Availability of *in-situ* reinforced active-transient liquid phase bond with good wettability for 70 vol. % SiCp/A356 composite using Al-Mg-Ga-Ti interlayer

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Abstract: For transient liquid phase bonding of as-cast 70 vol. % SiCp/A356 composite, Al-24Mg-16Ga-1Ti (wt.%) active braze was designed by adding Ti as melting point increaser (MPI) for Al into Al-Mg-Ga active alloy near eutectic composition to produce *in-situ* reinforcement by precipitating a fine and dispersive Ti-containing intermetallic compound as reinforcement within the solid-solution bond seam after isothermal solidification. Bonding was performed at three temperature levels of quasi-melting (450 °C), just fully melting (500 °C), and over melting (550 °C) for the braze under 0.5 ~ 1.75 MPa in argon. The braze was able to produce fine (less than 1 μm) and dispersive Ti-containing precipitated phase (Al-2Mg-1Ga-0.5Ti, at.%) as *in-situ* reinforcement only at 500 °C under medium pressure (1~1.5 MPa), increasing bond seam microhardness by 30 Hv over the matrix. The braze robustly showed good wettability for each temperature due to the matrix dissolution and detectable amorphous reaction products containing Mg and Ga on SiC surface. Higher temperature caused coarsening of needle-like Si-containing Al-Si-Ti precipitated phase while higher pressure produced blocky Al₃Ti with a shell of Mg and Ga. Sound joint produced at 500 °C under 1.5 MPa with 118.6 MPa shear strength (98.8% parent composite) fractured completely within the composite.

Keywords: Transient liquid phase bonding; Metal matrix composites (MMC); Melting point increaser (MPI); Interlayer composition design; *In situ* reinforced bond seam

1. Introduction

High volume fraction ceramic particle reinforced Al metal matrix composites (Al-MMCs) can be used as electronic packaging material due to low coefficient of thermal expansion and as structural material due to high abrasion resistance. However, Al-MMCs show poor wettability for fusion welding processes because of detrimental reaction products between SiC particle reinforcement and superheated molten Al matrix (Al₄C₃) [1]. Therefore, brazing and transient liquid phase bonding (TLP) were extensively attempted. In comparison to conventional brazing, TLP bonding using an interlayer containing sufficient melting point depressant (MPD) offers the following benefits: (1) omitting of flux for removing oxide film owing to undermining of substrate oxide caused by the diffusion of MPD into substrate [2-10], (2) achievement of solid-solution bond seam by isothermal solidification, and (3) lower bonding temperature for avoiding potential thermal degradation of substrate materials. The micro-interface between Al-MMCs composite and metallic interlayer can be subdivided into two types: matrix/metal (M/M) and particle/metal (P/M) interfaces [11]. TLP bonding of Al-MMCs suffers from particle segregation and poor wettability at P/M interface. To inhibit particle segregation, thinner Cu interlayer and Al-containing interlayer

were recommended. To improve the wettability at P/M interface, mechanical route (introducing ultrasonic vibration by Yan and Xu [12-18]) and metallurgical route (adding active element by the present authors [19-23]) were reported. The TLP bonding process for Al-MMCs using an interlayer containing the active element capable of reacting with ceramic reinforcement, e.g., Ti, is called active-transient liquid phase bonding (A-TLP) by the current author [19-23].

In recent years, some researchers succeeded in reinforcing bond seam by externally adding ceramic particle into monolithic interlayer [24-31]. In contrast, *in-situ* reinforcing phase formed within the bond seam has clean interface and resultant excellent wettability with bond seam matrix [32]. To obtain fine, dispersive, stable and refractory intermetallic compound (IMC) as *in-situ* reinforcing phases in TLP bond seam, the current authors proposed that melting point increaser (MPI), which must have a higher melting point than Al and has no eutectic reaction with Al (e.g., Ti or Zr), should be added into interlayer together with MPD via uniform precipitation of Al-MPI IMC phase during isothermal solidification, and succeeded in achieving fine Al-Si-Ti IMC phase as *in-situ* reinforcement for TLP bond seam for 10 vol. % SiCp/A356 composite using Al-Cu-Ti braze foil [20].

