*Central European Journal of Energetic Materials*
ISSN 1733-7178; e-ISSN 2353-1843
Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

*Cent. Eur. J. Energ. Mater.* 2017, *14*(3): 547-558; DOI: 10.22211/cejem/75605

# Characterization and Application of Al/Ni Reactive Multilayers in Exploding Foils

**Tao Wang, Qingxuan Zeng,* Mingyu Li****

*State Key Laboratory of Explosion Science and Technology,*
*Beijing Institute of Technology,*
*5 South Zhongguancun Street, Haidian District, Beijing, 100081, China*
*E-mail: *zengqingxuan@bit.edu.cn, **mingyuli@163.com*

**Abstract:** A self-propagating reaction achieved by initiating an Al/Ni reactive multilayer foil can generate significant heat. The interdiffusion rate of the reactants plays an important role in the foils properties and is mainly affected by premixing and the bilayer thickness. The present research aims to characterize Al/Ni multilayer foils and to investigate their influence on an exploding foil initiator. Samples with different bilayer thicknesses were fabricated by magnetron sputtering. The heat released and the flame velocity were characterized. Foils with a stored energy of about 1100 J/g were prepared and the heat released revealed the existence of a 4 nm premixing layer. The analytical model proposed by Mann was employed to match the measured flame velocities; the fitted model showed good agreement with the experimental results. To make a comparison, Cu and Al/Ni exploding foils with the same bridge size were fabricated and tested in the identical discharge circuit. The results showed that the energy deposition ratio of an Al/Ni foil was 67-69%, while the value for Cu was only 39-45%, which indicated that Al/Ni multilayers could effectively increase the energy utilization of an initiator. Larger average flyer velocities were also observed with the Al/Ni initiators.

**Keywords:** energetic material, Al/Ni reactive multilayer foils, bilayer thickness, exploding foil initiator

## 1 Introduction

Over the last few decades, there has been considerable interest in reactive multilayer foils (RMFs) because of several potential applications, including room-temperature bonding [1, 2], initiators [3, 4] and thermal batteries [5]. The alternating nanoscale layers increase the interfacial contact area and decrease the diffusion length between the reactants. Among these RMFs, Al/Ni multilayers have received extensive attention due to their impressive underlying energy and a desirable reaction speed of about 10 m/s [6, 7].

Thin-film reactions in Al/Ni RMFs have been extensively studied. Ma *et al*. [8] first prepared Al/Ni RMFs and observed the exothermic reaction triggered by an electrical pulse. Analytical models coupling atomic diffusion and thermal transport equations provided an effective means for evaluating the flame velocity of the Al/Ni foils, based on bilayer thickness and premixing spacing [9, 10]. In-situ [11, 12] and ex-situ [13, 14] characterization offered details about the reaction mechanism. Research on Al/Ni RMFs as local heat sources for soldering and ignition has received increasing attention [15, 16]. The exploding foil initiator (EFI) shows excellent safety and reliability, especially in harsh conditions including high electromagnetic pulses or intense stray currents [17]. The bridge foil is vaporized and becomes a plasma when a short burst of a large pulse current is loaded. The flyer accelerated by the plasma can initiate a secondary explosive by its high kinetic energy. Cu is the most widely used bridge foil in EFIs due to its superior electrical burst behaviour, nevertheless the high operating voltage of former Cu EFIs requires a large size and a high price for the initiator. As a heat-producing and highly reactive material, Al/Ni RMFs may be crucial in reducing the operating voltage of EFIs. However, little attention has been paid to the application of Al/Ni exploding foil initiators.

The objective of the present research was to characterize Al/Ni RMFs and investigate their influence on EFIs. We report here on Al/Ni multilayer samples with different bilayer thicknesses. The thermal effects and flame velocities were tested separately using Differential Scanning Calorimetry(DSC) and high-speed videos. The analytical model proposed by Mann [9] was employed to match the experimental data. In the EFI experiments, an oscilloscope was used to obtain the voltage and current waveforms of the discharge circuit. Polyvinylidene fluoride (PVDF) films were used to measure the average velocities of the flyers. Comparisons were made between Cu and Al/Ni RMF initiators.

