Influence of a Ni Foil Interlayer on Interface Properties of Mg-Clad Al Materials by Vacuum Roll Bonding

Jinfeng Shu, Tomiko Yamaguchi* and Yuya Hara

Department of Materials Science, Graduate School of Engineering, Kyushu Institute of Technology, Kitakyushu 804-0015, Japan

Mg-clad Al materials with a Ni foil interlayer were successfully prepared by vacuum roll bonding at a 450°C rolling temperature and 25% reduction, and the effects of the Ni foil interlayer on the interfacial properties were investigated. The clad materials with the Ni foil interlayer only formed the Mg₂Ni intermetallic compound with a 0.9 μm thickness on the Mg–Ni interface, which was smaller than that of the intermetallic compounds (Al₁₂Mg₁₇ of 1.77 μm and Al₃Mg₂ of 7.76 μm) formed on the Al–Mg interface without the interlayer. The bonding strength of the interface increased from 0.79 MPa to 10.46 MPa. The growth characteristics of the Mg₂Ni intermetallic compound on the Mg–Ni interface after heat treatment were investigated. The growth activation energy of Mg₂Ni was 157.24 kJ/mol, which is higher than that of Al₃Mg₂ which mainly affects the Al–Mg interfacial bonding strength. Therefore, the thickness of the Mg₂Ni was thinner and the interfacial bonding strength was greater after the vacuum roll bonding. [doi:10.2320/matertrans.MT-M2019277]

(Received September 27, 2019; Accepted February 3, 2020; Published March 6, 2020)

Keywords: vacuum roll bonding, Mg-clad Al materials, Ni foil interlayer, intermetallic compounds, growth activation energy

1. Introduction

Magnesium (Mg) alloys are currently some of the most widely used eco-friendly structural materials because of their high specific strength, low density and recyclability. They have broad application prospects in the fields of automobiles, aerospace and biomaterials. However, Mg alloys have a poor corrosion resistance due to their electrochemical property.^{1–4)} Therefore, improving the corrosion resistance of Mg alloys and the application scope have become a very meaningful subject to investigate.

In recent years, clad materials have attracted much attention because of their excellent overall performance. They can be used to manufacture industrial parts with different internal and external properties. Therefore, they are widely used in mobile phones, automobiles, aviation, electronics and medical devices.^{5–7)} Aluminum (Al) is a low density and high strength material with an excellent corrosion resistance. It can effectively improve the corrosion resistance of Mg alloys by cladding an Al layer on the Mg alloy's surface to avoid direct contact between the Mg alloys and corrosion medium.^{8,9)} Roll bonding is one of the important technologies for preparing clad materials.¹⁰⁾ However, the surfaces of the Al and Mg sheets are prone to form oxide films during the hot roll bonding, which leads to difficulties in joining dissimilar materials. In this study, in order to avoid the formation of oxide films, the roll bonding will be carried out in a vacuum environment.

During the process of preparing Mg-clad Al materials by roll bonding, Al–Mg intermetallic compounds are easily formed on the Al–Mg interface. The intermetallic compounds are hard and brittle, which significantly affects the interfacial bonding strength.¹¹⁾ In order to reduce the influences of the Al–Mg intermetallic compounds, the most effective method is to avoid direct contact between the Al and Mg sheets by adding an interlayer.⁶⁾ Since the growth activation energies of the intermetallic compounds Mg₂Ni and MgNi₂ which may be formed on the Mg–Ni interface, and Al₃Ni and

Al₃Ni₂ which may be formed on the Al–Ni interface are higher than those of the Al–Mg intermetallic compounds, the adverse effect on the interface properties may be smaller.^{12–16)} Therefore, Ni foil was used as the interlayer to study the effects of the Ni interlayer on the Al–Mg interface in this study.

