Enhancement of mechanical properties for Mg-9Li-1Zn alloy by accumulative roll bonding

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Abstract
The microstructures and mechanical properties of LZ91 alloys by accumulative roll bonding (ARB) were firstly investigated. The results show that the cycle numbers of ARB process and preheating temperatures before ARB process have an impact on the microstructures and mechanical properties of LZ91 alloys. The microstructures of LZ91 sheets after ARB perform two main phases of $\alpha$ and $\beta$ in the fibrous form. ARB process can drive more numbers of crystallographic planes for $\alpha$ and $\beta$ phases in LZ91 alloys to proof the grain refinement after ARB. More cycle numbers of ARB on LZ91 sheets makes the fibrous microstructures finer and increases the mechanical properties such as hardness and ultimate tensile strength due to the mechanism of strain hardening and grain refinement.

1. Introduction

LZ91 alloy, containing elements of 9 wt\% Li, 1 wt\% Zn, and Mg balance, is one of the magnesium alloys with a very low density of 1.48 g cm$^{-3}$ \cite{1, 2}. This ultra-light alloy is full of potential for applications in computer, electronics, automobile, military and aviation industries. The microstructure of LZ91 is dominated by two main phases: HCP Mg-rich $\alpha$ phase with poor formability and BCC Li-rich $\beta$ phase with better ductility. However, dual-phase LZ91 alloy still has low strength for applications, which stimulates researchers to enhance the mechanical properties of LZ91 alloys \cite{3, 4}.

In terms of magnesium alloy plates, normally melted in the inert argon gas and produced by rolling processes, their strengthening mechanism includes solid-solution, grain-refinement and secondary-phase precipitations to improve the mechanical properties of magnesium alloys. Al and Zn are the most common alloying elements for Mg-Li base alloys \cite{5, 6}. For the LZ91 plates, which are processed by rolling and heat treatments, their microstructure and mechanical behavior was investigated. Cold working presented hardening effect, and the hardness was proportional to the extent of cold working as well as the age hardening effect \cite{7}. The texture of $\alpha$-Mg and $\beta$-Li phases can be elongated along the rolling direction \cite{8}. However, the enhancement of mechanical properties by a traditional rolling process still has the limitations for LZ91 plates applied in a structure material \cite{9}.

Accumulative roll bonding (ARB), developed firstly by Prof. Saito’s team in Japan in 1998 \cite{10–12}, is a promising method to achieve ultrafine-grained metallic sheets or plates, which usually have high strength and moderate ductility. ARB normally has been performed at room temperature to have the 50\% reduction of thickness for each rolling pass. In order to enhance the interface bonding quality, ARB process has been also modified by elevating rolling temperatures. So far, ARB technique has been applied for many alloys such as the plain low-carbon steel \cite{13}, austenitic steel \cite{14}, pure nickel \cite{15}, $\alpha$-brass alloy \cite{16}, titanium alloy \cite{17} and many kinds of aluminum alloys: AA1235 \cite{18}, AA2024 \cite{19}, AA5083 \cite{20} and AA8006 \cite{21}. In 2016, R. Wu \textit{et al} firstly reported the fine-grained and high-strength Mg-8Li-3Al-1Zn (LAZ831) alloy by ARB. The 5-cycle ARB-processed sheet possesses the small grain size (about 3 um) and great mechanical properties (tensile strength 287 MPa, elongation 12.5\% and hardness 77.2 HV) \cite{22}. In 2017, Mg-3Al-1Z (AZ31) alloy with relatively low Al...
content is a suitable alloy for the sheets fabrication and also for ARB process. Anisotropy of mechanical and thermal properties of AZ31 sheets prepared by ARB technique was investigated [23]. They also proposed different deformation mechanisms of AZ31 by ARB operate during tensile deformation in the rolling and transversal directions in 2018 [24]. R. Wu’s group also proposed the deformation mechanism of Mg-5Li-1Al (LA51) alloy during the ARB process. Strain hardening and grain refinement dominate the strengthening of Mg-5Li-1Al alloy [25]. Recently, the effect of rolling temperature on AZ31 alloy with ARB had been investigated for microhardness and interface bonding strength [26].