## 2 Materials and Methods

### 2.1 Fabrication

Al/Ni RMFs of different bilayer thicknesses were fabricated by magnetron sputtering from Al and Ni targets (both of purity ≥ 99.5%, Chengdu Ultra-Pure Applied Material, China). The alternating nanoscale layers were achieved by

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

programmed shuttering of the targets. Samples with the same total thickness of approximately 6.2 μm but different bilayer thicknesses, including 40 nm, 55 nm, 70 nm, 90 nm, 120 nm and 180 nm were prepared by a magnetron sputter (Discovery 635, Denton Vacuum, USA). The process chamber was evacuated to about $5.0 \times 10^{-7}$ Torr prior to fabrication, while the pressure of Ar (purity ≥99.999%) was 2.2 mTorr during deposition. All of the RMFs had an overall composition of 50Al/50Ni, and were covered with a Ni layer in both sides. Due to its magnetic properties, a Ni film is difficult to deposit with a conventional magnetron system. To eliminate this problem, the sputter was provided with a much stronger magnetic field in the preparation of the Ni layers, however this method generated more heat on the Ni target and the substrates. In order to suppress the premixing, it is important to maintain a low substrate temperature during the fabrication procedure. As no active cooling system of the substrate is present in our sputter, an interval time after each Ni layer was set from 0.5 h to 2 h to minimize excess heat accumulation and to keep the temperature in the process chamber below 26 °C. During the interval time, the high voltage between target and substrate was cut off, and the process chamber was evacuated to an ultrahigh vacuum again.

An Al/Ni RMF with a 450 nm bilayer thickness was chosen to fabricate the EFIs and the preparation technology was similar to the above. Specifically, as a result of its large film thickness, fabrication of each Ni layer was divided into two stages, half-thickness Ni was deposited in each stage and a 2.5 h time interval was applied after each half-thickness Ni layer. Cu and Al/Ni RMF exploding foils were fabricated by a shadow mask (machining tolerance ≤0.01 mm, Ningbo Dongsheng Integrated Circuit Component, China), and the sizes of the bridge foils were 350 μm (length) × 350 μm (width) × 3.79 μm (thickness). The EFIs were tested with the same discharge circuit with a lumped inductance of 84 nH. A 25 μm thick polyimide layer was employed as the flyer and the barrel, with a diameter of 440 μm, was made of SU-8 photoresist.

## 2.2 Characterization of properties

The reactive performance of Al/Ni was characterized by its heat release and flame velocity. The deposition energy and average flyer velocity of EFIs were compared between Cu and Al/Ni exploding foils.

The micromorphological characterization was performed by SEM (S4800, Hitachi, Japan). Both surface and cross-section images were observed. 1-2 mg of free-standing foils were analyzed by DSC (DSC404F3, Netzsch, Germany). In each DSC scan, the sample was heated from 30 °C to 600 °C, at 20 °C/min in a flowing high-purity Ar atmosphere.

To acquire the flame velocity, Al/Ni RMFs deposited on 25.4 × 76.2 mm

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

glass slides were placed in a rough vacuum chamber at less than $10^3$ Pa, and ignited at one end with an electric spark generated by a 90 V DC power source. The propagation process of the flame front was recorded by a high-speed camera (FastCAM-APX RS, Photron, Japan) at 1500 fps and 2 μs exposure time per frame. The thermal loss between the RMF and substrate can be ignored due to the low thermal conductivity of the slide in the experimental process. The analytical model proposed by Mann was employed to evaluate the reaction velocities as a function of bilayer thickness and premixing thickness.

