

Article

Challenges and Latest Developments in Diffusion Bonding of High-Magnesium Aluminium Alloy (Al-5056/Al-5A06) to Stainless Steels

Amir A. Shirzadi ^{1,2,*} , Chengcong Zhang ^{3,4}, Muhammad Zeeshan Mughal ¹  and Peiyun Xia ⁴¹ School of Engineering and Innovation, The Open University, Milton Keynes MK7 6AA, UK; zeeshan.mughal@open.ac.uk² School of Materials and Metallurgy, Wuhan University of Science and Technology, Wuhan 430081, China³ International Research Institute for Steel Technology, School of Sciences, Wuhan University of Science and Technology, Wuhan 430081, China; zhangcc0202@163.com⁴ Technical Centre, Shanghai Aerospace Equipment Manufacturer, Shanghai 200245, China; xypei@126.com

* Correspondence: amir.shirzadi@open.ac.uk

Abstract: The aim of this work was to investigate the challenges associated with bonding Al-Mg alloys and develop a new method for bonding these alloys to steels. During an extensive R&D project, over 80 attempts, using 11 methods, were made to bond Al-6 wt.% Mg alloy (Al-5056/Al-5A06) to two types of stainless steels (heat-resistant 1Cr18Ni9Ti and conventional 316). Wide ranges of temperature (500 °C to 580 °C), pressure (0.5 MPa to 10 MPa) and time (1 min to 2 h) were used when direct diffusion bonding of these alloys. Then, effects of using various interlayers and brazing foils were investigated. The interlayers used in this work were gallium, pure titanium, copper and aluminium foils, aluminium 6061 alloy sheets, aluminium-silicon brazing foils, zinc and zinc alloy foils as well as an active brazing foil (known as Incusil-ABA containing silver, copper, indium and titanium). Several complex and multi-stage processes, using up to 3 different interlayers in the same joint, were also developed and assessed. Examination and assessment of the bonded samples, including failed attempts, paved the way of developing new methods for bonding these dissimilar materials. A number of samples with tensile strengths from 200 MPa to 226 MPa were made by using complex combinations of 2 or 3 interlayers and triple-stage bonding cycles. The highest recorded bond strength was 226 MPa in the as-bonded condition. This value is above the measured yield strength (134 MPa) and about 93% of the measured ultimate strength (243 MPa) of the parent Al-Mg alloy after it was subjected to the same bonding cycle. Since the use of complex processes was not feasible for bonding large components, a simpler and more practical bonding cycle was also developed in the project. Using the simpler process, joints with tensile strengths around 90 MPa could be made. This article also sheds light on the difficulties associated with brazing and soldering aluminium alloys with a high magnesium content.

Keywords: diffusion bonding; Al-Mg alloys; dissimilar joints; aluminium steel welding

Citation: Shirzadi, A.A.; Zhang, C.; Mughal, M.Z.; Xia, P. Challenges and Latest Developments in Diffusion Bonding of High-Magnesium Aluminium Alloy (Al-5056/Al-5A06) to Stainless Steels. *Metals* **2022**, *12*, 1193. <https://doi.org/10.3390/met12071193>

Academic Editor: Hardy Mohrbacher

Received: 8 June 2022

Accepted: 10 July 2022

Published: 13 July 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Aluminium alloys and steels are the most widely used engineering materials. Welding these materials is of immense interest to transport, construction, power generation, mining and energy industries. However, it is impossible to weld aluminium and steel by conventional fusion welding processes due to the distinct differences in their melting points, thermal expansion coefficients, densities, heat conductivities and so forth [1]. Furthermore, the formation of thick intermetallic compounds (IMCs), due to limited inter-solubility of the main elements (Al and Fe), results in dissimilar joints with inferior mechanical properties [2,3]. Therefore, solid-state joining methods, such as friction stir welding [4], diffusion bonding [5,6] and explosive welding [7], are considered to be the only viable techniques to join Al-Mg alloys to ferrous alloys.

Previous studies developed several new solid-state methods to join un-weldable dissimilar aluminium alloys and steels. Haghshenas et al., who combined friction welding and diffusion bonding to join Al-5754 to DP600 and 22MnB5 steels, reported the formation of the high-aluminium and high-iron intermetallics (Al_5Fe_2 and FeAl) [8]. Liu et al. carefully set up a friction stir welding (FSW) process to reduce the thickness of Al-Fe intermetallic down to a nanoscale [4]. Howlader et al. attempted the direct bonding of pure aluminium and steel at room temperature after ion sputter cleaning in an ultra-high vacuum [9]. Huang et al. proposed a new approach for joining thin aluminium and stainless steel sheets based on thermally assisted plastic deformation [10].

Hirose et al. used diffusion bonding to join various aluminium alloys to steels. They concluded that the reaction layers in Al-5000 aluminium-magnesium alloys grow much faster than in Al-6000 alloys when bonding these alloys to any steel. Consequently, the joints between Al-Mg and steel had very low strengths due to the rapid and irregular growth of the reaction layer caused by the high Mg content. The authors also reported that many samples fractured during the machining of tensile test specimens; hence, the joint strength could not be determined [5]. Hirosea et al. also studied the effect of magnesium content on the interfacial reaction of the diffusion bonded aluminium-steel joints. Their work showed that to obtain reasonable bond strengths, the magnesium content should be below 1.0 wt.% [6].

Al-5A06 alloy (equivalent to Al-5056) with a high magnesium content (5.8–6.8 wt.%) is one of the most widely used aluminium alloys in the transport and aerospace sectors due to its high specific strength and excellent corrosion resistance [11,12]. However, the high content of magnesium makes it one of the most difficult alloys to bond to other alloys. Very few attempts have been made to diffusion bond these family of aluminium alloy to steels. In this work, several methods to diffusion bond Al with 6 wt.% Mg alloy to stainless steel were developed, and the resultant microstructures and mechanical properties of the bonded sample were investigated.

2. Materials and Methods

An aluminium alloy, with about 6 wt.% magnesium (Al-5A06) and a heat-resistant stainless steel (1Cr18Ni9Ti), were used in most bonding trials. The compositions of these base materials are given in Table 1. Early results showed that conventional 316 stainless steel exhibits very similar properties to heat-resistant stainless steel (1Cr18Ni9Ti) when bonded to the Al-Mg alloy.

Table 1. Chemical compositions (wt.%) of base alloys used in this work.

Chemical Composition	Mg	Mn	Si	Fe	Zn	Cu	Ti	Al	Cr	Ni	C	S	P
5A06	5.8–6.8	0.5–0.8	0.4	0.4	0.2	0.1	0.02–0.1	Bal.	-	-	-	-	-
1Cr18Ni9Ti	-	≤2.0	≤1.0	Bal.	-	-	0.5–0.8	-	17.0–19.0	8.0–11.0	≤0.12	≤0.03	≤0.035

The interlayers used in this work were liquid gallium, pure titanium coating, titanium, copper and aluminium foils, aluminium 6061 alloy sheets, aluminium-silicon brazing foils, pure zinc and zinc alloy foils, as well as an active brazing foil (known as Incusil-ABA containing silver, copper, indium and titanium). The thickness of all coatings was measured using the standard ball crating method.

Disc-shape samples with diameters between 10 mm to 25 mm and lengths from 5 mm to 25 mm, depending on the intended post-bonding evolution, were bonded. The faying surfaces of all samples were ground using 600 Grit emery paper and rinsed in isopropyl alcohol before loading them in the diffusion bonder. The samples were bonded in a vacuum (<0.01 Pa) using a purpose-built diffusion bonder capable of monitoring and controlling the bonding temperature and pressure. The diffusion bonder used in this work is designed by the first author and located in main campus of The Open University (Milton Keynes, UK). Wide ranges of temperature, pressure and time were used when bonding these alloys

with or without an interlayer. A large number of bonding trials were needed to develop and optimise a successful bonding method.

The microstructural analyses were conducted using an optical microscope made by Brunel Microscopes Limited and the SEM Zeiss crossbeam 550 system equipped with Oxford Instruments Ultim Max detector at The Open University, Milton Keynes, UK. The tensile strengths of selected samples were measured using Instron 5969 universal testing machine at The Open University. The effects of using various interlayers and brazing foils were investigated. Several complex and multi-stage processes, using up to 3 different interlayers in the same joint, were also developed and assessed. The specimen setups and bonding conditions are detailed below.

2.1. Method I: Direct Diffusion Bonding without Using an Interlayer

Previous work has shown that limited or no reaction occurs between aluminium and steel when bonding them below 450 °C. Therefore, the first bonding trial was carried out at 500 °C under constant pressure of 5 MPa in a vacuum below 10^{-2} Pa (10^{-4} mbar). The heating rate was 60 °C per minute for most samples except those bonded around the melting point of the Al alloy which required fast heating and short bonding times. All of the samples were furnace cooled, and the vacuum chamber was vented when the sample temperature dropped to 150 °C or lower.

