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Influences of solid-state brazing conditions on microstructure and tensile shear force of Ag-Cu-P brazed copper sheets

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Abstract. This study aimed to examine the tensile shear force and microstructure of copper sheets when Ag-Cu-P filler metal was applied for the joints involved in the production of laminated micro-channel array (LMA) devices. The properties of the microstructure of the Ag-Cu-P/copper brazed joints were examined following a process which included furnace brazing, with the parameters comprising temperature, holding time, and loading pressure. Analytical tools included an energy dispersive spectrometer (EDS), scanning electron microscope (SEM) and X-ray diffraction analysis (XRD). In addition, tensile tests were performed to measure the tensile shear force. It was difficult to observe the Cu_3P which forms along with the layer between the Ag-Cu-P filler metal and the copper sheet when the brazing temperature was only 580 ℃ since this allowed gaps to form within the joint, thus producing very low tensile shear force results. When the brazing temperature is increased to 620 ℃ along with longer holding time and increased pressure, this allows a thicker phase of Cu_3P to form in the brazing joint. With a holding time of 30 minutes and pressure of 12.173 kPa, tensile shear force was maximized since this condition produced thick Cu_3P phase. It is apparent that it is possible to produce a brazed joint which has an average shear force of 736.27 N under these conditions as described above. Within the joint could be found a Cu_3P phase with no brittle phase.

1. Introduction

The large-scale processing of mass and energy through laminated micro-channel array (LMA) devices is known as micro-channel process technology (MPT). [1–2]. In comparison with conventional fluidic technology the most significant advantage of MPT is the very high the ratio of surface area to volume, permitting heat and mass transfer to take place very quickly within the micro-channels as the distances over which diffusion taking place are much shorter. This allows micro-channels to provide energy and chemical systems of reduced weight and size [3]. MPT is applied to an ever greater extent in numerous industrial application, such as the recuperation of waste heat, and portable heat exchanger [4].

In recent years, MPT technology has been made using mechanical processes, or by selective laser melting. Under mechanical methods, the structures are made by cutting sheets very accurately and then joining the stacked sheet layers to make the MPT device by fusion joining or by brazing and diffusion bonding [5]. In fusion welding, sheets are joined around the edges, ensuring that the micro-channels are not inevitably in direct contact with each other, and thus the total welding area is not large. These particular MPT devices are not expected to endure high pressure, while a further weakness is the development of the heat affected zone (HAZ), which can undergo considerable change to the bonded material microstructure, which ultimately causes the loss of mechanical properties [6].

Earlier studies have presented a number of methods such as, diffusion bonding or fusion welding. These approaches have all been used to form the joints in manufacturing MPT devices [7–8]. The most widely used approach in MPT production is diffusion bonding. This method is widely used because it ensures that the micro-channels are tightly held together. In addition, there is no liquid phase involved in the process, and the procedure can employ parameters which do not result in changes to the microstructure. However, lengthy bonding time is required for diffusion bonding.

In this study, the solid-state diffusion brazing of copper sheets using Ag-Cu-P filler metal is proposed as another alternative for LMA fabrication. The research involves brazing using varying parameters of holding time, brazing temperature, and loading pressure. The microstructure and tensile shear force of the brazed joint were assessed using SEM and EDS, while tensile tests were performed to evaluate the tensile shear force of the joint. The effects of changes in the three parameters upon the characteristics of the joints were subjected to detailed examination. The relationship between the microstructure and the mechanical properties of the joint is discussed in this paper. The results of this study should prove to be useful in establishing the set of manufacturing processes which can subsequently be employed in the development of a range of LMA devices.

