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Effect of In and Pr on the Microstructure and Properties of Low-Silver Filler Metal

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Abstract: The novel low-silver 12AgCuZnSn filler metals containing In and Pr were used for flame brazing of copper and 304 stainless steel in this study. The effects of In and Pr content on the melting temperature, wettability, mechanical properties and microstructure of 12AgCuZnSn filler metal were analyzed. The results indicate that the solidus and liquidus temperatures of filler metals decrease with the addition of In. Trace amounts of Pr have little impact on the melting temperature of the low-silver filler metals. In addition, the spreading area of filler metals on copper and 304 stainless steel is improved. The highest shear strength of brazed joint is 427 MPa when the content of In and Pr are 2 wt.% and 0.15 wt.%, respectively. Moreover, it is observed that the trace amount of Pr significantly refines the microstructure of brazed joint matrix. A bright $Pr_3Cu_4Sn_4$ phase is found in filler metal and brazing seam when the contents of In and Pr are 5 wt.% and 0.5 wt.%, respectively.

Keywords: low-silver filler metals; melting temperature; wettability; microstructure; mechanical properties

Citation: Wu, J.; Xue, S.; Zhang, P. Effect of In and Pr on the Microstructure and Properties of Low-Silver Filler Metal. *Crystals* **2021**, *11*, 929. https://doi.org/10.3390/cryst11080929

Academic Editor: Umberto Prisco

Received: 14 July 2021
Accepted: 8 August 2021
Published: 11 August 2021

1. Introduction

Silver-based filler metals have a diverse range of applications in aerospace, electrical appliances, automobile industries, refrigeration applications navigation and military application in view of their distinguished brazing properties, such as, low melting point, high joint strength, excellent liquidity, strong corrosion resistance and other positive features [1,2]. In the past few decades, AgCuZnCd series alloys have been used for brazing dissimilar joints of ferrous metals and copper alloys due to their excellent comprehensive properties [3]. The addition of Cd to AgCuZn system reduces the solidus and liquidus temperature and the melting range and improves the fluidity of the alloys on the substrate [4]. However, the fumes produced by cadmium-containing silver brazing filler metals during brazing processes are highly toxic to humans, which has caused the application of AgCuZnCd series brazing alloys to be greatly restricted [5]. Furthermore, the high cost of silver is also a major factor restricting the wide application of silver brazing alloys. Therefore, it is of interest to develop cadmium-free and low-silver brazing filler metals as the alternatives [6,7].

The decrease of Ag content will seriously affect the brazing performance of silver-based filler metals [8,9]. Thus far, many attempts have been made to solve the problem and to produce cadmium-free low-silver alloys that have similar characteristics of low melting point, excellent fluidity and high joint strength, several novel brazing filler metals have been developed, such as AgCuTi [10], AgCuZnMnNi [11], AgCuZnSn [12,13], AgCuZnSnIn [14], AgCuZnSnCe [15] and AgCuZnSnGa [16], which have resulted in the initial application and development of Cd-free silver-based filler metals in green manufacturing. Lai [17] has reported that the addition of Ga, Sn, In, and Ni can optimize the microstructure and mechanical properties of 30AgCuZn filler metal, but the Ag content is still very high.

Indium element is a fifth as expensive as silver, and the melting point of Indium is only 156.6 °C. The appropriate amount of indium can dissolve in copper and silver to form silver-based and copper-based solid solutions with lower melting points [18]. China is the absolute largest country in rare earth reserves, and the price is much lower than that of silver; meanwhile, rare earth elements are known as "industrial gold", which can significantly enhance the properties of metals [19]. In this study, 12AgCuZnSn filler metals with designed additions of In and Pr were prepared. The effect of In and Pr addition on the melting characteristics, wettability and microstructure of novel brazing alloys was investigated; meanwhile, the microstructure, mechanical property and fracture morphologies of the brazing joints were studied.

2. Materials and Methods

Pure Ag, Cu, Zn, Sn, In (99.9 wt.% purity) and Cu-10Pr master alloy were used as raw materials and melted in a medium frequency furnace (frequency 600 Hz, power 110 kW), and borax was used as the cover material to prevent the melting alloys from oxidizing. Then, the molten alloys were poured into a steel mold to get cast ingots, and all the cast ingots were drawn into a wire with a 1.9 mm diameter for brazing. The designed compositions of the novel low-silver filler metals used in the present study are listed in Table 1.

