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Development of a Novel Low-Silver Cu-P Brazing Filler Metal Bearing Sn

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Abstract: The flame brazing of H62 brass using a novel, low-silver Cu-P brazing filler metal was investigated in this study. The effect of the addition of a trace amount of Sn on the microstructure and properties of Cu-7P-1Ag filler metals was analyzed by means of X-ray diffractometer, scanning electron microscopy and energy dispersive spectrometer. The addition of trace Sn led to a decrease in the solidus and liquidus temperatures of Cu-7P-1Ag filler metals. Meanwhile, the spreading performance of the filler metals on a H62 brass substrate was improved. The microstructure of the low-silver, Cu-P brazing filler metal was mainly composed of α-Ag solid solution, α-Cu solid solution and Cu_3P; an increase of Sn content led to the transformation of the microstructure of the joints from a block to a lamellar structure. When the Sn content was 0.5 wt. %, the shear strength of the joint at room temperature reached 348 MPa, and the fracture morphologies changed from a cleavage to a quasi-cleavage structure.

Keywords: low-silver Cu-P filler metals; melting temperature; spreading performance; microstructure; mechanical properties

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1. Introduction

Cu-P brazing filler metals are extensively used to braze copper and its alloys in the aerospace, electronics, energy, transportation, military, automobile, and other industries due to their superior performance, such as low melting point, good wettability, low price and self-brazing property when brazing pure Cu [1,2]. Elemental P can greatly reduce the melting temperature of the filler metal and increase the spreading area on the substrate [3]. However, the content of P in the Cu-P brazing filler metals is relatively high, and the alloy matrix contains a large amount of brittle Cu_3P compounds, resulting in poor plasticity at room temperature [4]. In addition, the melting temperature of Cu-P brazing filler metal is still too high to braze copper alloys with low melting points, such as brass. Therefore, several novel Cu-P brazing filler metals have been developed, such as Cu-P-Ni [5,6], Cu-P-Ag [7], Cu-P-Sn [8,9], Cu-P-Zr [10] and Cu-P-RE (rare earth) [11], some of which have been used in the manufacturing industry for brazing copper alloys.

Compared with Cu-P brazing filler metals, Cu-P-Ag series alloys show lower melting points and better wettability. Meanwhile, brittle Cu_3P compounds in the brazing filler metal matrix can form eutectic structures with α-Ag solid and α-Cu solid solutions, which greatly improves the plasticity and processing properties of the brazing filler metals. As such, this approach has been widely used in the electronics industry [12]. However, the price of silver is extremely high and volatile, which limits the application of high-silver Cu-P-Ag filler metals in middle- and low-end manufacturing. The melting point of Sn, which is a relatively cheap metal, is only 231.9 °C. Adding Sn to Cu-P brazing filler metals can greatly decrease their solidus and liquidus temperatures and improve their flowability [13]. Furthermore, appropriate amounts of Sn can dissolve in copper to form copper-based solid solutions. The strengthening effect of these solutions can improve the mechanical

properties of Cu-P brazing filler metals. However, Sn has limited solid solubility in copper, and excessive Sn can form brittle intermetallic compounds with Cu, negatively affecting the mechanical properties of the filler metals [14].

In order to reduce the cost of high-silver Cu-P brazing filler metals, in this study, Cu-7P-1Ag filler metals with various Sn additions were produced. The effect of Sn addition on the melting characteristics and spreading performance of novel Cu-7P-1Ag brazing alloys was investigated, and the microstructures and phase were analyzed. Additionally, the mechanical properties and fracture morphologies of brazed joints were studied.

2. Materials and Methods

Pure Cu, Ag and Sn, as well as Cu-14P master alloy, were used as raw materials. These metals were melted in a medium frequency furnace (frequency 600 Hz, power 110 kW). The molten alloy was subsequently held for 20 min and then poured into a 50mm diameter steel mould. Finally, all the cast ingots were extruded (Hydraulic Press, Xinke Y32-315) into wire with a 1.9 mm diameter for brazing. The designed and actual compositions of the Cu-7P-1Ag-*x*Sn filler metals used in present study are listed in Table 1.

Table 1. Designed and actual compositions of the Cu-7P-1Ag-*x*Sn filler metals (wt. %).

No.	Cu	P		Ag		Sn	
		Designed	Actual	Designed	Actual	Designed	Actual
1	Bal.	7.00	6.96	1.00	1.02	0	0
2	Bal.	7.00	6.99	1.00	0.98	0.10	0.11
3	Bal.	7.00	7.03	1.00	1.00	0.30	0.31
4	Bal.	7.00	6.95	1.00	0.98	0.50	0.52
5	Bal.	7.00	6.93	1.00	1.05	0.70	0.70
6	Bal.	7.00	7.06	1.00	1.03	1.00	1.04

The commercially supplied H62 brass substrates used in the present work were processed into plates with the dimensions of 40 mm × 40 mm × 1 mm for the spreading test and 60 mm × 25 mm × 3 mm for the shear test. All filler metals and specimen surfaces were polished by SiC papers in order to remove the oxide layer, and then ultrasonically cleaned by ethanol.