2. Experimental Procedure

The materials used in the experiment were magnesium alloy AZ31 sheets (Mg sheets) with the size of 1.5 mm × 65 mm × 200 mm, pure aluminum A1050 sheets (Al sheets) with the size of 3 mm × 80 mm × 300 mm and 10 μm thick pure Ni foil. In order to prevent the bending of the Al and Mg sheets during rolling, steel SS400 sheets (Fe sheets) with the size of 8 mm × 80 mm × 300 mm were used for rolling together with the Al and Mg sheets. The Al and Mg sheets were annealed for 30 min at 350°C and 400°C, respectively. Before the experiment, the oxide films were removed from the Al and Mg sheet surfaces by a steel brush and 2000# sandpaper, respectively. Using Ni foil as the interlayer of the Al and Mg sheets, the materials were heated to 450°C by high frequency induction under a 6×10^{-3} Pa vacuum environment and entered the bonding rolls. The heating time was 30 min to ensure that there was enough time for heat conduction to keep the temperature between the Al and Mg sheets consistent. The reduction was set to 25% (refers to the total reduction of the Al and Mg sheets). After rolling, the clad materials were cooled in a vacuum. Figure 1 shows a schematic diagram of the vacuum rolling equipment used in the experiment. The effects of the Ni foil interlayer on the Al–Mg interfacial properties were investigated by scanning electron microscopy (SEM), energy dispersive x-ray spectrometry (EDS), electron probe x-ray micro-analyzer (EPMA), X-ray diffraction (XRD) and a TCE-N300 tensile testing machine. As a comparison, the experiments without the Ni foil interlayer were also carried out under the same conditions. Figure 2 shows the tensile test of the interfacial bonding strength.

*Corresponding author, E-mail: yamaguchi.tomiko374@mail.kyutech.jp

Fig. 1 Schematic diagram of the vacuum rolling equipment.

The Mg-clad Al materials with the Ni foil interlayer prepared by vacuum roll bonding were heat treated at 623 K (350°C), 673 K (400°C) and 703 K (430°C) in a muffle furnace. The growth characteristics of the intermetallic compounds on the interface after heat treatment were investigated.

3. Results and Discussion

After vacuum roll bonding without any interlayer, the interfacial properties were analyzed. Figure 3(a) shows an SEM image of the Al–Mg interface. It can be seen that about 10 μm a reaction layer was formed on the Al–Mg interface. The EDS line-scanning for the interface was performed, and the results are shown in Fig. 3(b). It is shown that the reaction layer was divided into two layers with different

Al–Mg compositions. Based on the Al–Mg equilibrium phase diagram,¹⁷⁾ the atomic ratio of Al to Mg in the reaction layer connecting the Mg side is close to $\text{Al}_{12}\text{Mg}_{17}$, and that of Al to Mg in the reaction layer connecting the Al side is close to Al_3Mg_2 , with a thickness of 1.77 μm and 7.76 μm , respectively. This indicated that two layers of intermetallic compounds, $\text{Al}_{12}\text{Mg}_{17}$ and Al_3Mg_2 , were formed on the Al–Mg interface after vacuum roll bonding. The bonding strength of the interface was 0.79 MPa by the tensile test on the clad materials. The intermetallic compounds are hard and brittle, and the hardness of the Al_3Mg_2 and $\text{Al}_{12}\text{Mg}_{17}$ reached HV415 and HV383, respectively.¹⁸⁾ Considering that the intermetallic compounds layer on the Al–Mg interface have a significant influence on the bonding strength, the bonding strength was very low.

The morphology of the fracture surface for the Al–Mg interface without the interlayer is shown in Fig. 4(a). The typical morphology of the brittle cleavage fracture is observed on the fracture surface, which is relatively flat and there are some cleavage steps. Based on Fig. 4(b), the atomic percentages of the Al and Mg elements on the fracture surface are 60.16% and 39.18%, respectively, which are close to Al_3Mg_2 . Therefore, it can be inferred that the fracture mainly occurred in the Al_3Mg_2 layer during the tensile test and was brittle fracture. As shown in Fig. 3(b), considering that the thickness of the Al_3Mg_2 is greater than that of $\text{Al}_{12}\text{Mg}_{17}$, and the hardness of Al_3Mg_2 is also greater than that of $\text{Al}_{12}\text{Mg}_{17}$, the formation and propagation of cracks mainly occurred in the Al_3Mg_2 layer.

Fig. 2 Tensile test of the interfacial bonding strength.

Fig. 3 (a) SEM image of the Al–Mg interface without the interlayer and (b) EDS line-scanning profile for the interface.