The bridge foil was connected to a 0.22 μF capacitor through a metal foil gap switch [18], and an oscilloscope (DPO3034, Tektronix, USA) was used to monitor the voltage and current waveforms of the circuit. The deposition energy could be gained by time integration of U(t)·I(t) in the first half discharging cycle of the current waveform and the energy deposition ratio was equal to the deposition energy divided by the initial stored energy of the capacitor. The PVDF film was placed at the end of the barrel to measure the average velocity of the flyer. Figure 1 shows the details of EFI test device.

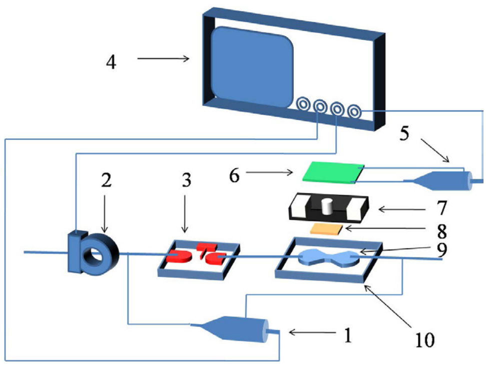


**Figure 1.** Schematic of the EFI test device: 1. high voltage probe, 2. Rogowski coil, 3. metal foil gap switch, 4. oscilloscope, 5. voltage probe, 6. PVDF film, 7. barrel, 8. flyer, 9. bridge foil, 10. substrate

## 3 Results and Discussion

### 3.1 Micromorphology and heat release

The SEM images of the Al/Ni reactive multilayers with 180 bilayer thickness are shown in Figure 2. The images demonstrate that the film is compact and uniform, while the layer structures are homogeneous, and the interfaces between the layers are sharp.

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

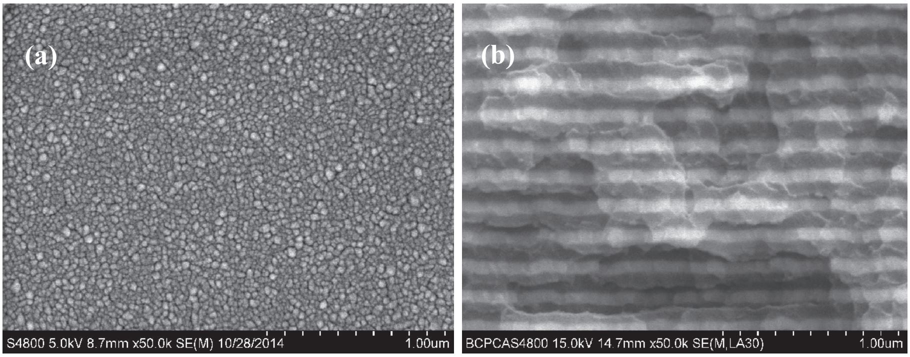


**Figure 2.** SEM micrographs for Al/Ni multilayers with a 180 nm bilayer thickness, (a) surface; (b) cross section

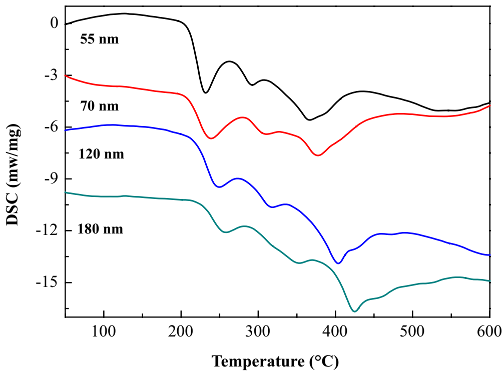


**Figure 3.** DSC curves for samples with 55 nm, 70 nm, 120 nm and 180 nm bilayer thicknesses

**Table 1.** Analysis of DSC curves with different bilayer thicknesses

| Bilayer thicknesses [nm] | 55 | 70 | 120 | 180 |
|---|---|---|---|---|
| Heat released [J/g] | 1085 | 1137 | 1193 | 1198 |
| The first peak [°C] | 231.7 | 237.7 | 246.4 | 256.9 |
| The second peak [°C] | 292.8 | 310.0 | 315.6 | 352.1 |
| The third peak [°C] | 365.0 | 377.1 | 403.4 | 424.4 |
| Predicted premixing thickness [nm] | 3.97 | 3.62 | 3.55 | 4.97 |