2. Experiment

2.1. Materials and methods

For the solid-state brazing, the base material used in the study was commercially pure copper at a thickness of 0.350 mm. The samples were divided into rectangular plates with 10×27.7×0.35 mm in dimensions. Ag-Cu-P with a thickness of 0.250 mm was employed as the filler metal. In the experiment, rectangular plates of the filler metal were cut to a size of 10×5×0.25 mm. Furnace brazing was carried out using a BOREL Swiss SA No. S0908 model. The solid-state brazing process was performed under a hydrogen atmosphere to prevent the oxidation. Following the solid-state brazing process, the microstructure was examined and the composition of the brazing joints was assessed using SEM (JEOL model JSM-6510LV, Japan). An EDS (Inca, Oxford, UK) was also employed for the analysis. Testing of the tensile shear force of the brazing joints was carried out at room temperature and a crosshead speed of 0.1 mm/min, using Shimadzu AG-100 universal testing machine. The phases in the filler metal and brazed joints were identified using XRD (Bruker D8 Discover), and Cu Kα was chosen as the X-ray source.

3. Results and Discussion

3.1. Ag-Cu-P filler metal microstructure

The microstructure of Ag-Cu-P filler metal is presented in Figure 1 (a-e). Among the phases shown, the white one is associated with silver, the light gray color comprises of copper, and the dark gray area is Cu_3P compound. Chemical composition analysis can be employed to determine the exact constituents and distribution of the major phases, as displayed in the Table 1.

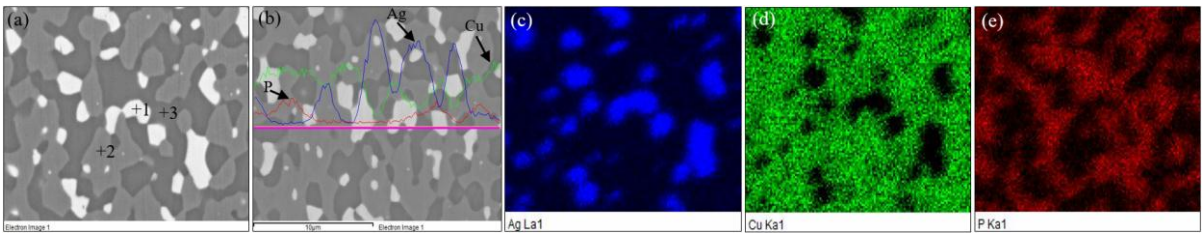

Figure 1. (a) Characteristics of the Ag-Cu-P filler metal microstructure; (b) Profile analyses of the filler metal by EDS linear scanning; Element distribution maps for the filler metal generated via EDS, including (c) Silver; (d) Copper and (e) Phosphorus.

3.2. Brazed joint microstructure characterization

The microstructure of the brazed joint between the copper sheet and the filler metal is presented in Figure 2. The brazing conditions consisted of a temperature of 620 ℃ for 30 minutes at a loading pressure of 12.173 kPa. In this case the wetting of the filler metal with the copper sheet was very good, and no holes or cracks were found in the joint as shown Figure 2 (a-b). There are two distinct parts of the brazing joint, which are denoted using the Roman numerals I and II as shown in Figure 2 (a-b). The section marked as I consists of the continuous reaction layer which is nearest to the copper sheet, while the section marked II forms the center of the joint. The two parts, I and II consist mainly of the silver phase (in white, denoted by 1), while the copper phase is light gray denoted by 2, and the dark gray phosphorus phase is shown at point 3. The compositions of the marked zones shown in Figure 2 (b) tabulated in Table 1.

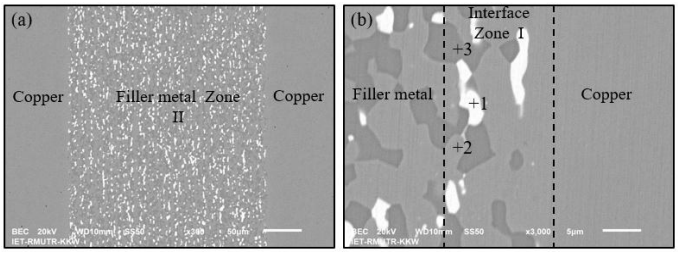

Figure 2. Brazed joint morphology (a-b), including (b) marked zones of the chemical composition for a brazed joint under conditions of 620 ℃, 30 minutes, and 12.173 kPa.