ARB is an effective and low-cost technique to enhance the mechanical properties of Mg alloys. In this report, ARB technique applied on LZ91 alloy has been firstly investigated. One- or five-cycle ARB rolling passes with roller 135 °C were applied on LZ91 plates. Different pre-heating temperatures (150 °C or 400 °C) for soaking time of 45 s have been also discussed. The characterizations of their microstructures, mechanical properties of LZ91 alloys were conducted for different ARB parameters.

2. Materials and methods

Original LZ91 plates were received from the factory in the form of extruded sheets with a size of 315 mm long, 213 mm wide, and 5.2 mm thick. The chemical composition of our LZ91 alloy is the same as our previous paper in the reference [7]. The mechanical properties of original LZ91 have the hardness of 56.7 HV, ultimate tensile strength of 177.5 MPa, and elongation of 45.5%. The LZ91 plate was cut into 30 mm wide blank with the same length. The blank was heated at 150 °C for 60 s and conducted 50% reduction rolling for three times to get the final thickness of 0.65 mm. The rolling processes were performed by Two High Hot Rolling Mill Model of DBR100x130L (Daito Seisakusho Co., Ltd) with the rollers diameter of 100 mm, and the roller speed was adjusted at 2.15 m s\(^{-1}\). The rolled blank was cut into 100 mm-long pieces. Then, ARB process was performed for two layers pack for each rolling pass as the schematic diagram shown in figure 1. One surface of each piece was cleaned by a steel brush. Two cleaned surfaces were stacked and roped by copper wires before rolling. Then it was heated at the temperature 150 °C or 400 °C for 45 s in the air by an electric furnace and then provided 50% reduction rolling process with the roller temperature of 135 °C. A two-layer roll bonded plate was done for one cycle. Meanwhile, to get a 32-layer plate, the cycle of the sequential steps must be repeated for five times. In order to study the impact of ARB temperatures and rolling cycles on LZ91 alloys, the same procedures were conducted for two and 32 layers by one- and five-cycle rolling passes at the pre-heating temperatures of 150 and 400 °C, respectively.

After ARB process on LZ91 sheets, the microstructures and mechanical properties of LZ91 were analyzed. The microstructure examinations include optical metallographic test, scanning electron microscopy (SEM), and x-ray Diffraction (XRD). Before the optical metallographic test and SEM, the samples were prepared by grinding, polishing and etching treatments. The optical microscopy is Olympus BX 41M-LED, and SEM is Hitachi S-3400. Meanwhile, X’Pert PRO X-ray diffraction system was used to check the phases of LZ91. The mechanical properties of LZ91, including hardness and tensile strength, were observed by FM-310e Vickers.
microhardness tester and Shimadzu AG-1 100kN tensile tester, respectively. Indentations were performed on the polished surface of plate specimens. Gauge dimension of the tensile specimens, carried out along the rolling direction, is 10 mm in length, 6 mm in width and 0.65 mm in thickness.

3. Results and discussions

3.1. Microstructure analysis

Figure 2 shows the microstructures of two different conditions of LZ91 plates, named as received and fully annealed conditions before ARB process. Two main bonding strength of interface is the combined effect of radial pressure and tangential shear stress to improve interfacial quality of LZ91 samples without delamination. During the ARB process, the magnification of the tensile specimens, carried out along the rolling direction, is 10 mm in length, 6 mm in width and 0.65 mm in thickness.

Figure 2. Optical micrographs of LZ91 before ARB process in longitudinal rolling and transverse directions for the conditions as received (a) and (a’) and fully annealed (b) and (b’).

After one-cycle or five-cycle ARB process at pre-heating temperatures of 150 °C or 400 °C, the microstructures of LZ91 alloys are performed by the optical micrographs in the magnifications of 50X in figure 3 to see the full image of four samples. Figures 3(a) and (a’) show images of the one-cycle ARB-processed sheet at 150°C along rolling and transverse directions, respectively. Figures 3(b) and (b’) are the images of the one-cycle ARB-processed sheet at 400°C. Figures 3(c) and (c’) perform the images of five-cycle ARB-processed sheet at 150 °C. Figures 3(d) and (d’) are the images of five-cycle ARB-processed sheet at 400 °C.