The DSC traces for the samples with typical bilayer thicknesses are displayed in Figure 3. Heat release, peak temperature and predicted premixing thickness are listed in Table 1. The total thermal effects calculated from the DSC data are close to the values in references [19, 20]. The exotherm of the samples

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

was reduced continuously with decreasing bilayer thickness, which should be ascribed to a larger volume fraction of premixing. Three exothermic peaks were observed; the third peak became dominant with increases in the bilayer spacing. The peak temperature rose slightly with increasing bilayer spacing, from 231.7 °C to 256.9 °C for the first peak for example, which suggested that longer diffusion distances were indispensable for the Al and Ni atoms to interdiffuse and fully react. The heat released can be used to figure out the premixing thickness, according to the following equation [19]:

$$\Delta H = \Delta H_0 \left( 1 - \frac{2\omega}{\sigma} \right) \tag{1}$$

where $\Delta H$ is the measured heat of reaction, $\Delta H_0$ is the enthalpy of Al/Ni formation, $\omega$ is the premixing thickness at each Al/Ni interface and σ is the bilayer thickness. For the Al/Ni RMFs prepared in this research, the predicted premixing thickness $\omega$ was about 4 nm.

## 3.2 Flame velocities

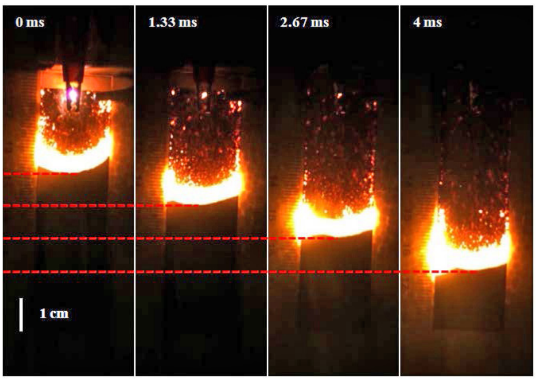


**Figure 4.** Images of the flame front propagating along the Al/Ni RMF

The combustion process was captured by high speed photography. Representative images of the flame front propagating along the foil are displayed in Figure 4. The combustion front was quite flat and the propagation length could be recognized easily in each image. The flame velocities were obtained by linear least squares fitting of the experiment data. The correlation coefficients were generally higher than 0.99, which indicated a steady propagation speed of the reaction in the Al/Ni multilayers. An example of a 90 nm bilayer thickness RMF is shown in Figure 5,

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

with a 7.46 m/s flame velocity and 0.9978 correlation coefficient. The flame velocities of each foil are plotted in Figure 6 as a function of bilayer thickness. It is evident that the flame velocity decreased with increasing bilayer thickness.

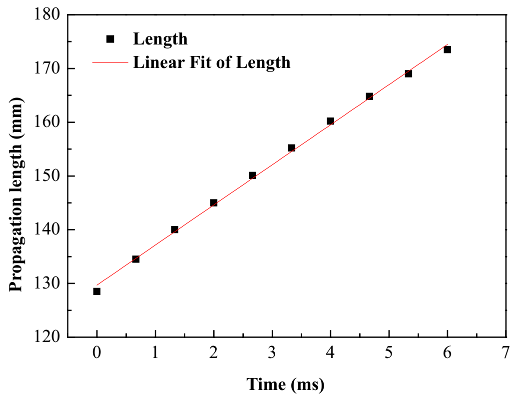


**Figure 5.** Linear fitting of the propagation length versus time for a 90 nm bilayer thickness RMF

Mann's models have been widely used and constantly improved upon in velocity predictions [9, 10], the model providing an effective means of evaluating the flame velocity of an RMF based on the bilayer thickness and premixing spacing. According to the model, the flame propagation process can be described as atomic diffusion and thermal conduction. The flame velocity can be considered as an analytical solution of the simplified atomic diffusion and thermal transport equations:

