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Coating Design for Controlling β Phase IMC Formation in Dissimilar Al-Mg Metal Welding

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Abstract

The brittle intermetallic compound (IMC), β -Al₃Mg₂, grows very rapidly at the interface during dissimilar welding of Al to Mg. This currently prevents acceptable joints being produced between these two important light alloys. In the present study, the use of Al-alloy inter-layer coatings has been investigated to address this problem, for application to friction welding processes. Thermodynamic principles were employed to aid design of the coatings. Two types of coatings were selected: i) a Si-rich coating, which promoted the slower growing and more stable Mg₂Si phase; ii) an Al-Zn-rich solid solution coating, which replaced the undesired Al₃Mg₂ phase with a ternary (Al,Zn)₄₉Mg₃₂ intermetallic compound. In statically annealed diffusion couples both coatings were found to inhibit the growth of Al₃Mg₂. Refill friction stir spot welding (RFSSW) experiments also showed that both coatings were effective in reducing interfacial reaction and can improve the mechanical properties of Al - Mg dissimilar joints.

1. Introduction

Today's automobile industry is facing ever more demanding emissions targets together with consumer pressure for greater safety and performance [1]. The estimated effect of a reduction in vehicle mass on CO₂ emissions is ~ 4.7 g CO₂ per 100 kg's. When combined with the rising cost of fuel, this has aroused wide interest in replacing traditional car structural materials, like steel, with lighter metals, such as Al and Mg [2,3]. In addition, in the most efficient new vehicles designs are based on multi-material bodies, where the best attributes of different materials can be exploited in a single structure. This new direction of the automotive industry presents major challenges in joining dissimilar metals as well as in joining metals to composites.

Direct joining of aluminum to magnesium alloys has been widely reported to be very difficult, with virtually all welding methods producing poor weld properties that suffer from low toughness [4]. This is even true of lower temperature solid state joining techniques, like friction spot welding (FSW) [5]. The problem mainly lies in the rapid formation of several Al - Mg intermetallic compounds (IMCs) at elevated temperature, which can severely embrittle the joining interface. In particular, the β -Al₃Mg₂ phase is reported to have the highest growth rate among the common Al - Mg compounds [6,7] and is the phase in which cracks are most prone to initiate, owing to its low strength and highly brittle nature [8]. Therefore, the β -Al₃Mg₂ compound is the phase in the Al - Mg system that it is most desirable to avoid to improve joint performance. Previous studies have suggested that thin barrier coating inter-layers, such as PVD Mn, can be effective in controlling the Al-Mg interfacial reaction, but unfortunately such coatings are brittle themselves and can readily become damaged by friction welding processes [9]. Hence, better coatings are still required to improve the mechanical properties of Al - Mg dissimilar welds.

In this work, to address the intermetallic reaction problem, thermodynamic calculations have been used to select two ductile Al-alloy coatings; i) a Si-rich coating; and ii) an Al-Zn-rich solid solution coating. Both of these coatings were chosen as they are predicted to change the reaction path and avoid the formation of the β phase. Long-time annealing and refill friction stir spot welding (RFSSW) experiments were carried out in each case to study the intermetallic growth behavior in both static and welding conditions.

2. Experimental

An AZ31 Mg alloy and several different Al-alloy sheets were used in this research, the compositions of which are shown in Table I. All the sheets had a similar thickness of around 1 mm. To test the concept of a Si-rich coating, a commercial Al sheet was used, produced by the 'FusionTM' process, which had an AA6016 core with a 0.1 mm thick ~ 10% Si brazing alloy (AA4045) cladding layer. To investigate the concept of an Al-Zn-rich solid solution coating, sheets of the correct predicted composition were prepared by casting and hot rolling, and then homogenized at 460°C. The effect of the two coating materials on the growth behavior of the intermetallic compounds was compared by using isothermal heat treatments on diffusion couples made with the same AZ31 Mg alloy. The samples were produced by first lightly welding them together using a 2kW Sonobond ultrasonic welder, with a very short weld time of 0.2 sec. Owing to their different melting points, the couple produced between the Al-Si alloy and Mg sheet was heat treated at 400°C, whereas the Al-Zn alloy and Mg couple was annealed at 360°C, before for microstructure analysis. The Refil-Friction Stir Spot welding (RFSSW) technique was adopted to test the performance of the coating materials under previously optimized welding condition: rotation speed of 2500 rpm, welding duration 1 sec, with the AZ 31 Mg alloy placed on the top, and a plunge depth of XX mm. Baseline uncoated AA6111 - AZ31 samples were also prepared under the same conditions for comparison purposes.

