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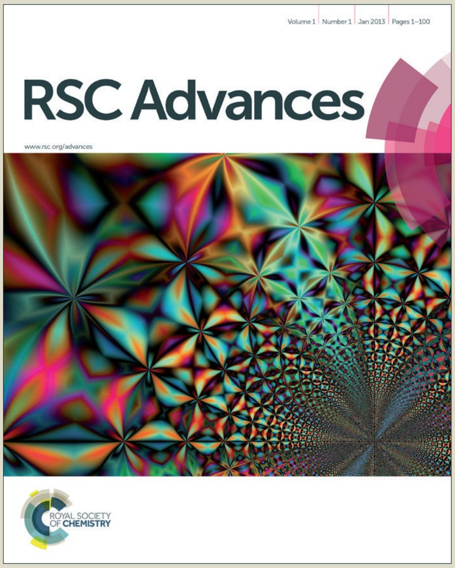

Journal Name

ARTICLE

Effect of thickness of interfacial intermetallic compound layers on the interfacial bond strength and the uniaxial tensile behaviour of 5052 Al/AZ31B Mg/5052 Al clad sheets

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Jian-jun Zhang,[ab] Wei Liang*[a] and Hai-tao Li[bc]

The thickness of intermetallic compound (IMC) layers at interface would significantly influence the interfacial bo strength, and the interfacial bond strength would further affect the tensile behavior of 5052 Al/AZ31B Mg/5052 Al tri-laminate structural clad sheets fabricated by hot rolling. In this manuscript, the relations among the thickness of IMC layers produced by post-roll annealing, the interfacial bond strength and the tensile behavior of clad sheets were investigated. No reactive diffusion phases were observed in the as-rolled clad sheets and in the rolled clad sheets annealec at 473K for 1h. When annealing temperature at 573K for 0.5 h, 1 h, 2 h, 4 h, 8 h, new reaction diffuse phase layers with various thickness are formed at interface. Two types of reaction layers, viz., Al_3Mg_2 and $Al_{12}Mg_{17}$,adjacent to the 5052 A side and AZ31B Mg side, respectively, are identified by EDS analysis. The effects of thickness of IMC layers on the norma bond strength and the shear bond strength were investigated. Uniaxial tensile tests of the clad sheets with and without IMC layers were investigated to reveal the relationships between the tensile behavior and the bond strength. Meanwhile, the fractured process of IMC layers and the delaminated process of tri-laminate composite sheets were also discussed during the uniaxial tensile testing.

1. Introduction

Up to this point, our world faces the main global environmental challenge of a changing climate. Lightweight design of materials and structures is being regarded as a key tactic for improving climate and lowering greenhouse gases emissions.[1-4] Magnesium alloys are being increasingly used in the automotive and aerospace industries due to their weight loss and high specific strength.[1] However, their poor corrosion resistance, poor formability, and high production cost severely hinder their wider application in industries.[5] One of the major weakness of magnesium alloys in many applications is their poor surface corrosion resistance. Over the years, various surface treatment techniques have been developed and applied to protect Mg alloys against the environmental corrosion. An important and effective way of overcoming the flaws is to clad magnesium alloys with aluminium alloys, which have excellent corrosion resistance and high specific strength, by different fabricated technology.

Various fabricated techniques of laminated metal composites composed of similar or dissimilar components have been developed. Such as hot pressing,[6, 7] diffusion bonding,[8-11] hot roll bonding,[12-15] cold roll bonding,[16-19] warm roll bonding,[20, 21] explosive welding,[22] accumulative roll bonding,[23] laser welding,[24] twin-roll casting,[25] ultrasoni welding,[26] Ultrasonic spot welding,[27] friction stir welding,[28, 29] equal channel angular extrusion.[30] Among all the mentioned solid-state joining techniques, the roll bonding is the mos economical and productive manufacturing process and has been widely used.

The mechanical behaviours of 5052 Al/AZ31B Mg/5052 A are mainly governed by the interfacial microstructure and interfacial bond strength. Different interfacial microstructure including a certain thickness of IMC layers were formed along the Al/Mg interface by different post-annealing temperature and different post-annealing time. In this study, the interfacial microstructures, the interfacial bond strength and the uniaxial tensile behaviours of tri-laminate 5052 Al/AZ31B Mg/5052 A were investigated to elucidate the relations among them.

