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Mechanical and metallurgical properties of diffusion bonded AA2024 aluminium alloy and commercial grade copper

G. Mahendiran[1], V Balasubramanian[2] and T. Sethilvelan[3]

[1]Sri Ganesh College of Engineering & Technology Mullodai, Puducherry India

[2]Departmental of Manufacturering Engireering, Annamalai University, Annamalai Nagar – (P.O.), India – 608 002

[3]Department of Mechanical Engineering, Pondicherry Engineering College, Pondicherry. India – 605 014.

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ABSTRACT

This paper reveals the mechanical and metallurgical properties of the dissimilar joints of AA2024 aluminium alloy and commercial grade copper alloy fabricated by diffusion bonding process. Three diffusion bonds with varying diffusion layer thickness were fabricated and study the effect of diffusion layer thickness on mechanical and metallurgical properties. Mechanical properties such as lap shear strength, bonding strength and hardness are evaluated, Metallurgical characteristics such as diffusion layers thickness, phases in diffusion layer, elemental composition of diffusion layers were analysed. From this investigation, it is found that the diffusion bond with a medium diffusion layer thickness (in between very minimum to very maximum) exhibits superior mechanical properties.

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Introduction

Product requirements in the aircraft, missile, electronics, nuclear, aerospace and commercial fields have given rise to many new and demanding service conditions. To meet the stringent requirements of these exacting operations, it is not only necessary to develop new material and also it is important to find new methods to fabricate them into useful engineering components. One such fabrication technique is solid phase joining process [1]. Wide ranges of processes are now grouped under the generic term of solid phase joining, among this diffusion bonding method alone join two surfaces by interatomic diffusion and microscopic deformation without any distortion and distinguishable interfaces [2].

Diffusion bonding is relatively simple joining process which is controlled by three important process parameters, which are bonding temperature, bonding pressure and holding time and these three parameters are interrelated and thus have an effect on each other. The bonding temperature should be between 50% and 70% of the melting point of either of the lowest melting point material.

The use of elevated temperature will aid the interdiffusion of atoms across the interface of the bond and assist surface deformation. The bonding pressure should be high enough to ensure a tight contact between the joining surfaces, moreover it should be sufficient to aid in the deformation of surface asperities and to fill all the voids in the bonding zone. Also, the holding time should be sufficient for an intimate contact to be formed and for he diffusion process to take place. However, excessive holding time may lead to degradation of physical and chemical properties of the bonds [3].

Copper (Cu) and Aluminium (Al) alloys have good characteristics such as high electrical conductivity, thermal conductivity, machinability and low casting costs and hence widely applied in electronics and electrical power industry, electrical appliances, machinery and automobiles [4]. The application of copper and aluminium alloys to heat exchange for cooling the electronic component is the most common. Many times the parts being cooled are attached to Cu or Al base plates and its leads to make an attempt to join copper-aluminium dissimilar materials.

However, the refractory oxide films of Cu and Al results in inclusion at the weld metal during fusion welding. Moreover, the conventional fusion welding technique causes several thermal cracking and easy formation of brittle intermetallic in the joints produced.

This makes the performance of joints very poor and makes it very difficult to obtain satisfactory joints. Therefore, Cu/Al dissimilar materials are to be joined by diffusion bonding technique [5].

This paper reveals the mechanical and metallurgical properties of the dissimilar joints of AA2024 aluminium alloy and commercial grade copper alloy fabricated by diffusion bondingg process.

Experimental Work

AA2024 Al alloy and commercial grade copper alloy plates of 5 mm thickness were cut to the required dimensions (50 mm x 50 mm) by power hacksaw cutting and grinding. The chemical composition and mechanical properties of base metals are presented in Table 1. The optical micrographs of base metals are displayed in Fig.1.