Table 1. The compositions of the marked zones from Figure 2 (b) and Figure 4 (a, f and k).

Solid-state brazing condition	Position	Composition (at. %)			Possible phase
		Ag	Cu	P	
Filler metal	1	89.00	10.44	0.56	Ag
	2	0.92	96.03	3.05	Cu
	3	0.05	70.36	29.59	Cu_3P
580 ℃; 10 min; 8.656 kPa	1	64.09	28.88	7.03	Ag
	2	1.47	95.87	2.66	Cu
	3	8.27	70.12	21.61	P
600 ℃; 30 min; 12.173 kPa	1	56.37	42.97	0.65	Ag
	2	0.79	79.70	19.51	Cu
	3	1.66	85.79	12.54	P
620 ℃; 30 min; 12.173 kPa	1	85.35	14.07	0.58	Ag-rich
	2	1.85	95.35	2.80	Cu-rich
	3	0.57	74.43	25.01	Cu_3P

Figure 3 (a-d) presents the EDS results in the form of compositional maps of the brazed joint along with the linear scanning profiles. The elemental spatial distribution can thus be specified. It is apparent that the brazed joint contains a continuous Ag-rich phase, a phase which is Cu-rich and the Cu_3P phase. On the basis of the binary Cu-P phase diagram along with the atomic ratio of the microstructure of the brazed joint, it can be stated that the reaction layer may comprise both Cu_3P and CuP_2 phase [9], However in this study there was no evidence of the formation of CuP_2 phase at the brazed joint. In this case it can be seen from the binary phase diagram for Ag-Cu that there was no intermetallic phase, thus confirming that Ag-Cu formed a solid solution and indicating that brazing can take place without the formation of an intermetallic phase. In the formation of Cu_3P, it is was occurred from phosphorus atoms in filler metal diffused towards the interface, and copper atoms in base material diffused towards the interface, thus leading to the formation of Cu_3P compound.

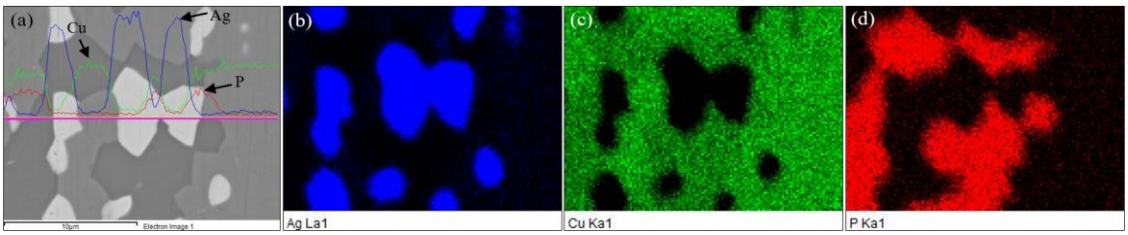

Figure 3. Distribution images of the corresponding elements area for brazed copper joints using Ag-Cu-P filler metal at 620 ℃ for 30 min and 12.173 kPa: (a) Linear scanning profiles; (b) Ag phase distribution map; (c) Cu phase, and (d) Cu_3P compound.

3.3. The influence of parameters for brazing temperature, holding time and loading pressure upon joint microstructure

Figure 4 (a, f and k) presents the SEM micrographs of the brazed copper joints which used Ag-Cu-P filler metal. The temperature, holding time, and loading pressure varied in each case. It is also apparent that the joint between the Cu substrate and the filler metal contained micro-voids throughout the interface, calling into question its efficiency, as shown in Figure 4 (a). This occurred as a result of the conditions: at a low temperature, short holding time, and low loading pressure (580 ℃ for 10 min and 8.656 kPa), it is not possible for the filler alloy to fully react with the copper sheet to create the continuous reaction layer. When the solid-state brazing conditions are improved (600 ℃ for 30 min and 12.173 kPa) the voids become less significant and it is possible to distinguish a continuous reaction layer, as shown in Figure 4 (f). When the conditions are further intensified (620 ℃ for 30 min and 12.173 kPa) the cracks disappear altogether, as shown in Figure 4 (k). Furthermore, the grain size of the region phase showed clear growth, and thus it can be inferred that the filler metal was able to fully react with the copper to create the continuous reaction layer, and consequently to form a Cu_3P compound (dark gray) of a suitable thickness.