Table 1. Designed composition of the novel filler metals (wt.%).

Filler Metals No.	Ag	Cu	Zn	Sn	In	Pr
1	12.0	Bal.	38.3	1.5	1.0	0.1
2	12.0	Bal.	37.8	1.5	2.0	0.1
3	12.0	Bal.	37.8	1.5	2.0	0.15
4	12.0	Bal.	37.7	1.5	2.0	0.3
5	12.0	Bal.	37.6	1.5	2.0	0.5
6	12.0	Bal.	36.1	1.5	5.0	0.5

The solidus and liquidus temperatures of the brazing alloys were determined using differential thermal analysis (DTA, HCR-1, HENVEN, Beijing, China) with the heating rate and nitrogen flow rate being 10 °C/min and 200 mL/min, respectively. The test plates of 304 stainless steel and commercial pure copper supplied for the spreading test and brazing were machined to plates with dimensions of 40 mm × 40 mm × 2 mm and 60 mm × 25 mm × 3 mm, respectively. The spreading test was performed according to China's National Standard GB/T 11364–2008 [20]. The novel silver filler metal (0.2 g) was placed on surface of the specimen covered with FB102, which was heated at 850 °C for 1 min in an electrical resistance furnace. The spreading area was photographed from above using a digital cameraand calculated using Image-Pro Plus software (Media Cybernetics, Rockville, MD, USA).

Automatic oxy-acetylene torch method was used in this study for brazing copper and 304 stainless steel. The shear strength of joints with an overlap length of 2 mm was tested on an electronic universal testing machine according to the China's National Standard GB/T 11363-2008 [21]; the constant loading rate of 5 mm/min under room temperature was applied in this study. All the filler metals and joined surfaces were polished by SiC papers and ultrasonically cleaned by acetone prior to braze. To ensure the accuracy and reliability of the results, the wettability and shear strength tests were done five times under the same condition.

The brazing filler metals and joints interface layer specimen were etched about 7–8 s with a solution of (($(NH_4)_2S_2O_8$ (15 g) + H_2O (100 mL) + $NH_3{\cdot}H_2O$ (2 mL)) after grinding and polishing, and then, the mounted specimens and fracture morphologies of copper/304 stainless steel brazing joints were checked using the energy-dispersive (EDS, Bruker Nano XF Lash Detector 5010, Billerica, MA, USA) and scanning electron microscopy (SEM, ZEISS Σ IGMA 500, Oberkochen, Germany). In addition, the phase composition of the brazing alloys was performed using an X-ray diffractometer (XRD, Bruker D8 Advance, Billerica, MA, USA) with Cu Kα radiance.

3. Results

3.1. Melting Temperature of the Novel Low-Silver Filler Metals

The differential thermal analysis (DTA) curves of 12AgCuZnSn-*x*In-*y*Pr brazing filler metals are shown in Figure 1, and the solidus temperatures (T_s) and liquidus temperatures (T_l) are marked by arrows in DTA traces (the red line is the extrapolated baseline and the tangent to the maximum slope point of the peak leading edge, and the intersection point of the red line is T_s and T_l, respectively). As we can see from Figure 1b–e, when the Pr content was increased from 0.1 wt.% to 0.5 wt.%, the T_s and T_l of the filler metals decreased by 6 °C and 3 °C, respectively, the results show that the addition of trace Pr has little impact on the melting temperature of the novel low silver filler metals. Similar results were reported by Lai [22] when the effect of rare-earth element Pr on the properties of 30AgCuZnSn filler metal was investigated.