Tele: 04142-234345

E-mail addresses: mahe1967@yahoo.com

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(a) AA2024 Aluminium alloy

(b) Commercial grade Copper alloy

Fig.1 Optical micrographs of base metals

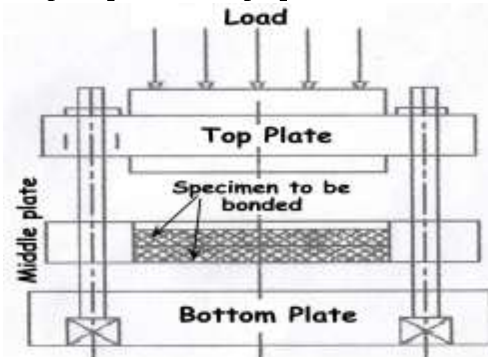


Fig.2 Configuration of the diffusion bonding die setup

The polished and chemically treated specimens of a pair (Mg/Al) were stacked in a die made up of 316L stainless and the entire diffusion bonding die setup, shown in Fig.2. and its inserted into a vacuum chamber. The heating chamber was fitted with super Kant Hal resistance heating wire band contains a water cooling coil to protect the O-rings and vacuum seals. The chamber is a PID (Programmable interface device) controlled furnace with a maximum temperature of 1473 K at an accuracy of ±1 K.

The temperature was measured using Platinum/Platinum-Rhodium thermocouple and with a non-indicating safety controller. The furnace is fitted with a removable bellows and a central shaft (Nimonic rod) that transmits the load to the specimens to be diffusion bonded. The central shaft has a groove along its length to suck the air from the chamber. The bottom side is flat and can be fixed in the hydraulic press. The size of the inside chamber is 100 mm in die and 100 mm in height. The furnace is insulated with high quality; lightweight, ceramic fiber blankets to improve the efficiency of the system.

The constant load or constant diffusion pressure was applied using a servo-controlled hydraulic press with maximum capacity 10 Ton. The pre determined pressure was applied using a pressure relief valve and the press was turned to auto mode after the required load was reached. A pressure switch is actuated by turning the press in to auto mode, which maintains the pressure level with an accuracy of ±100N. A hydraulic ram transfers the load to the central shaft of the heating a chamber and the specimens fixed in the dies. The load was measured using a load cell fixed in the hydraulic ram. Care was taken to fix the axis of the hydraulic ram and the central shaft of the heating chamber in linear axis.

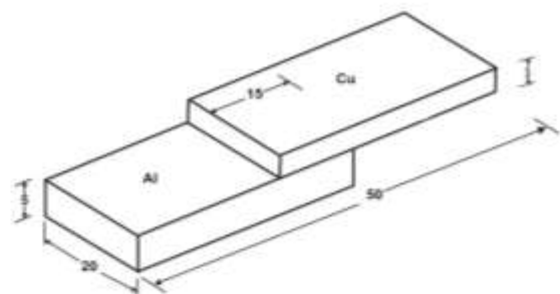
Water cooling is essential to maintain the temperature of the vacuum seals bellows at 473K. The cooling system has two tanks connected to heating chamber. The warm water (at 325 K) coming out of the heating furnace is pumped to the upper cooling tank and recirculated to bottom tank to maintain the inlet temperature at 300 K. The cooled water is then circulated to the heating chamber through a 0.25 hp pump.

A vacuum system containing a rotary pump connected in series is attached to the heating chamber, to maintain a vacuum level of 10^{-3} mm of Hg. The prepared specimens were heated up to the bonding temperature using induction furnace with a heating rate of 25°C/min; simultaneously the required pressure was applied. After that the samples were cooled to room temperature and then removed from the chamber. By this procedure, Al/Cu joints were fabricated using different combinations of bonding temperature, bonding pressure and holding time.

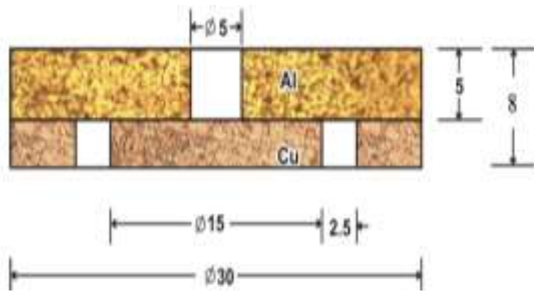
Lap shear tensile test was performed to evaluate shear strength of the joints and ram tensile test was conducted to evaluate bonding strength. As the joints were not large enough for normal lap shear testing and ram tensile testing a non-standard test was devised to measure the shear strength and bonding strength of the bonds. Similar specimens were used by other investigators also [6, 7].