Fig. 4 (a) Morphology of the fracture surface for the Al-Mg interface without the interlayer and (b) EDS analysis from the fracture.

Fig. 5 (a) SEM image of the Al-Mg interface with the Ni foil interlayer after vacuum roll bonding and (b) EDS analysis from reaction layer on the Mg-Ni interface.

The SEM image of the interface after vacuum roll bonding using Ni foil as the interlayer is shown in Fig. 5(a). No reaction layer was observed on the Al-Ni interface. However, a reaction layer with about a 0.9 μm thickness was observed on the Mg-Ni interface. Figure 5(b) shows the elemental contents in the Mg-Ni interfacial reaction layer. The reaction layer contains almost no Al element, and the atomic ratio of Mg to Ni 2.03 is almost equal to the stoichiometric ratio of Mg_2Ni according to the Mg-Ni equilibrium phase diagram.¹³⁾ Therefore, the Mg_2Ni intermetallic compound was formed on the Mg-Ni interface after vacuum roll bonding. After adding the Ni foil interlayer, the direct contact between the Al and Mg sheets was prevented, and the Al-Mg intermetallic compounds, Al_3Mg_2 and $\text{Al}_{12}\text{Mg}_{17}$, were not observed. In addition, the thickness of Mg_2Ni (0.9 μm) formed on the Mg-Ni interface is much smaller than that of the intermetallic compounds ($\text{Al}_{12}\text{Mg}_{17}$ of 1.77 μm and Al_3Mg_2 of 7.76 μm) formed on the Al-Mg interface without an interlayer.

Figure 6 displays the distribution of the Al, Ni and Mg elements on the Al-Mg interface with the Ni foil interlayer after vacuum roll bonding. Similar to the results of Fig. 5, the Mg_2Ni layer with a 0.9 μm thickness was observed on the Mg-Ni interface. In addition, the diffusion layers of the Ni element were observed on both sides of Mg and Al, and it was obvious that the diffusion rate of Ni on the Mg side is much faster than that on the Al side.

Figure 7(a), (b) shows the fracture surface morphologies of the Al-Mg interface with a Ni foil interlayer after the tensile test. The bonding strength of the interface reached 10.46 MPa. Compared to the interface without any interlayer

(0.79 MPa), the bonding strength of the interface was significantly improved. Most areas of the fracture surface appear to have the morphological characteristics of ductile fracture (area A), and there are some small dimples in the fracture surface. In addition, there are also some morphological characteristics of brittle fracture on the fracture surface (area B). The elemental contents of the Mg, Al and Ni in area A and area B are shown in Fig. 7(c) and (d), respectively. Whether in the ductile fracture area A or the brittle fracture area B, the main elements present are Mg and Ni, and the atomic percentages of Mg and Ni in area B are 59.88% and 36.48% respectively, which is close to that of Mg_2Ni . Therefore, the fracture occurred on the Mg-Ni interface during the tensile test, and only a small amount occurred in the Mg_2Ni layer. This indicated that although Mg_2Ni was formed on the Mg-Ni interface, it does not play a major role in the fracture process due to the small layer thickness.

In Fig. 8, the XRD peaks from the Al and Mg sides of the fracture surface for the Al-Mg interface with Ni foil interlayer are displayed. In addition to the diffraction peaks of the main elements, there are also diffraction peaks of Ni and Mg_2Ni on both sides of the fracture surface. Furthermore, the diffraction peaks of Mg appear on the Al side, while the diffraction peaks of Al are not observed on the Mg side. These results show that the fracture occurred only on the Mg-Ni interface, which is the same as the results obtained in Fig. 7. A schematic diagram of the interface fracture is shown in Fig. 9.

The Mg-clad Al materials with a Ni foil interlayer were heat treated at 623 K (350°C), 673 K (400°C) and 703 K

Fig. 6 EPMA area analysis of the Al-Mg interface with the Ni foil interlayer after vacuum roll bonding.

Fig. 7 (a), (b) Fracture surface morphologies of the Al-Mg interface with a Ni foil interlayer; (c), (d) EDS analysis from area A and area B.

