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Effect of Annealing Temperature on the Microstructure and Mechanical Properties of the Al/Mg–8Li–3Al–1Zn/Al Composite Plates Fabricated by Hot Rolling

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Abstract—Al/Mg–Li/Al composite plates were fabricated by hot rolling with 50% thickness reduction at 673 K and annealed at 473–673 K for 1 h. The microstructure and mechanical properties of the composite plates under different annealing temperatures were investigated. The results show that the composite plates with a good bonding interface are fabricated. The diffusion layer consisting of Al_3Mg_2, $Al_{12}Mg_{17}$, AlLi, and $MgLiAl_2$ has formed at the interface after annealing at and above 623 K, while the interfacial bonding strength increases with the annealing temperature. The microhardness of the interface increases rapidly when the annealing temperature is above 573 K. The elongation of the composite plate increases with annealing temperature. The tensile strength decreases when annealing temperature is above 573 K.

Keywords: composite plates, hot rolling, anneal, diffusion layer, interface bonding

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1. INTRODUCTION

With the advantages of low-density, good machinability, excellent castability, good damping ability and electromagnetic shielding, high specific stiffness, and specific strength, etc., Mg–Li alloy is becoming more and more attractive [1]. It has widespread applications in the fields of aerospace, electronic communication, and defense industry [2, 3]. However, Mg–Li alloy has a crucial drawback of poor corrosion resistance [4]. It is known that aluminum and aluminum alloys possess better corrosion resistance due to the inert oxide film, and they are also extensively used as lightweight materials [5, 6]. Accordingly, the sandwich composite materials of Al/Mg–Li/Al are found to possess both the advantages of Mg–Li alloys and Al alloys.

It is an effective way to improve the comprehensive properties of the material by the preparation of composite plates [7]. There are many ways to prepare composite plates, such as explosive welding, roll bonding, roll-cast composite, vacuum diffusion welding, and so on [8–10]. Among these methods, roll bonding has the advantages of simple process, security, and low processing cost [11].

In the composite plate prepared by roll bonding, the joint interface is an important item that determines the bonding strength. The annealing temperature after rolling is one of the most important parameters that affect bonding strength.

As for Mg alloys/Al composite plate, Y.L. Guo [12] used warm rolling process to fabricate pure Al/Mg–9.5Li–2Al alloy clad plates. The results show that the interface bonding strength increases with the annealing temperature and attains its maximum value of 20 MPa when the annealing temperature is 623 K. Hiroaki Matsumoto fabricated the Pure Al/Mg–9.5Li composite plates by cold rolling, the results show that, when the composite plates have been annealed at and above 523 K, the phase of Al_3Mg_2 appears at the interface [13]. The microstructure and mechanical property of 5052/Mg–9.5Li–2.1Al composite plates at different annealing temperature was studied by Z. Chen, the results show that, when the annealing temperature is 623 K, the composite plates possess the best mechanical property [14].

However, the microstructure evolution of interfaces under the different annealing temperature is still not clear entirely. Mg–8Li alloy possesses duplex structure (α-Mg and β-Li) and presents a good ductil-

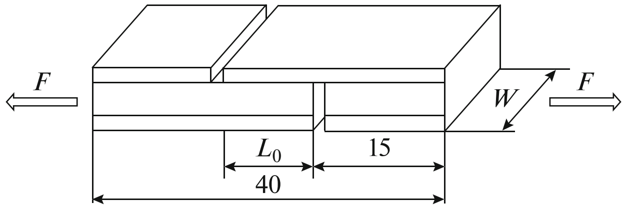

Fig. 1. Schematic of the specimen for bonding strength testing.

ity with a moderate strength. Al and Zn are the commonly used alloying elements to improve the mechanical properties of Mg–Li alloy. In this paper, Mg–8Li–3Al–1Zn was chosen as investigation object. It was cladded with Al by hot rolling. The composite plate was annealed under different temperature, the effect of temperature on the microstructure and mechanical properties was investigated.