The lap shear tensile specimens, as shown in FIg.3a were prepared from the Al/Cu diffusion bonded joints by a electric spark line cutting machine (Make: ELECTROICA, Japan; Model: Super Cut-734) was used at a cutting speed of 1.5 mm/min. Ram tensile specimens, as shown in Fig.3b were prepared from the Al/Cu diffusion bonded joints by a spark erosion machine (Make : ELEKTRA, Japan; Model : Cut-500). The ram tensile test setup is shown in Fig.3c. Both lap shear and ram tensile test were carried out in 100 kN capacity servo controlled universal testing machine (Make: FIE-BLUESTAR, India; Model: UNITEK 94100) at a constant ram speed of 5 mm/min. Reproducibility of data was verified by repeating the evaluation procedure.

A transverse cross section of the specimen was extracted from each joint and it was subjected to conventional metallographic preparations to reveal the various features of the joints. The microstructural analysis was carried out to measure the thickness of diffusion layer at the interface of the joints using light optical microscope (Make: MEIJI, Model : MIL-7100) and scanning electron microscope (Make : JOEL, Japan, Model : 5610 LV). The aluminium side was etched with Keller's solution and copper side was etched with a solution containing ethanol, concentrated HCl and $FeCl_3$ to reveal the microstructure.



a) lap shear tensile test specimens



b) Ram tensile test specimens

Fig.3 Dimensions of lap shear tensile and ram tensile test specimens

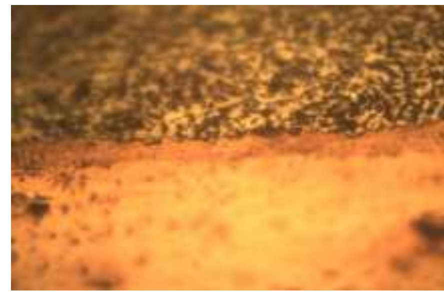
Vickers's microhardness testing machine (Make: SHIMADZU, Japan; Model: HMV-T1) was employed with 0.5 kg load for measuring the diffusion layer hardness. Five readings were taken along the interface of the joint at close promixity distance and mean values are used for further analysis. Microhardness was also measured across the joint (normal to the interface region) for the selected specimens to understand the hardness variation.

Energy Dispersive Spectrum (EDS) analysis was carried out using scanning electron microscope (Make: JEOL and Model : 5610LV) at high magnification to estimate the weight percentage of elements, which are diffused at the interface zone and its adjacent sides of the bonded joints. To identify the phase constitution near the interface of the diffusion bonded joints, samples were cut from the transition region of both the sides and XRD analysis was carried out. The XRD analysis was carried out in Theta-Theta (Vertical type), D/Max (Make : RIGAKU, Japan; Model : ULTIMA-III) with copper target under a working voltage of 40 kV and 40 mA working current. Scintillation counter detector was used with a scan range 3 to 154 deg (min. setup size 0.0002 deg). The results obtained are compared with data from the Joint Committee on Powder Diffraction Standards (JCPDS).

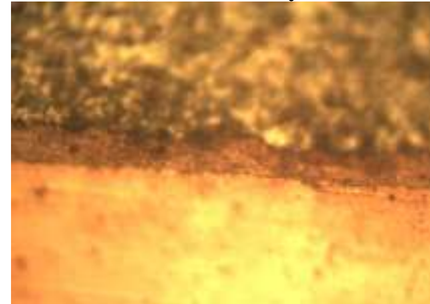
Results

The interface of Al/Cu bonds was analysed using OM, SEM, EDS and XRD instruments to reveal the characteristics of interface region. The observing orientation was perpendicular to the axis of the diffusion bonded joint. Fig.4 displays the OM and SEM micrographs of interface of the joints fabricated using the process parameters presented in Table 2. Microhardness was measured at the joint interface at five different locations and the average value is presented in Table 2. Shear strength and bonding strength of the joints were evaluated and the values (average of 3 results) are tabulated Table 2. EDS analysis was carried out at the interface and the results are presented in Fig.5. XRD analysis was carried out at the interface and results are presented in Fig.6. From the results presented in Table 2 and the Figs.4-6, the following observations are obtained.