For high volume fraction SiC reinforced Al-MMCs, the present study aims to optimize interlayer composition design route (i) by adding substantial amounts of gallium (29 °C melting point, 20 wt.% solubility in aluminum) to significantly decrease melting point of the active braze, soften heated braze [33], and especially to assist Mg to wet matrix and SiC at low temperature in the early bonding stage, (ii) by increasing Mg (as active MPD) content to extend actual reaction time for wetting at each P/M interface during isothermal solidification [34-37], and especially (iii) by adding active MPI (e.g. Ti) for Al into braze to improve wettability at P/M interface and to simultaneously reinforce bond seam by *in-situ* producing small and dispersive Ti-containing IMC particulates as reinforcement. The modified A-TLP process being able to achieve both favorable wettability and *in-situ* reinforcement within bond seam could be called *in-situ* A-TLP bonding process, in which the “in-situ” and “A” terms are used to describe bond seam and interface features, respectively. Effects of interlayer composition, temperature and pressure on wettability and availability of *in-situ* reinforced bond seam in A-TLP bonding of high volume fraction SiC particle reinforced aluminum matrix composite were investigated.

2. Experimental materials and procedures

The materials used were 70 vol. % SiCp/A356 composites, produced by vacuum infiltration of molten matrix into a pre-sintered bulk. The as-cast parent composite was cut into slices of a dimension of 7.5×7.5×3 mm and 15×15×3 mm by wire cutting, used as top and bottom pieces in lap configuration, respectively. The novel active braze foil of Al-24Mg-16Ga-1Ti of 60 ~ 70 μm in thickness (near Al-32Mg-16Ga eutectic composition, wt.%) was developed by inductively melting pure Al, Mg, Ga, and Al-5Ti master alloy at 750 °C in flowing argon for 30 min, immediately followed by rapid solidification with a rotating copper wheel. Compared with Al-24Mg-1Ti of melting range from 447 °C (solidus temperature, T_S) to 521 °C (liquidus temperature, T_L), the developed Al-24Mg-16Ga-1Ti foil showed quite low melting range from 383 °C (T_S) to 497 °C (T_L) due to the addition of a lot of Ga. The newly developed interlayer foil is characterized by combined addition of Mg (as active MPD) and Ti (as active MPI), too low solidus temperature (being able to provide liquid Mg as active element at so low temperature below 450 °C of Al-Mg eutectic temperature) and the absence of Cu and Si as MPD.

Before bonding, the specimen surface was abraded with dry 240 grit diamond abrasive disk, dry 240 and 600 grit silicon carbide abrasive papers in sequence, and then ultrasonically cleaned in alcohol. According to the measured melting range of the Al-24Mg-16Ga-1Ti braze foil (383 ~ 497 °C), bonding temperature was set at three levels: quasi-melting (450 °C), just fully melting (500 °C), and over melting (550 °C). Such low bonding temperatures (450 and 500 °C in quasi or just melting state) were selected to avoid expulsion of liquid braze out of interface before wetting reaction. Also, the quasi-melting state condition would be beneficial to evaluating the feasibility of wetting reaction between the composite and Mg and/or Ga at low temperature, because at 450 °C (Al-Mg eutectic temperature, higher than Al-Mg-Ga ternary eutectic temperature of 400 °C) large amounts of Mg and Ga could be in liquid state, permitting the two active elements to wet SiC, although some Ti atoms were still in solid state in the form of a IMC. In contrast, the over melting state condition (550 °C) was selected to facilitate wetting reaction for each active element (including Ti) [22].

For bonding pressure, during the bonding process for 30 min in flowing Ar of 5 L/min, high pressures (0.5, 1, 1.5, 1.75 MPa) were applied to the bond assembly to avoid Mg oxidation by introducing close contact between the interlayer with high Mg content and the composite with high hardness and creep strength. Several copper blocks were placed on the top of the assembly to apply the above compressive stress to the faying surface during heating cycle. The used bonding apparatus was schematically given in a previous article [21].

After bonding, back-scattered electron (BSE) and energy dispersive x-ray spectroscopy (EDS) imaging were used to investigate alloying element distribution, wettability and *in-situ* reinforcing phase formation within the solid-solution bond seam newly formed after isothermal solidification. For the best joint with such a bond seam consisting of the newly precipitated IMC particulate phase containing Ti and a solid solution matrix, Vickers microhardness of the bond seam and A356 matrix was measured with 10 gf for 15 s to identify whether it could be *in-situ* reinforcing phase for bond seam. Also, the wetting reaction between the bond seam and SiC particle (P/M interface) was examined by transmission electron microscopy (TEM). The joints were evaluated by shear test at a loading speed of 0.5 mm/min, and the fracture path was examined to verify the wettability and reinforcing effect of the *in-situ* formed fine and dispersive Ti-containing IMC particulate phase on bond seam matrix.