Design of the Al-Si and Al-Zn coatings was aided by the CALPHAD software PANDAT, with the database PanAl2008, and the diffusion simulation package DICTRA. Imaging of the joint interfaces was carried out with an FEI Quanta 650 SEM. Phase identification of the intermetallics was performed by high resolution EBSD with a step size of 0.1 μm , and TEM micro-diffraction on sections removed from the interface by FIB milling. Lap shear tests were carried out on an Instron testing machine to characterize the joining performance of the friction welds.

Table I. Compositions of the Al and Mg alloys employed this work (wt. %)

	Si	Zn	Fe	Cu	Mn	Ti	Cr	Ni	Mg	Al
AA6016	1-1.5	0.2	0.5	0.2	0.2	0.15	0.1	-	0.25-0.6	Balance
AA4045	9.88	-	0.15	-	-	0.02	-	-	-	Balance
Al-Zn alloy	-	52	-	-	-	-	-	-	-	Balance
AA6111	0.63	-	0.05	0.75	0.05	0.06	0.06	-	0.75	Balance
AZ31	0.017	1.03	0.0045	0.0036	0.2	-	-	0.0003	Balance	2.51

3. Results

3.1 Interfacial Characterization of the Si-Rich Coating

The Mg_2Si phase has a low density, high hardness and has high stability in both Al and Mg and has previously been investigated as a candidate reinforcement for forming in-situ composites in both Al and Mg matrices [10,11]. Thermodynamic equilibrium calculations of the Al-Mg-Si system confirmed that the Mg_2Si should be preferably formed over any of the other possible Al-Mg IMC binary phases [12]. Therefore, a Si rich coating was predicted to be a strong candidate for changing the normal reaction path seen in Al - Mg dissimilar welds away from the undesirable rapidly-growing $\beta\text{-Al}_3\text{Mg}_2$ phase.

The typical microstructure of the AA4045 Al-Si clad layer on the Al sheet used in this study, prior to welding, is illustrated in Fig.1. Small globular silicon particles of around 2-4 μm in diameter can be seen to be distributed uniformly in the coating layer, which is about 100 μm thick. The cladding layer also contains some Fe rich primary particles.

Fig.1. Microstructure of the AA4045 clad layer on the as-received AA6016 aluminum ‘Fusion’TM, sheet product; (a) the whole layer, (b) Si particles at a higher magnification.

Fig.2 compares SEM images of the interface region in the Si rich coated Al sheet, after isothermal treatment at 400°C for 3hrs, when joined to the Mg sheet with that of the uncoated AA6111 Al – Mg baseline alloy combination. It can be seen that in the joint with the Si coating the overall reaction layer appears about two thirds as thick, (50 μm , compared to 75 μm) which indicates that the concept of introducing a Si rich coating has been successful. In addition, while the IMC reaction layer between the uncoated standard Al alloy and the Mg alloy is comprised of two continuous sub-layers of different compounds, the thinner reaction layer in the coated Al - Mg joint appears to be made up of a single phase on the Mg side of the joint, above a more complex irregular sub layer, containing second phase particles.

In the baseline couple the reaction behavior is similar to that already reported [13]. The two phases present were readily identified as a thinner layer of γ , which forms first in the reaction sequence on the Mg side of the joint, and β , which forms later, but grows more rapidly after nucleating on the Al side of the joint (see [13,14]). In comparison with the Si rich coating, EDX and TEM micro diffraction analysis indicated that the single phase layer adjacent to the substrate was also $\gamma\text{-Al}_{12}\text{Mg}_{17}$, but this was thicker than in the baseline couple and extended down into the region where second phase particles were present (Fig.2a). EDX mapping (Fig. 3) and TEM micro-diffraction analysis (Fig. 2c) showed that the second phase particles in the IMC layer that had grown into the Al-Si cladding were Mg_2Si particles that had formed by reaction between Mg diffusing into the surface and the Si particles distributed in the cladding layer. The lower part of this irregular multi-phase region also contained residual Si particles (Fig. 3). Although analysis is currently incomplete, the β phase has not so far been detected in the IMC reaction between the

Si-rich cladding material and the Mg alloy, with the matrix in the irregular multi-phase region still being predominantly γ phase.