To observe more clearly the microstructure of LZ91, figure 4 shows the optical micrographs in the higher magnification as 500X for four different samples. We can find clearly fibrous microstructures of LZ91 sheets and good interface bonding quality. The center of the specimens is the roll bonding interface for one-cycle ARB. Those interfaces after ARB are not clear, which means the well bonding for LZ91 alloys after ARB process. The soaking time of 45 s preheating treatment at 150 °C or 400 °C, and ARB with the roller at 135 °C could also maintain the interfaces bonding quality of LZ91 samples without delamination. During the ARB process, the bonding of interface is the combined effect of radial pressure and tangential shear stress to improve interfacial bonding strength [27]. Moreover, bright area (α phase) is not easy to find for five-cycle ARB process sheet with pre-heated at 150 °C in figures 3(c) and (c’), which shows the finer grain and homogeneous structure for this samples.

Besides the observations of optical micrographs on LZ91 after ARB process with four different parameters, figure 5 shows the SEM images with higher magnification for all samples in the rolling direction, respectively. The fibrous microstructure consists of α phase in brighter area and β phase in darker area.

For one-cycle or five-cycle ARB-processed sheets with preheating temperature 150 °C in figures 5(a) and (c), the fibrous microstructure of five-cycle ARB looks finer than the one of one-cycle ARB. The grain of α-phase was still continuous along the rolling direction as shown in figure 5(a). After five-cycle ARB process (much higher plastic deformation), the continuous α-phase’s morphology changed to become thinner and discontinuous grain as shown in figure 5(c). Similarly, the ARB-processed sheets with preheating at 400 °C, the microstructures...
are shown in figures 5(b) and (d). Some fine and discontinuous grains are also observed in figure 5(d). The same results as the ARB process on other magnesium alloys [22, 23, 25], more cycle numbers of ARB can make more grain refinement for LZ91 alloys.

For the effect of preheating temperature on the microstructure, the fibrous microstructure of Mg-rich α phase was thinner for higher preheating temperature in comparison with one-cycle ARB-processed samples preheated at 150 °C in figure 5(a) and at 400 °C in figure 5(b). For five-cycle ARB-process samples preheated at 150 °C in figure 5(c) and 400 °C in figure 5(d), fibrous microstructure of LZ91 alloy became not clear, and grain of α phase became blunt for the high preheating temperature. For grain refinement of magnesium alloys, elevating temperature is necessary for ARB because of the low plasticity of hexagonal magnesium alloys at lower temperature [23].

3.2. XRD analysis

Figure 6 shows the XRD patterns for six different conditions: fully annealed (FA), original LZ91 plate (as received), one-cycle ARB at preheating temperature of 400 °C (1xarb400), five-cycle ARB at preheating temperature of 400 °C (5xarb400), one-cycle ARB at preheating temperature of 150 °C (1xarb150) and five-cycle ARB at preheating temperature of 150 °C (5xarb150). All the patterns show that LZ91 specimens contain two main α and β phases. The fully annealed condition shows five planes for α phase: (100), (002), (101), (110), and (103) as well as two planes for β phase: (110) and (211). The original plate appears three more planes (102), (112), and (004) for α phase, as well as one more plane (220) for β phase. After ARB process, there are 11 peaks for α phase, and each peak indicates the certain planes (100), (002), (101), (102), (110), (103), (200), (112), (201),
Another phase, $\beta$, has 4 peaks including (110), (200), (211), and (220) planes. From the XRD results, we can find that the ARB process has an impact on the crystallographic orientation of LZ91 alloys, which is manifested in the number of peaks. More and more crystallographic orientation can be observed because of

**Figure 4.** Optical micrographs of LZ91 alloys along the longitudinal rolling and transverse directions: (a) and (a') one-cycle ARB pre-heated at 150 °C; (b) and (b') one-cycle ARB pre-heated at 400 °C; (c) and (c') five-cycle ARB pre-heated at 150 °C; (d) and (d') five-cycle ARB pre-heated at 400 °C.

