

#### Effect of copper based filler composition on the strength of brazed joint

## A. J. Sulaiman H.<sup>1</sup>, W. N. W. M. N. Hissyam<sup>1</sup>, Aiman M. H.<sup>1</sup>, M. Ishak<sup>1</sup> and T. Ariga<sup>2</sup>

 <sup>1</sup>Faculty of Mechanical Engineering, Universiti Malaysia Pahang 26600 Pekan, Pahang, Malaysia,
<sup>2</sup>Department of Materials Science, School of Engineering Tokai, University Kanagawa, Japan Phone: +6094246216; Fax: +6094246222 \*Email: aimanmh@ump.edu.my

## ABSTRACT

Compositions of molten brazing filler material are considering as important factors that influence the strength of brazed joint, in addition to effects of brazing temperature have been carried out. Generally, the composition of copper-based filler and temperature effect the strength of the brazed joint. Vacuum brazing of copper-based filler was performed using pure copper plate as its based metal. The brazed joint was obtained at different temperatures, 680°C and 730°C, with same holding time, which is 30 minutes. MBF 2005 (Cu, 5.7wt.%Ni, 9.7wt.%Sn, 7.0wt.%P), MBF 2002 (Cu, 9.9wt.%Ni, 4.0wt.%Sn, 7.8wt.%P) and VZ 2250 (Cu, 7.0wt.%Ni, 9.3wt.%Sn, 6.3wt.%P) alloys were used as brazing filler materials. The intermetallic compound (IMC) and mechanical properties of the joints are investigated using tensile test. The result shows that the interfacial microstructure of the brazed joints is Cu<sub>3</sub>P and (CuNi)<sub>2</sub>P. As the brazing temperature increase from 680°C to 730°C, the IMC become thicker leading to decrease in joining strength due to brittle IMC formation. The maximum shear strength reached 112 MPa at 680°C for MBF 2002 and 129 MPa at 730°C for MBF 2005. Hence, it is found that the composition of filler effects the strength of brazed joint.

Keywords: Intermetallic compound (IMC); shear strength; temperature; copper-based filler.

## INTRODUCTION

Brazing of copper joint is widely used in the thermal alternative materials because of its high thermal conductivity, strength and service temperature [1]. Brazing also suitable for small parts and when high joint strength is required. According to the American Welding Society (AWS), the strength of a brazed joint can meet or exceed that of metals being joined [2]. Previous studies have reported that copper and its alloys can withstand high temperature brazing process without a substantial loss in strength [3]. Filler material properties, wetting and spreading by molten filler and interfacial microstructure reaction between molten filler and spreading ability of molten brazing filler metallic alloy give high influence to the brazing joint strength. Other than that, brazing temperature are also found to influence the microstructure and the brazing joints in such that higher temperature results in higher joining

strength [4-7]. This is because the thickness of reaction layer to material increase with the rise of brazing temperature [8]. It was found that the brazing temperature has a strong influence over the bond strength and the electrochemical behaviour. This is mainly due to the different interfacial morphologies and reaction products corresponding to a certain brazing condition [9].

As much as temperature important in brazing, the composition of the filler alloys also play significant role in brazing. Different percentage of composition may influence the wetting ability of the filler on base material. Fluxing is used except when an atmosphere is specifically introduced into furnace to perform the same shielding gas, but in this study the Argon gas used as 'Flush' agent, to remove all the excess oxygen inside the tube furnace. Furnace are either batch or continuous type with possibly atmosphere controls and should have automatic time and temperature [10, 11]. Apart from these factors, the microstructure of the joint has been carried out to investigate the joint strength of the copper based fillers. Nevertheless, brazing has many parameters, which can decide the perfectly brazed joint. There are many parameters which affect the brazing characteristic of a joint, there a gap that explained the relationship between the individual element in fillers of copper brazing and the brazing temperature that can relate to strength of brazing. Hence, in this study the relationship of the elements in fillers and the strength of brazing will be conducted. The present work investigated elements in the filler compositions particularly in copper-based filler and brazing temperature with a view to strength of brazing joint.