The samples bonded at 500 °C had very low strength and failed during handling. Increasing the bonding temperature to 510 °C, 530 °C and 540 °C did not improve the bond strength noticeably. An examination of the fractured surfaces revealed the presence of very limited reactions between the two alloys. This is in contrast with the results of previous work on diffusion bonding 6000 series aluminium to 316 stainless steel, where continuous and adherent intermetallic formed at such high temperatures. It appears that the presence of magnesium oxide, a very stable refractory ceramic, on the surface of the Al-Mg alloy acted as a diffusion barrier preventing the formation of Al-Fe intermetallics.

A further increase in the bonding temperature to 550 °C or 560 °C, which is close to the nominal solidus temperature of the alloy (568 °C), led to the decomposition and disintegration of the Al-Mg alloy under the applied load—see Figure 1. Despite using such high bonding temperatures, there was a very limited reaction between the Al-Mg alloy and stainless steel, and consequently, the joint strengths were very low.

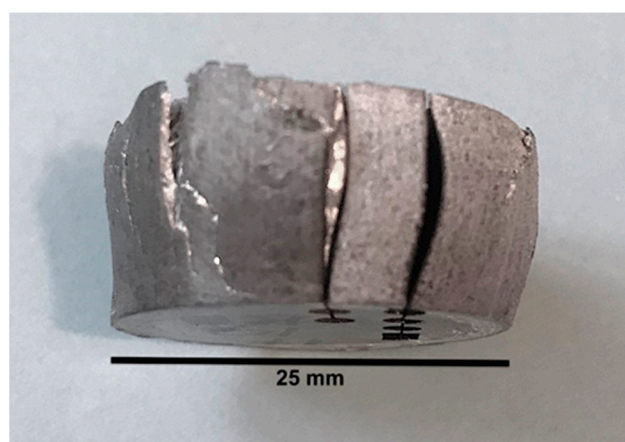


Figure 1. Disintegration of Al-Mg alloy during solid-state bonding it to stainless steel at 560 °C. Almost no bond formation occurred despite using a temperature close to the melting point of the Al-Mg alloy.

2.2. Method II: Gallium-Assisted Diffusion Bonding

Previous work showed that gallium can modify or even remove the surface oxides on certain alloys [13]. A small amount of gallium was used to remove or modify the magnesium oxide on the Al-Mg surface before repeating the same bonding steps used in

Method I. Although the gallium treatment enhanced the reaction between the two alloys, all of the samples broke during handling or machining.

Two samples containing much more liquid gallium in their joint interfaces were bonded at and above 540 °C. The bonding cycle had to be interrupted due to sudden melting of the Al-Mg alloy. Figure 2 shows a partially melted Al-Mg alloy bonded to a steel disc. The solidus and liquidus temperatures of the Al-Mg alloy are 568 °C and 638 °C, respectively, and the observed sub-solidus melting was due to the Ga-Mg eutectic reaction, as explained below.

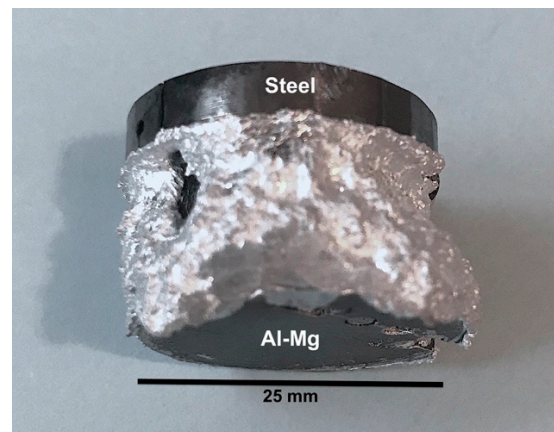


Figure 2. The sudden melting of Al-Mg alloy due to migration of Mg to the surface and its eutectic reaction with liquid gallium.

The phase diagrams in Figure 3 show that gallium goes through eutectic reactions with magnesium and aluminium. However, it is unlikely that the sub-solidus melting was due to Al-Ga eutectic reaction for the following reasons. Firstly, gallium has a higher solubility in aluminium than in magnesium: hence, a much higher amount of gallium would be required to form a large amount of liquid seen in Figure 2. Secondly, any liquidation due to Al-Ga eutectic could have occurred at a lower temperature. Thirdly, the formation of liquid on the exterior of the Al-Mg alloy is unprecedented and never seen when gallium brazing of aluminium alloys with a low magnesium content, e.g., Al-6000 series [13].

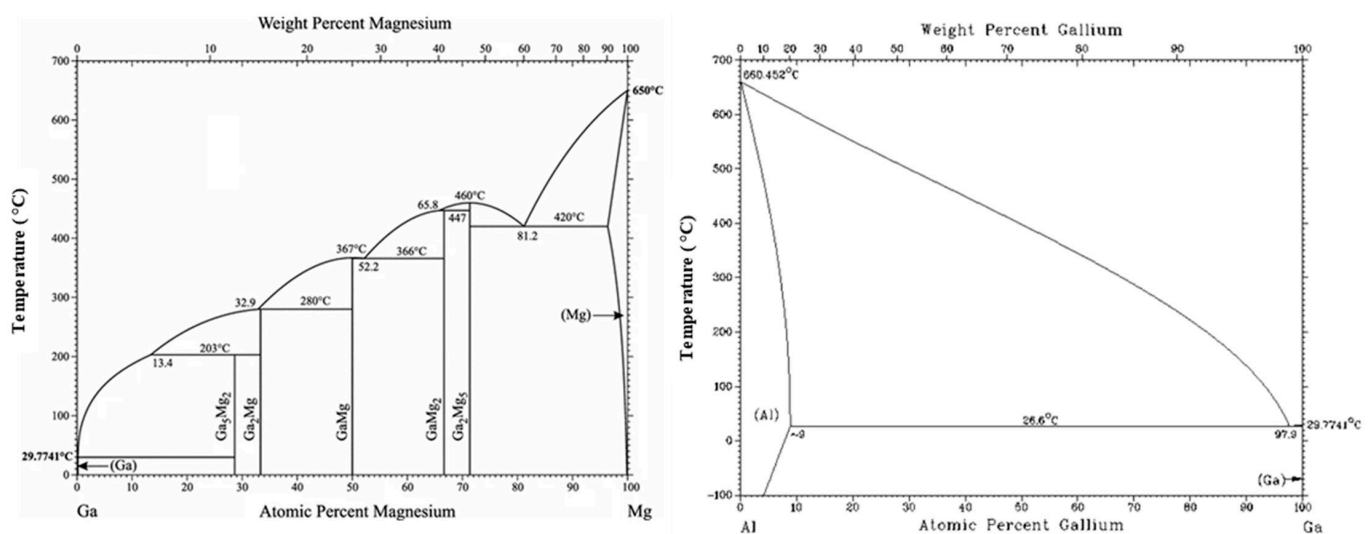


Figure 3. Phase diagrams of Ga-Mg (left) and Al-Ga (right) show eutectic reactions may occur well below the melting point of the Al-Mg alloy (solidus 568 °C) used in this project.

It is a well-established fact that magnesium can diffuse to the surface of the Al alloys containing more than 0.5 wt.% Mg during high temperature vacuum brazing. Given the 10 times more Mg in the Al-Mg alloy (6 wt.%), it is plausible that the migration of Mg on the surface and its reaction with gallium resulted in the sudden melting of the alloy.

Gallium-assisted bonding at lower temperatures failed to produce strong bonds, and the use of gallium was discontinued.

2.3. Method III: Diffusion Bonding Using Titanium

The effect of titanium, as an interlayer, to bond Al-Mg alloy to stainless steel was investigated. The titanium was in the form of a 4-micron thick coating or 1 mm thick overlayer deposited on the steel by cold metal spraying process.

The bonding temperatures were mainly between 510 °C to 560 °C, and for a short time at 570 °C which is just above the solidus temperature of the Al-Mg alloy (568 °C). Despite using such high temperatures, there was no reaction between the titanium and the Al-Mg alloy. The presence of stable magnesium oxide and lack of chemical affinity to titanium made the bond formation impossible.

2.4. Method IV: Transient Liquid Phase (TLP) Bonding Using Pure Copper

Bonding copper to stainless steel is a well-developed process for manufacturing heat exchangers. Copper-based alloys are also used for brazing various structural alloys because of their good wettability and adhesion. On the other hand, copper is a “melting point depressant” for aluminium owing to Al-Cu eutectic reaction at 548 °C. Therefore, it was interesting to investigate the possibility of bonding the Al-Mg to stainless steel using a thin layer of pure copper.

Previous attempts on TLP bonding of aluminium 6000 series to stainless steel 316 were unsuccessful because of poor wettability of the Al-Cu eutectic liquid on the stainless steel. The temperature of the eutectic phase (around 550 °C) is too low to break up the chromium oxide on the stainless steel and form a metallurgical bond. This is consistent with the fact that stainless steels are usually immune from “liquid metal attacks” if its surface oxide remains intact. Therefore, as explained below, a new 2-stage TLP bonding process was developed in this work.

First, a stainless-steel disc (Ø25 mm) was copper-cladded by diffusion bonding of a 100-micron thick copper foil. The cladding was carried out at 950 °C for 10 min under 1 MPa pressure. Then, the copper side of the stainless-steel disc was assembled with a matching Al-Mg disc before inserting them inside a graphite crucible. Induction heating was used to heat the crucible while simultaneously monitoring the joint and crucible temperatures. Figure 4 shows the TLP bonding setup used in this work.

