispersive spectrometer (EDS). The current pulse was derived from the firing CDUs of the Al/Ni multilayer foils with a capacitance of 0.22 μF at voltages from 3.5 kV to 4.1 kV. Both the current flowing through the multilayer foil and the potential difference or voltage, across it due to its combined properties must be determined. The current and voltage were measured using Rogowski coil and a high voltage probe, respectively. The plasma electron temperature generated by samples was measured by the streak emission spectroscopy with 100 ns gate width. The testing baseline delay was controlled by digital delay generator (DG535). Photonic Doppler velocimetry (PDV) was used to measure the flyer velocity from the electrical
transduction units. The frequency difference between the original laser and return laser was converted to an electrical signal by a fibre optic receiver, which was then recorded by an oscilloscope with a bandwidth of 32 GHz.

3. Results and Discussion

3.1. Structure and Layered Morphology
The thin layer of Al was first deposited on the substrate to prevent oxidation after the deposition process. For the Al/Ni multilayers, the stoichiometric ratio was 1 in this work. Each modulation period was 4000 nm (type D: 2.4 μm/1.6 μm, type E: 1.6 μm/2.4 μm 1 period), 500 nm (type C: 300 nm/200 nm, 8 periods)). The final thickness of all samples was approximately 4 μm. Figure 3 shows the TEM images of the sample of types C. Al and Ni layers were alternately deposited using a magnetron sputtering process. The TEM images of the sample showed that the Al and Ni layers had distinct boundary lines between the Al/Ni multilayers. The thicknesses of the Al and Ni layers were 300 nm and 200 nm, respectively. The EDS patterns of type C (Figure 3c and 3d) confirmed the presence of each deposited layer of Al and Ni. The modulation period of the reactive multilayers is a crucial parameter because it significantly influences the chemical reactions of Al/Ni, namely, the reaction rate, heat flow, and reaction mechanism [13]. Therefore, the properties of Al/Ni multilayers with different modulation periods are different in high-heat rating by a pulse of current, which is investigated here.

![Figure 3. FIB image of the (a) and TEM image of the (b) and (c) EDS pattern of Al and (d) EDS pattern of Ni](image)

3.2. Electrical Performance of Multilayers
Figure 4 shows a representative current and voltage history for the discharge of the storage capacitor through the type C sample (Al/Ni), wherein the capacitor was initially charged to 3.5 kV. The current initially rises, rapidly heating the multilayer foil. This heating increases the resistance, as exhibited by the increase in voltage across the foil. When the temperature is higher than the vaporization temperature of the multilayer films, the resistance increase is sufficiently high to cause “exploding”. “Exploding” is assumed to be the time at which the maximum voltage is measured. The vaporized multilayer films are ionized by the voltage, thereby creating plasma and causing an inflection in the current. The plasma formed by the multilayer foil re-establishes a path of low resistance, causing the voltage to drop towards zero. The spare energy from the capacitor continues to discharge towards the created plasma, thus increasing the current.
Figure 4. Current and voltage history for Al/Ni multilayer foil of type C, with the storage capacitor initially charged to 3.5 kV.

Figure 5 shows the exploding time and voltage of different samples at charging voltages ranging from 3.5 kV to 4.1 kV. As expected, the exploding voltage increased with the increasing charging voltage, while the exploding time decreased. The type C sample (Al/Ni multilayer foil) has a higher exploding voltage and shorter exploding time for the same charging voltage levels than samples A and B. Figure 6 shows the time-integrated electrical energy traces from the current-voltage waveforms at the charging voltage of 3.5 kV. Type C (Al/Ni multilayer foil) shows a higher electrical absorbed energy, indicating that adding further Al/Ni multilayers influences the electrical exploding process, resulting in different electrical parameters, such as exploding time, exploding voltage, exploding current, and the absorbed electrical energy.

Figure 5. Exploding parameters vs. capacitor charging voltage levels for different samples in Figure 1: (a): exploding time, (b): exploding voltage
Figure 6. Absorbed electrical energy vs. time for different samples in figure 1 with the storage capacitor initially charged to 3.5 kV

Above the results, the exploding process was related to the structure of the foil. Many references have been proved that the improved voltage-induced plasma was related to the chemical energy stored in the Al/Ni multilayers. In order to deeply understand the factors to influence the exploding process, the type D and E was designed with the same room-temperature resistivity and chemical energy. The plasma electron temperature was measured by streak emission spectroscopy of Type D and E. Figure 7 shows the spectrogram of Al atomic lines using sample D and E at 3.5 kV with a distance of 10 mm above the samples. The intensity of emission spectrum for type D is much higher than type E, indicating much easier for ionization of Al/Ni structures. The Al I lines around the wavelength 396 nm were the strongest in the spectrogram. The spectral lines at 308.216 nm, 309.271 nm, 394.407 nm and 396.152 nm were used to estimate the electron temperature of the samples.