2. Experiments procedures

2.1. Raw materials

Commercial AZ31B Mg alloy plates and 5052 Al alloy sheet were used as substrate and cladding, respectively. Dimensio of AZ31B plates were cut parallel to rolling direction of a

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[a] College of Materials Science and Engineering, Taiyuan University of Technology, Taiyuan 030024, China.
[b] School of Science, North University of China, Taiyuan 030051, China.
[c] Institute of Mining Technology, Taiyuan University of Technology, Taiyuan 030024, China.
† Corresponding author. Tel.: +86 351 6018398; fax: +86 351 6018398.
E-mail addresses: liangwei@tyut.edu.cn, zlsyspaper@126.com (W. Liang).

received commercial AZ31B Mg plate to 150 mm × 60 mm × 2.8 mm, and the 5052 Al sheets were also cut to rolling direction of as-received commercial 5052 Al sheet to 320 mm × 70 mm × 2.8 mm. The chemical composition of the AZ31B Mg plate and 5052 Al sheet used in this research were listed in **Table 1**.

2.2. Fabrication of the 5052 Al/AZ31B Mg/5052 Al composite plates

All the pre-bonding surfaces of tri-laminate 5052 Al/AZ31B Mg/5052 Al clad sheets were degreased using acetone and mechanically ground by grit SiC papers to eliminate the contaminated surface and the oxidation film. And then the AZ31B Mg plate were placed in the folded 5052 Al in sequence of 5052 Al/AZ31B Mg/5052 Al, the seven same stacks were heated at 673 K for 15 minutes in furnace, and then, hot rolled to 2.16 mm in 40% reduction. And then, each prepared Al/Mg/Al laminated metallic composite was performed different post-annealed process. In this paper, experimental mill with twin-roller in 130 mm diameter and 260 mm width was used, and the rolled speed was 0.0628 m/s (10 rpm/min).

2.3. Annealing treatments

To investigate the relations among the different thickness of IMC layers at interface caused by post-annealing, the interfacial bond strength and the uniaxial tensile behaviours of tri-laminate 5052 Al/AZ31B Mg/5052 Al. for the prepared s the hot rolling process. To obtain different interfacial microstructures with and without reactive diffusion phases, different annealing treatment was conducted. The annealing temperature and holding time were selected at 473K + 1 h and at 573K+0.5 h, 573K+1 h, 573K+2 h, 573K+4h and 573K+8 h, respectively.

2.4. Microstructure characterization

To examine the interfacial microstructure, the specimens cut parallel to the experimental rolling direction. The interfacial microstructures were examined by a MIRA 3 Field Emission Scanning Electron Microscope (SEM) equipped with an energy dispersive X-ray spectrometer (EDS).

Table 1 Specifications of the commercial Al 5052 and AZ31B Mg sheets used in this research.

Material	Chemical composition (at%)			
5052 Al	2.45 Mg	96.33 Al	0.25 Si	0.11 Zn
AZ31B Mg	95.24 Mg	3.20 Al	0.12 Si	0.92 Zn

2.5. Mechanical properties tests

2.5.1 Bond strength

Enough interfacial bond strength is needed for almost all of the laminates and cladding plates used as structural materials because it strongly affects the overall mechanical behaviour of these composite materials. In general, diffusion necking would occurred in plastic stage and localized necking would arise in fracture stage during the uniaxial tensile process.[32] For the laminated composites or the clad sheets composed by different materials, which is of different mechanical properties. The different level of deformation of component layer in thickness direction and in width direction would cause interfacial separated trend in normal direction and in tangential direction. Hence, the normal bond strength and shear bond strength are starkly different. In order to investigate the effect of different interfacial bond strength on the uniaxial tensile behaviours, two types of testing methods were performed to measure the interfacial normal bond strength and the interfacial shear bond strength. To test the normal bond strength, the 10 mm diameter wafers was cut from the prepared composite laminates, then, four-point bending with adhesive butt joint was conducted according to our previous work.[30] Shear test was conducted by the lap joint with 10 mm length and 10 mm width.