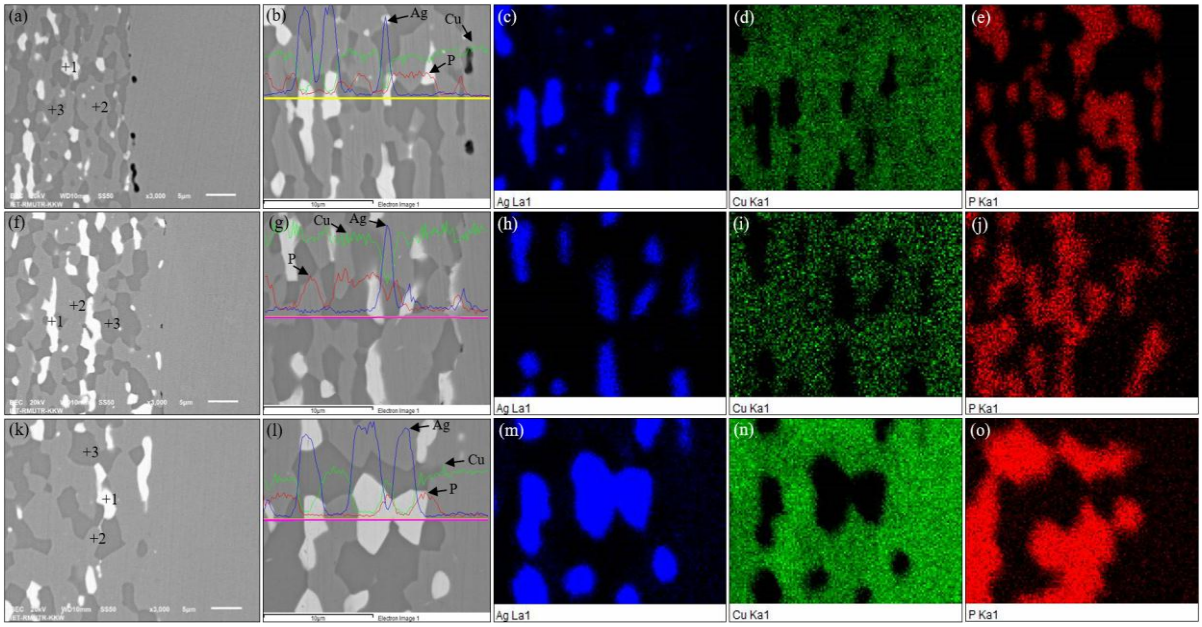

Figure 4. SEM micrographs showing the linear scanning profile and profiles of the EDS compositional maps of the brazed copper joint using Ag-Cu-P filler metal under varying solid-state brazing conditions: (a-e) 580 ℃ for 10 min and 8.656 kPa; (f-j) 600 ℃ for 30 min and 12.173 kPa, and (k-o) 620 ℃ for 30 min and 12.173 kPa.

Figure 4 (b-e, g-j and l-o) presents maps showing EDS composition and linear scanning profiles for brazed joints formed under varying solid-state brazing conditions. The images show the formation of Cu-rich and Ag-rich phases along with Cu_3P upon the brazed joints. The details of the elemental composition of the results from different brazing conditions are provided in Table 1 and can be seen also in Figure 4 (l-o). The result confirms that the alloy primarily consists of Cu-based solid solution, Ag-based solid solution and Cu_3P compound. Figure 5 (a-d) shows the XRD pattern of filler metal and brazing specimens after solid-state brazing with copper sheet on the experimental brazing variables encompassed the brazing temperature, holding time and loading pressure at 580 ℃ for 10 min and 8.656 kPa, 600 ℃ for 30 min and 12.173 kPa and 620 ℃ for 30 min and 12.173 kPa respectively. Obviously, Cu_3P is formed from the beginning of forming process of the filler metal. However, after solid-state brazing at high temperature (620 ℃ for 30 min and 12.173 kPa) grains of Cu_3P phase were larger.