3. Results and discussion

3.1 Availability of *in-situ* reinforced A-TLP bond with good wettability at different temperatures

Fig. 1 shows the effects of temperatures on *in-situ* reinforced bond seam formation and wettability under constant pressure of 1 MPa. The retained thickness of bond seam decreased to 60, 48 and 20 μm at 450 °C, 500 °C and 550 °C from initial thickness of the braze foil of 70 μm, respectively, showing that the active braze foil could not be completely expelled out of interface, but high temperature led to more obvious expulsion of liquid phase. Especially, the developed Al-24Mg-16Ga-1Ti active braze foil was able to produce *in-situ* reinforcement particulates within the bond seam only when it was in just fully melting state (500 °C), and it showed good wettability for each bonding temperature.

For the availability of *in-situ* reinforcement within A-TLP bond seam, only at 500 °C, fine (less

than 1 μm) and dispersive Ti-containing IMC particulate phase (Al-2.0Mg-1.0Ga-0.5Ti, at.%) was formed via precipitation of Ti as MPI within the newly formed bond seam of Al-based solid solution matrix containing a little Mg (Al-1.1Mg-0.3Ga, at. %), as shown in Fig.1(d, f). Actual Ti content in the precipitated IMC particulate phase should be higher than the detected value of 0.5 at.% due to the too small size (less than 1 μm) of the Ti-containing IMC phase compared to the much larger reaction volume of the EDS composition readings. While at low temperature of 450 $^{\circ}\text{C}$, a coarse network phase containing more Mg content (Al-17.6Mg-6.1Ga-1.1Ti, at.%) was present within the bond seam due to the uncompleted isothermal solidification, leading to embrittlement of the bond seam. And also, at high temperature of 550 $^{\circ}\text{C}$, cluster of coarse needle-like Al-35Si-18Ti (at.%) phase, which was characterized by high Si content, high Ti content and coarse size ($\sim 10\ \mu\text{m}$ in length), was formed in the bond seam of Al-1.5Si (at.%) solid solution (Fig. 1(g, h)). The fact of the presence of Si in the bond seam confirmed that the A356 matrix has been remarkably dissolved caused by diffusion of Mg and Ga as MPD into the A356 matrix. Nevertheless, the Mg and Ga diffused into the matrix was hard to observe like in Fig. 1a due to (i) rapid diffusion rate of Mg and Ga in the matrix at higher temperature, and (ii) reduction in the source for Mg and Ga by preferential expulsion of the low melting point part of braze with higher contents of Ga and Mg under a pressure (see small thickness of the retained bond seam in Fig. 1g). Also, the pressure-induced partitioning of the melting interlayer by preferentially expelling the low melting point portion with higher contents of Ga and Mg in the braze would be sufficient for increasing Si and Ti contents in the thinned bond seam and thus for forming coarser reinforcements with higher Si and Ti contents. Accordingly, both of the excessive matrix dissolution and expulsion of partial liquid phase at higher temperature of 550 $^{\circ}\text{C}$ should be responsible for the coarsening of the Si-containing Al-Si-Ti IMC phase. The above results showed that bonding temperature also had a significant effect on the composition, size and distribution of MPI-containing IMC phase within A-TLP bond seam via varying the retained liquid amount and the actual liquid composition during isothermal solidification, depending on expulsion behavior of braze and the dissolution behavior of matrix.

The presence and refinement of the *in-situ* precipitated Ti-containing IMC particulate phase only when the braze was in just fully melting state (500 $^{\circ}\text{C}$) can be analysed as follows. According to the model of *in-situ* reinforcement formation within TLP bond seam during isothermal solidification just for MPI-containing interlayer, which was proposed by Zhang et al (the current author) [20], the presence of the precipitated Ti-containing IMC phase should result from the concentration-precipitation-termination-engulfment cycle, namely when local concentration of liquid MPI (Ti) at isothermally solidified front increases to a critical value with the movement of liquid-solid interface during isothermal solidification, an Al-MPI IMC would be precipitated. While MPI-free interlayer is unable to uniformly distribute IMC because liquid MPD (an eutectic former) would be pushed towards bond center by isothermally solidified front, leading to the absence of precipitation of Al-MPD IMC in the early formed isothermal solidified region of solid-solution matrix. In contrast, Al-MPD IMC would be present only at center line region with a large size. For the refinement of the Ti-containing IMC particulate phase at 500 $^{\circ}\text{C}$, the possible reasons could be summarized as follows: (i) thick retained liquid phase (48 μm) after expulsion would be beneficial to dispersing liquid MPI, (ii) so low temperature would be beneficial to decreasing the critical value of Ti concentration for precipitation at local area at isothermally solidified front, and (iii) the limited matrix dissolution resulting from pressure-induced

partitioning of the melting interlayer and low temperature inhibited the presence of a number of Si atoms in liquid phase.