2.5.2 Uniaxial tensile

Uniaxial tensile test is a fundamental and important measurement for obtaining the engineering stress-strain curves of laminated metallic composites. From the stress-strain curves can get yield strength, ultimate strength, and elongation. To obtain the above mentioned curves of Al/Mg/Al annealed at different condition, dog-bone specimens according to ASTM-E8M sized with a parallel length of 60 mm (gauge length 50 mm) and width of 12.5mm were cut parallel to the rolling direction from the prepared clad sheets. Uniaxial tensile tests were conducted at room temperature using a CMT5205 electronic universal testing machine under a quasi-static strain rate $1\times10^{-4}s^{-1}$. An extensometer with a gauge length of 50 mm was used to measure the strain.

To verify the reproducibility and the validity, three specimens in each condition were tested for uniaxial tensile test. For each bond strength test, five repeatable specimens were used in this paper.

3. Results and discussion

3.1. Evolution of interface microstructures

Fig. 1 shows SEM images of interfacial microstructures of cross-section of 5052 Al/AZ31B Mg/5052 Al clad sheets along the rolled direction in the conditions of as-rolled and different annealed process. No new phase layers were being observed at the interface for the specimens of as-rolled and annealed at 473K for 1h as shown in **Fig. 1(a, b)**. Especially when the annealed temperature at or under 473K, regardless of how long annealed time was increased, no new phase layer formed at the interface according to our previous research.[12] When raised the annealed temperature from 473K to 573K and changed the annealed time from 0.5 h to 8 h resulted in different thickness of IMC layers at the interface as shown in **Fig. 1(c-g)**.

Fig. 2 shows the thickness of IMC layers change with the annealed time when annealed at 573K. From which can be seen that the formation of IMC layers decelerated with increasing the annealed time. Note that annealing temperature at 473K for 1 h was not plotted in this figure because of similar microstructure compared to as-rolled specimen. Obviously, there are two different phase IMC layers when annealed at 573K for 0.5 h, 1 h, 2 h, 4 h and 8 h, the each layer thickness and total thickness of IMC were measured and plotted in **Fig. 2.** EDS line scan and point scan were performed to identify these layers, as shown in **Fig.3**. The compositions in the interest region A and B were analyzed by EDS point scan as shown in **Table 2**. It suggests that the layer adjacent to 5052 Al side is Al_3Mg_2 and that close to AZ31B Mg side is $Mg_{17}Al_{12}$ according to Al-Mg binary phase diagram.[33] The divisional morphology can be clearly distinguish from the typical EDS line scan across a interface of 5052/AZ31B of a sample annealed at 573K for 8 h as shown in **Fig. 3**. From which can be seen clearly that the thickness of Al_3Mg_2 is about 10 times over than that of $Mg_{17}Al_{12}$. The diffusion constant and activation energy for the formation of Al_3Mg_2 are 1.1×10^{-6} m^2/s and 85.5 kJ/mol, respectively, and those for $Mg_{17}Al_{12}$ are 0.18 m^2/s and 165 kJ/mol,[34] respectively. Comparison with $Mg_{17}Al_{12}$, Al_3Mg_2 has more fast diffusion rate and more low activation energy that is responsible for the difference of thickness of two intermetallic compound layers.

3.2. Normal bond strength and shear bond strength

Fig. 4 shows the normal bond strength and shear bond strength varying with the thickness of IMC layers of tri-laminate clad sheets 5052 Al/AZ31B Mg/5052. The normal bond strength revealed the ability of resisting interfacial separation in the normal direction; the shear bond strength demonstrated that the capable of resisting interfacial slip in the tangential direction. The values of normal bond strength increased from 15.37MPa to 20.73MPa with IMC layers thickness increasing from 0μm to 13μm, and decreased from 20.73MPa to 15.7MPa with IMC layers thickness increasing from 13μm to 24μm. From the change trend of normal bond strength and shear bond strength, it can be seen that there exists a suitable thickness of IMC layers. This can obtain greatest interfacial normal bond strength when the thickness of IMC layers is 13μm. However, the situation is somewhat difference for the relationship between shear bond strength and thickness of IMC layers compared to the relationship between normal bond strength and thickness of IMC layers. From **Fig. 4**, it can be seen that the shear bond strength reached the maximum value 55.05MPa at 4.2μm thickness.