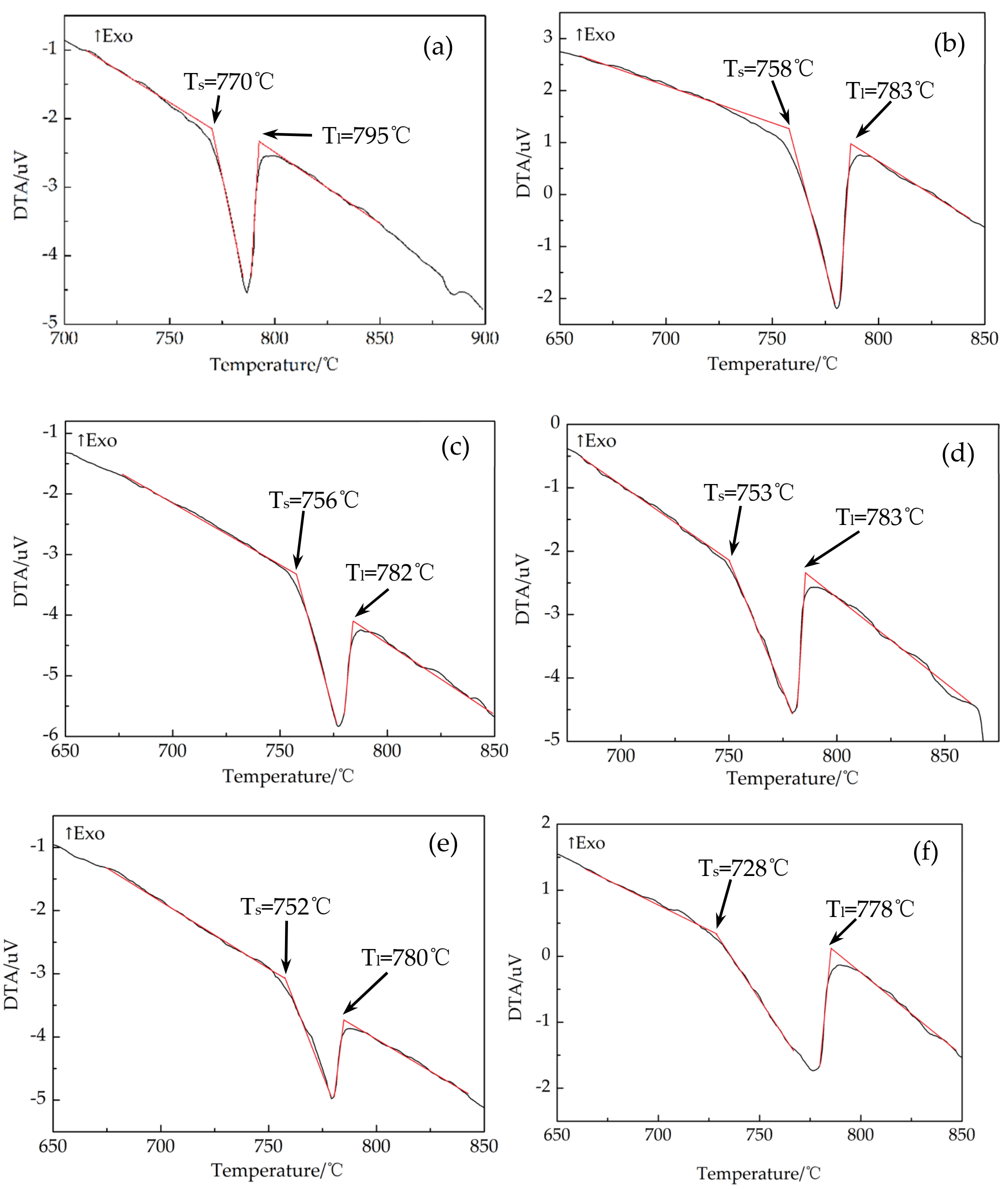

Figure 1. DTA curves of the novel low-silver filler metals: (**a**) 12AgCuZnSn-1In-0.1Pr, (**b**) 12AgCuZnSn-2In-0.1Pr, (**c**) 12AgCuZnSn-2In-0.15Pr, (**d**) 12AgCuZnSn-2In-0.3Pr, (**e**) 12AgCuZnSn-2In-0.5Pr, (**f**) 12AgCuZnSn-5In-0.5Pr.

However, the T_s and T_l of the filler metals were suppressed by increasing In content, as shown in Figure 1f, when the contents of In and Pr are 5 wt.% and 0.5 wt.%, the T_s and

T_l of the brazing filler metal decreased to 728 °C and 778 °C, respectively. The distinct decrease of both T_s and T_l of the filler metal is mainly attributed to the fact that the melting point of In is only 156.61 °C. Appropriate amounts of In can dissolve in copper and silver to form silver-based and copper-based solid solutions with lower melting points, which greatly reduce the melting temperature of low-silver filler metals [14].