For wettability, the developed Al-24Mg-16Ga-1Ti active braze foil was able to wet the composite well at each bonding temperature. Even in quasi-melting state of the braze (at 450 °C some Ti atoms should still be in solid state in the form of a IMC), no gaps were observed at both M/M and P/M interfaces at 10,000 magnification (Fig. 1a-c). The favorable wettability at M/M interface was attributed to slight dissolution of A356 matrix adjacent to bond seam, which was achieved by the Mg diffusion into matrix (see mapping scanning analysis for Mg in Fig. 1a). At the dense P/M interface, the presence of Mg at entire P/M interface (5 at.% for major gray zone with little other active elements and 10 at.% for minor white zone) and a detectable Mg-containing covering layer on SiC surface (Fig. 1c) showed that Mg was able to wet the SiC particles. At 500 °C, gaps were hard to observe at both M/M and P/M interfaces. On the other hand, the dissolution of matrix was obvious and isothermal solidification of bond seam matrix was completed, which were beneficial to wetting matrix and eliminating Mg-containing brittle phase, respectively. In over-melting state of the braze (550 °C), the active braze was not expelled completely although the thickness of bond seam was reduced to 20 µm. For M/M interface, large amounts of Si present in the coarse needle-like Al-Si-Ti phase demonstrated that the A356 matrix has been significantly dissolved. The partial expulsion of molten interlayer and enhanced diffusion of Mg at higher temperature led to absence of residual Mg in matrix adjacent to bond seam. At the produced dense P/M interface, only trace amount of Ti (0.5 at.%) with Si (41.1 at.%) and C (2 at.%) was detected, but Ga and Mg could not be detected. This is because Ti (as MPI) is hard to diffuse into A356 matrix and then is able to react with SiC for sufficient time, while Mg and Ga can be consumed by pressure-induced partitioning of the melting interlayer and can readily diffuse into A356 matrix as MPD at higher temperature. The difference of distribution of Mg, Ga and Ti at dense P/M interface at different temperatures suggested that Ga and Mg could improve wettability at P/M interface primarily at lower temperature range (450~500 °C), while Ti could be able to react with SiC at high temperature for long time. As a competitive mechanism, enhanced diffusion of Mg and Ga into matrix at higher temperature would consume the amount of Mg and Ga to react with SiC.

For relatively detailed wettability examination at P/M interface, TEM analysis on the joint produced at 500°C under 1.5 MPa showed that a quite thin amorphous reaction zone of ~100 nm in thickness was formed between bond seam and SiC particle (Fig. 2), where considerable Mg (12 at.%) and Ga (3 at.%), 9 times higher than that within bond seam matrix, were detected. The fact that Mg and Ga tended to arrange to the SiC surface suggested that Mg played the most important role in improving wettability at P/M interface at low temperature with the aid of Ga by reducing Ts from 450 °C (Al-Mg eutectic temperature) to 383 °C and softening the active braze foil in the early stage of heating. So, the achievements of excellent wettability at P/M interface and *in-situ* reinforcing phase formation within bond seam can be attributed to Mg (as active MPD) and Ti (as active MPI) additions, respectively.

3.2 Availability of *in-situ* reinforced A-TLP bond with good wettability under different pressures

Fig. 3 shows the effects of pressure on *in-situ* reinforced bond seam formation and wettability at

the constant temperature of 500 °C. Overall, it can be seen that the developed Al-24Mg-16Ga-1Ti was able to produce *in-situ* Ti-containing reinforcing phase within bond seam of Al-based solid solution matrix under medium pressure (1 and 1.5 MPa). The retained thickness of bond seam decreased to 50, 48, 30, 25 µm at 0.5, 1, 1.5 and 1.75 MPa from initial braze foil thickness of 70 µm respectively, showing that the developed Al-24Mg-16Ga-1Ti was not completely expelled out of interface under each pressure, but high pressure led to more obvious expulsion of molten braze.