(a) Thin layer



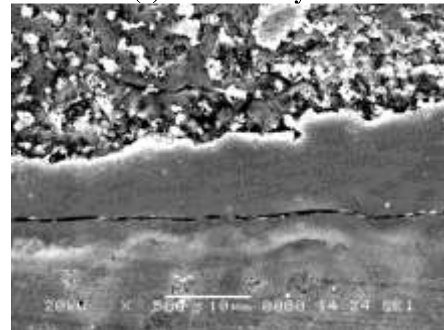
(b) Medium layer



(c) Thick layer

(d) Thin layer

(d) Medium layer



(e) Thick layer

Fig.4 Optical and SEM micrographs of Al/Cu bonds (a-c: OM; d-f; SEM)

(i) A very thin layer of thickness 1.39 μm was formed under the bonding temperature of 425^{0}C, holding time of 10 minutes and hardness of 50 Hv and exhibited shear strength and bonding strength of 26 MPa and 42 MPa respectively. The interface contains 42.9% of Cu and 57.07.% of Al along with the intermetallic phases of $MgAl_2O_4$, Al_2O_3, SiO_2, FeN, AlCuFe.

(ii) A medium thick diffusion layer of 7.99 µm was formed under the bonding temperature of 500^0C, holding time of 50 minutes and bonding pressure of 12 MPa. This interface recorded a hardness of 74 Hv and exhibited shear and bonding strength of 58 MPa and 98 MPa, respectively. The interface contains 60.6% of Cu and 39.4% of Al along with the intermetallic phases of BC, FeB, BN, FeC, $MgAl_2O_4$ and Fe_2O_3.

(iii) A very thick diffusion layer of 17 µm was formed under the bonding temperature of 550^0C, holding time 90 minutes and bonding pressure of 16 MPa. This interface recorded a hardness of 100 Hv and exhibited shear and bonding strength of 32 MPa and 54 MPa, respectively. The interface contains contains 63.05% of Cu and 36.95% of Al along with the intermetallic phases of $MgAl_2O_4$, SiO_2, Fe_7C_3, BC, BN, $MgAl_2O_4$ and $MgSiO_4$.

(iv) Bonding temperature, bonding pressure and holding time have directly proportional relationship with diffusion layer thickness (DL) and interface hardness (IH). Interface hardness is directly proportional to diffusion layer thickness.

(v) Diffusion of copper atoms should a directly proportional relationship with bonding temperature, bonding pressure and holding time. This is evident from weight percentage of aluminium elements at the interface (Table 2 – EDS results).

(vi) Maximum shear strength and bonding strength were exhibited by the joint which contained medium diffusion layer thickness (8 µm). Both the joints with thin (2 µm) and thick (12 µm) diffusion layers exhibited lower shear strength and bonding strength compared to the joints with medium diffusion layer.

(vii) Presence of BC and BN at the interface of the medium diffusion layer enhances the hardness and strength of the joints compared to joints with thin and thick layers.

Microhardness was also measured across the interface of Al/Cu joints (perpendicular to the diffusion layer) at the different locations and the results are presented in Fig.7. EDS analysis was also carried out at three different locations and the results are presented in Table 3. From the microhardness and EDS results, following inferences can be obtained.

(i) Hardness is maximum at the interface, irrespective of thickness of the diffusion layer. This may by due to the formation of intermetallic compounds at the interface.

(ii) Thick diffusion layer recorded maximum hardness at all locations. This may be due to higher level of bonding temperature, bonding pressure and holding time used to fabricate the joints.

(iii) Very near to the interface region (approximately 1mm from the interface region on both the sides), an appreciable reduction in hardness was recorded in all the joints. This may be due to the depletion of respective atoms, which are diffused into the interface region to form intermetallic compounds. This was confirmed by EDS results presented in Table 3.

(iv) The atomic radii of Cu and Al are 0.128 nm and 0.143 nm respectively. Since, the atomic radius of 'Cu' is less than 'Al', 'Cu' migrates more faster towards 'Al' side than 'Al' towards 'Cu' side in Al/Cu joints.