#### METHODS AND MATERIALS

#### **Material Preparation**

The filler alloy that have been chosen are custom-made from company which is had copperbased composition. The composition of these filler consist of Copper, Nickel, Tin and Phosphorus. Table 1 shows the chemical composition in copper-based fillers alloys. Commercially (99.99%) pure copper was selected as the base material for brazing. As received 2 mm diameter pure copper plates were cut sectioned by using wire-cutting machine into 60x10 mm samples. The surfaces of the samples are grit using 800 grit paper, in order to remove any coating or dirt on the samples surface. Then samples were cleaned with ethanol prior to processing. The purpose of the ethanol usage on cleaning process is to remove any residual oil contamination on the samples surfaces. The specimens were placed in the jig with butt joint configuration; this is prior to the tensile test. Figure 3 illustrate the schematic diagram for lap joint configuration on the jig. The purpose of jig is used to ensure the consistency of the result and repeatable for testing is accessible. The filler placed in between 2 base metal. After that, the first specimen placed inside the tube furnace with heating temperature of 680°C and 730°C and its holding time is 30 minutes. The specimen allowed cooling until the furnace temperature reached under 100°C. The process repeated for the second specimen with temperature of 730°C. Figure 4 shows the temperature profile for the brazing process at 730°C.

Effect of copper based filler composition on the strength of brazed joint

Table 1. Chemical composition of copper-based filler alloy				
Filler metal	Cu (wt.%)	Ni (wt.%)	Sn (wt.%)	P (wt.%)
MBF 2002	77.6	5.7	9.7	7.0
MBF 2005	78.3	9.9	4.0	7.8
VZ 2002	77.4	7.0	9.3	6.3

Copper plate and Filler metals



Grind copper plate using gritt paper



Clean copper plate and filler metal using ethanol



Stack the copper plate on the jig



Immersed in ultrasonic bath for 3 minutes Figure 2. Process flow for material preparation.



Figure 3. Schematic diagram for lap configuration joint inside the jig



After the brazing done, for the microstructure part, the specimens undergo the cold mounting process by using epoxy powder. Since the pure copper is soft, cold mounting is preferable than using hot mounting. After the cold mounting fully cured, the specimens were grinded from 320 grit until 4000 grit until the surface of the joint appears. The polishing process also has its own sequence and direction starting with 1  $\mu$ m until 0.5  $\mu$ m of diamond compound liquid with the selected polishing cloth. The specimen with mirror-look surface can proceed

with an etching process. Etching is the process of revealing microstructure details on samples after the grinding and polishing step.



Figure 5. Vacuum Furnace setup

This process will reveal the microstructure properties such as the microstructure phase, shape and size of grain boundaries. Pure copper has 99.9 % of copper element and stated as a smooth metal. The etching process of the pure copper uses 10 % of the hydrochloric acid (HCL) solution. The chemical compound hydrochloric acid is the aqueous (water-based) solution of hydrogen chloride gas. The HCL solution put in the beaker mixed with 90 % of methanol. The polished samples are soaked into the solution for 1.5 seconds. The microstructure revealed and the samples cleaned by using distilled water. The microstructure and phase composition of joints interfacial were examined using scanning electron microscope (SEM) equipped with an energy dispersive X-ray (EDX) spectrometer. The strength of the joining is identified by using the tensile test. The test is conducted by using the INSTRON Universal Testing machine that is able to support the strength up to 50 kN. The specimen is prepared in accordance to ESTM E8 specimen standards. The strain versus stress curve obtained from this testing is based on speed of 5 mm/min. The ultimate tensile strength (UTS) value is used as the response in the statistical analysis.

## **RESULTS AND ANALYSIS**

## Microstructure

The dominant element at each point in figure 6 detected by EDX are listed in table 2 respectively. Based on EDX result tabulated in table 2, it shows that the point marked as point 3 at top interface was enriched with Cu (91.86 at%) and small amount of Sn (3.35 at%), P (2.03at.%) elements. Nevertheless, it can be identified that point marked as point 1 and point 2 had high amount of P (23.34 at%) and (23.25 at%) which is in the diffusion area. Then, it can be highlighted that the P is diffusing towards the top of the joint is fastest than

Ni and Si element. From table 2, it was observed that the light grey point marked as point 3 is enriched with Sn (3.35 at.%), Cu (91.86 at.%) and P (2.03 at.%). The possible IMC phase for point 1 and 2 are Cu<sub>3</sub>P and for point 3 as Cu substrate (Sn-rich).