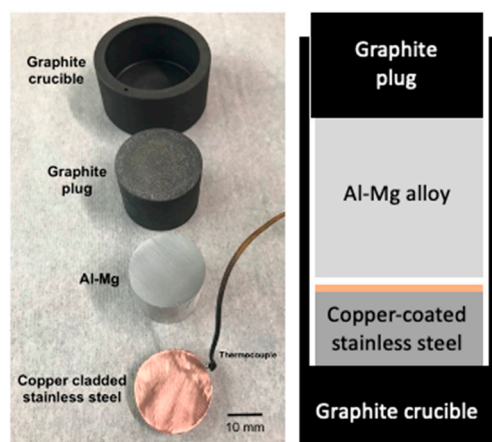


Figure 4. A graphite crucible was used to heat the Al-Mg and Cu-cladded stainless steel. The bonding pressure was applied by the graphite plug placed on top of the assembled sample.

Since the exact melting point of the joint interface was unknown, the temperature was gradually increased until a sudden drop in the pressure occurred at 517 °C due to liquidation at the joint. Figure 5 shows the extent of the reaction between the copper and the Al-Mg alloy during the TLP bonding process. The copper/magnesium phase diagram shows two eutectic reactions at 552 °C and 486 °C, which may explain the observed decomposition of the Al-Mg alloy. Admittedly, the presence of Cu, Al and Mg, of which only Cu is present in a pure state, imposes limitations on using binary phase diagram(s) to predict the start and completion of the transient liquid phase bonding process.

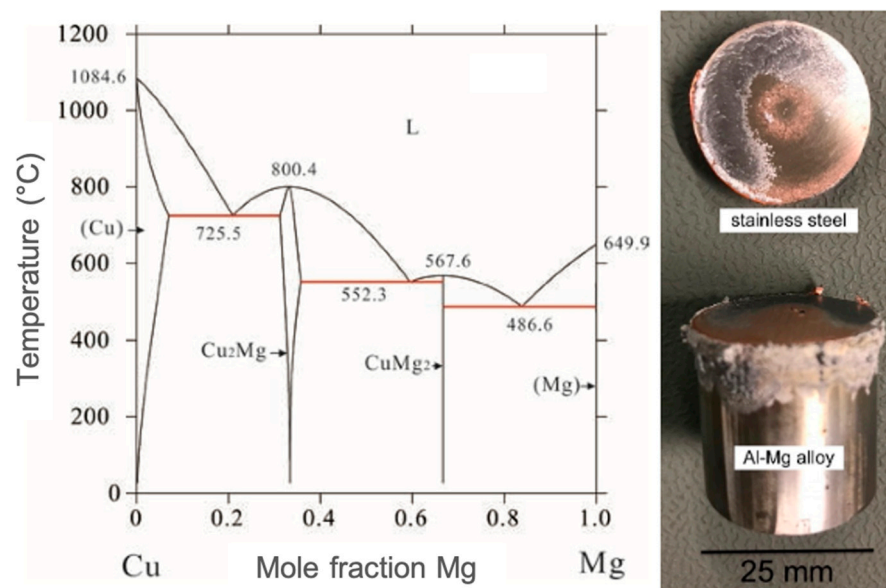


Figure 5. Decomposition of the Al-Mg alloy in the presence of copper just below 520 °C is consistent with the low temperature eutectic reactions seen in the corresponding phase diagram.

A comprehensive investigation was undertaken to determine the microstructure and composition of the TLP bonded sample. As expected, the reaction zone around the Al-Mg alloy mainly consists of Al, Mg and Cu, i.e., the “frosted-looking” area around the sample shown in Figure 5.

The elemental maps in Figure 6 show the higher affinity of copper to magnesium than copper to aluminium. This is clear that the aluminium-rich oval phase in Figure 6 hardly contains any copper.

More detailed EDX point analysis confirmed that the concentration of Mg can reach up to 12 wt.% in the dendritic phase (see Figures 6 and 7).

As a result of having very low bond strengths, TLP bonding using copper interlayer was terminated.

2.5. Method V: Liquid Phase Bonding Using Zinc Interlayer

The melting point of zinc (420 °C) is well-below that of the Al-Mg alloy (568 °C). In order to avoid the decomposition of Al-Mg alloy during the bonding cycle, a 0.1 mm thick pure Zn interlayer was used to bond Al-Mg to stainless steel at 400 °C in 1 h under ~1.5 MPa pressure. This led to partially melting of Al-Mg, as a result of eutectic reaction which occurs around 340 °C between pure zinc and magnesium.

The bonding temperature was further reduced to prevent excessive melting of the Al-Mg alloy. Despite repeated attempts and using various conditions, all efforts to bond Al-Mg alloy to stainless steel below 400 °C failed because of poor zinc wettability on the stainless steel surface.

In addition to the above issues, zinc has a very low vapour pressure at high temperatures. Therefore, zinc-bonding proved very problematic due to the evaporation of zinc and contamination of the vacuum chamber.

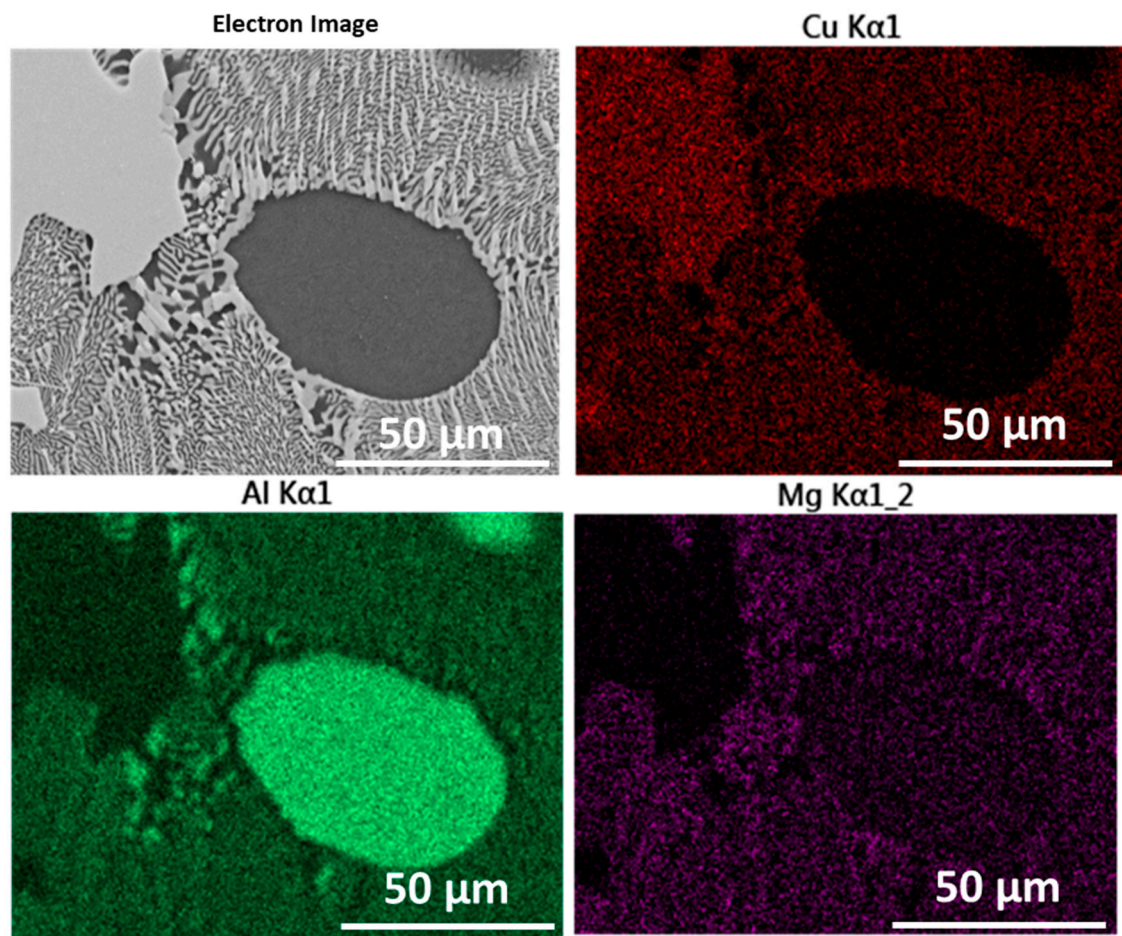


Figure 6. Elemental mapping of the reaction zone of the sample shown in Figure 5. The bias affinity of Mg to Cu led to low temperature melting at the joint interface and the decomposition of the Al-Mg alloy.