2. EXPERIMENTAL

The Mg–8Li–3Al–1Zn alloy was melted in an induction furnace under the protection of argon atmosphere. The materials used for preparation of Mg–8Li–3Al–1Zn alloys were commercial pure (CP) magnesium, CP lithium, CP aluminum, CP zinc. The materials were loaded to a graphite crucible. After the materials were fully molten, the melt was poured into a permanent mold. To eliminate segregation, the as-cast alloy was homogenized at 473 K for 12 h under the protection of MgO powder. Then it was cut into specimens 4 mm thick. The chemical composition of the alloy was analyzed by the methods of inductively coupled plasma spectroscopy to be 8.35% Li, 3.26% Al, 1.27% Zn (wt %), and Mg the balance. The 1050Al plate was obtained commercially of which the initial thickness was 10 mm. The chemical composition of the 1050Al was established by inductively coupled plasma spectroscopy as 0.04% Cu, 0.03% Mn, 0.03% Mg, 0.04% Zn, 0.03% Ti, 0.05% V (wt %), and Al the balance. The thickness reduction of 1050Al plates during cold-rolling processes was 90%. Then the 1 mm thickness 1050Al cold rolling plates and 4mm thickness Mg–8Li–3Al–1Zn plates were used for roll bonding. All plates were cut into 5 mm × 10 mm rectangles, with cleaning the surface by mechanical and chemical methods. Then the plates were fixed with rivets in the sequence of 1050Al/Mg–8Li–3Al–1Zn/1050Al for hot rolling at 673 K. The thickness reduction of composite plates by hot rolling was 50% (6–3 mm, 15% per pass). Due to that the recrystallization temperature of Mg–8Li–3Al–1Zn was near 573 K and the size of specimens was small, the specimens were annealed at 473–673 K for 1 h. Finally, the specimens were cooled in the air.

Microstructure of specimens was observed with LEICA DMRIM optical microscopy (OM) and JEOL JSM 6480A scanning electron microscopy (SEM). Before observation, the side of Mg–8Li–3Al–1Zn constituent was etched with a solution containing ethanol, picric acid, acetic acid, and water, whereas the side of 1050Al was etched with Keller's solution. The interface chemical compositions of specimens were measured with the JEOL INC250 energy dispersive spectroscopy- (EDS) and Rigaku TTR-III X-ray diffraction (XRD) attachments. The scanning line is 42 microns long, the midpoint of which is the midpoint of the interface. The mechanical properties of the specimens were measured on a WDW3050 universal electronic tensile machine with the 0.96 mm/min speed at room temperature in accordance with the standard GB/T 228.1-2010. The microhardness was measured with a HXS-1000Z microhardness tester, the loading force was 50 g, and the holding time, 15 s. And the Microhardness values of 1050Al and Mg–8Li–3Al–1Zn alloys were collected in an interval of 20 μm. The dimensions of the specimens for interface bonding strength test are shown in Fig. 1. The bonding strength can be calculated by the equation of $\tau_{\max} = \frac{F_{\max}}{wL_0}$, where $\tau_{\max}$ is the bonding strength, $F_{\max}$ is the tensile strength load, w is the width of specimens and L_0 is the distance of slits.

3. RESULT AND DISCUSSION

3.1. Effect of Annealing Temperature on Microstructure

Figure 2 shows the OM images of Mg–8Li–3Al–1Zn in the states of as-rolled and subsequently annealed at different temperatures for 1 h. The Mg–8Li–3Al–1Zn alloy is composed of two phases. A blocky α(Mg) phase (white part), uniformly distributes in the β(Li) phase (grey part) [15]. There is a small amount of twins in the as-rolled alloy, as is shown in Fig. 2a. When the annealing temperature is 473 K, a mass of annealing twins form in the α(Mg) phase, as is shown in Fig. 2b. The amount of the annealing twins decreases with the increase in temperature, as is shown in the Figs. 2c and 2d, which can be explained that twins are "swallowed" gradually by the recrystallization grains. When the temperature reaches 623 K, the twins almost disappear, while the recrystallization occurs and the grain size becomes larger with the increase in temperature, as is shown in Figs. 2e and 2f.