The EDX point analysis carried out with the same brazing temperature and holding time, but this time by changing the filler from MBF2002 to MBF 2005. The elemental distribution at marked point in figure 7 which are tabulated the dominant element in table 3, respectively. The joint marked as point 5 and 6 consist of enrich element of Cu-P, which is Cu (74.54 at.%), P (22.53 at%) and Cu (73.88 at.%), P (23.42 at.%). As can be seen from the figure, round phosphorus poor dendrites in a matrix of Cu–P are observed. Dendrites have a uniform distribution in the matrix owing to proper brazing conditions [12]. Joint marked as point 4 indicates that there are possibility for IMC phase which is (CuNi)<sub>2</sub>P with distribution element of Cu (35.45 at%), Ni (49.46 at.%) and P (19.59 at%). However, there is no Sn detected in point 4, 5 and 6 due to the Sn has diffuse out of filler metal into the Cu substrate and then Sn is not detected in all the marked point in table 3.

For filler VZ 2250, the same brazing parameters used to obtain the microstructure analysis. The microstructure grain is shows in figure 8 and its major element present on the interface are tabulated in table 4. Based on EDX results and figure 8, the block light grey marked as point 9 is enriched with P (23.04 at%), Cu (75.73 at%) and a small quantity of Sn (0.35 at%). The less dark grey phase marked as point 7 is enriched with Ni (46.60 at%), P (20.30 at%) and Cu (30.10 at%). For point 8 with dark grey at top of the interface is enriched with Sn (5.40 at%), Cu (92.40 at%) and fewer amount of P (1.12 at%). It can identify that possible phase for point 8 is Cu<sub>13.7</sub>Sn<sub>86.3</sub> (Sn-rich), point 7 is an IMC having the chemical formula (CuNi)<sub>2</sub>P and for point 9 IMC is Cu<sub>3</sub>P. Brittle IMCs such as Cu<sub>3</sub>P and (CuNi)<sub>2</sub>P are produced during crystallization with increasing the brazing temperature and they will certainly degrade the mechanical properties of the joints [12].

Since all the reaction are between copper to copper, then most of the element diffuse during brazing are Cu-Ni-P. With increasing either brazing temperature or time, the filler metals melt and the liquid filler metals begin to spread and flow. The solid base metals react and continuously melt into filler metals until reaching the maximum saturated concentration, so the dissolution stops [12]. Concurrent, elemental P and Ni speed up the diffusion towards the base metal through liquid phase channel, which contribute to the formation of Cu-solid solution containing elements Sn, P, Ni [15-17].



Figure 6. SEM image of the joints brazed at 730°C for 30 min using MBF2002 filler

Element		At	%	
	Cu	Sn	Ni	Р
Point 1	75.21	-	-	23.34
Point 2	75.68	-	-	23.25
Point 3	91.86	3.35	-	2.03

Table 2. Element composition on brazing joint for MBF 2002



Figure 7. SEM image of the joints brazed at 730°C for 30 min using MBF2005 filler

Element		A	t %	
	Cu	Sn	Ni	Р
Point 4	35.45	-	49.86	19.59
Point 5	74.54	-	-	22.53
Point 6	73.88	-	-	23.42

Table 3. Element composition on brazing joint for MBF 2005



Figure 8. SEM image of the joints brazed at 730°C for 30 min using VZ 2250 filler

Element		At	: %	
	Cu	Sn	Ni	Р
Point 7	30.10	-	49.60	20.30
Point 8	92.04	5.40	-	1.12
Point 9	75.73	0.35	-	23.04

Table 4. Element composition on brazing joint for VZ 2250



Type of Copper-based Filler Figure 9. Intermetallic compound layer thickness

According to Figure 9, the result can be simplified that the thickness of IMC increase with increasing the temperature. The IMC thickness between these 3 copper-based fillers increase gradually, the graph line shows the different of VZ2250 tremendously differ to MBF 2005 and MBF 2002. The effect of the IMC thickness on the tensile strength [18,19], on the shear strength [22-24] and on the fracture toughness [24]. The increasing in spreading and wetting angle cause in increasing in IMC layer thickness. The higher of IMC thickness will lead the joining become more brittle. Intermetallic compound formations are the indication of metallurgical and its define strength and reliability of the joining. There is a negative correlation between the IMC thickness and the tensile strength and a positive correlation between the IMC roughness and the tensile strength [25].