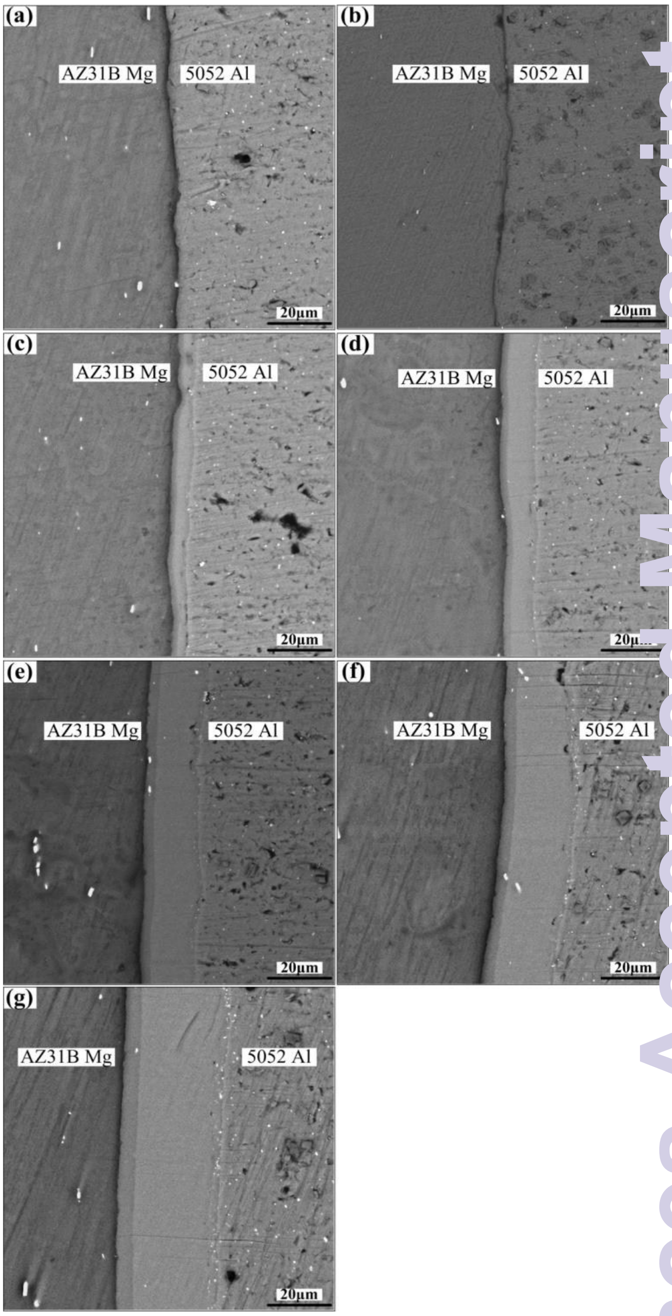

Fig. 1 Typical SEM images of interfacial microstructure of 5052 Al/AZ31B Mg/5052 tri-layer clad sheets of (a) as-rolled, (b) 473K+1 h, (c) 573K+0.5 h, (d) 573K+1 h, (e) 573K+2 h, (f) 573K+4 h and (g) 573K+8 h.

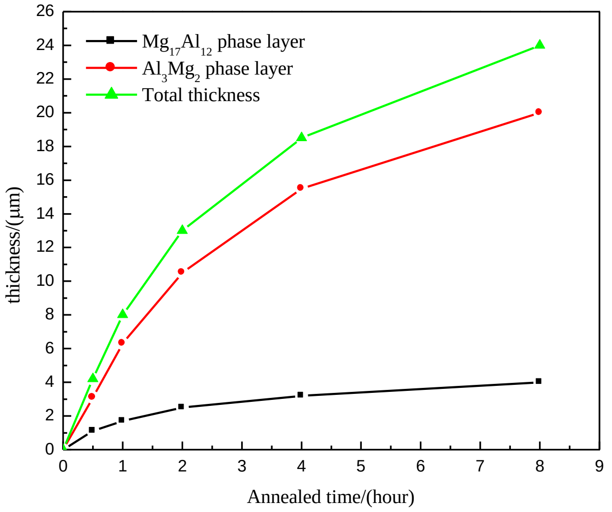

Fig. 2 Effect of annealing time on the thickness of individual layer thickness and total thickness at anneal temperature 573K and annealed time from 0.5 h to 8 h.

Table 2 Chemical compositions (at%) of interest region determined by EDS point scan.