The solidus and liquidus temperatures of the brazing alloys were determined using a differential scanning calorimeter (DSC, Netzsch STA 449F, NETZSCH Group, Selb, Germany) under a nitrogen atmosphere with heating at a rate of 10 °C/min. The spreading test was carried out according to China's National Standard GB/T 11364-2008 [15]. First, 200 mg of each novel filler metal (diameter: 1.9 mm) was placed on the surface of the H62 brass substrates covered with FB102 (40 wt.% of KF, 25 wt.% KBF_4, 35 wt.% of B_2O_3), which was then heated at 750 °C for 1 min in an electrical resistance furnace. The spreading area was measured using the Image-Pro Plus software.

The phase and microstructure of the Cu-7P-1Ag-*x*Sn filler metals were examined using an X-ray diffractometer (XRD, Bruker D8 Advance, Bruker, MA, USA, Range: 10°–90°, Speed: 6°/min, Voltage: 40 kV, Current: 150 mA) and a scanning electron microscope (SEM, ZEISS Σ IGMA 500, ZEISS, Oberkochen, Germany) equipped with an energy dispersive spectrometer (EDS, Bruker Nano XF Lash Detector 5010, Bruker, MA, USA). The flame brazing method was used in this study to braze the H62 brass, and the shear strength of joints with an overlap length of 2 mm and joint clearance of 0.07 mm was tested on an electronic universal testing machine, according to the China's National Standard GB/T 11363-2008 [16]. At least five specimens were tested for each experimental condition and the averages of the tested results were calculated. In addition, the fracture morphologies of H62 brass brazed joints were characterized using a scanning electron microscope (SEM).

3. Results

3.1. Thermal Properties and Spreading Performance of Cu-7P-1Ag-xSn Filler Metals

Figure 1 shows the thermal properties of Cu-7P-1Ag-*x*Sn filler metals. The results indicate that the solidus temperature (T_s) and liquidus temperature (T_l) of the alloys decreased with increasing Sn content. Compared with Cu-7P-1Ag, the T_s and T_l of Cu-7P-1Ag-1Sn filler metal were reduced to 560 °C and 751 °C, respectively, representing decreases of 13.2% and 5.2%, respectively. The decrease of both T_s and T_l was likely due to the fact that the melting point of Sn is only 231.9 °C. Appropriate amounts of Sn could dissolve in copper to form copper-based solid solutions with lower melting temperature, which is beneficial for the brazing of the H62 brass substrate. The effect of alloying elements on the thermal properties of solder has been reported elsewhere [17–19], and its mechanism is similar to that described in this study. Notably, the T_s of the brazing filler metal was 560 °C when the amount of Sn addition exceeded 0.5 wt.%, indicating that the Cu-P-Ag-Sn quaternary eutectic structure of the novel low-silver filler metal had begun to emerge [20].

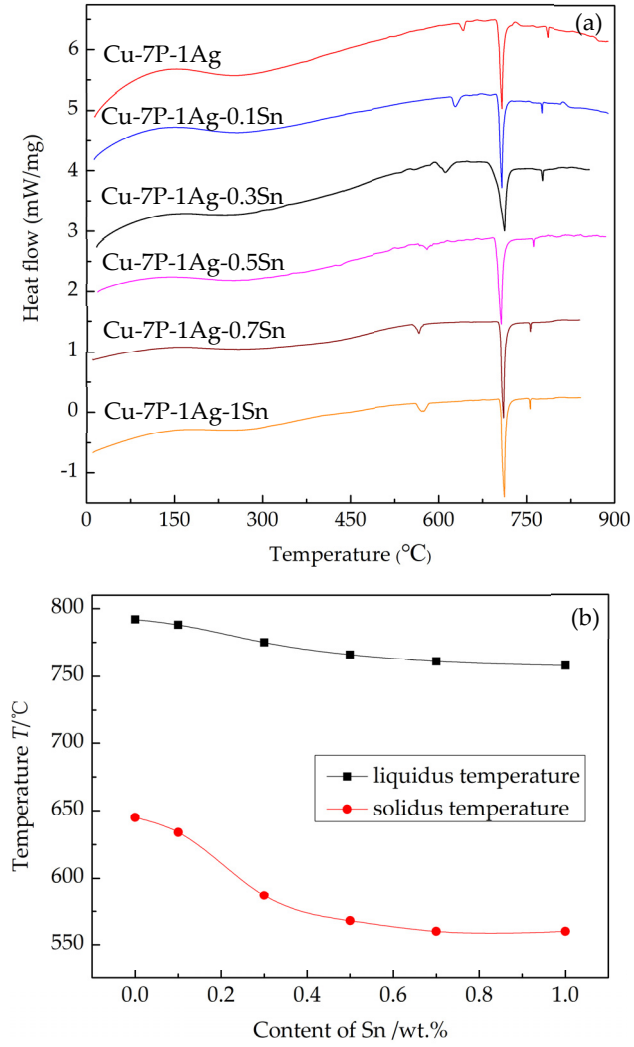

Figure 1. Melting behavior of Cu-7P-1Ag-*x*Sn filler metals: (**a**) DSC melting curves of the filler metals, (**b**) The change trend of T_s and T_l.