Discussion

The formation of diffusion layer depends on atomic diffusion. When the bonding temperature was increased to the required level, the joining processes have allowed the diffusion of all elements from both the sides quickly. This fact promotes the chemical joint (in all welding condition) between materials, when inter-diffusion between the materials is provided without the formation of voids and brittle phases such as intemetallic compounds. These findings are in agreement with Fick's second law, a partial differential equation describes that the rates at which atoms are redistributed in a material by diffusion [8]. The composition, extent, nature and properties of the phases originated during the welds, will control the resulting mechanical properties. The intermetallic compound grows steadily and gradually via enhanced temperature, at the bond region of dissimilar metal joints.

Element	Weight%
Al K	57.07
Cu K	42.93
Total	100.00

(a) Thin layer

Element	Weight%
Al K	39.70
Cu K	60.30
Total	100.00

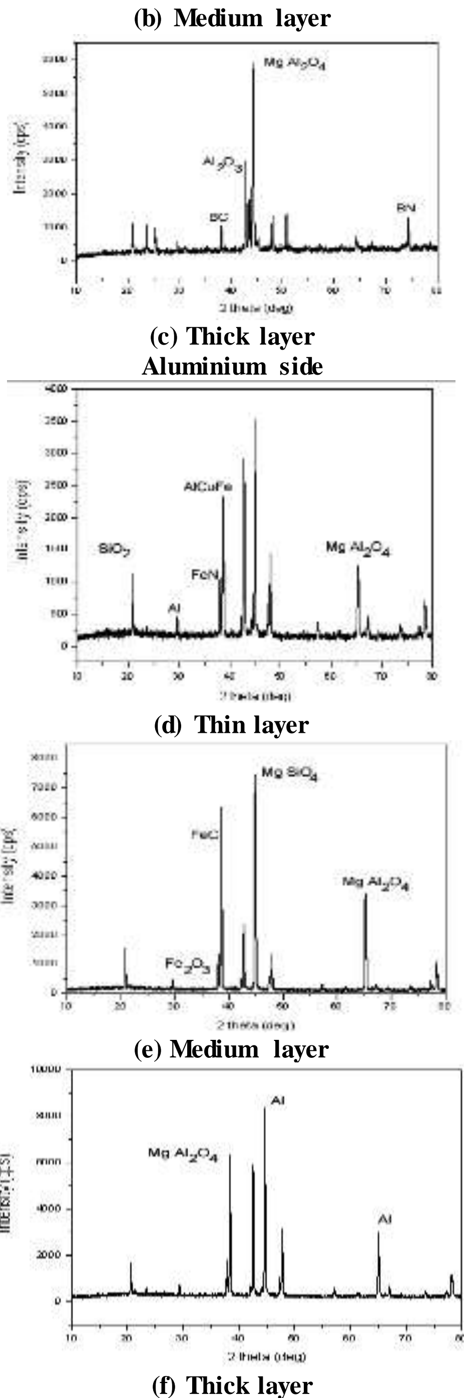
(b) Medium layer

Element	Weight%
Al K	36.95
Cu K	63.05
Total	100.00

(c) Thick layer

Fig.5 EDS results at the interface region of Al/Cu bonds Copper side

(a) Thin layer



(b) Medium layer

(c) Thick layer
Aluminium side

(d) Thin layer

(e) Medium layer

(f) Thick layer

Fig. 6 X-ray diffraction (XRD) patterns for Cu/Al bonds

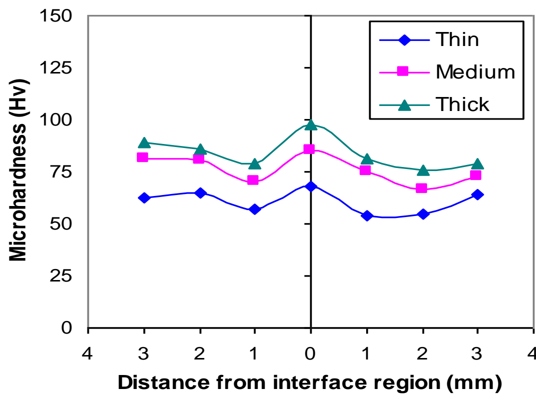


Fig.7 Microhardness survey across the interface region

The particle distribution of intermetallic compounds has no harmful effects on the joint performances; moreover, it can strength the joints. The intermetallic compounds have never joined up and formed a whole body; they also have no effect on the plasticity and strength of joints. But once they connect and thickness grows beyond 5 μm, the plasticity and strength of joints will obviously be decreased.