Figures 3 and 4 shows SEM images and EDS elemental distributions of Al and Mg in the interface of 1050Al/Mg–8Li–3Al–1Zn/1050Al composite plates under the conditions of the as-rolled state and states subsquently annealed at different temperatures for 1 h. There does not exists any crack and flaw at the bonding interface. When the temperature exceeds 573 K, some cracks and flaws appear at the bonding interface. There exists a small diffusion layer at the interface when the annealing temperature is 623 K. However, when the annealing temperature is 673 K, the diffusion layer rapidly grows. When the annealing temperature is low, diffusion does not happen because of the

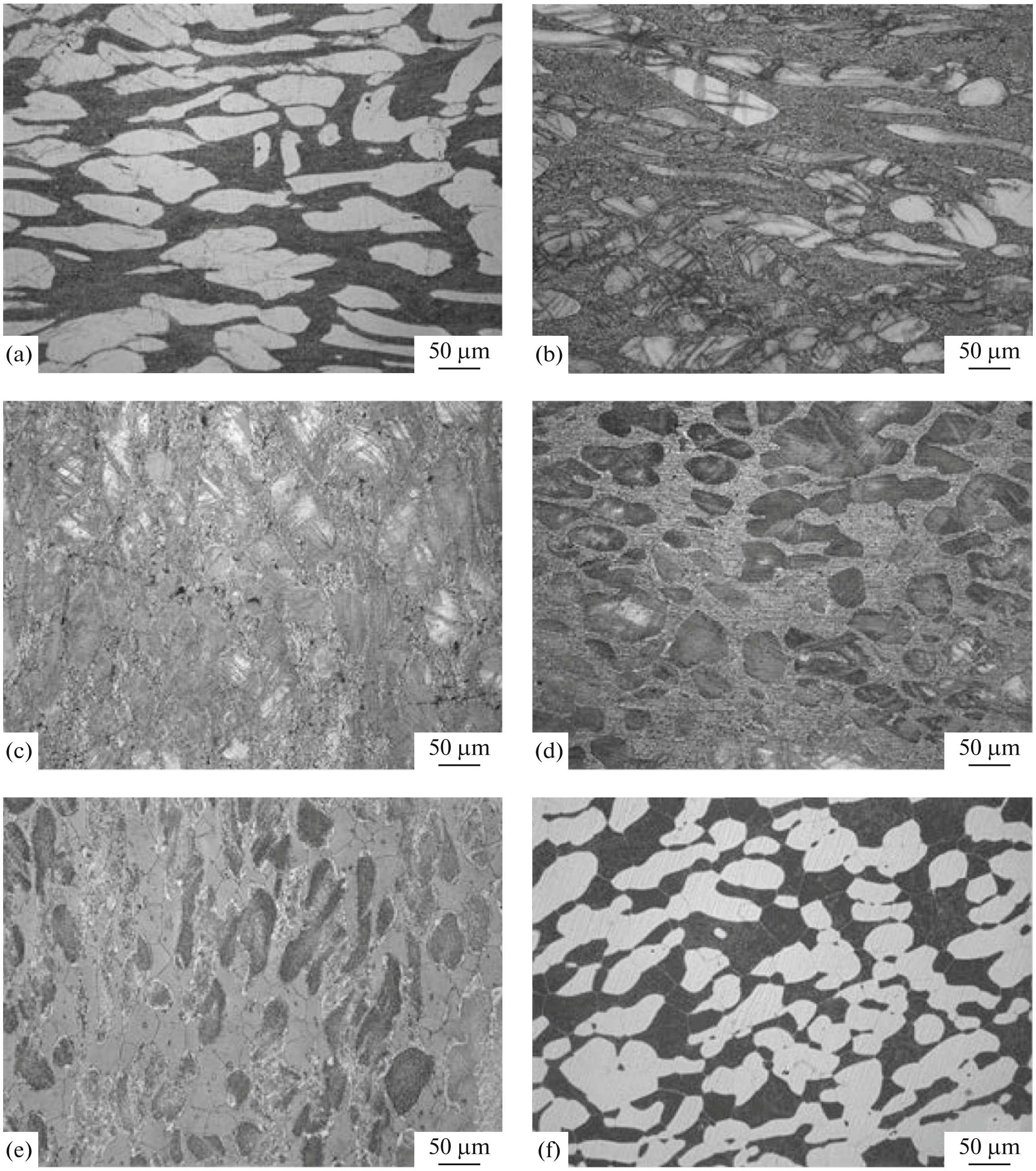

Fig. 2. OM images of the Mg–8Li–3Al–1Zn constituent in the states of (a) as-rolled, and annealed at (b) 473, (c) 523, (d) 573, (e) 623, and (f) 673 K for 1 h.

surface energy. With the increases in annealing temperature, the active atoms break through the diffusion energy barrier of interface, to form a diffusion layer. According to the diffusion equation, with the diffusivity $D = D_0e^{-Q/RT}$, the relationship between diffusion flux and annealing temperature is exponential. Therefore, the interfacial transition layer expands rapidly with the increase in temperature.