In general, the spreading area of liquid brazing filler metals on a substrate is usually used to evaluate the fluidity: the larger the spreading area, the better the fluidity [21]. Figure 2 presents the spreading test results of Cu-7P-1Ag-*x*Sn filler metals on H62 brass substrates with the aid of FB102 flux at a temperature of 750 °C. As shown in the figure, with an increase in Sn content from 0 to 1 wt.%, the spreading area of the filler metals on

H62 brass improved significantly, i.e., the spreading area of Cu-7P-1Ag-1Sn filler metal was about 13% larger than that of the Sn-free one. However, when the Sn content reached 0.5 wt.%, the growth rate of the spreading area began to slow down. Therefore, it can be inferred that the improvement in spreadability was due to the decrease of the melting temperature of the filler metals. Under the same brazing process parameters, the lower the melting point of the brazing filler metal, the greater the superheat and the lower the surface tension of the liquid alloy, which improves the spreadability of the filler metal on the substrate [22].

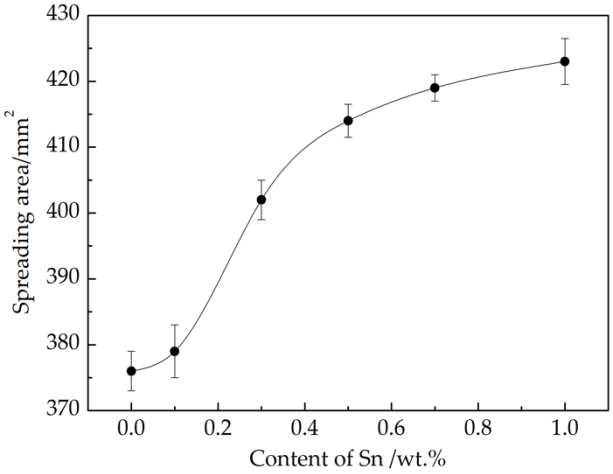

Figure 2. Spreading areas of Cu-7P-1Ag-*x*Sn filler metals on H62 brass plates.

3.2. Microstructure of Cu-7P-1Ag-xSn Filler Metals

Figure 3 shows the X-ray diffraction (XRD) patterns of Cu-7P-1Ag-*x*Sn filler metals. As we can see from the picture, when the Sn content was less than 0.7 wt.%, the novel filler metals consisted of three phases, i.e., the α-Ag solid solution phase, α-Cu solid solution phase and Cu_3P compound phase, in which the brittle structure of Cu_3P negatively influenced the mechanical properties of the alloy. Therefore, the P content in Cu-P brazing filler metals should be closely monitored [23,24]. Notably, the solid solubility of Sn in Cu is limited at room temperature, and when the content of Sn reaches 1 wt.%, the diffraction peak of the $Cu_{20}Sn_6$ intermetallic compound phase appears.

Figure 5 shows a high magnification SEM image of Cu-7P-1Ag-0.5Sn filler metal. According to the Cu-P binary phase diagram, the maximum solid solubility of P in Cu is 3.5 at.%, and the P content in Cu_3P and eutectic phase (α-Cu + Cu_3P) is 25 at.% and 15.7 at.%, respectively. Combining the results of the XRD patterns in Figure 3, it may be inferred that the dark region (named as C) is a eutectic structure of Cu_3P and α-Cu solid solution, the grey region (named as A) is a mixed phase of α-Cu primary phase and eutectic phase (α-Cu + Cu_3P), and the white region (named as B) is a mixed phase of α-Ag solid solution, α-Cu solid solution and Cu_3P. Notably, the Sn content in the B region was higher than in the A and C regions, which indicated that the Ag has a higher affinity for Sn. The solidification process of the Cu-7P-1Ag-0.5Sn alloy occurred in three stages. First, the α-Cu primary phase with a higher melting point formed. Second, as the temperature decreased, the eutectic phase increased with the precrystallized α-Cu particles as the nucleation core. Finally, Ag-rich and Sn-rich phases with low melting points formed at the end.