For the availability of *in-situ* reinforcement within A-TLP bond seam, just under the medium pressure levels (1 and 1.5 MPa), fine and dispersive Ti-containing IMC particulate phase (Al-2.2Mg-1.1Ga-0.3Ti, at.%) was formed via precipitation of Ti as MPI within the bond seam of Al-based solid solution matrix containing a little Mg (Al-1.3Mg-0.3Ga, at.%) (Fig. 3c~e and Fig. 3f and h)). As a result, the measured Vickers microhardness of the bond seam (84 Hv) produced under 1.5 MPa was higher than that of A356 matrix (50 Hv), as shown in Fig. 3g. The facts of (1) fine and dispersive nature of the Ti-containing IMC particulate phase (Fig. 3h) and (2) higher microhardness of the bond seam (Fig. 3g) suggested that the small Ti-containing IMC particulate phase can be considered as *in-situ* reinforcement for the bond seam of Al-based solid solution matrix. While under lower pressure of 0.5 MPa, Ti-containing precipitated phase within bond seam was not observed due to the dispersion of liquid Ti in thick retained liquid phase, and under higher pressure of 1.75 MPa, coarsening of Ti-containing precipitated phase within bond seam was observed due to concentration of liquid Ti in thin retained liquid phase. Interestingly, under 1.75 MPa, large amounts of blocky phase (20~50 µm) consisting of Al₃Ti core and a thin shell with a small amount of Ga (2.4 at.%) and Mg (0.5 at.%) were present in the bond seam, implying that the formation of *in-situ* reinforcing phase could be divided into two steps: previous precipitation of refractory Al-Ti IMC phase and subsequent precipitation of Mg and Ga around the previously precipitated refractory Al-Ti IMC phase.

For wettability, also just under medium pressure levels (1 and 1.5 MPa), the absence of gaps at M/M and P/M interfaces confirmed that the developed Al-24Mg-16Ga-1Ti interlayer was able to wet the composite well. While under lower (0.5 MPa) and higher pressure (1.75 MPa), although entire M/M interface was dense, a number of interfacial gaps (e.g. 10~50 µm in length by 10 µm in width for 1.75 MPa) were obviously observed at some P/M interfaces. The results showed that the bonding pressure had considerable effect on wettability at P/M interface. It could be explained as high pressure-induced partitioning of the melting interlayer with a wider melting range. In detail, higher pressure induced obvious partitioning of the melting interlayer by preferentially expelling low melting point portion of the melting braze (mainly consisting of Mg and Ga) out of interface (Fig. 3k), resulting in (i) loss of Mg and Ga as active elements for SiC during incubation period for wetting reaction and (ii) decrease in contacting time (e.g. actual wetting time) of SiC fixed at surface with liquid phase during isothermal solidification duration. Lower pressure would (1) reduce actual pre-contacting area at interface in early stage during heating cycle because the high volume fraction SiC particle (70 vol.%) reinforced Al matrix composites possess quite high hardness and quite high creep strength, and (2) weaken the effect of pressure on mechanically puncturing oxide film on the molten braze foil with high Mg content. Moreover, in addition to TEM observation result, the presence of gaps at P/M interface in the joint produced under 1.75 MPa (despite large amounts of the retained Ti being sufficient to form blocky Al₃Ti, Fig. 3i and j) demonstrated that in preference to Ti, Mg and Ga can act as active elements for SiC at such a low temperature of 500 °C once again.

Based on the information presented for different pressures and temperatures, the possible constituent for wetting reaction at P/M interface, reaction degree and reaction products can be roughly summarized. The melting range of the Al-24Mg-16Ga-1Ti foil employed was 383 °C ~ 497 °C. At 500 °C, compared with refractory Ti, not only Ga but also Mg was fully and preferentially activated in liquid state, thus both of them tended to react with SiC because EDX analysis results showed that Mg and Ga were concentrated at P/M interface rather than within the bond seam. Furthermore, the amorphous feature of the reaction product observed in TEM analysis showed that the wetting reaction was insufficient for the formation of a stable intermetallic, such as Mg₂Si [35,36].