Interest region	Mg	Al
A	41.2	58.6
B	55.6	44.1

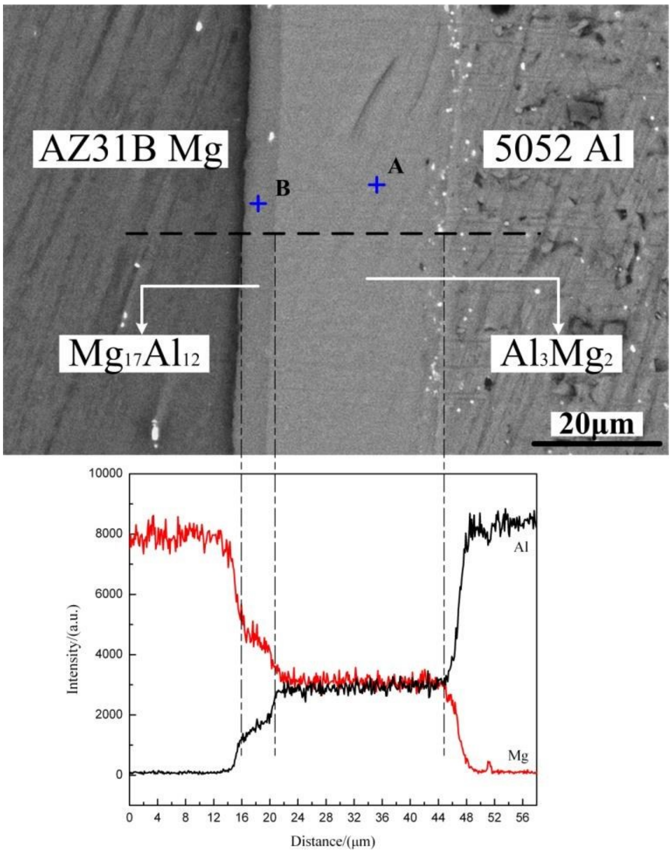

Fig. 3 Interface microstructure of tri-layer clad sheet 5052 Al/AZ31B Mg/5052 annealed at 573K for 8 h and EDS line scan analysis by the dashed line.

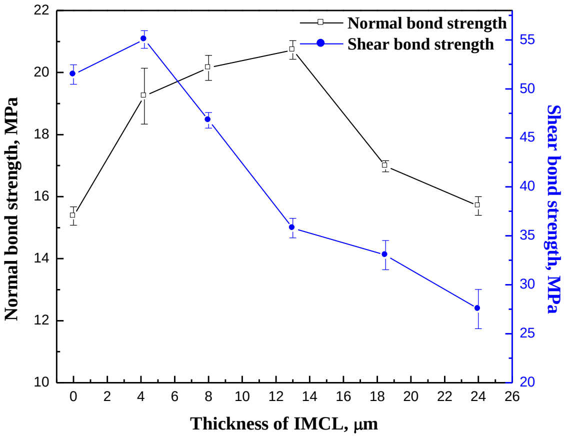

Fig. 4 Interface normal bond strength and shear bond strength of tri-laminate clad sheets 5052 Al/AZ31B Mg/5052 with different thickness of IMCLs.

After that, the magnitude of shear bond strength decreased fast with increasing the IMC layers thickness. From the above comprehensive assessment, we can conclude that the IMC layers thickness of 4.2μm is an optimal thickness for overall bond strength because the shear stress is major factor in delamination during tensile test. The further reason will be discussed at the section of uniaxial tensile behaviours of tri-laminate clad sheets.

3.3. Uniaxial tensile behaviour

A series of uniaxial tensile tests were conducted on the specimens fabricated by the same hot-rolling process and the different post-hot-rolling procedure. Typical engineering stress-strain curves were plotted and are shown in **Fig. 5**. And the yield strength, ultimate strength and elongation of each specimen were all summarized in **Fig. 6**. With the increasing thickness of IMC layers, the elastic modulus has almost no change according to rule of mixtures due to the thick fraction

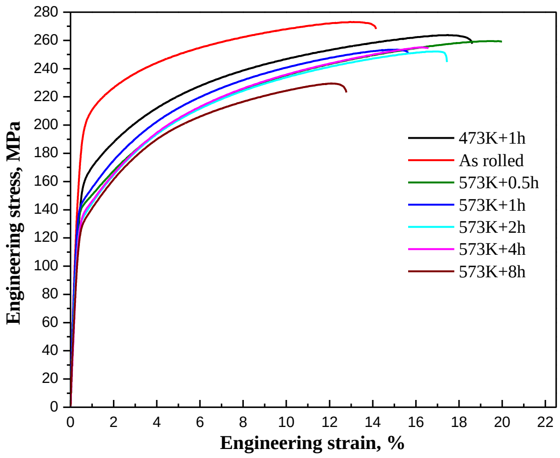

Fig. 5 Engineering stress-strain curves of tri-laminate clad sheets 5052 Al/AZ31B Mg/5052 under different post-annealed conditions.