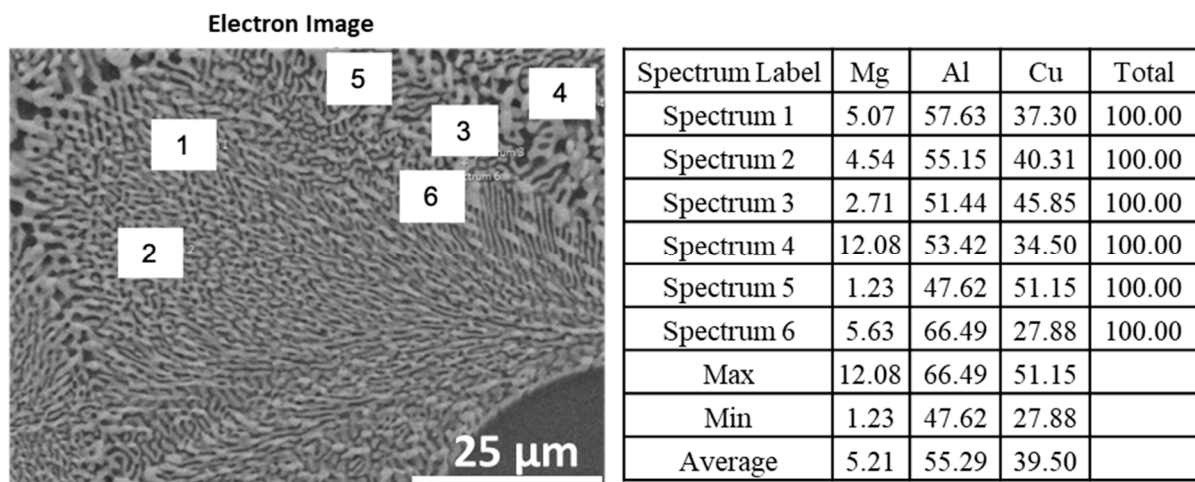


Figure 7. High concentration of Mg up to 12 wt.% was found in six randomly selected points in the reaction zone of the sample shown in Figure 5.

2.6. Method VI: Using Zn-Based Soldering Alloy (TECHNO-WELD)

TECHNO-WELD is a commercial Zn alloy containing aluminium, copper, manganese and magnesium, which is used for flame-soldering various aluminium alloys. It must be noted that flame-soldering of aluminium requires mechanically abrading the aluminium

surface to ensure adequate wetting by molten solder. Nevertheless, with a melting point of 380 °C (40 °C below that of pure zinc), TECHNO-WELD seemed to be an attractive option to join the Al-Mg alloy to steel.

Despite the absence of abrading and any flux, the Al-Mg alloy was bonded to stainless steel using TECHNO-WELD and at about 400 °C in a vacuum under 0.5 MPa pressure. However, all 2-mm diameter tensile test rods, extracted from the bonded samples broke during machining or snapped when bend tested. The fracture surfaces of the samples bonded using zinc or zinc alloys are quite similar and show the lack of adequate wetting and formation of non-uniform brittle phases.

2.7. Method VII: Using Multi-Interlayers of Cu, Zn and Al

Several attempts were made to bond the Al-Mg alloy to stainless steel using more than one interlayer. Two exemplary cases are as follows:

Example 1:

Stage 1: Bonded Cu foil (0.1 mm) to stainless steel at 1050 °C, 1 MPa, 10 min;

Stage 2: Bonded Zn foil (0.1 mm) to Cu from Stage 1 at 410 °C, 1 MPa, 30 min;

Stage 3: Bonded Al-Mg alloy to Zn from Stage 2 at 330 °C, 1 MPa, 30 min.

Example 2:

A few samples were also bonded using 7-micron Cu foil on the steel side and 20-micron Al foil on the Al-Mg side at 550 °C, which is just above the eutectic temperature of Al-Cu, i.e., 548 °C.

The bonding temperatures in the above stages were chosen based on the corresponding phase diagrams and previous work experience. Despite achieving reasonably strong bonds in some cases, the formation of intermetallics resulted in brittle joints. Due to the very poor ductility, low strengths and complexity of the method, the use of multi-interlayers was abandoned.

2.8. Method VIII: Fast Brazing Using Al-Si Foils

Al-Si alloys are extensively used to join aluminium to aluminium, e.g., in car radiators and air conditioners. Silicon based fluxes such as NOCOLOK® are used in manufacturing aluminium kitchen pots with stainless steel bases. Aluminium with 12 wt.% Si goes through eutectic reaction at 577 °C, which is far below the melting point of most aluminium alloys. Since the Al-Mg alloy's solidus temperature (568 °C) is 9 °C below the eutectic temperature of Al with 12 wt.% Si, it was not possible to silicon-braze the Al-Mg alloy to stainless steel without melting the alloy. However, a few attempts were made with very short bonding times to minimise damage to the Al-Mg alloy when using an Al-Si brazing foil. The sample was quickly heated to 580 °C under 1 MPa and kept at that temperature for less than one minute or until liquidation occurred. The Al-Mg part was slightly decomposed and reacted with the molten Al-Si alloy. Nevertheless, the time was too short to fully wet the stainless steel surface and form a strong bond.

2.9. Method IX: Active Diffusion Brazing

Incusil-ABA is an Active Braze Alloy (ABA) of silver, copper, indium and titanium suitable for joining ceramics as well as certain alloys, e.g., stainless steels. The composition (wt.%) of Incusil-ABA is 59.0% Ag, 27.25% Cu, 12.5% In and 1.25% Ti. The presence of Indium brings down the melting point below those of all other active brazing alloys. However, the solidus and liquidus temperatures of Incusil (605 °C and 715 °C) are still above the solidus temperature of the Al-Mg alloy (568 °C). Nevertheless, potential benefits of fast heating rates (200 °C per minute) and very short bonding times (less than a minute) when using Incusil interlayer were assessed in this project.

In short, brazing cycles just above the Incusil's solidus temperature failed to form a bond with stainless steel because of poor wetting. On the other extreme, brazing at higher temperatures close to the Incusil's liquidus temperature resulted in very firm joints but at the cost of damaging the Al-Mg alloy.

2.10. Method X: Cladding Al-Mg Alloy with Pure Al to Reduce Migration of Mg into the Joint Interface

The previous methods showed that the presence and further migration of Mg onto the surface of the Al-Mg alloy prevented achieving satisfactory joints with stainless steel. For instance, the Mg-rich alloy did not show any affinity even to titanium at high temperatures (Method III). Increasing the bonding temperature up to the melting point of Al-Mg alloy had an adverse effect on the bulk Al-Mg alloy—see Figure 1. In addition, Mg led to low temperature melting of the Al-Mg alloy in an uncontrollable manner when using Cu or Zn interlayers.

Therefore, it seemed reasonable to build a “Mg barrier” between the Al-Mg alloy and steel to prevent the problems mentioned above. Pure aluminium foils and coatings were considered potential “Mg barriers” to limit the migration of Mg and improve bond strength.

The faying surface of Al-Mg samples was ground with 1200 Grit emery paper and thoroughly cleaned before magnetron deposition of 6-micron thick pure aluminium. The coated side of the Al-Mg sample was directly diffusion bonded to stainless steel at temperatures between 500 °C to 550 °C under a constant load between 1 MPa to 10 MPa. The bonding time was about 30 min for all samples. Similar cycles were repeated after inserting a titanium foil between the aluminium coating and stainless steel.

The samples containing pure aluminium coating had very low bond strength, regardless of the bonding parameters. Full SEM and EDX analyses were conducted on the joint cross sections and fractured surfaces. The unexpected outcome of EDX analysis revealed that a large amount of Mg migrated and passed through the 6-micron thick pure aluminium. Figure 8 shows the aluminium coating was too thin to stop Mg reaching the faying surface of the stainless steel.

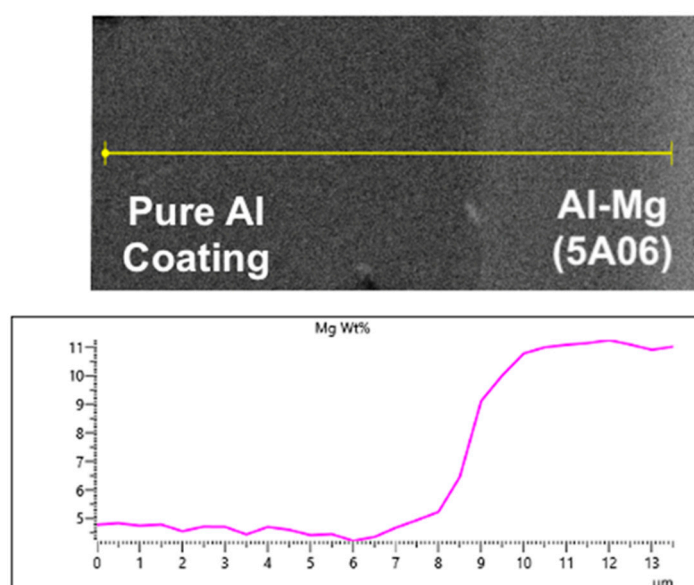


Figure 8. Magnesium from the Al-Mg alloy had a long-range diffusion into thin pure aluminium coating.

The presence of such a high amount of magnesium prevented successful bonding of the Al-Mg to stainless steel, as repeatedly observed in the previous methods.

Figure 8 also revealed that the concentration of Mg was up to 11 wt.% on free surfaces, which is above the expected values (6 wt.%). It was concluded that migration of magnesium occurred during magnetron coating and subsequent bonding cycle. In order to ascertain the unexpected high concentration of Mg, EDX analysis was carried out on the fractured surfaces. The EDX graph in Figure 9 reconfirmed that a very high amount of Mg is present on the fractured surface too.