3.3 Joint shear strength and fracture paths

3.3.1 Joint shear strength

For the effect of temperature on joint shear strength (Fig. 4a), regardless of pressure (1 and 0.5 MPa), the maximum shear strength (90.6 and 53.6 MPa) was achieved when the active braze was in just fully melting state (500 °C). Moreover, joint shear strength could be further improved for each temperature by 20~40 MPa by increasing pressure from 0.5 to 1 MPa (Fig. 4a). A more detailed investigation of the effect of pressure on joint shear strength showed that the maximum shear strength could be increased once more by continuously increasing pressure up to 1.5 MPa, which reached 118 MPa (Fig. 4b), quite close to that of the as-cast parent composite of 120 MPa (as high as 98% of parent composite shear strength). But joint shear strength tended to decrease as exceeding 1.5 MPa. The results showed that (i) the newly developed Al-24Mg-16Ga-1Ti braze is dramatically superior to previous Al-33Cu-6Mg-1Ti braze [22] in joint strength (118 vs 62 MPa), and (ii) unlike monolithic metal, the maximum shear strength can be achieved in slight melting state of the active braze (500 °C) under medium pressure (1.5 MPa).

3.3.2 Effect of temperature on fracture path

Fig.5 and Fig.6 show the fracture path of the joints produced at three temperature levels (quasi-melting, just fully melting and over melting for Al-24Mg-16Ga-1Ti braze) under 1 MPa for 30 min at low and high magnifications, respectively. The total length of the fracture path was 7.5 mm, equal to the size of top component. Three kinds of possible fracture paths (within composite, within bond seam and along bond/composite interface) were summarized in Table 1 with joint shear strength for comparison. Only at 500 °C (in just fully melting state), favorable fracture path within parent composite could be partially available up to 4.5 mm (Fig.5b and Fig.6c and d), accompanying with partial fracture path along interface between bond seam and parent composite. For the joint produced at lower (450 °C) and higher (550 °C) temperatures, the fracture paths could not propagate into the composite but were partially within bond seam and partially along the interface between bond seam and parent composite (see Table 1). The reasons for the fracture path within bond seam at 450 °C and 550 °C were the presence of brittle network Si-free compound due to the uncompleted isothermal solidification and coarse Al-Si-Ti phase due to the remarkable dissolution of matrix, respectively. The reasons for the fracture path along the interface between bond seam and parent composite were poor wettability at P/M interface due to insufficient extent of reaction on SiC at 450 °C and insufficient amounts of active elements of Mg and Ga at 550 °C (caused by expulsion). So, temperature had an effect on actual wetting degree, final bond seam microstructure and fracture path primarily by changing actual Mg and Ga content in liquid braze

during isothermal solidification (depending on both matrix dissolution and expulsion of low melting point portion of braze) and activated degree for wetting reaction. Although at 500 °C, proper Mg and Ga content after expulsion and limited dissolution of matrix enabled fracture to propagate partially into the composite matrix, the presence of fracture path along bond/composite interface indicated that wettability could not be improved very well only by varying temperature to balance expulsion with residue of the braze.

Table 1 Effect of temperature on distribution of fracture path (1 MPa×30 min)*

Temperature / °C	Within composite	Within bond seam	Along interface	Shear strength / MPa
450	-	5.3 mm (71%)	2.2 mm (29%)	49.4
500	4.5 mm (60%)	-	3 mm (40%)	90.6
550	-	3.8 mm (51%)	3.7 mm (49%)	61.4

(* 450 °C: quasi-melting, 500 °C: just fully melting, 550 °C: over melting; Total length of fracture path was 7.5 mm)

3.3.3 Effect of pressure on fracture path

Fig. 7 and Fig. 8 show the fracture path of the joints produced under different pressures at 500 °C for 30 min at low and high magnifications, respectively. The primary fracture path can be divided into two types: within composite and along interface, as summarized in Table 2. The absence of the fracture path within bond seam showed that Mg and Ga could readily diffuse into A356 matrix at 500 °C, resulting in solid-solution bond seam matrix.

Only under 1.5 MPa, favorable fracture path within composite adjacent to bond seam was completely available (Fig. 7c and Fig. 8c and d). As a result, this joint showed the maximum shear strength of 118.6 MPa (98 % of parent composite), as shown in Fig. 4b. The favorable fracture path and such high strength efficiency indicated that both the interfacial bonding and the bond seam were so strong, comparable to the composite adjacent to bond seam. As shown in Fig. 3f and h, when the braze was in just fully melting state at 500 °C and medium pressure (1.5 MPa) was used, the strong interfacial bonding can be attributed to excellent wetting result without interfacial gaps, and the strong bond seam can be attributed to the newly formed Al-based solid solution matrix after isothermal solidification and the presence of dispersive precipitation of fine Ti-containing particulate phase as *in-situ* reinforcing phase for the bond seam. Such strong interfacial bonding at 500 °C under 1.5 MPa showed that the bonding condition combination could offer proper (i) actual Mg and Ga content in liquid phase during isothermal solidification, and (ii) reacting period and extent by balancing between (a) expulsion and residue of the braze and (b) matrix dissolution and inhibiting coarsening of Ti-containing phase.