**Figure 5.** SEM images of LZ91 alloys along the longitudinal rolling direction: (a) one-cycle ARB pre-heated at 150 °C; (b) one-cycle ARB pre-heated at 400 °C; (c) five-cycle ARB pre-heated at 150 °C; (d) five-cycle ARB pre-heated at 400 °C.
the grain refinement effect for LZ91 sheets by ARB. Similarly, R. Wu’s group observed the grain of ARB-processed LA51 refined by selected area electron diffraction as the number of diffraction spots increases [25].

3.3. Mechanical properties

Figure 7 shows the Vickers hardness measurements of fully annealed, as received, and four ARB-processed sheets. ARB-processed sheets have higher hardness rather than full annealed and as received plates. The hardness of LZ91 sheets increases as the number of ARB process for both preheating temperatures. For the samples with preheating temperature 150 °C, Vickers hardness number (VHN) is 66.2 for one-cycle ARB. VHN becomes 73.8 for five-cycle ARB. Whereas, the samples preheated at 400 °C have VHN of 58.3 for one-cycle ARB and 67.1 for five-cycle ARB. The hardness of ARB-processed plates with preheating at 400 °C is lower than the one with preheating at 150 °C.

For the tensile test, the Engineering stress-strain curve is shown in figure 8. It also performs the values of ultimate tensile strength (UTS) and elongation for all samples. The tensile strength of LZ91 was enhanced by ARB process and the number of cycles. For the samples preheated at 150°C, the UTS values are 235.4 MPa for one-cycle ARB and 290.2 MPa for five-cycle ARB. Meanwhile, the samples preheated at 400 °C, the UTS values are 221.5 MPa and 246.5 MPa for one-cycle and five-cycle ARB, respectively. On the contrary, the elongation of LZ91 declines as the rising cycle numbers of ARB. The elongation is 22.8% for one-cycle ARB and 15.7% for five-
cycle ARB for preheating temperature 150 °C. Furthermore, we can find the preheating treatment at 400 °C improve the elongations of ARB-processed LZ91 sheets.

According to the results, the influence of ARB parameters on mechanical properties of LZ91 is discussed as following:

3.3.1. Effect of ARB cycle number
The mechanical properties of LZ91 such as hardness and UTS had been enhanced by more cycle number of ARB process. Such mechanical properties improvement on LZ91 are typical of the strain (work) hardening/strengthening effects due to plastic deformation of this alloy leading to dislocation multiplication in each grain [28, 29]. Microscopically, the finer fibrous structures shown in figures 5(c) and (d) are the evidence of higher plastic deformation and grain refinement. XRD measurement is also a simple way to indirectly verify the grain refinement of LZ91. According to the Hall-Petch relationship, the finer grain size gets the stronger strength of magnesium alloys [30].

3.3.2. Effect of pre-heating temperature:
For the samples with the same ARB cycles, the hardness and UTS of ARB LZ91 samples preheated at 400 °C are lower than those at 150 °C. The pre-heating temperature of 400 °C is higher than the recovery and recrystallization temperature of magnesium alloys [9]. The dislocations interaction from plastic deformation could be eliminated by the pre-heating treatment at such temperature, so the hardness and UTS decrease in the cases. On the contrary, the ductility of LZ91 increases after ARB process with preheating temperature 400 °C. The same effect of rolling temperature on AZ31 sheets with ARB also observed [26].

4. Conclusions
The mechanical properties LZ91 alloys had been improved by the ARB technique due to the strain hardening and grain refinement effects. Vickers hardness and UTS value can reach 73.8 and 290.2 MPa, respectively, for five-cycle ARB-processed sheet at preheating temperature 150 °C. The microstructure of LZ91 with different ARB parameters (cycle numbers and preheating temperatures) was also investigated in the work. More ARB cycle provided more plastic deformation to make the fibrous microstructure finer and increase the mechanical properties. Preheating treatment at the temperatures 150 °C or 400 °C before ARB can both improve the interface bonding performance of LZ91. The ARB process with preheating at higher temperature can also improve the ductility of LZ91 alloys. However, the hardness and strength decrease because of the recovery and recrystallization for the preheating temperature of 400 °C. Moreover, the plastic deformation of LZ91 alloys by ARB technique can drive more number of crystallographic planes of α and β phases, which is a simple way to proof the grain refinement effect.
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References