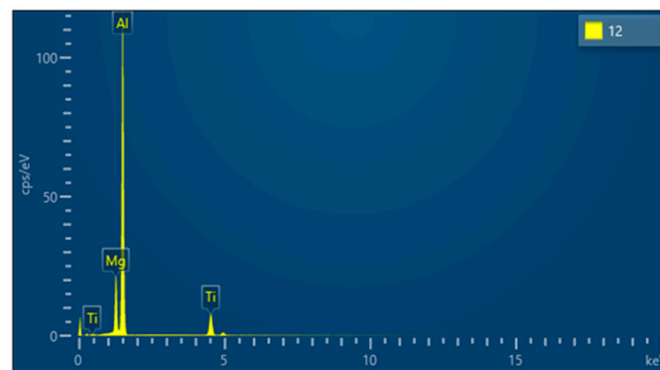


Figure 9. Up to 11 wt.% Mg was detected on the fracture surface of a bonded sample despite using pure aluminium coating as a diffusion barrier.

2.11. Method XI: Use of Al-6061 Plate as a Buffer Layer

Given the observed high diffusivity of Mg, it was decided to replace the thin pure aluminium coating with a much thicker plate of Al-6061. The specimen setup is schematically shown in Figure 10.

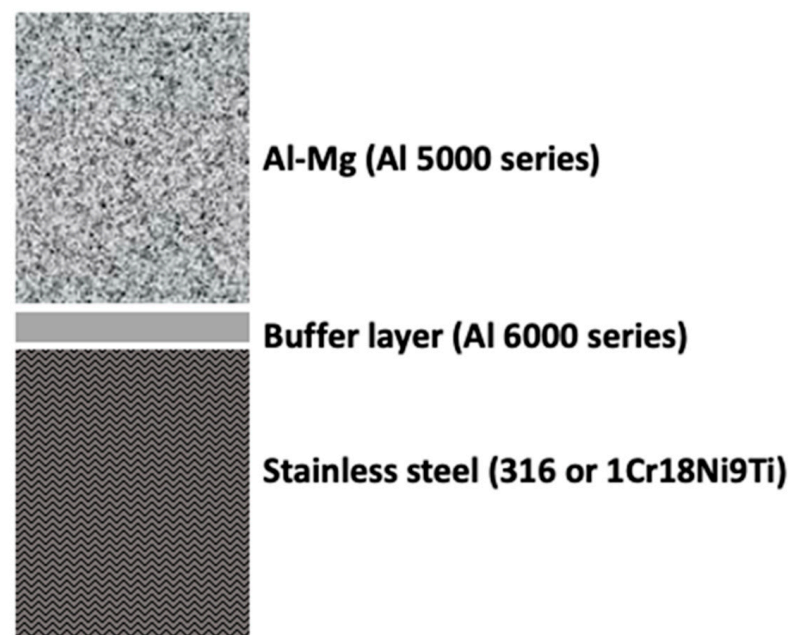


Figure 10. A plate of Al-6061 was inserted between Al-Mg alloy and steel to create a buffer layer and prevent migration of Mg into joint interface.

In the first stage, Al-6061 plates with 0.5 mm to 1.0 mm thicknesses were diffusion bonded to the Al-Mg alloy at 520 °C to 550 °C within 1 h. Once the parameters for diffusion bonding of Al-6061 to Al-Mg (5A06) were determined, the clad side of the Al-Mg alloy was joined to the stainless steel.

However, solid-state bonding of the Al-Mg alloy to other aluminium was also problematic. Figure 11 shows the extent of the decomposition of Al-Mg alloy after solid-state bonding it to conventional Al-6061 alloy. Pre-bonding heat treatment of Al-Mg (normalising) in air reduces the rate of Mg migration on the alloy's surface, hence reducing alloy's disintegration in the subsequent bonding cycle.

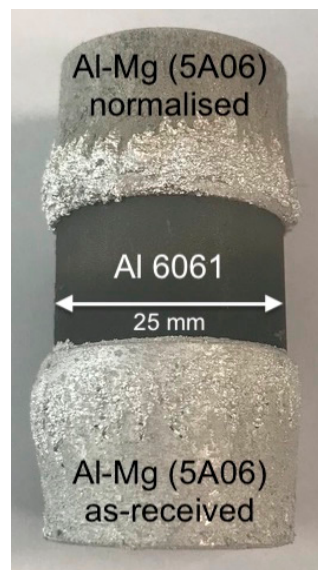


Figure 11. Diffusion bonding of Al-Mg to Al-6061 in vacuum resulted in decomposition of Al-Mg when bonding temperature exceeded $\sim 550^\circ\text{C}$, while Al 6061 remained intact.

Examination of the Al-Mg/Al-6061 samples reconfirmed the long-range diffusion of Mg into the Al-6061. Figure 12 shows Mg diffused into Al-6061 more than 0.5 mm deep.

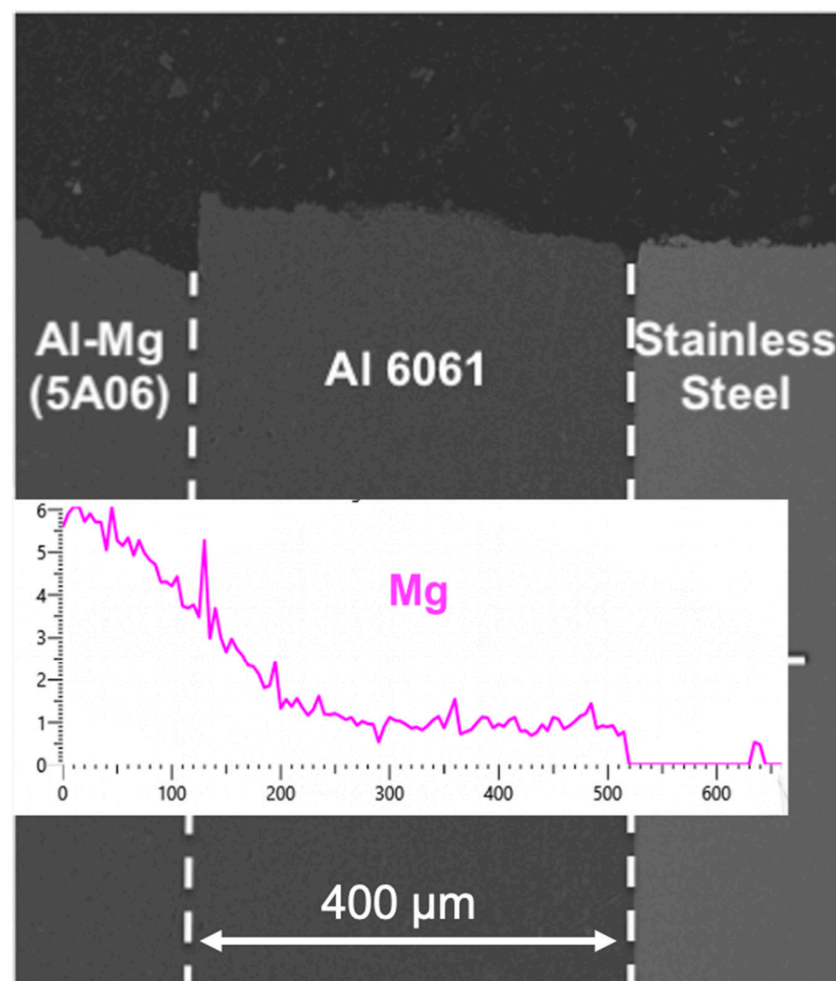


Figure 12. EDX mapping shows Mg diffuses up to 0.5 mm deep into the Al-6061 interlayer.

Based on the elemental analysis of various joints made using Al-6061 interlayer, it was concluded that at least a 1 mm thick diffusion barrier is required to prevent Mg from reaching the aluminium-steel interface.

3. Candidate Methods for Joining Al-Mg to Stainless Steel on a Laboratory Scale

A large number of bonding trials and subsequent joint assessments had to be carried out to overcome the above-mentioned challenges. Finally, the following methods for bonding Al with 6% Mg to stainless steel were developed on a laboratory scale. The proposed bonding processes and results of tensile tests are summarised in Figure 13.