The elemental distributions at interface of 1050Al/Mg–8Li–3Al–1Zn/1050Al composite plate is shown in Fig. 4. When annealing temperature is higher than 573 K, it shows an obvious mutual diffusion between Mg and Al. To measure the phases at the interface, XRD patterns at the surface of separated plates from the composite were taken. The results are shown in Fig. 5, the reaction phases at the interface of composite plate include $Mg_{17}Al_{12}$, Al_3Mg_2, AlLi, and $MgLiAl_2$. The phases $Mg_{17}Al_{12}$, AlLi, and $MgLiAl_2$ exist at the interface near the side of Mg–8Li–1Zn alloy, and the phases Al_3Mg_2, AlLi, and $MgLiAl_2$ exist

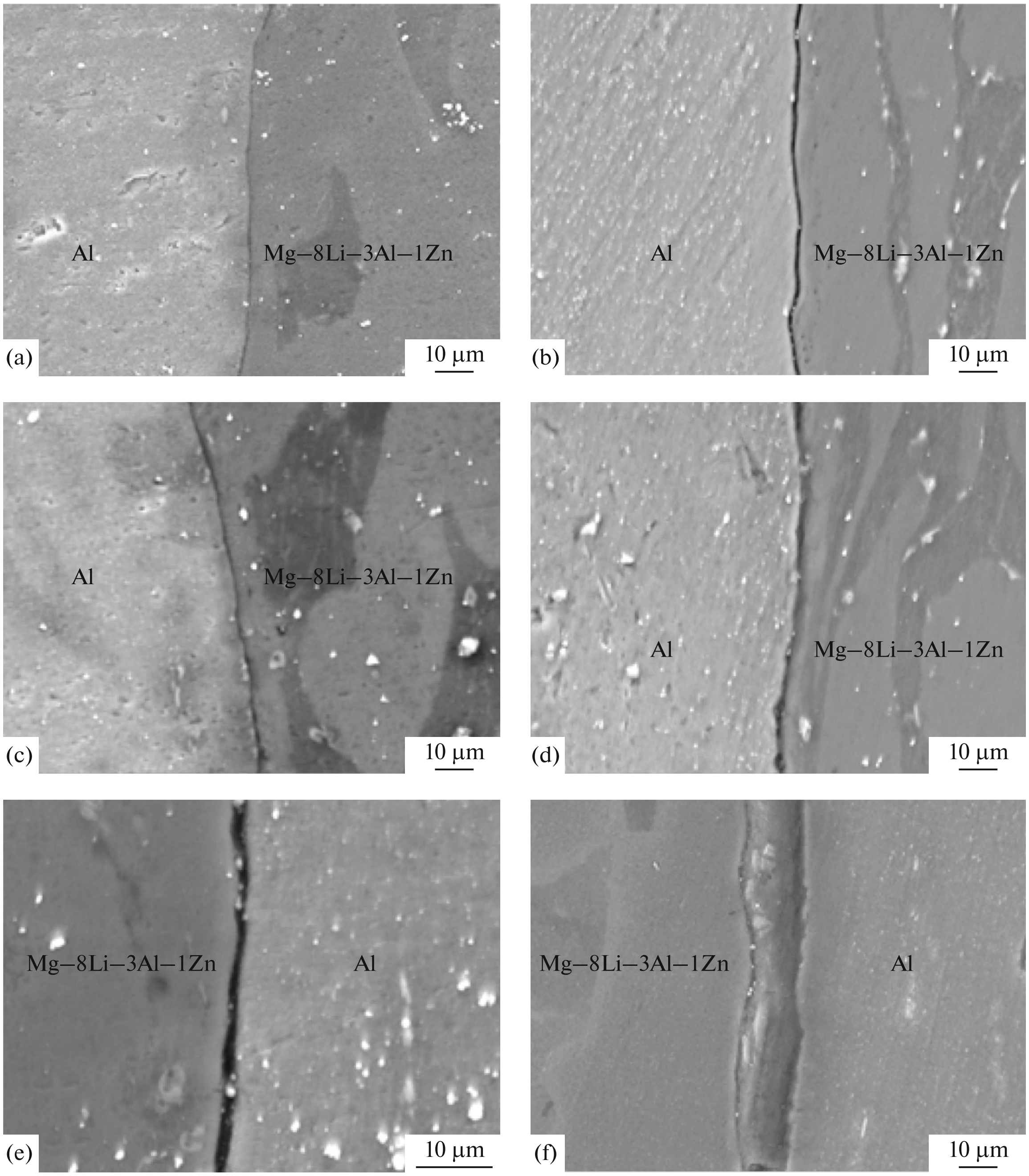

Fig. 3. SEM graphs of Al and Mg at the interface of composite plate in the states of (a) as-rolled, and annealed at (b) 473, (c) 523, (d) 573, (e) 623, and (f) 673 K for 1 h.