Fig. 8 XRD peaks from the Al (a) and Mg (b) sides of the fracture surface for the Al-Mg interface with Ni foil interlayer.

Fig. 9 Schematic diagram of the Al-Mg interface fracture with a Ni foil interlayer.

(430°C). Figure 10(a), (b) shows the SEM image of the interface after heat treatment at 673 K for 2 h and 703 K for 6 h, respectively. The thickness of the reaction layer on the Mg–Ni interface grew from 0.9 μm to 1.24 μm after the heat treatment at 673 K for 2 h, and no reaction layer was observed on the Al–Ni interface. However, the thickness of the reaction layer on the Mg–Ni interface grew from 0.9 μm to 7.29 μm after heat treatment at 703 K for 6 h, and a reaction layer with a 2.94 μm thickness was also observed on the Al–Ni interface. The EDS line-scanning of the interface was performed, and the results are shown in Fig. 10(c) and (d). The atomic ratio of Mg to Ni in the Mg–Ni interfacial reaction layer is close to Mg_2Ni after heat treatment at 673 K for 2 h and 703 K for 6 h, indicating that even after heat treatment at 703 K for 6 h, the reaction layer on the Mg–Ni interface contains only Mg_2Ni . There are two intermetallic

compounds, Mg_2Ni and MgNi_2 , based on the Mg–Ni equilibrium phase diagram.¹³⁾ The absence of MgNi_2 in the reaction layer of the Mg–Ni interface indicated that the diffusion temperature was not high enough and the time was not long enough. From Fig. 10(d), in the reaction layer formed on the Al–Ni interface after heat treatment at 703 K for 6 h, the atomic ratio of Al to Ni is close to 3, which indicated that the intermetallic compound, Al_3Ni , with a 2.94 μm thickness was formed on the Al–Ni interface according to Al–Ni equilibrium phase diagram.¹²⁾

Figure 11(a) shows the relationship between the Mg_2Ni thickness and time at different heat treatment temperatures. At any temperature, the thickness of the Mg_2Ni is proportional to the square root of the time. The thickness of the growing Mg_2Ni phase after heat treatment can be described by:¹⁹⁾

$$X^2 = 2Dt \quad (1)$$

where X is the thickness of the Mg_2Ni layer, D is the growth rate constant and t is the heat treatment time. The growth of Mg_2Ni clearly followed parabolic kinetics, suggesting that the growth was diffusion controlled. Based on the results of the marker experiments,¹⁴⁾ Mg was the dominant diffusing element in the Mg_2Ni . Using eq. (1) and Fig. 11(a), the growth rate constants D were calculated for each temperature, which were $4.61 \times 10^{-5} \mu\text{m}^2/\text{s}$, $6.60 \times 10^{-4} \mu\text{m}^2/\text{s}$, $1.35 \times 10^{-3} \mu\text{m}^2/\text{s}$ at 623 K, 673 K and 703 K, respectively. There is a linear relationship between $\ln D$ and reciprocal of the temperature, as shown in Fig. 11(b). The temperature dependence of the growth rate constant for Mg_2Ni should follow the Arrhenius relation expressed by:¹⁹⁾

$$\ln D = -(Q/R) \times (1/T) + \ln D_0 \quad (2)$$

where Q is the growth activation energy, T is the temperature (in kelvin), R is the universal gas constant, and D_0 is a

Fig. 10 SEM image and EDS line-scanning profile of the interface after heat treatment for clad materials with the Ni foil interlayer at 673 K for 2 h (a), (c) and 703 K for 6 h (b), (d).

Fig. 11 (a) Relationship between the Mg₂Ni thickness and time at different temperatures and (b) temperature dependence of the growth rate constant for Mg₂Ni.

constant. The growth activation energy of Mg₂Ni can be calculated from eq. (2) and Fig. 11(b): $Q = 157.24$ kJ/mol. M.Y. Tsai¹⁴) studied the solid–solid interfacial reaction in the Mg–Ni binary diffusion couples and the reaction temperature was at 673, 703, 723, and 753 K. The growth activation energy of Mg₂Ni was determined to be 120 kJ/mol. This different result may be due to the different temperature range of the study. The growth activation energy of Mg₂Ni is greater than that of Al₃Mg₂ (85.5 kJ/mol¹⁶) which mainly affects the interfacial bonding strength, so the Mg₂Ni layer formed on the interface after vacuum roll bonding was thinner, which improved the interfacial bonding strength.