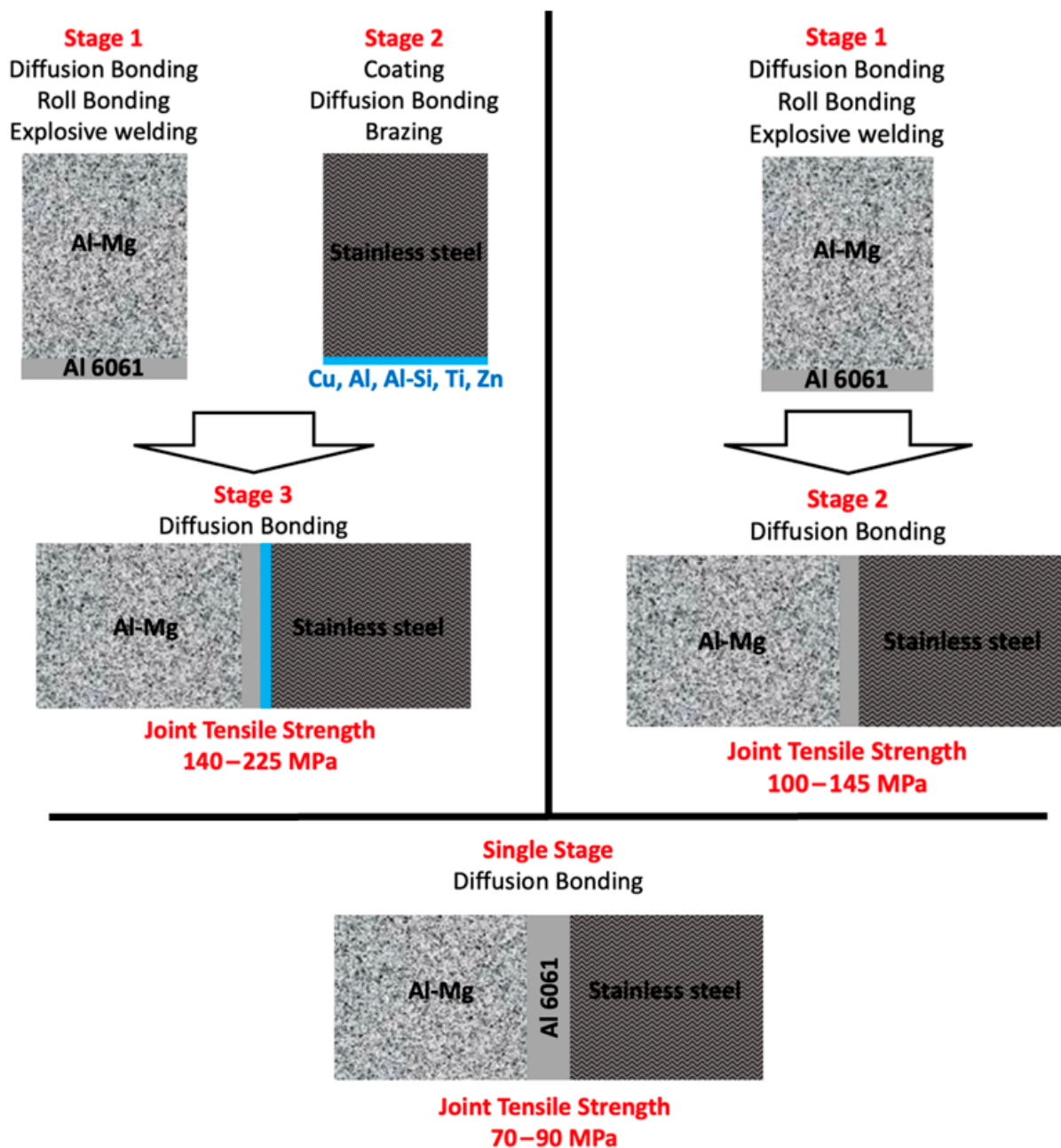


Figure 13. Specimen setups for triple (top left), double (top right) and single stage (bottom) bonding cycles developed in this work; the greater the complexity of the bonding process, the higher the bond strength.

Each proposed method has its own advantages and restrictions. Therefore, the suitability of a process for bonding larger components must be evaluated based on the size, costs as well as the capacity and availability of diffusion bonding equipment.

Using a complex cycle and multi-interlayers, bonds with tensile strengths up to 226 MPa were made. Figure 14 shows the stress vs. displacement of a tensile-tested sample made using a triple stage bonding cycle. The measured tensile strength of unbonded parent Al-Mg alloy was 243 MPa in the as-bonded condition. Figure 14 also shows that the elongation (not the strain) of the bonded bi-metal sample was less than that of the monolithic parent aluminium alloy. This is partly because half of the bonded sample was made of non-deforming steel. Therefore, in Figure 14, the elongation of the bonded sample should be multiplied by 2 before comparing it with that of the parent alloy.

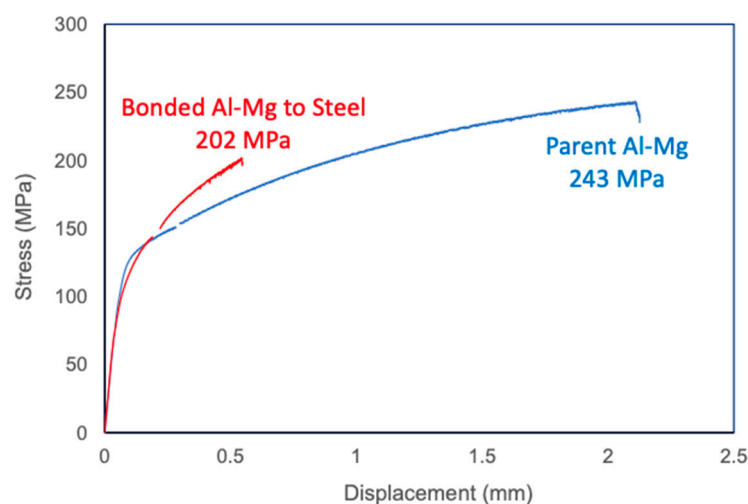


Figure 14. Bonds with tensile strengths above the yield point of Al-Mg (5A06) alloy were achieved using complex and multi-stage processes. Note that the relative elongations are not comparable directly—see the text for more detail.

4. Optimised Process for Diffusion Bonding Large Parts

Figure 15 shows a bonded sample cross-section when using a 1 mm thick Al-6061 interlayer and the corresponding hardness profile. The gradual change in the hardness across the interface of two aluminium alloys is consistent with the observed diffusion of Mg from the Al-Mg alloy into the Al-6061 buffer layer. Figure 16 shows the results of a detailed examination of the sample using SEM. The joint interfaces of Al-Mg/Al-6061 were free from large voids or discontinuities when bonding trials were carried out above 510 °C. However, the bonding temperature should not exceed 555 °C to avoid high temperature deterioration of the Al-Mg alloy, as shown in Figures 1 and 11.

Figure 16 also shows that the steel to Al-6061 joint, as expected, contained thin and mostly continuous Al-Fe intermetallic phase. The formation of thicker and undesirable intermetallics must be avoided to prevent joint embrittlement.

The line EDX scan across both joint interfaces are shown in Figures 17 and 18. The most remarkable finding throughout this project concerns the migration of Mg towards the surface of the joint, which is clearly seen in Figure 17. The same phenomenon was seen in the sample shown Figure 11, where the decomposition of Al-Mg alloy occurred close to the joints. Clearly, there is a sharp jump in the Mg concentration at the Al-Mg/Al-6061 interface, which was also found in the sample shown in Figure 12.

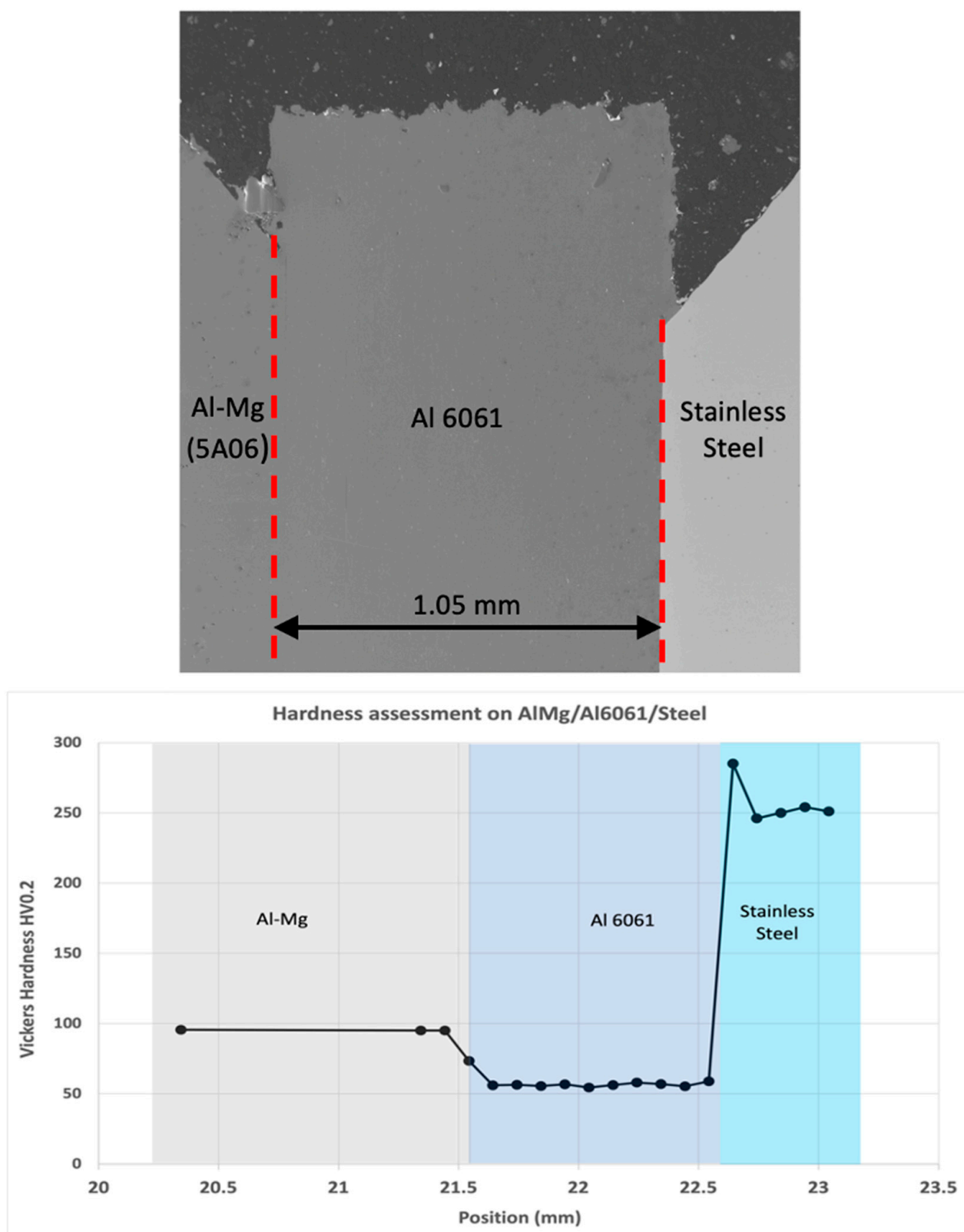


Figure 15. Cross-section (SEM) and hardness profile of bonded Al-Mg (5A06) to stainless steel (1Cr18Ni9Ti).