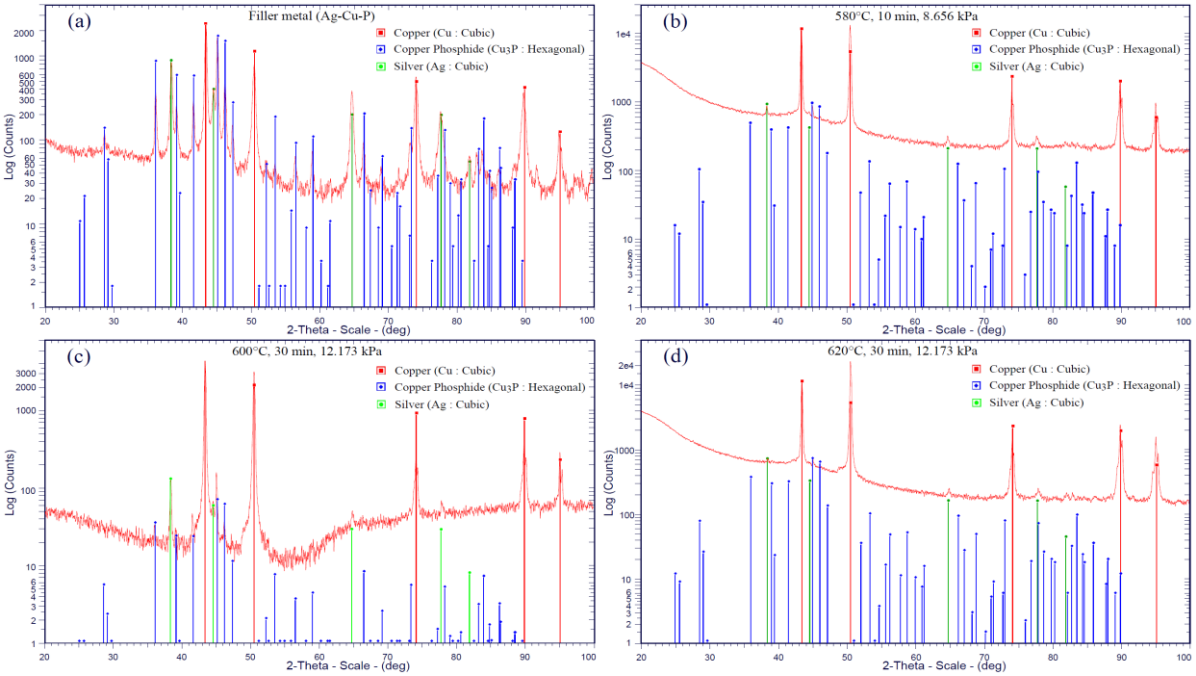

Figure 5. XRD pattern of filler metal and brazed joints after solid-state diffusion brazing under variable of brazing temperature, holding time and loading pressure respectively. (a) XRD pattern of filler metal, (b) XRD pattern of brazed joint after solid-state brazing at 580 ℃ for 10 min and 8.656 kPa, (c) 600 ℃ for 30 min and 12.173 kPa and (d) 620 ℃ for 30 min and 12.173 kPa.

3.4. The mechanical characteristics of the joints

Evaluation of the tensile shear force was carried out in order to determine the properties of the brazed joint. The various brazing conditions involved temperatures of 580 ℃, 600 ℃, and 620 ℃; 10 min, 20 min, and 30 min, and 4.713 kPa, 8.656 kPa and 12.173 kPa. There is a clearly apparent increase in the tensile shear force as the temperature, holding time, and loading pressure are increased. Analysis of the microstructure revealed that voids were present in the joints at low temperatures, short holding times, and low loading pressure, indicating relatively weak bonding (Figure 4 (a and f)). At higher temperatures and loading pressures with a longer holding time, the voids disappear and stronger joints are the result.