$$v_x^2 = \left( \sum_{n=odd} \kappa_n^2 / a_n^2 \right)^{-1} \frac{4\lambda^2 \mathrm{R} T_f^2 A}{E(T_{f0} - T_0)} \exp\left(-E/RT_f\right) \tag{2}$$

where $\kappa_n$ and $a_n$ are the Fourier coefficient and eigenvalue of the sine series for $C_0(y)$, respectively, $C_0$ denotes the initial composition related to $\omega$ and $\sigma$, $\lambda$ is the thermal diffusion coefficient, R is the gas constant, $E$ is the activation energy for atomic diffusion, $A$ is the Arrhenius prefactor, $T_f$, $T_0$ and $T_{f0}$ are final temperature of the reacted foil, the initial temperature of the foil and the adiabatic temperature of the reaction, respectively. The parameters were selected from reference [9] and are listed in Table 2. The final temperature of the reacted foil $T_f$ was evaluated from the heat released. The activation energy $E$ and Arrhenius prefactor $A$ was adjusted to get a better agreement with the experimental results.

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

**Table 2.** Parameters selected in Mann's model

| Parameter | $\lambda$ [m$^2$/s] | $\rho$ [kg/m$^3$] | $T_0$ [K] | $T_{f0}$ [K] |
|---|---|---|---|---|
| Value | $2.18 \times 10^{-5}$ | 5800 | 298 | 1958 |

Commercial software MATLAB was employed to optimize the parameters of $E$ and $A$. Mann's model was found to be very consistent with the velocity results from the Al/Ni RMFs fabricated in this research, as $E = 122.1$ kJ/mol and $A = 0.1$; a high correlation coefficient exceeding 0.98 was obtained. Comparisons between the fitted model and the experimental flame velocities, as well as an ideal model ignoring any premixing layer and phase change during the reaction are displayed in Figure 6. For the foils with a small bilayer thickness (for example, 40 nm), the actual propagation speeds of the combustion fronts were much lower than the ideal results. This tendency may be attributed to a larger percentage of premixing layer. The premixing region was assumed to be fully reacted [9, 19] and atomic diffusion was reduced by the presence of an intermetallic compound at the interfaces between each layer. This compound also reduces the available energy and the maximum flame temperature of the sample, which significantly reduces its reaction velocity. At a thicker bilayer (ranged from 55 nm to 120 nm), premixing has little effect on the flame velocity due to its low content. Therefore, the actual propagation speeds were close to the fitted model, as well as the ideal value. However, combustion could not be self-sustained for RMFs with a much larger bilayer (180 nm for example), owing to its heat generation being slower than dissipation. The fitted model shows good agreement with the measured data, when the bilayer thickness is in the range 55 nm to 120 nm. The model obtained is now available for the design of an Al/Ni RMF with a specified flame velocity.

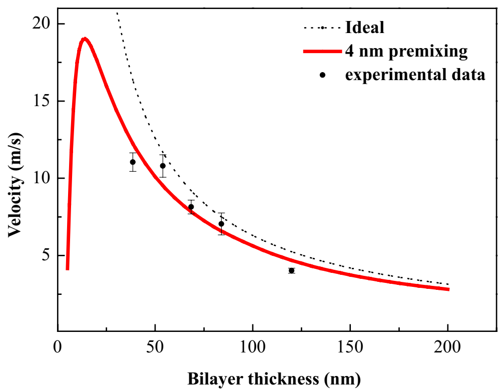


**Figure 6.** Comparisons between the fitted model, the ideal model and the experimental results for different bilayer thicknesses