Fig.2. Interface characterization of the Al(Si) clad material (a) and unclad Al (b) joined to Mg AZ31 after annealing at 400°C for 3 hrs; (c) identification of the 2nd phase in the Al(Si)-Mg IMC layer, (d) EDX line profile along the red line shown in (a).

Fig.3. Elemental mapping of the interfacial area of the Al(Si) cladding - Mg joint after heat treatment at 400°C for 3h; (a) BSE image; (b) Mg; (c) Al; (d) Si.

3.2 Interfacial Characterization of the Al-Zn-Rich Coating

Zn is a common alloy element in both Al and Mg alloys. It has the advantage over Si that at elevated temperatures it is highly soluble in Al (~ 83 wt. % Zn at 381°C) and therefore will be homogeneously distributed in a coating. Moreover, the presence of Zn can help enhance the corrosion resistance of Mg alloys. Based on this reasoning, an Al-Zn solid solution coating was

designed by CALPHAD aided simulation to avoid the formation of the β phase, by again changing the reaction product to an alternative more stable compound. Fig.4 shows a calculated Al-Zn-Mg isothermal section at 360°C. The gray zone on the diagram corresponds to the composition range where the β - Al_3Mg_2 phase is stable. It can also be seen that, with a high enough Zn content one binary phase, MgZn_2 , and two ternary phases, τ -(Al,Zn) $_{49}\text{Mg}_{23}$ and ϕ - $\text{Al}_2\text{Mg}_5\text{Zn}_2$, can also be produced. Diffusion profiles between Al-Zn alloys with various Zn contents and the Mg alloy AZ31 were simulated using DICTRA at 360°C, which is in the range of typical peak temperatures reached during friction welding Al and Mg alloys. The result of such an analysis for an Al(52 wt. %Zn) - Mg couple is marked with a red line in Fig.4. It can be seen that this profile touches the tip of the phase fields in which Al_3Mg_2 exists, implying that at this temperature this is the minimum Zn content required to eliminate β . Moreover, the line goes through phase fields containing several binary and ternary phases (namely the γ - $\text{Al}_{12}\text{Mg}_{17}$, MgZn_2 , τ and ϕ phase), indicating that these intermetallics would be expected to be preferentially formed during any inter-diffusion across a weld interface.

Fig.4. Al-Zn-Mg isothermal section at 360°C with concentration profiles predicted by DICTRA for a diffusion couple between Al(52%Zn) and AZ31.

In Fig.5 the microstructures of the joint interface between the Al(52%Zn) coating to Mg and the baseline Al - Mg couple are depicted after annealing at 360°C for 24 hrs. It can be seen that both interfaces exhibit a three-layer IMC structure. In both cases the IMC layer adjacent to the Mg was indexed as γ - $\text{Al}_{12}\text{Mg}_{17}$. As before, in Fig. 2b in the standard Al to Mg joint, a uniform thicker layer of β - Al_3Mg_2 phase was found to have formed next to the Al side of the joint, (Fig.5b). In comparison, at the same location in the Al(Zn) - Mg joint (Fig.5a), EBSD and TEM micro-diffraction confirmed the presence of the ternary phase τ -(Al,Zn) $_{49}\text{Mg}_{32}$, and no Al_3Mg_2 was detected, confirming the predictions from the diffusion simulations. This means that the less favored intermetallic phases can be successfully prevented during welding, or diffusion bonding, Al to Mg by using an Al-Zn-rich coating based on the composition predicted above. A third IMC layer was also observed in both samples that has yet to be identified, which appears dark in the phase maps. This phase is likely to be $\text{Al}_{30}\text{Mg}_{23}$ which is known to be stable in the temperature range 250-410°C and is difficult to identify owing to its complex rhombohedra crystal structure and the stoichiometry of $\text{Al}_{30}\text{Mg}_{23}$ [14,15].