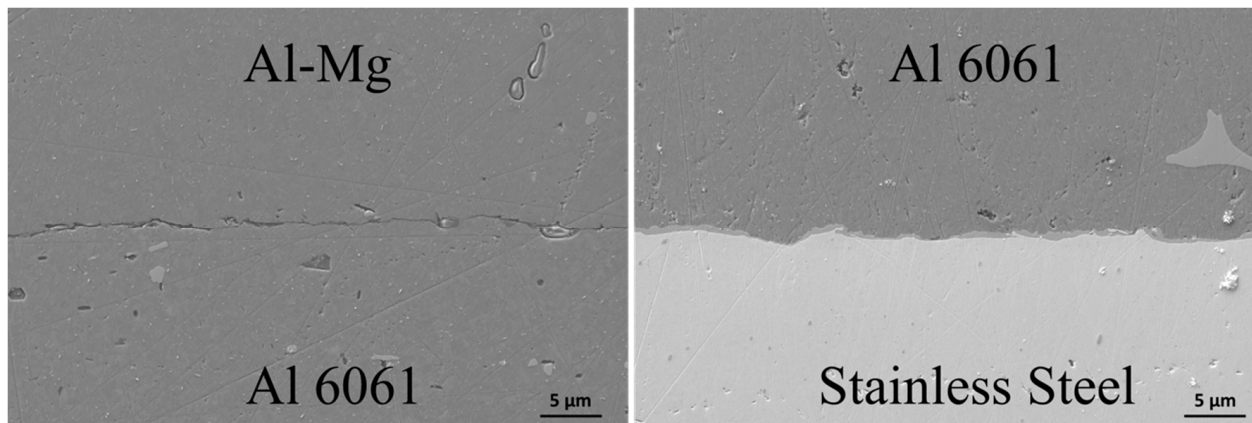


Figure 16. SEM micrographs show joint continuity in a bonded Al-Mg (5A06) to stainless steel (1Cr18Ni9Ti) sample. The presence of Al-6061, as a buffer layer, prevented the migration of Mg toward the surface of stainless steel.

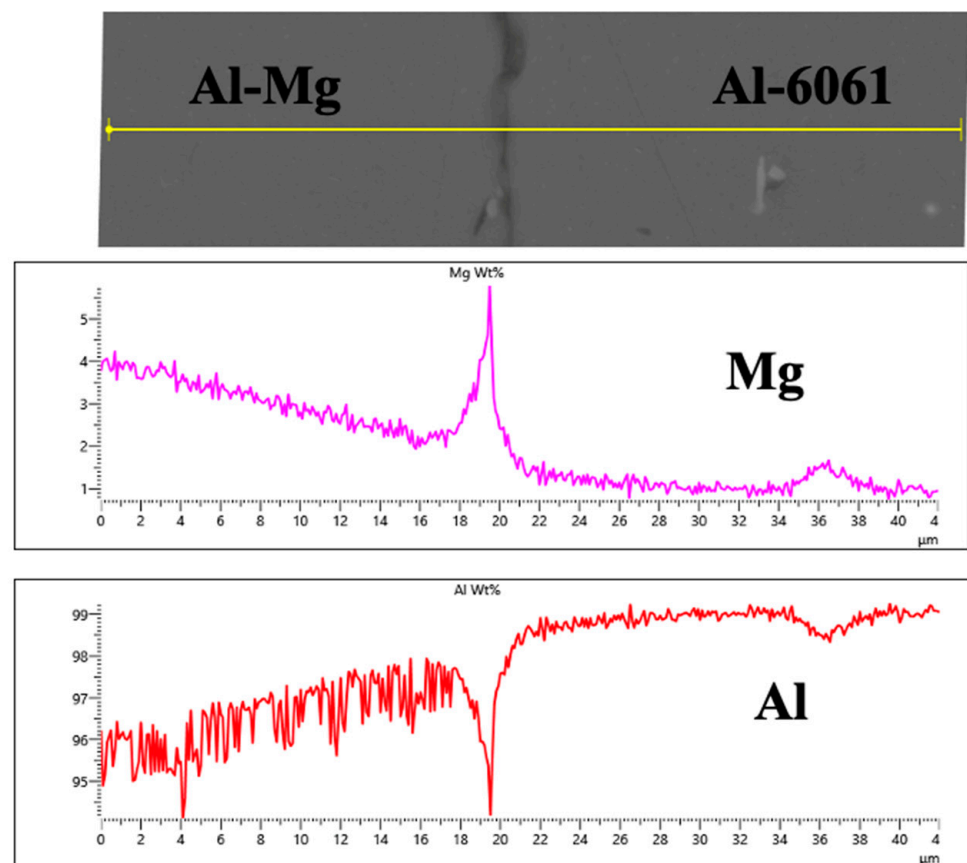


Figure 17. Migration and high concentration of Mg at joint interfaces occur when diffusion bonding or brazing Mg-rich aluminium alloys.

The sudden increase in the concentration of magnesium at the joint interface indicated the possibility of so-called “reverse diffusion”, where the solute diffuses towards the region with a higher concentration. This is to be studied in future work.

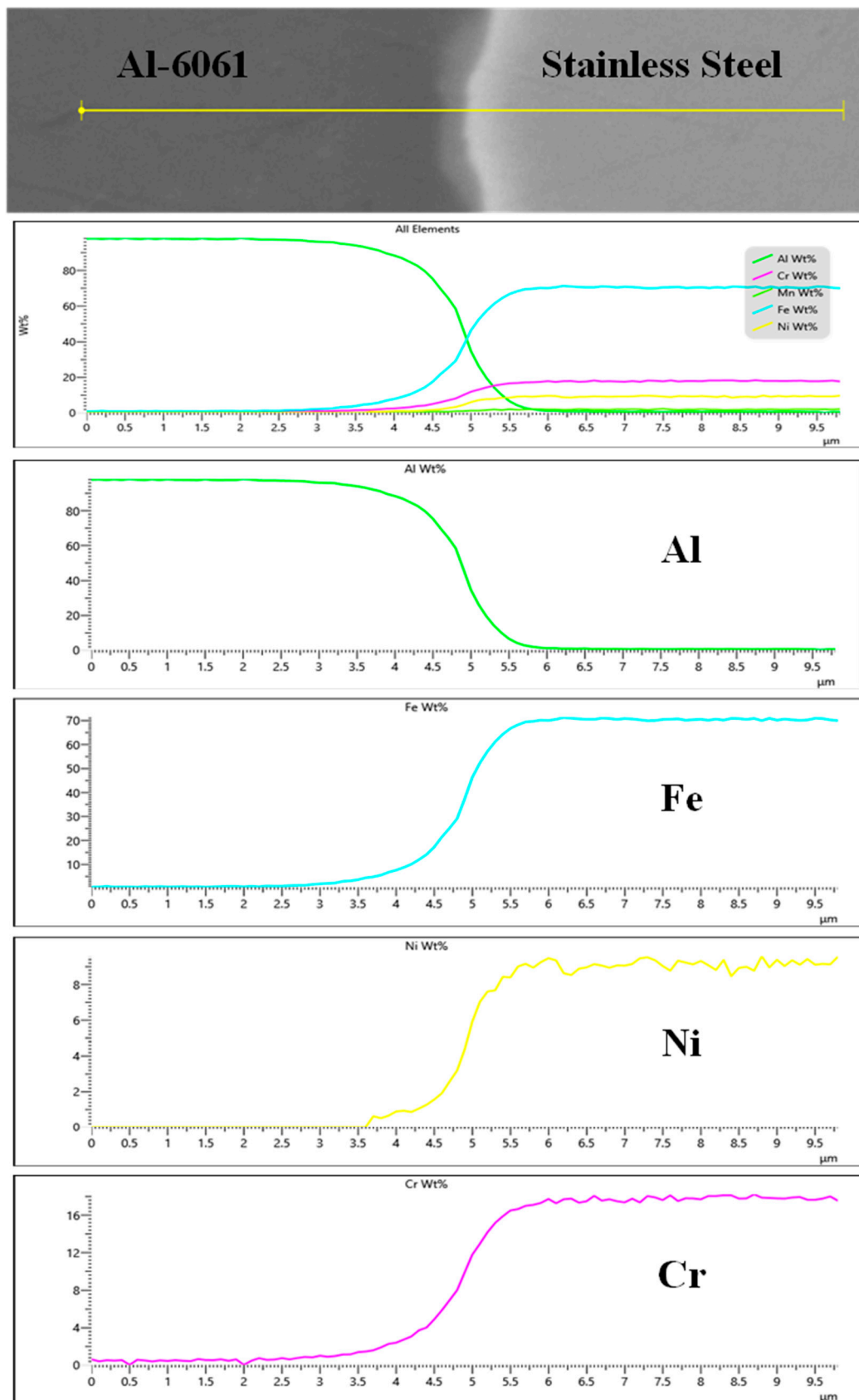


Figure 18. Distribution of main elements around an Al-6061/steel joint. Smooth transitions in the concentration profiles were achieved using the optimum bonding temperature and time developed in this work.