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

## 3.3 EFI experiments

The capacitor was initially charged to 1306 V; the measured voltage and current waveforms of the exploding bridges are shown in Figure 7. The deposition energy and energy deposition ratio in the first half discharging cycle were calculated and are listed in Table 3. Compared with Cu, an Al/Ni EFI burst much earlier and became much closer to the peak current time, the reasons for this being speculated to be due to the following aspects: (1) the resistivity of an Al/Ni RMF was higher than Cu, so the bridge was heated faster due to strong electrocaloric effects and reached the melting point earlier; (2) the self-propagating reaction was initiated in the Al/Ni foils and resulted in a rapid exploding of the bridge. The closer peak voltage and peak current time led to a higher energy deposition by the Al/Ni EFIs.

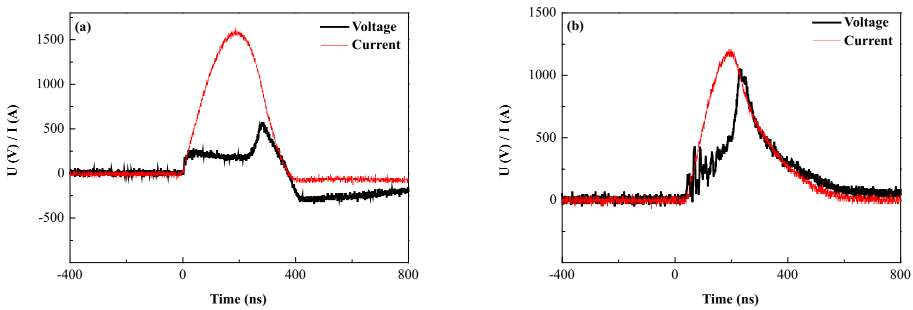


**Figure 7.** The measured voltage and current waveforms of the exploding bridges, (a) Cu bridge; (b) Al/Ni bridge

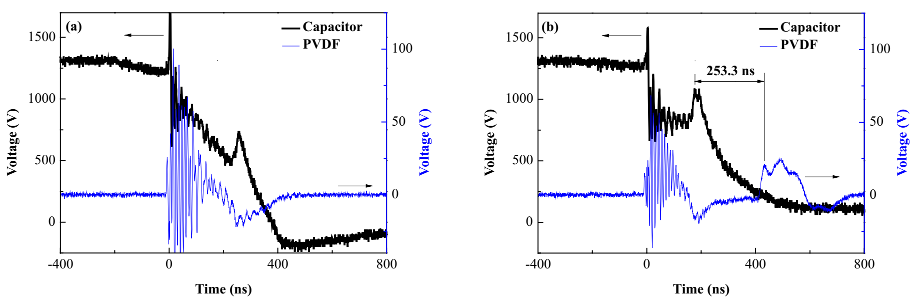


**Figure 8.** Signals of the average velocity measurement, (a) the bridge without flyer; (b) the bridge with flyer

The thickness of the SU-8 photoresist barrel was measured with a screw-thread micrometer and a PVDF film was placed at the end of the barrel to measure the average velocity of the flyer. Figure 8 shows the typical signals of the EFIs without and with a flyer layer; the first peak of the piezoelectric response after

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

the foil exploded was considered to be the time when the flyer impacted the PVDF film. In these experiments, the measured average flyer velocities of Al/Ni RMF exploding foils with 493 μm and 496 μm barrels were 1946.31 m/s and 1859.77 m/s respectively, while the corresponding results for Cu foils with 481 μm and 490 μm barrels were 1751.00 m/s and 1717.49 m/s, respectively. The thickness of the barrels was nearly equal, however Al/Ni EFIs tended to have a higher flyer velocity. We believe that the larger energy deposition and stored chemical energy of an Al/Ni RMF contribute to the higher average velocity of the flyer.