of IMCL layers in total thickness is ~1%. Despite the as-rolled specimen and the 473K+1h specimen have similar interfacial microstructure, there is a significant difference in stress-strain behaviour due to the recovery, recrystallization and atom diffusion at interface caused by post-annealed. For the series specimens of annealed at 573K with different annealed time, the varying thickness of IMC layers started to form at interface, the magnitude of thickness were plotted in **Fig. 2.** The 573K+0.5 h specimen with 4.2μm total thickness of ICM layers has the maximum elongation, the moderate yield strength and ultimate strength among the annealed at 573K series specimens. From the engineering stress-strain curve of as-rolled specimen, it can be seen that the yield strength and the ultimate strength reached the maximum value among all the tested specimens. The elongation is, however, relative low due to the work hardening effect produced by hot rolled. Although the interfacial microstructure of 473K+1h specimen is similar to the as-rolled sample shown in **Fig. 1(a)** and **(b)**, slight atom-to-atom diffusion but no reactive diffusion phases occurred at the interface due to the post-annealed procedure. Thus, more excellent interfacial bonded property of 473K+1h specimen was obtained than that of as-rolled specimen. Other clear evidences are the increased interfacial bond strengths shown in **Fig. 4**, the normal bond strength and the shear bond strength of 473K+1h specimen are all higher than those of as-rolled specimens. Post-annealed process result in decreasing of yield strength and ultimate strength due to disappearing of work hardening effect, but the higher interfacial bond strengths guaranteed the larger elongation due to that interfacial delamination would not occurred during the larger amount of plastic strain. With the increasing of IMC layers thickness for specimens of annealed temperature at 573K for 1 h, 2 h, 4 h and 8 h. The mechanical behaviour of those tri-laminate clad sheets deteriorated because the delamination may be occurred at elastic stage or plastic flowing stage (containing nominal yield stage and strain hardening stage) in different tensile stages.

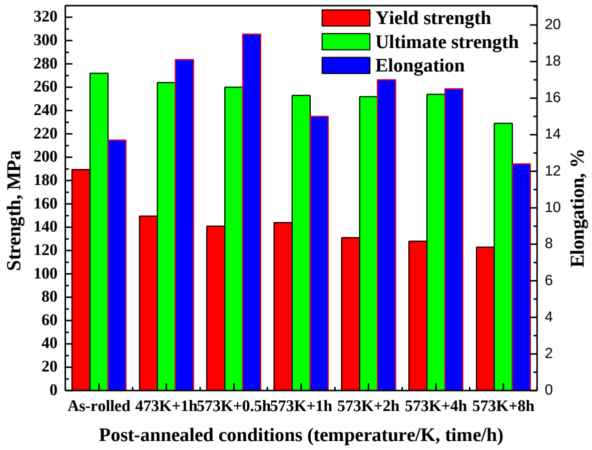

Fig. 6 Effects of annealed temperature and annealed time on the yield strength, ultimate strength and elongation of tri-laminate clad sheets.

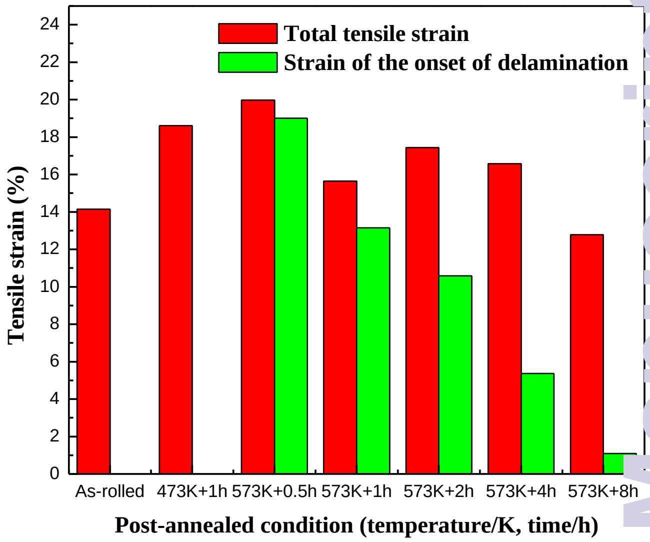

Fig. 7 Total tensile strain and strain of the onset of delamination of tri-laminate clad sheets under different post-annealed temperature and post-annealed time.