In the confirmation test process, a total of six samples were brazed. Table 2 shows the outcome in the high brazing sample, representing the optimal brazing conditions. The tests confirmed that all samples brazed under optimal conditions achieved tensile shear force results which are considered to fall within the range of higher tensile shear force.

Table 2. Testing to confirm the optimal solid-state brazing conditions.

Specimen (no.)	Tensile shear force (N)	Specimen (no.)	Tensile shear force (N)
1	743.771	4	744.565
2	746.838	5	713.015
3	731.611	6	737.842

4. Conclusions

The current study assessed the properties and microstructure of the brazed joints between copper sheets and Ag-Cu-P filler metal which are potentially employed in the production of LMA devices. The process involved solid-state brazing at temperatures of 580 ℃, 600 ℃ and 620 ℃, with loading pressure at 4.713 kPa, 8.656 kPa, and 12.173 kPa, and holding times of 10 min, 20 min, and 30 min. The findings allowed certain conclusions to be proposed, as described below:

To obtain the greatest tensile shear force on average, the parameters were set as follows: brazing temperature 620 ℃; holding time 30 minutes; loading pressure 12.173 kPa. This resulted in average tensile shear force of 736.27 N. The analysis using EDS and SEM showed that it was difficult to form a reaction layer between the copper sheet and the Ag-Cu-P filler metal at the lower temperatures of 580 ℃ and 600 ℃, lower holding time of 10 min and 20 min and lower loading pressure of 4.713 kPa and 8.656 kPa leading to gaps in the joint and low shear force. However, under optimal conditions (brazing temperature 620 ℃; holding time 30 minutes; loading pressure 12.173 kPa) Cu_3P phase was produced and apparent at the brazing joint between the filler metal and the copper sheets, leading to much better results on tensile shear force and a stronger joint.

References

[1] Paulraj P and Paul B K 2011 Metal microchannel lamination using surface mount adhesives for low-temperature heat exchangers *Journal of Manufacturing Processes* vol 13 pp 85–95.
[2] Paul B K and Lingam G K 2012 Cooling rate limitations in the diffusion bonding of microchannel arrays *Journal of Manufacturing Processes* vol 14 pp119–125.
[3] Paul B K, Kwon P and Subramanian R 2006 Understanding limits on fin aspect ratios in counter flow microchannel arrays produced by diffusion bonding. *Journal of Manufacturing Science and Engineering* vol 128 pp 977–983.
[4] Eluri R and Paul B K 2012 Silver nanoparticle-assisted diffusion brazing of 3003 Al alloy for microchannel applications. *Materials and Design* vol 36 pp 13–23.
[5] Kanlayasiri K, Paul B K 2004 A nickel aluminide microchannel array heat exchanger for high-temperature applications *Journal Manufacturing Process* vol 6 pp 17–25.
[6] Basuki W W, Kraft O and Aktaa J 2012 Optimization of solid-state diffusion bonding of Hastelloy C-22 for micro heat exchanger applications by coupling of experiments and Simulations *Materials Science and Engineering* A vol 538 pp 340–348.
[7] Eluri R and Paul B K 2012 Silver nanoparticle-assisted diffusion brazing 3003 Al alloy for microchannel applications *Materials and Design* vol 36 pp 13–23.
[8] Cooke K O, Khan T I and Oliver G D 2012 Transient liquid phase diffusion bonding Al-6061 using nano-dispersed Ni coatings *Materials and Design* vol 33 pp 469–475.
[9] Okamoto H, Subramanian P R, Kacpzak L and Massalski T B 2001 Binary alloy phase diagram of Cu-P Second edition *The Materials Information Society* vol 2 pp 1449–1451.

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