**Table 3.** The deposition energy and energy deposition ratio for different EFIs

| Material | Energy deposition [J] | Energy deposition ratio [%] |
|---|---|---|
| Al/Ni | 0.12681948 | 67.59 |
| | 0.12922344 | 68.88 |
| | 0.12692928 | 67.65 |
| | 0.12654856 | 67.45 |
| Cu | 0.07359588 | 39.23 |
| | 0.07871892 | 41.96 |
| | 0.08378480 | 44.66 |

The Al/Ni RMF is able to lower the operating voltage of the initiators owing to its significant increase in energy deposition. A lower voltage would reduce the volume of the booster circuit as well as the initiator system. Furthermore, the larger average flyer velocity will create a higher pressure shock wave in the explosive placed at the end of its flight, which could enhance the detonation reliability of the EFIs.

## 4 Conclusions

In this paper, Al/Ni RMFs were prepared and characterized. The effects of bilayer thickness and proportion of premixing on the performance were examined. Exploding bridge foils of Cu and Al/Ni RMF were fabricated and tested.

This study indicates that the dependence of the stored energy on the bilayer thickness confirms the existence of a 4 nm premixing layer. A steady combustion velocity of an Al/Ni RMF was characterized and the experimental velocities matched Mann's analytical model well, with an activation energy $E = 122.1$ kJ/mol, Arrhenius prefactor $A = 0.1$ for bilayer thicknesses in the range 55 nm to 120 nm.

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

In addition, Al/Ni RMF could significantly increase the energy deposition and average flyer velocity of an EFI with the same discharge circuit. It can be confirmed that an Al/Ni RMF exploding foil is able to enhance the reliability of the initiator and reduce its volume.

This study indicates that specific properties of an Al/Ni RMF can be realized by adjusting the bilayer thickness, and promisingly, an increase in energy deposition and average flyer velocity will attract more attention on the application of Al/Ni EFIs.

## Acknowledgements

The authors gratefully acknowledge financial support from the National Defense Key Laboratory of Pyrotechnical S&R Technology (9140C370503150C37174). The authors are also grateful to the editors and reviewers for their effective work.

## References

[1] Wang, J.; Besnoin, E.; Duckham, A.; Spey, S. J.; Reiss, M. E.; Knio, O. M.; Powers, M.; Whitener, M.; Weihs, T. P. Room-temperature Soldering with Nanostructures Foils. *Appl. Phys. Lett.* **2003**, *83*(19): 3987-3989.

[2] Braeuer, J.; Besser, J.; Wiemer, M.; Gessner, T. A Novel Technique for MEMS Packaging: Reactive Bonding with Integrated Material Systems. *Sens. Actuators, A* **2012**, *188*(12): 212-219.

[3] Morris, C. J.; Mary, B.; Zakar, E.; Barron, S.; Fritz, G.; Knio, O. M.; Weihs, T. P.; Hodgin, R.; Wilkins, P.; May, C. Rapid Initiation of Reactions in Al/Ni Multilayers with Nanoscale Layering. *J. Phys. Chem. Solids* **2010**, *71*(2): 84-89.

[4] Yang, C.; Hu, Y.; Shen, R. Q.; Ye, Y. H.; Wang, S.; Hua, T. Fabrication and Performance Characterization of Al/Ni Multilayer Energetic Films. *Appl. Phys. A: Mater. Sci. Process* **2013**, *114*(2): 1782-1786.

[5] Poret, J. C.; Ding, M.; Krieger, F.; Swank, J.; Chen, G.; McMullan, C. Nanofoil Heating Elements for Thermal Batteries. *Proc. Army Science Conf.*, *26$^{th}$*, Orlando, USA **2008**, ADM002187.

[6] Rogachev, A. S. Exothermic Reaction Waves in Multilayer Nanofilms. *Russ. Chem. Rev.* **2008**, *77*(1): 22-38.