Fig. 7 shows macro-debonded strain by the in situ observatio For the as-rolled tensile specimens and 473 K+1 h specimens no delamination was observed before fracture. For the 573 K+0.5 h specimens, the delamination was occurred just before fracture, thus, a thin IMC layers (4.2 μm) would not deteriorate the interfacial bond properties. On the contrary, i would enhance effectively the comprehensive mechanical behaviours, such as elongation. This will be help in forming o clad sheets. For the specimens of 573 K+1 h, 2 h, 4 h and 8 h, the comprehensive mechanical properties, especia elongation, significantly became worse due to the thicken IMC layers, and the delaminated strain of the macro-crack observed by in situ is dramatically linearly decreased. **Fig.** shows the fracture profile of the tri- laminate clad sheets with

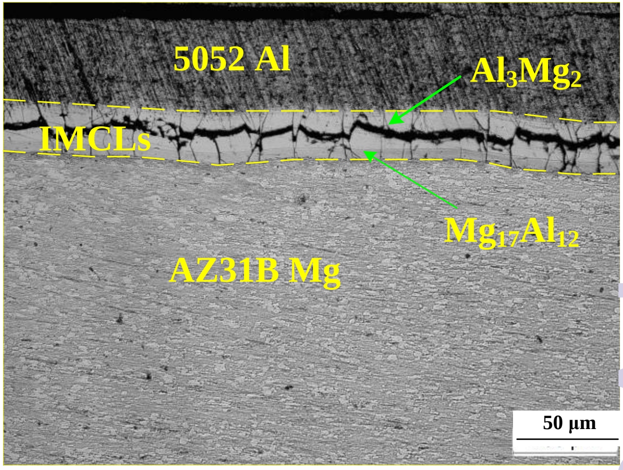

Fig. 8 Optical micrographs of fracture profile of IMCLs.

24µm thick IMC layers. From which can be conclude that the transverse crack will firstly occurred in IMC layers when the uniaxial tensile strength reached that of IMC layers , and then the longitudinal penetrative cracks were produced due to the interfacial shear stress by the mismatch mechanical properties of the clad 5052Al and AZ31B Mg.

4. Conclusions

This manuscript correlates interfacial IMC layers thickness-interfacial bond strength-uniaxial tensile behaviours of tri-laminate clad sheet 5052Al/AZ31B Mg/5052 Al, which fabricated by hot rolling process and with subsequent annealed at different temperature and different time. The following conclusions can be drawn:

(1) Tri-laminate sheets of 5052 Al/AZ31B Mg/5052 Al were successfully manufactured by hot rolling with excellent bond interface.

(2) The as-rolled and 473 K+1 h specimens, no IMC layers were observed to form at the interface between 5052 Al and AZ31B Mg. When the annealed temperature reached 573 K and varied the annealed time from 0.5 h to 8 h, different thickness of IMC layers were observed to form at the interface. The IMC layers were identified to be Al_3Mg_2 adjacent to 5052 Al side and $Mg_{17}Al_{12}$ adjacent to AZ31B Mg side.

(3) The interface bond strength varied with the change of different thickness of IMC layers. The interfacial normal bond strength reached maximum value 20.73MPa when the IMC layers thickness is 14µm, the interfacial shear bond strength reached maximum value 55.05MPa when the IMC layers thickness is 4µm, the shear bond strength dominate in bond strength during the tensile test.

(4) A certain thin IMC layers is contributed to enhance the comprehensive mechanical properties.

(5) The IMC layers fractured in transverse crack first than produced longitudinal crack along the tensile direction due to the mechanical properties mismatch of 5052Al and AZ31B Mg.