[1] Chiu C H, Wang J Y and Wu H Y 2006 Mater. Trans. 47 966–70
[2] Wu R Z, Qu Z K and Zhang M L 2010 Rev. Adv. Mater. Sci. 24 35–43 http://phys.ipme.ru/wu.pdf
[3] Kumar V, Govind, Shekhar R, Balasubramaniam R and Balani K 2012 Mater. Sci. Eng. A 547 38–50
[4] Song J M, Wen T Y and Wang J Y 2007 Scripta Mater. 56 529–32
[5] Zou Y, Zhang L, Li Y, Wang H, Liu J, Liaw P K, Bei H and Zhang Z 2018 J. Alloys Compd. 735 2625–33
[6] Takada H, Kikuchi S, Tsukada T, Kubota K and Hatta N 1999 Mater. Sci. Eng. A 271 251–6
[7] Chiu C H, Wu H Y, Wang J Y and Lee S 2008 J. Alloys Compd. 460 246–52
[8] Liu W H, Liu X, Tang C P, Yao W, Xiao Y and Liu X H 2018 J. Magnes. Alloy 6 77–82
[9] Yuan Y, Guo Q, Sun J, Lin H, Xu Q, Wu Y, Song D, Jiang J and Ma A 2019 Metals 9 386
[10] Saito Y, Tsuji N, Usunomiya H, Sakai T and Hong R G 1998 Scripta Mater. 39 1221–7
[11] Saito Y, Usunomiya H, Tsuji N and Sakai T 1999 Acta Mater. 47 579–85
[12] Tsuji N, Saito Y, Usunomiya H and Tanigawa S 1999 Scripta Mater. 40 795–800
[13] Kubina T and Gubis J 2015 Mater. Technol. 49 521–5
[14] Jafarian H R, Mousavi Anijdan S H, Eivani A R and Park N 2017 Mater. Sci. Eng. A 703 196–204
[15] Zhang Y B, Mishin O V, Kamikawa N, Godfrey A, Liu W and Liu Q 2013 Mater. Sci. Eng. A 576 160–6
[16] Bohme M and Wagner M F-X 2018 Scripta Mater. 154 172–5
[17] Cojocaru V D, Raducanu D, Gordin D M and Cinca I 2013 J. Alloys Compd. 546 260–9
[18] Yu H L, Lu C, Tieu A K and Kong C 2014 Mater. Manuf. Process. 29 448–53
[19] Nasiri M, Reihanian M and Borhani E 2016 Mater. Sci. Eng. A 656 12–20
[20] Sheikh H and Payne BD 2011 O. J. Metal. 1 112–5
[21] Homola P, Slálová M, Čežek I and Procházka I 2006 Mater. Sci. Forum 503–504 281–6
[22] Wang T Z, Zheng H P, Wu R Z, Yang J L, Ma X D and Zhang M I 2016 Adv. Eng. Mater. 18 304–31
[23] Halmosova K, Trojanova Z, Dzugan J, Drozd Z, Minarik P and Knapek M 2017 IOP Conference Series: Materials Science and Engineering 219 012023
[24] Trojanova Z, Dzugan J, Halmosova K, Nemeth G, Minarik P and Lukac P 2018 Acta Phys. Pol. A 134 863–6
[25] Hou L, Wang T, Wu R, Zhang J, Zhang M, Dong A, Sun B, Betsolens S and Krit B 2018 J. Mater. Sci. Technol. 34 317–23
[26] Rao X X, Wu Y P, Pei X B, Jing Y H, Luo L, Liu Y and Ju J 2019 Mater. Sci. Eng. A 754 112–20
[27] Alizadeh M and Pavidar M H 2009 Mater. Des. 30 82–6
[28] Pérez-Prado MT, del Valle J A and Ruano O A 2005 Mater. Lett. 59 3299–303
[29] Ghalibbandi S M, Malaki M and Gupta M 2019 Appl. Sci. 9 3627
[30] Kim W J, Kim M J and Wang J Y 2009 Mater. Sci. Eng. A 516 17–22