Figure 4 shows SEM images of the studied Cu-7P-1Ag-*x*Sn filler metals. As we can see from Figure 4a, the microstructure of Cu-7P-1Ag filler metal showed thick dendrites intertwined with each other. As the Sn content in the brazing filler metal increased from 0 to 0.5 wt.%, the microstructure of the filler metal gradually changed from thick dendrites into tiny worm-like structure. When the Sn content reached 1 wt.%, the microstructure of the filler metal transformed into a bulk, worm-like mixed structure. Similar results were reported by Huang et al. [14] in their study on the effect of trace amounts of Sn on the microstructure of Cu-6.5P filler metals.

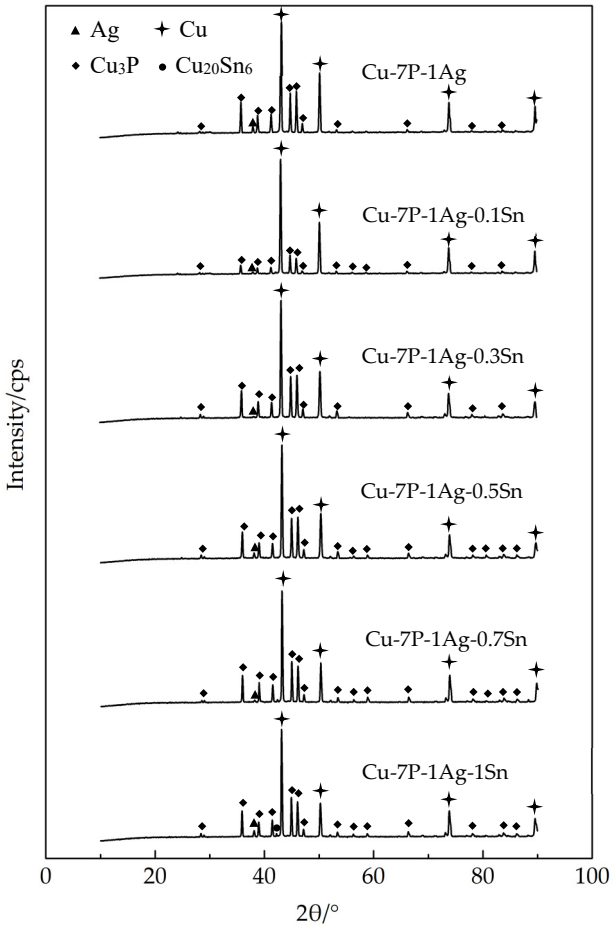

Figure 3. XRD patterns of Cu-7P-1Ag-*x*Sn filler metals.

3.3. Microstructure of the Brazed Joints

The characteristics of the reaction layer formed at the interface between the filler metals and substrate can significantly affect the mechanical properties of brazed joints [25]. In order to investigate the effect of Sn content in low-silver Cu-P filler metal on the performance of brazed joints, Cu-7P-1Ag filler metals with different Sn content were applied. Figure 6 shows the interfacial structures of the brass brazed joints using Cu-7P-1Ag-*x*Sn filler metals. It can be seen in Figure 6a that the brazing seam of Cu-7P-1Ag filler metal was mainly composed of the white, grey bulk and dark phases, and that the white phase was distributed around the grey bulk phase. When the content of Sn in the filler metal increased from 0 to 0.7 wt.%, the grey phase of the brazed joints gradually changed from a bulk to a lamellar structure. In contrast, as shown in Figure 6f, when the Sn content increased to 1 wt.%, some large grey phases were formed in the brazing seam, which may have negatively affected the mechanical properties of the brazed joints.

A high magnification SEM image of a Cu-7P-1Ag-1Sn brazing seam, the element mappings and an EDS analysis of point in (a) are shown in Figure 7. These data indicate that the element mappings of P and Ag overlapped with the dark and white phases, respectively, and that the Sn-rich phase surrounded the grey phase. Combined with the results of EDS, it can be inferred that the white phase (named as A) was a mixed phase of α-Ag, α-Cu and Cu_3P, the dark phase (named as B) was a eutectic phase of α-Cu and Cu_3P, and the grey phase (named as C) was a mixed phase of the α-Cu primary and eutectic phases (α-Cu + Cu_3P), which is similar to the phase characteristics of brazing filler metals. Notably, compared with the B and C regions in the brazing seam matrix, the P content of the reaction layer (named as D) which formed at the interface between the filler metals and

the H62 brass substrate was significantly reduced, and the content of Ag and Sn was higher, indicating that Ag and Sn had a higher diffusion coefficient in the H62 brass than P.