Acknowledgments

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References

1. T. M. Pollock, Science, 2010, 328, 986-987.
2. S. J. Davis, K. Caldeira and H. D. Matthews, Science, 2010, 329, 1330-1333.
3. M. McNutt, Science, 2013, 341, 435-435.
4. C. Ash, E. Culotta, J. Fahrenkamp-Uppenbrink, D. Malakoff, J. Smith, A. Sugden and S. Vignieri, Science, 2013, 341, 472-473.
5. B. Zhu, W. Liang and X. Li, Materials Science and Engineering: A, 2011, 528, 6584-6588.
6. X. Li, W. Liang, X. Zhao, Y. Zhang, X. Fu and F. Liu, Journal of Alloys and Compounds, 2009, 471, 408-411.
7. L. M. Zhao and Z. D. Zhang, Scripta Materialia, 2008, 58, 283-286.
8. M. H. M. Kouters, G. H. M. Gubbels and O. Dos Santos Ferreira, Microelectronics Reliability, 2013, 53, 1068-1075.
9. S. Kobayashi and T. Yakou, Materials science and engineering: A, 2002, 338, 44-53.
10. J. Shang, K. h. Wang, Q. Zhou, D. k. Zhang, J. Huang and J. q. Ge, Transactions of Nonferrous Metals Society of China, 2012, 22, 1961-1966.
11. A. Macwan, X. Q. Jiang, C. Li and D. L. Chen, Materials Science and Engineering: A, 2013, 587, 344-351.
12. C. Luo, W. Liang, Z. Chen, J. Zhang, C. Chi and F. Yan, Materials Characterization, 2013, 84, 34-40.
13. D. S. Zhao, J. C. Yan, Y. Wang and S. Q. Yang, Materials Science and Engineering: A, 2009, 499, 282-286.
14. X. P. Zhang, S. Castagne, T. H. Yang, C. F. Gu and J. T. Wang, Materials & Design, 2011, 32, 1152-1158.
15. G. P. Chaudhari and V. Acoff, Composites Science and Technology, 2009, 69, 1667-1675.
16. S. A. Hosseini, M. Hosseini and H. Danesh Manesh, Materials & Design, 2011, 32, 76-81.
17. I. K. Kim and S. I. Hong, Materials & Design, 2013, 49, 935-944.
18. S. Nambu, M. Michiuchi, J. Inoue and T. Koseki, Composites Science and Technology, 2009, 69, 1936-1941.
19. K. S. Lee, D. H. Yoon, H. K. Kim, Y. N. Kwon and Y. S. Lee, Materials Science and Engineering: A, 2012, 556, 319-330.
20. S.H. Kim, H. W. Kim, K. Euh, J. H. Kang and J. H. Cho, Materials & Design, 2012, 35, 290-295.
21. N. Zhang, W. Wang, X. Cao and J. Wu, Materials & Design, 2015, 65, 1100-1109.
22. M. M. Mahdavian, L. Ghalandari and M. Reihanian, Materials Science and Engineering: A, 2013, 579, 99-107.
23. M. Gao, S. Mei, X. Li and X. Zeng, Scripta Materialia, 2012, 67, 193-196.
24. J. H. Bae, A. K. Prasada Rao, K. H. Kim and N. J. Kim, Scripta Materialia, 2011, 64, 836-839.
25. L. Wang, Y. Wang, C. Q. Zhang, L. Xu, J. Robson and P. Prangnell, Materials Science Forum, 2014, 794-79[illegible], 416-421.
26. A. Macwan, V. K. Patel, X. Q. Jiang, C. Li, S. D. Bhole and D. L. Chen, Materials & Design, 2014, 62, 344-351.
27. M. Aonuma and K. Nakata, Materials Science and Engineering: B, 2010, 173, 135-138.

28. W. B. Lee, K. S. Bang and S. B. Jung, Journal of Alloys and Compounds, 2005, 390, 212-219.
29. X. B. Liu, R. S. Chen and E. H. Han, Journal of Materials Processing Technology, 2009, 209, 4675-4681.
30. J. Zhang, W. Liang, Y. Liu, X. Zhao, X. Li and B. Zhou, Materials Science and Engineering: A, 2014, 590, 314-317.
31. A. G. Magalhães, M. F. S. F. de Moura and J. P. M. Gonçalves, International Journal of Adhesion and Adhesives, 2005, 25, 313-319.
32. S. Okazawa, International Journal of Non-Linear Mechanics, 2010, 45, 35-41.
33. H. Baker, Materials Park, OH: ASM International, 1992. 1. 1, 1992.
34. S. Brennan, K. Bermudez, N. S. Kulkarni and Y. Sohn, Metallurgical and Materials Transactions A, 2012, 43, 4043-4052.