Also, the diffusion rate for Al atoms is much higher than that of Cu atoms at high temperature, an excess of Cu atoms diffuse across the interface into the aluminium side and forms cavities according to Kirkendall effect [9]. It leads to produce continuous cavities at the interface, resulting in a drastic reduction in the interface bonding strength level in the specimens bonded at maximum temperature.

The thickness of the intermetallic compound increases remarkably with holding time. Minimum holding time does not allow atoms to diffuse each other and hence diffusion layer thickness is very minimum. Increases of holding time to a maximum level increases grain boundary vibration this allows more atoms to diffuse on the other side. This leads to increase of diffusion layer thickness. Increase in hardness with increase in temperature and holding time can be attributed to the formation of intermetallic compounds as discussed above [10].

Effect of bonding temperature

Shear strength and bonding strength of the joints are increasing with increasing bonding temperature from 425^{0}C to 500^{0}C (Refer Table 2). At a low bonding temperature (425$_0$C), the shear strength and bond strength of the diffusion-bonded joint are low. This may be due to poor contact of the bonded surface and low thermal excitation. The bonding reaction is based on atoms diffusion and it is higher at higher bonding temperature. Bonding temperature improves the contact ratio and shear strength. Also, at low temperature, the flowability of metal is substantial yet yield strength. Also, at low temperature, the flowability of metal is substantial yet yield strength of the base materials still remains high which, leads to an incomplete coalescence of the mating surfaces [11]. Moreover, the grain boundary surface tension and grain boundary mobility are less at low temperature and these are the controlling factors for the initial movement of interface grain boundaries. At low temperature, boundary mobility is relatively low [12]. Also with increase in temperature (500^{0}C), the yeild strength of the joint decreases, this result in larger interfacial deformation, and the atomic diffusivity increases, results in easier and speeder chemical bonding. Therefore, the joint strength increases with increase in bonding temperature.

When diffusion-bonding temperature increases (500^{0}C), there is an increase in shear strength and bonding strength. Increase in diffusion bonding temperature promotes mass transfer of alloying elements across the interface, which is responsible for the increase in volume fraction of the reaction products; hence causes more embrittlement to the joints. However, plastic collapse of the mating surface asperities leads to intimated contact, which counter balances the embrittlement phenomena due to intermetallic phases; shear strength and bond strength naturally improves and attains maximum value [3].

In contrast, at high temperature the initial stages of bonding could involve migration of interface grain boundaries as above, the higher rate of grain growth would lead to rapid removal of evidence of the bond line and increases the strength nearer to the parent metal. Increase in the bonding temperature to maximum (550^{0}C), the thickness of the intermetallic compound increases

quickly. Quick increase in thickness of intermetallic compound leads to decrease in the strength and an increase in the brittleness of the joint. Further increase in temperature, the width of brittle intermetallics considerably increases and the embrittlement effect over-balances the positive effect obtained due to betterment in coalescence of faying surfaces [13]. So, both the shear and bond strength drops to a minimum value.

Effect of bonding pressure

From table 2, it can be inferred that the shear strength and bonding strength of the joints are increasing with increasing in bonding pressure, irrespective of bonding temperature and holding time. At low bonding pressure 8 MPa, shear strength and bonding pressure, contact is only at the protrusions on the bonded surface, so the contact rates and the strength of the bonded joint are lower. Generally, when the bonding pressure is applied, the points of contact between the two surfaces will expand almost instantaneously. When it is increased to 12 MPa, plastic deformation will develop at contact sites to increase the contact areas of clean surfaces and hence joint rate changes appreciably.