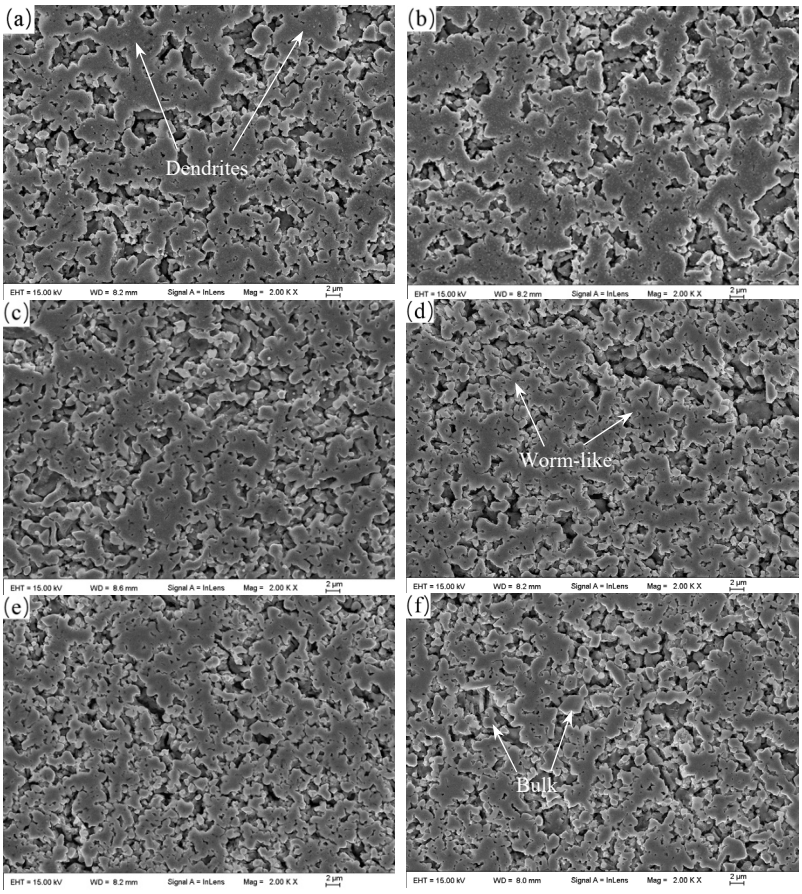

Figure 4. SEM images of Cu-7P-1Ag-*x*Sn filler metals: (**a**) Cu-7P-1Ag, (**b**) Cu-7P-1Ag-0.1Sn, (**c**) Cu-7P-1Ag-0.3Sn, (**d**) Cu-7P-1Ag-0.5Sn, (**e**) Cu-7P-1Ag-0.7Sn, (**f**) Cu-7P-1Ag-1Sn.

3.4. Mechanical Property of the Brazed Joints

The shear strength of the H62 brass lap joint was gradually enhanced when the content of Sn increased from 0 to 0.5 wt.%, as shown in Figure 8. Fractures occurred in the brazed joints of the H62 brass brazed specimens in all cases during shear tests. The results show that the peak shear strength of the joints reached a maximum of 348 MPa when the Sn content was 0.5 wt.%, which increased by 21.6% compared to the Sn-free one. The improvement of the shear strength of brazed joints with a trace amount of Sn may be explained by the microstructure of the brazing seam, as shown in Figure 7, where Sn had a higher diffusion coefficient in H62 brass than P, while Sn could be dissolved in Cu to form a Cu-based solid solution, the strengthening effect of which may have improved the shear strength of brazed joints.

However, the mechanical properties of the joints degenerated with further increases of Sn content. When the Sn content reached 1 wt.%, the shear strength of the H62 brass joint brazed with Cu-7P-1Ag-1Sn filler metal was reduced to 311 MPa. The decrease in shear strength may have been due to the form of $Cu_{20}Sn_6$ intermetallic compound. Zhao [26] reported that the hardness of $Cu_{20}Sn_6$ is extremely high, which easily become the stress concentration areas and deteriorate the mechanical property of the alloy.