at the interface near the side of 1050Al alloy. It is apperent that the diffusion of magnesium, lithium, and aluminum atoms occurs when the annealing temperature is 673 K.

3.2. Effect of Annealing Temperature on Interfacial Bonding Strength

Figure 6 shows the interfacial bonding strength at different annealing temperatures. The bonding strength increases with the annealing temperature. There are two main factors affecting the bonding strength of the composite plate. First, the residual stress between the solid-state bonded dissimilar materials can be reduced or removed through annealing [16]. The residual stress produced during deformation is unfavorable for bonding strength. Therefore, the increase of annealing temperature can increase the bonding strength. Second, when the annealing temperature reaches a certain value, the diffusion reaction

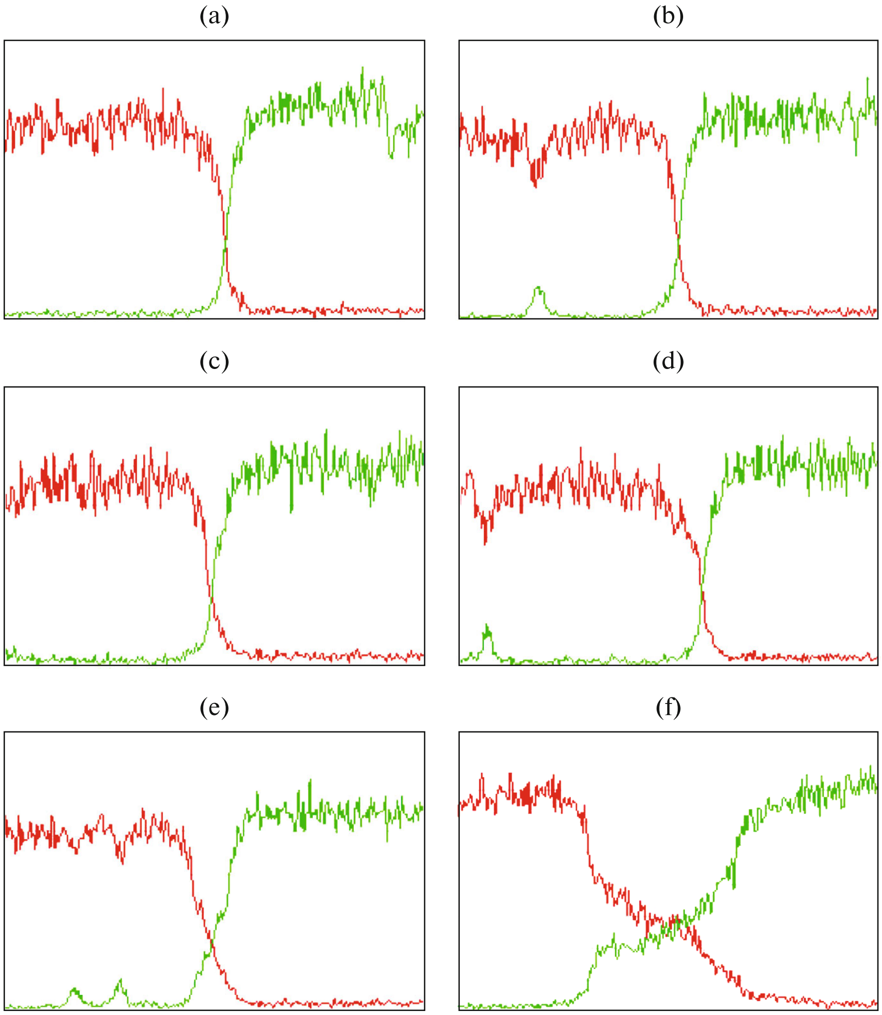

Fig. 4. EDS elemental distributions of Al (red) and Mg (green) at the interface of composite plate in the states of (a) as-rolled, and annealed at (b) 473, (c) 523, (d) 573, (e) 623, and (f) 673 K for 1 h.

happens, which improves the bonding strength. Therefore, the bonding strength of the composite plate increases as the annealing temperature increases in a certain range.