However, under too low (0.5 MPa) and too high (1.75 MPa) pressure, the favorable fracture path through the composite was not observed in the joints, as the fracture path in these joints was primarily along initial interface (Fig. 7a and Fig. 8a for 0.5 MPa, Fig. 7d and Fig. 8e for 1.75 MPa). According to the microstructure of these joints (Fig. 3b and i), the bad fracture path along interface can be attributed to a number of interfacial gaps at P/M interface. The presence of interfacial gaps at P/M interface, lower joint shear strength (53.6 MPa for 0.5 MPa pressure and 80.6 MPa for 1.75 MPa pressure), the bad fracture path along bond/composite interface and the absence of the fracture path within bond seam indicated that too low and too high pressure

primarily deteriorated wettability, but not bond seam matrix. As discussed in Section 3.2, the pressure would influence wettability by affecting (i) initial contact in solid state, (ii) mechanical disruption of oxide film on molten braze, and (iii) actual content of Mg and Ga in residual liquid phase after expulsion under considerable pressure required for initial contact.

Table 2 Effect of pressure on distribution of fracture path (500 °C × 30 min)*

Pressure / MPa	Within composite	Within bond seam	Along interface	Shear strength / MPa
0.5	-	-	7.5 mm (100%)	53.6
1	4.5 mm (60%)	-	3 mm (40%)	90.6
1.5	7.3 mm (97%)	-	0.2 mm (3%)	118.6
1.75	-	0.5 mm (7%)	7 mm (93%)	80.6

(* 500 °C: the Al-24Mg-16Ga-1Ti braze is in just fully melting state; Total length of fracture path is 7.5 mm)

4. Conclusions

The Al-24Mg-16Ga-1Ti (wt.%) active braze was developed for 70 vol. % SiCp/A356 composite to improve wettability and to produce *in-situ* reinforcement within a solid-solution bond seam after isothermal solidification. Also, the required bonding conditions were investigated. The main conclusions can be summarized as follows.

(1) The developed Al-24Mg-16Ga-1Ti active interlayer was able to produce *in-situ* reinforced A-TLP bond with good wettability (even at P/M interface) when the braze was in just fully melting state (500 °C) and medium pressure (1.5 MPa) was used. In this case, the maximum shear strength of 118.6 MPa (reaching 98.8% of parent composite shear strength) was achieved and the fracture path was completely within the parent composite matrix adjacent to the bond seam.

(2) The above mentioned *in-situ* reinforcement for bond seam, namely fine Ti-containing IMC phase being able to dispersively precipitate within newly formed bond seam of Al-based solid solution matrix after isothermal solidification, was obtained only at 500 °C under proper pressure (1~1.5 MPa), which was a fine (less than 1 μm) and dispersive Si-free Al-2Mg-1Ga-0.5Ti (at.%) precipitated particulate phase. The precipitation and refinement of the *in-situ* reinforcing phase within bond seam can be attributed to Ti addition as MPI and relatively low temperature (500 °C) at which the required critical value of MPI for precipitation would be low.

(3) The fine and dispersive nature of the Ti-containing IMC particulate phase, higher microhardness of the bond seam (by 30 Hv over the A356 matrix) and favorable fracture path within composite matrix demonstrated that the fine Ti-containing IMC particulate phase can be considered as *in-situ* reinforcement for bond seam of Al-based solid solution matrix containing a little Mg (Al-1.3Mg-0.3Ga, at.%).

(4) For the effect of temperature and pressure on the availability of *in-situ* reinforcing phase within bond seam, too high temperature (550 °C) led to coarsening of Al-Si-Ti phase within bond seam due to excessive matrix dissolution and expulsion, while too low temperature (450 °C) resulted in the uncompleted isothermal solidification of bond seam matrix. While for the 0.5 MPa case, Ti-containing precipitated phase within bond seam was not observed due to the dispersion of liquid Ti in thick retained liquid phase under lower pressure, and for the 1.75 MPa case, coarsening of Ti-containing precipitated phase within bond seam was observed due to

concentrating of liquid Ti in thin retained liquid phase under higher pressure.