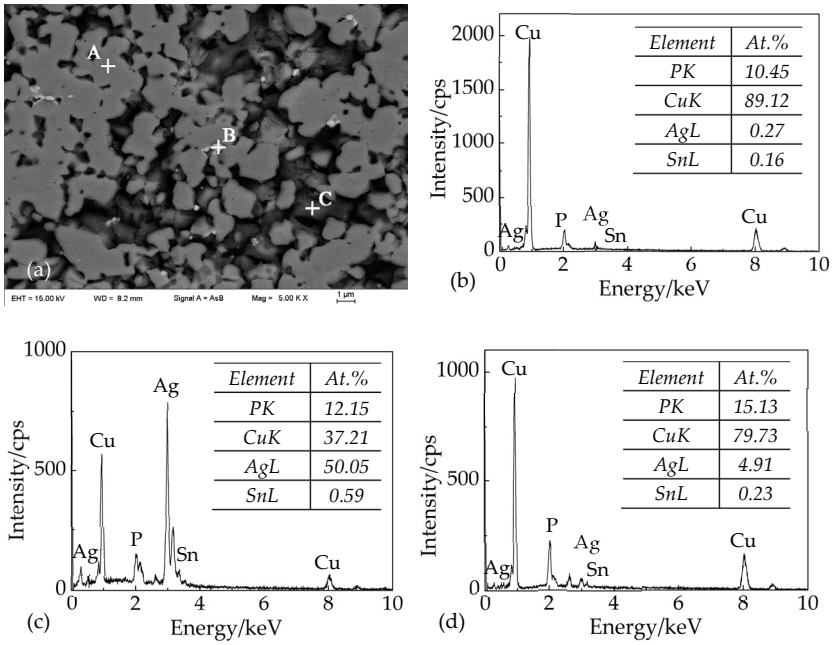

Figure 5. (**a**) High magnification SEM images of Cu-7P-1Ag-0.5Sn filler metals, (**b**) EDS analysis of point A in (**a**), (**c**) EDS analysis of point B in (**a**), (**d**) EDS analysis of point C in (**a**).

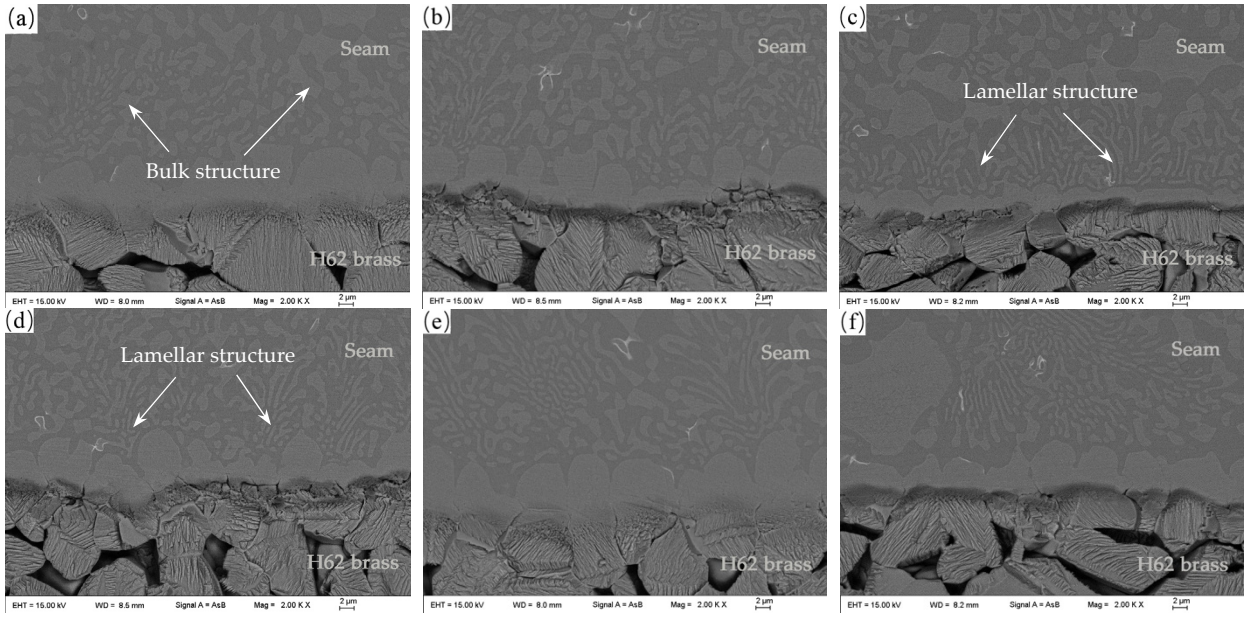

Figure 6. Interface microstructures of the brass brazed joints using different low-silver filler metals: (**a**) Cu-7P-1Ag, (**b**) Cu-7P-1Ag-0.1Sn, (**c**) Cu-7P-1Ag-0.3Sn, (**d**) Cu-7P-1Ag-0.5Sn, (**e**) Cu-7P-1Ag-0.7Sn, (**f**) Cu-7P-1Ag-1Sn.