3.3. Effect of Annealing Temperature on the Microhardness of Composite Plate

Figure 7 shows the microhardness value of composite plate and interface as a function of annealing temperature. The microhardness values of Mg–8Li–3Al–1Zn side and Al side both decrease as the annealing temperature increases. This can be attributed to the decrease of the internal stress during the annealing process. The microhardness of the interface decreases with the increase of annealing temperature when the annealing temperature is lower than 573 K, and it increases with the increase of annealing temperature when the annealing temperature is higher than 573 K. This may be due to the competition between the nucleation and growth of intermetallics and internal stress relaxation process. The intermetallics and internal stress have a positive effect on the microhardness. When the temperature is below 573 K, the effect of the decrease of internal stress on microhardness is greater than that of nucleation and growth of intermetallic compounds causing a downward trend of the microhardness. When the temperature is higher than 573 K, the effect of nucleation and growth of intermetallic compounds on microhardness is greater than that of internal stress, causing an upward trend of the microhardness.

3.4. Effect of Annealing Temperature on Tensile Properties

Figure 8 shows the tensile strength and elongation of the composite plate before and after annealing at different temperatures for 1 h. When the temperature is lower than 573 K, the elongation increases with the

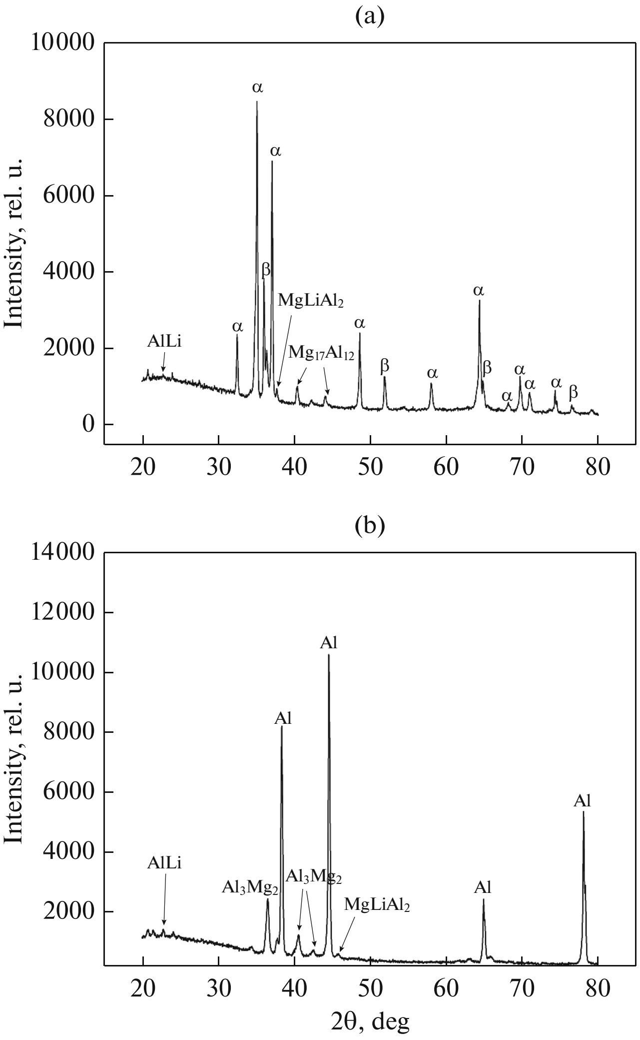

Fig. 5. The XRD patterns of the peeling surface of the composite plate annealed at 673 K for 1 h.

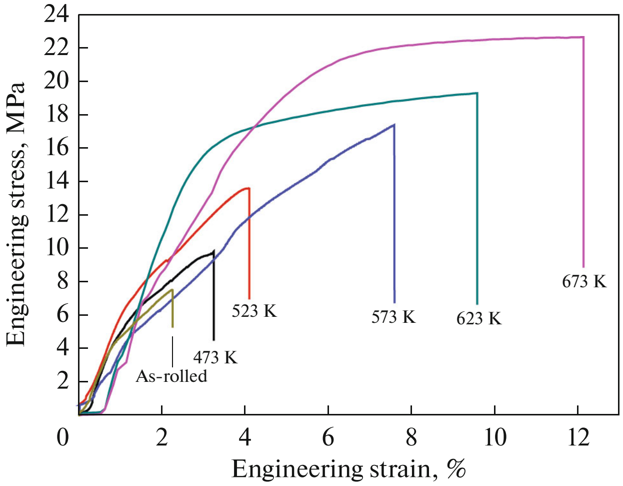

Fig. 6. Interfacial bonding strength of specimens of as-rolled and annealed at 473, 523, 573, 623, 673 K.