3.2. Wettability of the Novel Low-Silver Filler Metals

In general, the wettability is characterized by the spreading area of liquid brazing filler metals on the substrate, the greater the spreading area, the better the wettability [23]. Figure 2 shows the spreading areas of 12AgCuZnSn-*x*In-*y*Pr brazing filler metals on copper and 304 stainless steel substrates at the temperature of 850 °C with FB102 flux. It can be seen that appropriate amounts of In and Pr are beneficial to the spreading performance. When the content of Pr is 0.1%, the spread areas of the filler metals on the substrate increase with the increase of In content from 1% to 2%, the same result can be obtained by comparing No.5 and No.6 filler metals, which indicated that the addition of In can improve the wettability of the filler metals on the substrate.

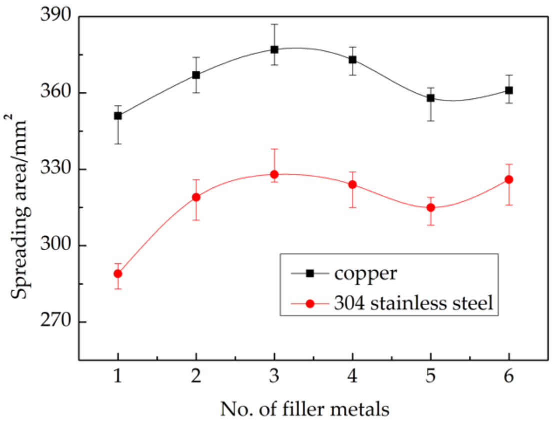

Figure 2. Spreading areas of low-silver filler metals on copper and 304 stainless steel plates.

Notably, the 12AgCuZnSn-2In-0.15Pr (No.3 in Figure 2) has the biggest spreading area among the novel low-silver filler metals and reaches 377 mm^2 and 328 mm^2 on copper and 304 stainless steel substrates, respectively. Pr is an active element, which can react preferentially to oxygen and accumulate to the surface of liquid filler metals; consequently, the surface tension of molten brazing alloys is decreased effectively. However, if the content of Pr in the brazing alloys is higher than 0.3 wt.%, the wettability begins to decrease.

Figure 3 shows the high magnification SEM microstructures and the elements mappings of the substrate surface after the spreading test on 304 stainless steel using 12AgCuZnSn-2In-0.5Pr filler metal, and the EDS of the points indicated in Figure 3 is shown in Table 2. As we can see from Figure 3a, there was a pre-spreading part on the leading edge of the molten brazing alloy, which consisted of a white phase (named as A region) and a gray phase (named as B region). The results of EDS shown in Table 2 and the elements mappings of the substrate surface in Figure 3 indicated that A and B regions were composed of Ag-In-Sn alloy and stainless-steel substrate, respectively; the preferentially spreading Ag-In-Sn phase liquid alloy greatly improves the wetting performance of the brazing filler metal on base metal. However, we have known from previous reports [24] that the wetting ring of 12AgCuZnSn-2In on 304 stainless-steel substrate was a regular circle, which consisted of a smooth white phase. In this study, the appearance of the wetting ring is extremely irregular, and the white phase shows a coarse honeycomb shape. The main cause may be that the excess Pr elements could form oxide slag, which would hinder the spread of the liquid brazing alloy on the substrate.