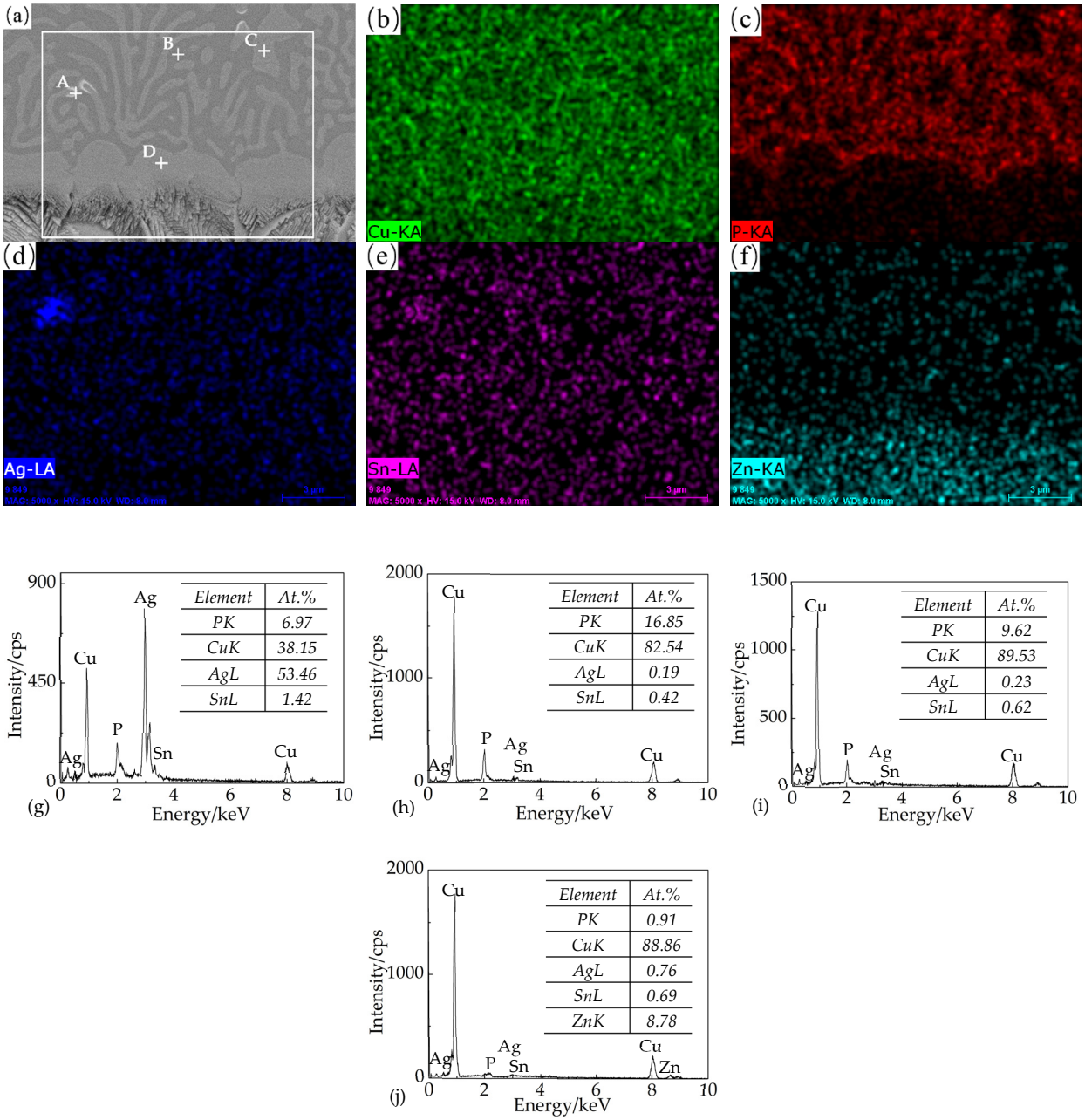

Figure 7. SEM images of interfacial microstructure of Cu-7P-1Ag-1Sn brazing seam and corresponding the element mapping of the area marked with white square in (**a**): (**b**) Cu, (**c**) P, (**d**) Ag, (**e**) Sn, (**f**) Zn, (**g**) EDS analysis of point A in (**a**), (**h**) EDS analysis of point B in (**a**), (**i**) EDS analysis of point C in (**a**), (**j**) EDS analysis of point D in (**a**).

The fracture morphologies of H62 brass joints brazed with Cu-7P-1Ag-*x*Sn filler metals are shown in Figure 9. The results show that the fracture morphology of the joint brazed with Cu-7P-1Ag filler metal presented the brittle characteristics of a river-like pattern, which indicated that a cleavage fracture had occurred. With the addition of Sn, secondary cracks and tearing edges began to appear in the fracture morphologies, which indicated that the fracture type had changed from cleavage into quasi-cleavage fractures, and thus, that the mechanical properties of brazed joints had improved. However, when the content of Sn reached 1 wt.%, the secondary cracks in the fracture morphology disappeared. The fracture morphology in Figure 9f shows a typical brittle fracture. All of the above analyses indicate that the content of Sn in Cu-7P-1Ag filler metal should be kept below 0.7 wt.%.