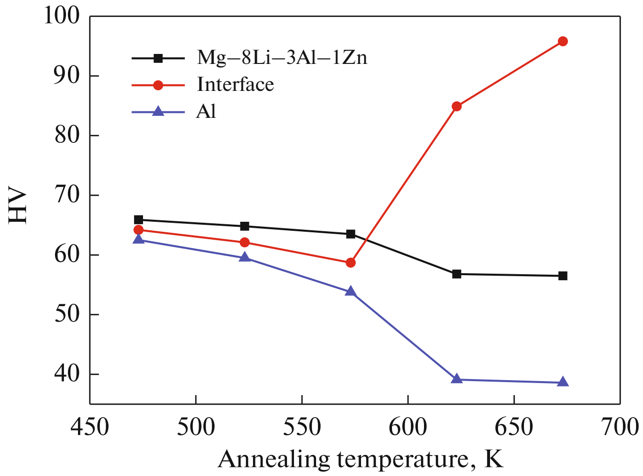

Fig. 7. Microhardness of each plate and interface as a function of annealing temperature.

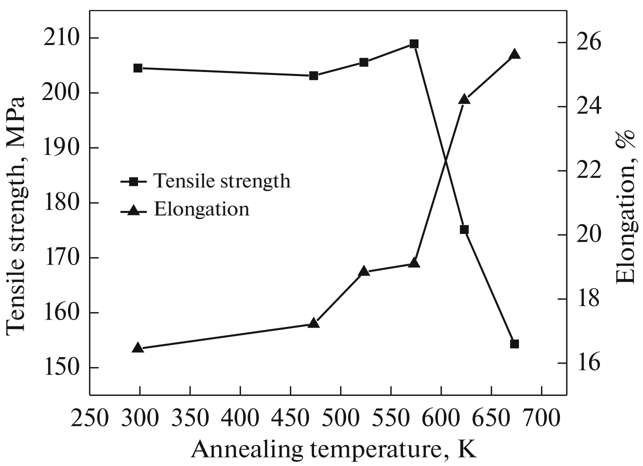

Fig. 8. Tensile strength and elongation of the composite plate before and after annealing for 1 h at different temperatures.

increase of temperature, while the ultimate tensile strength keeps virtually unaltered. The improvement of elongation can be attributed to no reaction phase existing at the interface layer and the decreases of residual stress. Moreover, due to the combined effects of hardening and softening induced by twins and recovery, the ultimate tensile strength keeps stable. In contrast, when the annealing temperature is higher than 623 K, the ultimate tensile strength decreases seriously with the increasing of annealing temperature. The improvement of elongation in tensile behavior is ascribed to restoration (recovery and recrystallization) of pure Al and Mg–8Li–3Al–1Zn constituents and to the disappearance of the hardening induced by the twins. The decrease of ultimate tensile strength is caused by the formation of continuous diffusion layer at the bonding interface. The layer consisting of intermetallic compound (a brittle phase of Al_3Mg_2) has poor plasticity, is easily crushed and fractured.

4. CONCLUSIONS

(1) Al/Mg–8Li–3Al–1Zn/Al composite plates were successfully fabricated by hot rolling with 50% reduction at 673 K. No cracks and flaws exist at the bonding interface.

(2) When the temperature exceeds 573 K, phase transition reaction occurs at the interface to form a transition layer, the main phases are Al_3Mg_2, $Al_{12}Mg_{17}$, AlLi, and $MgLiAl_2$.

(3) The bonding strength of an Al/Mg–8Li–3Al–1Zn/Al composite plate increases with the increase of annealing temperature.

(4) When the annealing temperature is lower than 573 K, the elongation increases with the increasing of annealing temperature, while the ultimate tensile strength keeps stable. When the annealing temperature is larger than 623 K, the elongation increases and the ultimate tensile strength decreases seriously with the increasing of annealing temperature.

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