#### **Tensile Strength of Copper-based Filler**

Table 5 show the average result of 3 sample of tensile test for all the fillers. As shown in table 5 and figure 10, the strength of the fillers may vary accordingly. This is due to the different dominant composition of weight percentage in each filler. The brazing temperature give influence towards each of the filler metal. From the table, strength of filler MBF 2005 and VZ 2250 slightly decrease approximately 1% with increasing temperature. Compared to other fillers; MBF 2002 is increase drastically with increasing temperature. Thus, based on the result it cannot prove that by increasing the temperature it will resulted higher tensile strength. The study done by Zhang et al [12], he stated that in his research, increasing the

temperature or dwelling time will cause drop on mechanical strength due to reduction of amount  $Cu_3P$  and  $(CuNi)_2P$  in intermetallic compound. One of the way to increase the mechanical strength, is to decrease the content of P to avoid the existence of block brittle phase  $Cu_3P$  on the promise of dissolving as much as possible base metal [18]. It can be related that the higher the IMC layer thickness cause the joint strength reduced sharply. Figure 11 illustrate the fracture surface for tensile specimen for MBF2002. It indicates that the fracture or necking occur at base metal due to high shear strength.



Copper-based filler Figure 10. Tensile Strength for copper-based filler

Table 5. Result for Tensile Strength				
Eillong	Tensile Strength (MPa)			
Fillers	680°C	730°C		
MBF 2005	112	110		
MBF 2002	92	129		
VZ 2250	91	89		



Figure 11. Fracture surface for tensile specimen

#### CONCLUSIONS

In this study, the copper-based fillers were joined with pure copper successfully by using vacuum furnace brazing. Too high in intermetallic compound can cause the joining to be more brittle due to reduction of amount  $Cu_3P$  and  $(CuNi)_2P$  in intermetallic compound. These higher in thickness may cause the decreasing in joint strength of copper brazing. This also can cause the joint strength decrease with increasing the temperature. Many study had proven that the increasing in temperature cause the joint strength become weak. Strength MBF 2005 and VZ 2250 decrease with increase in temperature. However, MBF 2002 higher in tensile strength while the temperature increase. Thus, the dominant weight percentage in filler composition affecting to the strength of the copper-based filler. Hence, as stated above, the pros and cons in filler composition gives big impact to the strength of brazed joint. In conclusion, the higher the temperature of brazing will give higher intermetallic compound thickness but decreases in tensile strength due to brittle IMC formation occur during the heating process. Moreover, the proper in weight percentage composition in filler alloys also is the factor that will improve the strength of the brazed joint.

#### **ACKNOWLEDGEMENTS**

The authors would like to thank to the Universiti Malaysia Pahang for laboratory facilities and financial assistance under Research Grant project RDU180382.