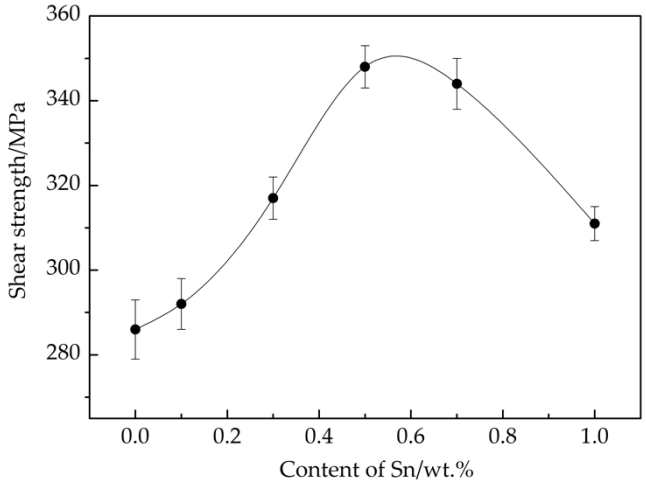

Figure 8. Shear strengths of the H62 brass lap joints using Cu-7P-1Ag-*x*Sn.

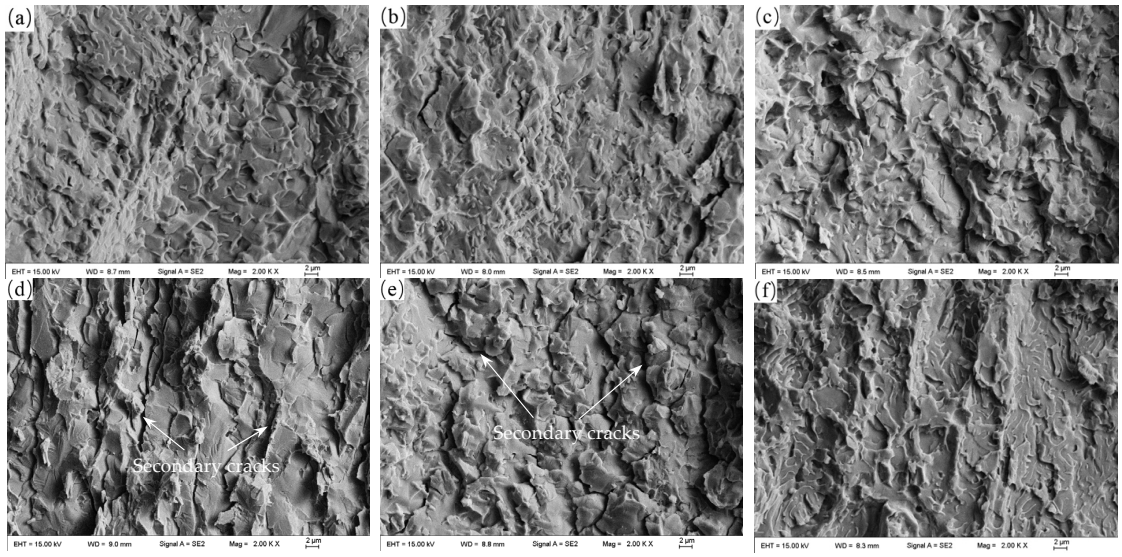

Figure 9. Fracture morphologies of the brazed joints using Cu-7P-1Ag-*x*Sn filler metals: (**a**) Cu-7P-1Ag, (**b**) Cu-7P-1Ag-0.1Sn, (**c**) Cu-7P-1Ag-0.3Sn, (**d**) Cu-7P-1Ag-0.5Sn, (**e**) Cu-7P-1Ag-0.7Sn, (**f**) Cu-7P-1Ag-1Sn.

4. Conclusions

The effect of Sn on the melting characteristics, spreadability, and microstructures of Cu-7P-1Ag brazing filler metals was studied. Additionally, the mechanical properties of H62 brass joints brazed with different low-silver Cu-P filler metals were investigated. The following conclusions can be drawn from this study:

(1) The solidus and liquidus temperatures of Cu-7P-1Ag-*x*Sn filler metals decrease with the addition of Sn. Meanwhile, the spreadability of the filler metals on H62 brass substrates is significantly improved.

(2) Cu-7P-1Ag-*x*Sn filler metals are mainly comprised of α-Ag solid solution, α-Cu solid solution and Cu_3P. $Cu_{20}Sn_6$ compound is formed in the matrix of Cu-7P-1Ag-1Sn filler metal. With the addition of Sn, the microstructure of the filler metal gradually changes from thick dendrites into tiny worm-like structures.

(3) With increasing Sn content, the microstructure of the brazed joints gradually changes from a bulk to a lamellar structure. Ag and Sn have higher diffusion coefficients into H62 brass substrate than P.

(4) The strength of H62 brass brazed joints presents a parabolic trend according to Sn content. A peak value of 348 MPa was achieved using Cu-7P-1Ag-0.5Sn filler metal. The fracture morphology of the joint brazed with Cu-7P-1Ag-0.5Sn showed quasi-cleavage fractures with secondary cracks and tearing edges, whereas when Cu-7P-1Ag-1Sn was studied, the fracture surface showed typical brittle fracture characteristics.

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