4. Conclusions

The Mg-clad Al materials with a Ni foil interlayer were prepared by vacuum roll bonding at a 450°C rolling temperature and 25% reduction. No intermetallic compound was formed on the Al–Ni interface, and the Mg–Ni interface formed the intermetallic compound Mg₂Ni layer with a 0.9 μm thickness, which was smaller than that of the intermetallic compounds (about 10 μm) formed on the Al–Mg interface without an interlayer. Compared to the Mg-clad Al materials without any interlayer, the bonding strength of the interface increased from 0.79 MPa to 10.46 MPa. The main reason is that the growth activation energy of Mg₂Ni (157.24 kJ/mol) is greater than that of Al₃Mg₂ which mainly affects the Al–Mg interfacial bonding strength, and there is no overly thick intermetallic compound layer on the interface.

REFERENCES

- 1) Z.P. Gu, Y.Z. Huang, Y. Wang, N.Y. Yuan and J.N. Ding: *Mater. Lett.* **252** (2019) 304–307.
- 2) T.M. Pollock: *Science* **328** (2010) 986–987.
- 3) H.A. Patel, D.L. Chen, S.D. Bhole and K. Sadayappan: *J. Alloy. Compd.* **496** (2010) 140–148.
- 4) H. Haferkamp, R. Boehm, U. Holzkamp, C. Jaschik, V. Kaese and M. Niemeyer: *Mater. Trans.* **42** (2001) 1160–1166.
- 5) Y. Yoshida, L. Cisar, S. Kamado and Y. Kojima: *Mater. Trans.* **43** (2002) 2419–2423.
- 6) K.S. Lee, J.S. Kim, Y.M. Jo, S.E. Lee, J. Heo, Y.W. Chang and Y.S. Lee: *Mater. Charact.* **75** (2013) 138–149.
- 7) H.L. Yu, C. Lu, A.K. Tieu, A. Godbole, L.H. Su, Y. Sun, M. Liu, D.L. Tang and C.L. Kong: *Sci. Rep.* **3** (2013) 2373.
- 8) X.P. Zhang, T.H. Yang, J.Q. Liu, X.F. Luo and J.T. Wang: *J. Mater. Sci.* **45** (2010) 3457–3464.
- 9) H. Matsumoto, S. Watanabe and S. Hanada: *J. Mater. Process. Technol.* **169** (2005) 9–15.
- 10) Q. Wang, X.S. Leng, J.C. Yan, W.B. Guo, Y. Fu and T.M. Luan: *J. Mater. Sci. Technol.* **29** (2013) 948–954.
- 11) Z.W. Xu, Z.W. Li, J.Q. Li, Z.P. Ma and J.C. Yan: *Ultrason. Sonochem.* **46** (2018) 79–88.
- 12) C. Colinet, P. Hicter and A. Pasturel: *Phys. Rev. B* **45** (1992) 1571–1580.
- 13) C.L. Tsao and S.W. Chen: *J. Mater. Sci.* **30** (1995) 5215–5222.
- 14) M.Y. Tsai, M.H. Chou and C.R. Kao: *J. Alloy. Compd.* **471** (2009) 90–92.
- 15) S.B. Jung, Y. Minamino, T. Yamane and S. Saji: *J. Mater. Sci. Lett.* **12** (1993) 1684–1686.
- 16) S. Brennan, K. Bermudez, M.S. Kulkarni and Y. Sohn: *Metall. Mater. Trans. A* **43** (2012) 4043–4052.
- 17) J.L. Murray: *Bull. Alloy. Pha. Diag.* **3** (1982) 60–74.
- 18) I.K. Kim and S.I. Hong: *Metall. Mater. Trans. A* **44** (2013) 3890–3900.
- 19) C.J. Li, Z.J. Yuan, R. Guo, W.D. Xuan, Z.M. Ren, Y.B. Zhong, X. Li, H. Wang and Q.L. Wang: *J. Alloy. Compd.* **641** (2015) 7–13.