Fig.5. Comparison of the interface microstructures in (a) the Al(52%Zn) – Mg and (b) the standard Al - Mg control joint following annealing at 360°C for 24 hrs. The inserts are from EBSD phase ID maps, where each phase has been indexed from its back scattered diffraction pattern.

3.3 RFSSW Tests

Following these promising initial studies the two coatings were tested under welding conditions, using the RFSSW method. For comparison purposes, baseline uncoated AA6111 - AZ31 welds were also prepared at the same time using identical welding parameters.

The typical morphology of a weld cross section following RFSSW is shown in Fig. 6 with the sleeve and pin plunging areas marked. These are the effective bonding regions where stirring took place and an IMC layers were observed. The average IMC thicknesses was measured near the weld center and near the edge (sites A & B in Fig. 6) to characterize the level of intermetallic growth under the pin and the sleeve, respectively, and the results are illustrated in Table II. Among all three combinations, the Al(Zn) coating produced the thinnest IMC layer, which was less than 30% of that in the baseline couple. For the Al(Si) - Mg couple, a similar IMC layer thickness was found at the weld centre (site A) comparing to that seen with the uncoated baseline; however, there was a 50% drop in the intermetallic thickness under the sleeve position site B, indicating that the coating layer had been effective in retarding the detrimental interfacial reactions. Finally, Fig. 7 shows the peak loads that the three weld combinations achieved in lap shear tests. It can be seen that both coatings have been beneficial in improving mechanical properties of the weld, although the improvement is quite modest.

Fig. 6. Typical cross section of weld zone in an uncoated Al and Mg dissimilar joints produced by RFSSW.

Table II. Average IMC thickness at site A and B marked in Fig. 6

Material	Average IMC thickness (μm)	
	Site A	Site B

Al - Mg	3.5	8.0
Al(Si) - Mg	3.4	3.9
Al(Zn) - Mg	0.7	2.1

Fig. 7. Peak load Measurements of the Al and Mg RFSSWs during lap shear tests

4. Discussion

The above results have shown that the approach of using thermodynamic predictions to select suitable coatings has some potential for reducing the detrimental effects of IMC formation, in dissimilar metal welds between Al and Mg. In both cases the systems selected have changed the reaction path avoiding the formation of the undesirable β -Al₃Mg₂ phase. Both coatings have also been shown to have had some effect on inhibiting the IMC layer growth in reflow-FSSWs.

During the Al - Mg reaction, γ -Al₁₂Mg₁₇ is the first intermetallic layer to grow, and β -Al₃Mg₂ only appears after a continuous layer of γ is formed [6]. In the Al-Mg-Si system, the inhibition of the more rapid growing β -Al₃Mg₂ phase was realized via a two-fold mechanism: i) in the initial stage of the IMC reaction, due to the Mg - Si reaction that produces Mg₂Si, Mg atoms diffusing into the aluminum matrix are consumed by the Si particles present in the coating, and their concentration is therefore reduced where β would normally nucleate near the Al- γ interface; ii) at a later stage, as Al₁₂Mg₁₇ and Mg₂Si grow simultaneously, when the Mg₂Si particles became more connected with each other they form a partial barrier layer which helps prevent more Al and Mg interdiffusion. Thus, the nucleation of the β phase was restricted which, owing to the lower growth rate of γ , also reduced the overall layer thickness.

In the Al-Mg-Zn system the diffusion simulation calculated the concentration profile between an Al-Zn coating alloy, of a given composition, and the AZ31 composition of the Mg alloy sheet. In Fig.4, β cannot form outside its phase field, because its Gibbs free energy of formation becomes higher than that of alternative compounds. Therefore, by selecting an Al-Zn alloy, where the interdiffusion line was tangential to the β phase field, in the calculated ternary diagram, the Al₃Mg₂ phase was replaced by the ternary compound (Al,Zn)₄₉Mg₃₂. In the static annealing tests this phase nucleated and spread once the γ -Al₁₂Mg₁₇ reached a critical thickness.