(5) For the effect of temperature and pressure on wettability, the developed Al-24Mg-16Ga-1Ti active braze robustly showed good wettability at both M/M and P/M interface for each temperature (even at 450 °C) under the moderate pressures (1 ~ 1.5 MPa). The wettability at M/M interface was achieved by A356 matrix dissolution. While the wettability at P/M interface was achieved by a quite thin amorphous reaction zone of ~100 nm in thickness formed between bond seam and SiC particle, where considerable Mg (12 at.%) and Ga (3 at.%), 9 times higher than that within bond seam matrix, were detected. Too low pressure (0.5 MPa) deteriorated wettability primarily due to the insufficient previous contact between the active braze with high Mg content and the composite with high hardness and high creep strength, while too high pressure (1.75 MPa) deteriorated wettability primarily due to pressure-induced partitioning of the melting interlayer with a wide melting range by preferentially expelling low melting point portion of the braze (mainly consisting of Mg and Ga). The effect of pressure on wettability is more significant compared with that on bond seam matrix microstructure.

(6) Both TEM observation result and the presence of gaps at P/M interface in the joint produced under 1.75 MPa (despite large amounts of the retained Ti being sufficient to form blocky Al_3Ti) demonstrated that Mg and Ga can act as active element for SiC at such a low temperature of 500 °C, in preference to Ti. While the active MPI of Ti not only inhibited the liquid expulsion under the moderate pressures, but also acted primarily as a nucleating agent of *in-situ* IMC reinforcement for solid-solution bond seam.

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Figure captions

- Fig.1. Excellent wettability without interfacial gap at each temperature, evolution of both bond seam matrix and in situ reinforcement with increasing temperature (1 MPa $\times$ 30 min): uncompleted isothermal solidification at 450 °C (a-c); achievement of fine and dispersive Si-free Ti-containing IMC phase at 500 °C (d-f); cluster of coarse needle-like Al-Si-Ti phase at 550 °C (g, h)
- Fig.2. TEM analysis showing quite thin amorphous reaction product ($\sim$ 100 nm in thickness) containing Mg and Ga at P/M interface (500 °C $\times$ 1.5 MPa $\times$ 30 min)
- Fig.3. Effect of pressure on wettability and in-situ reinforcing phase formation in bond seam (500 °C $\times$ 30 min): favorable wettability and lack of in-situ Ti-containing precipitated phase for 0.5 MPa (a, b), favorable wettability and dispersive in-situ Ti-containing precipitated phase for 1 and 1.5 MPa (c-h), poor wettability and coarsening of *in-situ* Ti-containing precipitated phase and obvious expulsion of liquid braze at fillet for 1.75 MPa (i-k)
- Fig.4. Effect of temperature (a) and pressure (b) on joint shear strength showing the maximum shear strength (quite close to the parent composite) can be achieved in just fully melting state of the active braze (500 °C) under medium pressure (1.5 MPa)
- Fig.5. Effect of temperature on joint fracture path at low magnification: partially within bond seam and partially along bond/composite interface at 450 °C (a) and 550 °C (c); presence of favorable fracture path within parent composite matrix adjacent to bond seam at 500 °C (b)
- Fig. 6. Effect of temperature on joint fracture path observed at high magnification at the locations indicated in Fig. 5: partially within bond seam and partially along bond/composite interface at 450 °C (a, b) and 550 °C (e, f); presence of fracture path within parent composite matrix adjacent to bond seam at 500 °C (c, d)
- Fig.7. Effect of pressure on joint fracture path at low magnification: along bond/composite interface at 0.5 MPa (a) and 1.75 MPa (d); partially and completely within parent composite adjacent to bond seam at 1 MPa (b) and 1.5 MPa (c), respectively.
- Fig.8. Effect of pressure on joint fracture path observed at high magnification at the locations indicated in Fig. 7: mostly along bond/composite interface at 0.5 MPa (a, b) and 1.75 MPa (e, f); completely within parent composite matrix adjacent to bond seam at 1.5 MPa (c, d)

Table captions

- Table 1 Effect of temperature on distribution of fracture path (1 MPa $\times$ 30 min)*
- Table 2 Effect of pressure on distribution of fracture path (500 °C $\times$ 30 min)*