Figure 7. Schematic illustration of the emission spectra of the different samples under 3.5 kV: (a): type D, (b): type E

Figure 8 shows the electron temperature profiles for different delay times for type D and E with the charging voltage 3.5 kV. As the delay time increased from 1000 ns to 3000 ns, the electron temperature of the type D decreased from 7970 K to 5465 K. The electron temperature of the type E decreased from 7094 K to 5290 K. The results indicated that the electron temperature of Al/Ni samples is higher than Ni/Al layer with the charging voltage 3.5 kV. But the small electron temperature difference of the voltage-induced plasma between type D and E is visible. The difference between Al and Ni melting temperature gives rise to the phenomenon. When current passes through
the foil, the temperature caused by Joule heating quickly reaches the melting temperature of Al. However, the melting temperature of Ni is approximately 1453 °C, which is much higher than that of Al. Therefore, the upper Ni layer would prevent the volume expansion of the Al layer as a result of the higher electron plasma temperatures. But the influences of the melting temperature between Al and Ni were decreased as increased the charging energy.

Figure 8. Electron temperature plots of the voltage-induced plasma vs. the different times

To verify these results, the resistance of type C combined with type A and B was calculated from the current-voltage waveforms shown in Figure 9. The resistance curves are consistent with the discharging voltage. The time of the peak voltage corresponds to an inflection in the current, where the resistance of the foil reaches a maximum. The exploding process of the foil usually contains four stages, namely, solid heating, melting, and gasification. Figure 10 shows the time between the melting and gasification process of different types. The melting process of type C is found to require more time than that of types A and B, which can be attributed to the different melting points of Al and Ni. For the single-metal films, the exploding process is controlled by their material properties, resulting absorbing less electrical energy. For the vaporization process of the foil induced by a sharp rise in voltage or resistance, the difference between various samples is not visible.

Figure 9. Resistance vs. time for different samples in figure 1 with the storage capacitor initially charged to 3.5 kV
Fig. 10. Time vs. capacitor charging voltage levels for different samples in figure 1: (a): gasification phase, (b): melting phase

The results of our investigations indicated that several factors can contribute to the improved electrical absorption energy of Al/Ni multilayers relative to that of Al and Ni films alone. The difference in physical properties between Al and Ni films was a factor.

4. Flyer Velocity

Although the combined properties of Al/Ni multilayer films improve the absorption of electrical energy relative to that of Al and Ni films alone due to differences in their physical properties and heat energy, the acceleration ability of multilayer foil was investigated by measuring the flyer velocity. The expanding pressure originating in the foil due to the pulse of current accelerates the flyer layer away from the foil surface, causing a portion of the flyer layer to tear away and continue accelerating. Figure 11 shows the resulting velocity traces at charging voltages ranging from 3.5 kV to 4.1 kV for different samples. The acceleration and final velocity increased as the charging voltage increased for different samples. The type C sample, corresponding to the Al/Ni multilayer foils, appeared to have a flyer velocity between those of the type A and B samples. The experimental results shown in Fig. 10 suggest the underlying mechanism. The Al/Ni multilayer foil showed a longer melting process due to differences between the melting temperature and resistivity of Al and Ni in the corresponding Al and Ni foils. Exploding has long been assumed to be the process wherein the exploding metal overcomes confinement and drives the material around it. The flyer should be accelerated at the time of maximum voltage. Therefore, the melting and exploding process is not useful to the flyer velocity of the foil. Recently, however, William Neal et al. [14] simultaneously measured the current, voltage, and flyer velocity and performed high-speed imaging in experiments. Their study showed that the foil significantly expands before the time of peak voltage, which was traditionally assumed to be the point of exploding. Evidently, the material around the foil already has a velocity before the maximum voltage has been measured corresponding to the exploding process. However, the response process of the Al/Ni multilayer foil with the current pulse shows a longer melting time, which is not attributed to the flyer velocity. Furthermore, the expansion process as the metal material vaporizes approximates the isentropic process [15]. The adiabatic index of the expanded gas is different for the different samples. The adiabatic index of the Al film is much lower than that of the Ni film, which means that the pressure attenuation caused by the expansion of Ni metal is much faster than that for the Al film under the same distance. Therefore, the flyer velocity of the Al/Ni multilayer foil is lower than that of the Al foil. Compared to the Cu layers in exploding foil initiators system in our previous work [16], the samples integrated with Al/Ni multilayers exhibits superior performance in plasma generation characterized by flyer velocity due to higher absorbed electrical energy. Hence, there is much prospect of the application of energetic Al/Ni superlattice on micro or nano-plasma initiators to implement special function.
5. Conclusions
In this study, different foil samples were designed to investigate the combined properties and mechanism by adding Al/Ni multilayers to a foil and applying high-rate heating using a high-current pulse. The multilayer foils were fabricated using a wet etching process and magnetron sputtering. SEM images showed the distinct layer structure of the Al/Ni multilayer films. The combined properties of the Al/Ni multilayer films were investigated by directly measuring the current and voltage histories, measuring plasma electron temperature, and evaluating the flyer velocity as measured by PDV. It is found that the differences between the physical properties of the Al and Ni films is a factor to causing higher plasma electron temperature and absorb more electrical energy than the single Al or Ni film. However, the flyer velocity of Al/Ni multilayer foil is much lower than that of the Al foil because of the high adiabatic index of Ni, which means that the attenuation of the pressure caused by the Ni metal material expansion is much faster than that for the Al film.

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7. References
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