[7] Adams, D. P. Reactive Multilayers Fabricated by Vapor Deposition: A Critical Review. *Thin Solid Films* **2015**, *576*(16): 98-128.

[8] Ma, E.; Thompson, C. V.; Clevenger, L. A.; Tu, K. N. Self-propagating Explosive Reactions in Al/Ni Multilayer Thin Films. *Appl. Phys. Lett.* **1990**, *57*(12), 1262-1264.

[9] Mann, A. B.; Gavens, A. J.; Reiss, M. E.; Heerden, D. V. Modeling and Characterizing the Propagation Velocity of Exothermic Reactions in Multilayer

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland

Foils. *J. Appl. Phys.* **1997**, *82*(3): 1178-1188.
[10] Wang, L.; He, B.; Jiang, X. H. Modeling the Velocity of Self-propagating Exothermic Reactions in Multilayer Foils. *Combust. Sci. Technol.* **2010**, *182*(8): 1000-1008.
[11] Trenkle, J. C.; Koerner, L. J.; Tate, M. W.; Gruner, S. M.; Weihs, T. P.; Hufnagel, T. C. Phase Transformations during Rapid Heating of Al/Ni Multilayer Foils. *Appl. Phys. Lett.* **2008**, *93*(8): 081903-081903-3.
[12] Trenkle, J. C.; Koerner, L. J.; Tate, M. W.; Walker, N.; Gruner, S. M.; Weihs, T. P.; Hufnagel, T. C. Time-resolved X-ray Microdiffraction Studies of Phase Transformations during Rapidly Propagating Reactions in Al/Ni and Zr/Ni Multilayer Foils. *J. Appl. Phys.* **2010**, *107*(11): 113511-113511-12.
[13] Rothhaar, U.; Oechsner, H.; Scheib, M.; Müller, R. Compositional and Structural Characterization of Temperature-induced Solid-state Reactions in Al/Ni Multilayers. *Phys. Rev. B* **2000**, *61*(2): 974-979.
[14] Rogachev, A. S.; Vadchenko, S. G.; Mukasyan, A. S. Self-sustained Waves of Exothermic Dissolution in Reactive Multilayer Nano-foils. *Appl. Phys. Lett.* **2012**, *101*(6): 3307-3314.
[15] Simões, S.; Viana, F.; Vieira, M. F. Reactive Commercial Ni/Al Nanolayers for Joining Lightweight Alloys. *J. Mater. Eng. Perform.* **2014**, *23*(5): 1536-1543.
[16] Qiu, X.; Tang, R.; Liu, R.; Huang, H.; Guo, S.; Yu, H. A Micro Initiator Realized by Reactive Ni/Al Nanolaminates. *J. Mater. Sci.: Mater. Electron.* **2012**, *23*(12): 2140-2144.
[17] Varesh, R. Electric Detonators: EBW and EFI. *Propellants Explos. Pyrotech.* **1996**, *21*(3): 150-154.
[18] Lv, J. J.; Zeng, Q. X.; Li, M. Y. Metal Foil Gap Switch and Its Electrical Properties. *Rev. Sci. Instrum.* **2013**, *84*(4): 045101-045101-5.
[19] Wang, J.; Besnoin, E.; Duckham, A.; Spey, S. J.; Reiss, M. E.; Knio, O. M.; Weihs, T. P. Joining of Stainless-steel Specimens with Nanostructured Al/Ni Foils. *J. Appl. Phys.* **2004**, *95*(1): 248-256.
[20] Knepper, R.; Snyder, M. R.; Fritz, G.; Fisher, K.; Knio, O. M.; Weihs, T. P. Effect of Varying Bilayer Spacing Distribution on Reaction Heat and Velocity in Reactive Al/Ni Multilayers. *J. Appl. Phys.* **2009**, *105*(8): 083504-083504-9.

Copyright © 2017 Institute of Industrial Organic Chemistry, Poland