# REFERENCES

- [1] A Hasap, N Noraphaiphipaksa, C Kanchanomai. The Microstructure and strength of Copper Alloy Brazing joints. Welding Research 2014; 94: 116-123.
- [2] American Welding Society. Chapter 3. Committee on Brazing and Soldering Handbook Fifth Edition. ASM International;2007.
- [3] YL Shabtay, M Ainali, A Lea. New brazing processes using anneal-resistant copper and brass alloys. Material Design 2004; 25(1): 83-89.
- [4] Satoshi K, Tetsurou K, Yukiakira H and Yasuyuki M. Wetting and Spreading Behaviour of Molten Brazing Filler Metallic Alloys on Metallic Substrate. IOP Conference Series. Materials Science and Engineering 2014; 61 012017.
- [5] Guedes A, Pinto A and Vieira M. The effect of brazing temperature on the titanium/glass ceramic bonding. Material Processing Technology. 1999; 102(6): 92-93.
- [6] Jinglong L, Zengchan H, Jiangtao X, Fusheng Z and Wen-Ya L. Study on microstructure and strength of AgNi alloy/pure Al vacuum diffusion bonded joints. Material Design. 2009; 30(80): 3265-8.
- [7] Li J, Liu Y, Tan Y, Li Y, Zhang L, Wu S and Jia P. Effect of tin addition on primary silicon recovery in Si-Al melt during solidification refining of silicon. Journal Crystal Growth. 2013; 371: 1-6.
- [8] Yongtong Cao, Jiazhen Yan, Ning Li Yi Zheng, Chenglai Xin. Effects of brazing temperature on microstructure and mechanical performance of Al2O3/AgCuTi/Fe– Ni–Co brazed joints. Journal of alloy and compound. 2015: 30-36.
- [9] O C Paiva, M A Barbosa. Brazing parameters determines the degradation and mechanical behaviour of alumina/titanium brazed joints. Journal of Materials Science. 2000; 5(35) :1165-1175.
- [10] Robert W M. Joining of Materials and Structures. First published, the United States of America. Elsevier Butterworth–Heinemann. 2004, p349-387.
- [11] Flom Y and Wang L. Flaw Tolerance in Lap Shear Brazed Joints Part 1 Welding Journal, January 2004, p32-38.
- [12] Jing Zhang, Weiyuan Yu, Wenjiang Lu. Mechanical and microstructure of pure copper joint brazed with amorphous Cu68.5Ni15.7Sn9.3P6.5 filler metal. International journal of simulation, science, system and technology. 2014; 17(24): 1473-8031.
- [13] Zhang PL, Yao S, Ding M, Lu FG, Lou SN. Microstructural analysis in the vacuum brazing of copper to copper using a phosphor-copper brazing filler metal. International Journal of Materials Research. 2010; 101(11): 1436-144.
- [14] SH Chen, LQ Li, YB Chen. Joining mechanism of Ti/Al dissimilar alloys during laser welding-brazing process. Journal of alloys and compounds. 2011; 509(01): 891-8.
- [15] XG Song, J Cao, HY Chen. Contact reactive brazing of Ti53311S alloy using Cu foil as interlayer: Interfacial microstructure and joining properties. Materials and Design. 2013; 4(04): 895-901.
- [16] WQ Yang, TS Lin, P He. Microstructure and mechanical properties of ZrB2–SiC joints fabricated by a contact-reactive brazing technique with Ti and Ni interlayers. Ceramics International. 2014; 40(06):7253-60.

- [17] Li Yi-nan, Wang Chang-wen, Peng Zi-long, Yan Jiu-chun, Liu Xue-song. Dissolution behaviour of Cu in Cu-Ag and Cu-P brazing alloys using weld brazing. Transactions of Nonferrous Metals Society of China. 2011; 2(21): 394-399.
- [18] Lee HT, Chen MH. Influence of intermetallic compounds on the adhesive strength of solder joints. Material science engineering A Structure material properties microstructure processing. 2002; 333: 24–34.
- [19] Prakash KH, Sritharan T. Tensile fracture of tin-lead solder joints in copper. Material Science Engineering A – Structure material properties microstructure processing. 2004; 379: 277–85.
- [20] Lee HT, Chen MH, Jao HM, Liao TL. Influence of interfacial intermetallic compound on fracture behaviour of solder joints. Material Science Engineering A – Structure material properties microstructure processing. 2003; 358: 134–41.
- [21] Zhao J, Cheng CQ, Qi L, Chi CY. Kinetics of intermetallic compound layers and shear strength in Bi-bearing SnAgCu/Cu soldering couples. Journal Alloys Compound. 2009; 73: 382–8.
- [22] Liu P, Yao P, Liu J. Effects of multiple reflows on interfacial reaction and shear strength of SnAgCu and SnPb solder joints with different PCB surface finishes. Journal Applied Physics. 2009; 470: 188–94.
- [23] Hayes SM, Chawla N, Frear DR. Interfacial fracture toughness of Pb-free solders. Microelectronics reliability. 2009; 49: 269–87.
- [24] Tong An, Fei Qin. Effect of the intermetallic compound microstructure on the tensile behaviour of Sn3.0Ag0.5Cu/Cu solder joint under various strain rates. Microelectronics reliability. 2014; 54: 932-938.
- [25] WNWMN Hissyam, AM Halil, T Kurniawan, M Ishak and T Ariga. Effect of copper based filler composition on spreading and wetting behaviour. Material and Science Engineering. 2017; 238: 012020.