It is known that when welding dissimilar metals, after complete bonding is achieved, the toughness of a joint decreases rapidly with growth of an IMC layer at the interface [9]. Both the

Si-rich and Al-Zn-rich coatings have been shown to have some effect on reducing the IMC thickness in the welding zone. As a result, the peak loads that the coated samples achieved in lap shear tests were found to have improved, although this was to a relatively modest level. For example, for the Al(Zn) - Mg couple, the IMC layer thickness was reduced by 70%, although the joint mechanical properties increased by just 5%. This suggests that, although greatly improved, the IMC layer was still too thick to have a dramatic effect on the joint strength and that submicron reaction layer thicknesses will need to be achieved to lead to high performance joints.

5. Conclusions

In this study, following thermodynamic assessment, two Al-alloy coatings (Al-Si and Al-Zn) were selected to prevent the formation of the detrimental β -Al₃Mg₂ phase in Al-Mg dissimilar welding. Both coatings were shown to be successful in changing the IMC reaction path.

When using an Al-Si cladding layer, the Mg substrate reacted with the Si particles during annealing to produce a more stable Mg₂Si phase, which itself acted as a diffusion barrier. As a result, the β phase was prevented and only Mg₂Si and the slower growing γ -Al₁₂Mg₁₇ phase were observed, which also reduced the IMC layer thickness.

An Al-Zn-rich coating composition was predicted by computer aided thermodynamic and diffusion simulations of the Al-Mg-Zn ternary system. This completely suppressed the Al₃Mg₂ phase, which was replaced by the ternary IMC, τ -(Al,Zn)₄₉Mg₃₂.

Welding experiments showed that both coatings were effective in reducing the IMC layer thickness produced at the joining interface in Al-Mg Refil-FSSWs, however this only had a modest effect on improving the joint the mechanical properties.

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References

1. D. Carle and G. Blount, "The suitability of aluminium as an alternative material for car bodies," *Materials & Design*, 20 (5) (1999), 267-272.
2. E. Aghion et al., "The role of the magnesium industry in protecting the environment," *Journal of Materials Processing Technology*, 117 (3) (2001), 381-385.
3. E. Schubert et al., "Light-weight structures produced by laser beam joining for future applications in automobile and aerospace industry," *Journal of Materials Processing Technology*, 115 (1) (2001), 2-8.
4. J. Wang et al., "Microstructure of Al-Mg dissimilar weld made by cold metal transfer MIG welding," *Materials Science and Technology*, 24 (7) 827-831.
5. R. Zettler et al., "Dissimilar Al to Mg alloy friction stir welds," *Advanced Engineering Materials*, 8 (5) (2006), 415-421.
6. A. Panteli et al., "The effectiveness of surface coatings on preventing interfacial reaction during ultrasonic welding of aluminum to magnesium," *Matallurgical and Materials Transactions A*, 44 (13) (2013), 5773-5781.
7. L. Wang et al., "Controlling interfacial reaction during dissimilar metal welding of aluminium alloys," *Materials Science Forum*, 794-796 (2014), 416-421.

8. H. Marsumoto et al., "Fabrication of pure Al/Mg-Li cold plate and its mechanical properties," *Journal of Materials Processing Technology*, 169 (1) (2005), 9-15.
9. A. Panteli et al., "The effectiveness of surface coatings on preventing interfacial reaction during ultrasonic welding of aluminum to magnesium," *Metallurgical and Materials Transactions A*, 44 (13) (2013), 5773-5781.
10. J. Zhang et al., "Microstructural refinement in Al-Mg₂Si in situ composites," *Journal of Materials Science Letters*, 18 (10) (1999), 783-784.
11. M. Mabuchi and K. Higashi, "Strengthening mechanisms of Mg-Si Alloy," *Acta Materialia*, 44 (11) (1995), 4611-4618.
12. Y. Tang et al., "Thermodynamic description of the Al-Mg-Si system using a new formulation for the temperature dependence of the excess Gibbs energy," *Thermochimica Acta*, 527 (2011), 131-142.
13. D. Dietrich et al., "Formation of intermetallic phases in diffusion-welded joints of aluminium and magnesium alloys," *Journal of Materials Science*, 46 (2) (2010), 357-364.
14. J. Murray, "The Al-Mg (aluminum-magnesium) system," *Bulletin of Alloy Phase Diagrams*, 3 (2) (1982), 60-74.
15. S. Shakhshir and M. Medraj, "Computational thermodynamic model for the Mg-Al-Y system," *Journal of Phase Equilibria and Diffusion*, 27 (3) (2006), 231-244.