5. Conclusions

1. Diffusion bonding and brazing of Mg-rich aluminium alloys to stainless steels proved extremely difficult due to (a) high concentration of magnesium at the joint interface, (b) low melting point of Al-Mg alloys (5A06), and (c) formation of stable and tenacious magnesium oxide on the faying surfaces;
2. At least eleven bonding methods were used to join the Al with 6 wt.% Mg to stainless steels. Some methods required multistage processing to achieve bonds with tensile strengths up to 226 MPa;
3. The complexity and costs of the double or triple stage bonding cycles make these processes impossible or unviable for joining large industrial components. Any post-bonding heat-treatment to restore the Al-Mg initial mechanical properties would make the process even more complex and could deteriorate the joint performance. Therefore, the project was focused on optimisation of the bonding parameters for a single stage bonding cycle;
4. Based on the outcome of this project, the following bonding processes are recommended for joining the Al with 6 wt.% Mg alloy (5A06) to stainless steels in a vacuum ($<10^{-2}$ Pa).

Option 1: single-stage process:

Diffusion bonding of the Al-Mg alloy to stainless steel, using a 1 mm thick Al-6061 interlayer, should be carried out at 510 °C to 530 °C for 30 min to 1 h under 5 MPa pressure in a vacuum below 10^{-2} Pa. The exact bonding temperature and time depend on the size of the components and the heating rate. The bonded samples are expected to withstand tensile loads up to 90 MPa.

Option 2: double-stage process:

In the first stage, the Al-Mg alloy must be clad with an Al-6061 plate (minimum 1 mm thick) by diffusion bonding (550 °C, 2 h, 5 MPa) or other methods such as roll-bonding. In the second stage, the Al-6061 side is diffusion bonded to stainless steel using the same conditions given for the above single-stage process. The bonded samples are expected to withstand tensile loads up to 140 MPa.

Emerging new technologies and recommendation for future work: The use of more complex facilities, capable of removing surface oxides in an inert gas followed by diffusion bonding in vacuum without exposure to atmosphere, is recommended [14]. Impulse Pressure-Assisted Diffusion Bonding (IPADB) is a new and promising process to join alloys with tenacious and stable surface oxides [15]. Finally, Spark Plasma Sintering might be used to break up the magnesium oxide on the Al-Mg alloy and enhance its bonding to stainless steel [16].

Author Contributions: Conceptualization, A.A.S.; methodology, A.A.S.; validation, A.A.S. and C.Z.; formal analysis, A.A.S. and M.Z.M.; investigation, A.A.S., C.Z. and M.Z.M.; resources, A.A.S. and P.X.; data curation, A.A.S., M.Z.M. and C.Z.; writing—original draft preparation, A.A.S.; writing—review and editing, A.A.S. and M.Z.M.; project administration, A.A.S. and P.X.; funding acquisition, P.X. and A.A.S. All authors have read and agreed to the published version of the manuscript.

Funding: This project was funded by the National Natural Science Foundation of China (Grant No. 51905348), Shanghai International Science and Technology Cooperation Fund Project (Grant No. 19190730100) and The Open University, UK (Budget No. E-00477-25).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The key data are presented in the manuscript; for more details contact the corresponding author.

Acknowledgments: The authors are grateful for the technical assistance from the workshop engineers and electron microscopy unit at The Open University-UK.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Wang, P.; Chen, X.; Pan, Q.; Madigan, B.; Long, J. Laser welding dissimilar materials of aluminium to steel: An overview. *Int. J. Adv. Manuf. Technol.* **2016**, *87*, 3081–3090. [\[CrossRef\]](#)
2. Mathieu, A.; Shabadi, R.; Deschamps, A.; Suery, M.; Mattei, S.; Grevey, D.; Cicala, E. Dissimilar material joining using laser (aluminium to steel using zinc-based filler wire). *Opt. Laser Technol.* **2007**, *39*, 652–661. [\[CrossRef\]](#)
3. Song, J.L.; Lin, S.B.; Yang, C.L.; Ma, G.C.; Liu, H. Spreading behavior and microstructure characteristics of dissimilar metals TIG welding-brazing of aluminium alloy to stainless steel. *Mater. Sci. Eng. A* **2009**, *509*, 31–40. [\[CrossRef\]](#)
4. Liu, F.C.; Dong, P. From thick intermetallic to nanoscale amorphous phase at Al-Fe joint interface: Roles of friction stir welding conditions. *Scr. Mater.* **2021**, *191*, 167–172. [\[CrossRef\]](#)
5. Hirose, A.; Matsui, F.; Imaeda, H.; Kobayashi, K.F. Interfacial Reaction and Strength of Dissimilar Joints of Aluminum Alloys to Steels for Automobile. *Mater. Sci. Forum* **2005**, *475*, 349–352. [\[CrossRef\]](#)
6. Hirose, A.; Imaeda, H.; Kondo, M.; Kobayashi, K.F. Influence of Alloying Elements on Interfacial Reaction and Strength of Aluminum/Steel Dissimilar Joints for Light Weight Car Body. *Mater. Sci. Forum* **2007**, *539*, 3888–3893. [\[CrossRef\]](#)
7. Li, Y.; Hatsujiro, H.; Eiichi, S.; Zhang, Y.; Zhang, Z. Morphology and structure of various phases at the bonding interface of Al/steel formed by explosive welding. *J. Electron Microsc.* **2000**, *49*, 5–16. [\[CrossRef\]](#) [\[PubMed\]](#)
8. Haghshenas, M.; Abdel-Gwad, A.; Omran, A.M.; Gökçe, B.; Sahraeinejad, S.; Gerlich, A.P. Friction stir weld assisted diffusion bonding of 5754 aluminium alloy to coated high strength steels. *Mater. Des.* **2014**, *55*, 442–449. [\[CrossRef\]](#)
9. Howlader, M.M.R.; Kaga, T.; Suga, T. Investigation of bonding strength and sealing behavior of aluminium / stainless steel bonded at room temperature. *Vacuum* **2010**, *84*, 1334–1340. [\[CrossRef\]](#)
10. Huang, Z.; Yanagimoto, J. Dissimilar joining of aluminium alloy and stainless steel thin sheets by thermally assisted plastic deformation. *J. Mater. Process. Technol.* **2015**, *225*, 393–404. [\[CrossRef\]](#)
11. Zhao, Y.B.; Lei, Z.L.; Chen, Y.B.; Tao, W. A comparative study of laser-arc double-sided welding and double-sided arc welding of 6mm 5A06 aluminium alloy. *Mater. Des.* **2011**, *32*, 2165–2171. [\[CrossRef\]](#)
12. Lv, S.; Cui, Q.; Huang, Y.; Jing, X. Influence of Zr addition on TIG welding-brazing of Ti-6Al-4V to Al5A06. *Mater. Sci. Eng. A* **2013**, *568*, 150–154. [\[CrossRef\]](#)
13. Shirzadi, A.A.; Laik, A.; Tewari, R.T.; Orsborn, J.; Dey, G.K. Gallium-assisted diffusion bonding of stainless steel to titanium; microstructural evolution and bond strength. *Materialia* **2018**, *4*, 115–126. [\[CrossRef\]](#)
14. Habisch, S.; Böhme, M.; Peter, S.; Grund, T.; Mayr, P. The Effect of Interlayer Materials on the Joint Properties of Diffusion-Bonded Aluminium and Magnesium. *Metals* **2018**, *8*, 138. [\[CrossRef\]](#)
15. AlHazaa, A.; Haneklaus, N.; Almutairi, Z. Impulse Pressure-Assisted Diffusion Bonding (IPADB): Review and Outlook. *Metals* **2021**, *11*, 323. [\[CrossRef\]](#)
16. Alhazaa, A.; Albrithen, H.; Hezam, M.; Ali, M.; Alhwaimel, I.; Estournes, C. Interfacial Evolution of Al7075/Cu/Ti-6Al-4V Processed by Spark Plasma Sintering. *Metals* **2022**, accepted.