Further increase of bonding pressure to 16 MPa results in small decrease in shear strength and bonding strength. Increase in pressure influences re-crystallization temperature and deformation tends to enhance the contact of bond surface and rapid growth of re-crystallization. This will obviously increase the rate of interface contact and atoms are made to pass through this bonding interface. So, more diffusion paths are created due to movement of atoms. Copper has more melting temperature compared to Aluminium. Also, the diffusivity value of copper is greater than that of aluminium. This leads to increase in movement of more number of copper atoms towards the aluminium side. The movement of atoms will increase voids named as Kirkendall effect [9]. The voids produced during bonding would reduce the joint property. The property of the bonded joints also mainly depends on thickness of the intermetallic compounds, which was unaffected as pressure increases. The voids formed at the original interface will disappear as the contact area expands with time, because the stress within the contact zone will cause a plastic flow by either conventional creep or super plasticity. The smaller voids would be removed rapidly by diffusion [14].

Effect of holding time

High shear strength is obtained at a holding time of 50 minutes for Al/Cu bonds. Holding time has an effect on the creep of the protrusions and the quantity of atomic diffusion [15]. Shear strength and bonding strength of the joint increases with increasing holding time. If holding time is not sufficient to allow diffusion of atoms across the bond interface from the mating surfaces, the strength will be lower. The strength increased more rapidly with increasing holding time up to 50 minutes and then it decreases sharply. Longer holding times showed a continuing grain growth accompanied by a small increase in specific strength. The sharp decrease in strength was attributed to the growth of intermetallic compounds [16]. The thickness of the intermetallic compound increases remarkably with holding time and the tensile strength of the bond joint decreases.

Conclusions

The effect of diffusion bonding process parameters such as bonding temperature, bonding pressure and holding time on bond characteristics (diffusion layer thickness, interface hardness, shear strength and bonding strength) was analyzed in detail.

(i) The diffusion bonding process parameters such as bonding temperature, bonding pressure and holding time have shown that these parameters are directly proportional to the diffusion layer thickness and interface hardness.

(ii) Diffusion layer thickness has shown directly relationship with interface hardness. If diffusion layer thickness is lower, then the interface hardness is lower and vice-versa.

(iii) Maximum shear strength and bonding strength were exhibited by the joint which contained medium diffusion layer thickness (8 µm). The joints with thin (2 µm) and thick (12 µm) diffusion layers exhibited lower shear strength and bonding strength compared to the joints with medium diffusion layer.

(iv) A bonding temperature of 500^0C, bonding pressure of 12 MPa and holding time of 50 minutes are found to be the optimum diffusion bonding process parameters to attain maximum bonding strength in Al/Mg dissimilar joints.

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Table 1 (a) Chemical composition of the base metals

Base metal	*Si*	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Al	O	Pb	B	S
Aluminium (AA2024)	0.5	0.5	4.9	0.9	1.8	0.1	0.25	0.15	Bal	-	-	-	-
Copper (Commercial Grade)	-	0.007	Bal	-	-	-	-	-	0.14	0.092	0.001	0.018	<0.001

Table 1 (b) Mechanical properties of the base metals

Base Material	Density (Kg/m^3)	Ultimate Tensile Strength (MPa)	Elongation (%)	Shear Strength (MPa)	Hardness (Hv) @ 50g load
Aluminium (AA2024)	$2.7x10^3$	483	18	283	137
Copper (Commercial Grade)	$8.96x10^3$	344	14	323	65

Table 2 Characterisation results of Al/Cu bonds

Bond type	T (°C)	P (MPa)	t (min)	DL (μm)	IH (Hv)	SS (MPa)	BS (MPa)	EDS Results		XRD Results
								Cu	Al	
Thin	425	8	10	2	50	26	42	42.93	57.07	$MgAl_2O_4$, Al_2O_3,SiO_2, FeN,AlCuFe
Medium	500	12	50	8	74	58	98	60.30	39.70	BC, FeB, BN, FeC, Fe_2O_3, .$MgAl_2O_4$
Thick	550	16	90	12	100	32	54	63.05	36.95	$MgAl_2O_4$, SiO_2, Fe_7C_3, BC, BN, $MgAl_2O_4$

Table 3 EDS values taken across the interface region of Al/Cu bonds

Region	Thin		Medium		Thick	
	Cu	Al	Cu	Al	Cu	Al
Copper side (Location 1)	97.48	2.52	95.40	4.60	100.00	-
Interface (Location 2)	42.93	57.07	60.30	39.70	63.05	36.95
Aluminium side (Location 3)	5.46	94.54	9.92	90.08	9.39	90.61