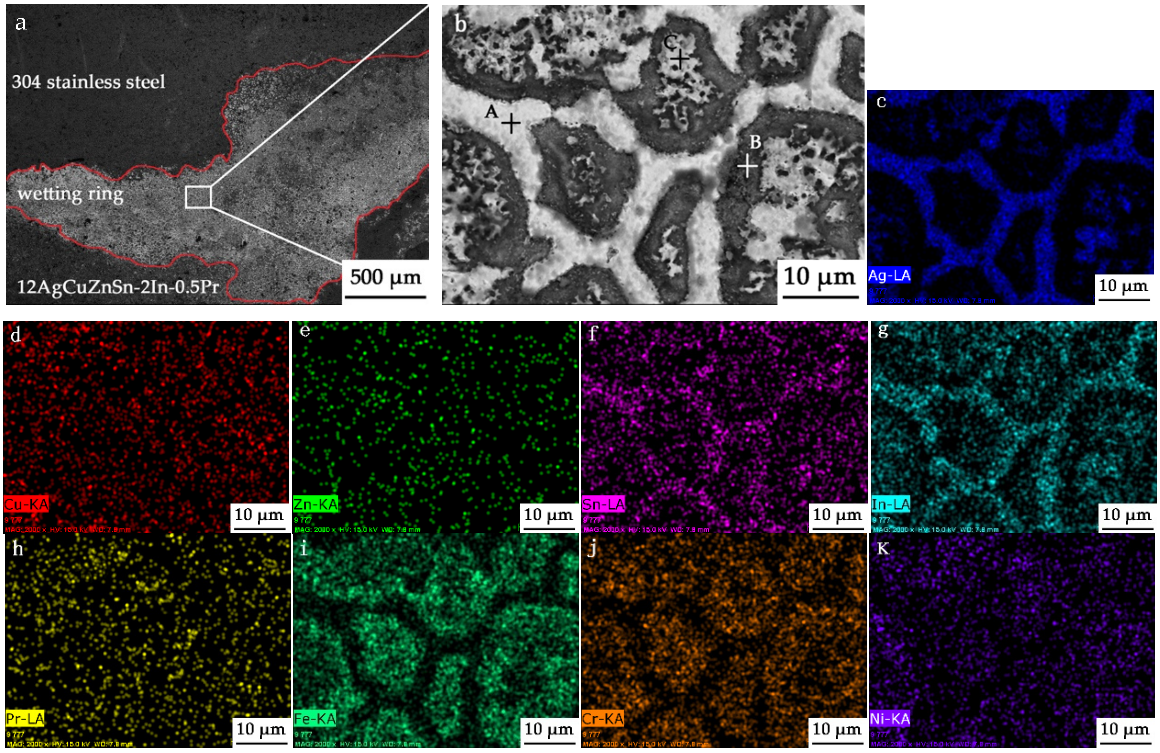

Figure 3. SEM image (**a**,**b**) of surface appearance after spreading of 12AgCuZnSn-2In-0.5Pr filler metal on 304 stainless steel and corresponding the elements mapping: (**c**) Ag, (**d**) Cu, (**e**) Zn, (**f**) Sn, (**g**) In, (**h**) Pr, (**i**) Fe, (**j**) Cr, (**k**) Ni.

Table 2. EDS analysis results of the points indicated in Figure 3b (at.%).

Points	Ag	Cu	Zn	Sn	In	Pr	Fe	Cr	Ni
A	70.64	10.34	2.55	2.72	8.82	0.68	2.12	1.44	0.69
B	0.06	0.65	0.92	0.07	0.11	0.14	70.57	19.85	7.63
C	59.39	6.30	1.48	1.43	5.65	1.76	18.02	4.23	1.74

3.3. Microstructure of the Novel Low-Silver Filler Metals

In order to clarify the phase constitution of the novel filler metals, X-ray diffraction (XRD) analysis was carried out under certain conditions, and Figure 4 shows the XRD patterns. The results of the XRD analysis suggest that 12AgCuZnSn-2In-0.15Pr and 12AgCuZnSn-2In-0.5Pr filler metals both contain three phases: Ag-based solid solution phase, Cu-based solid solution phase and CuZn compounds phase (β-CuZn and few γ-Cu_5Zn_8 phase). However, the diffraction peaks of Cu_4In and Ag_9In_4 intermetallic compounds phase arose when the contents of In and Pr were 5 wt.% and 0.5 wt.%, respectively, and previous studies [24] have noted that the Cu_4In and Ag_9In_4 intermetallic compounds would deteriorate the mechanical properties of the brazing alloy. Nevertheless, the content of Pr in the brazing alloys is still too little to be detected.

Figure 5 shows the SEM images of the novel low-silver filler metals as-extrude, and Table 3 shows the EDS analysis results of the points indicated in Figure 5. As we can see from Figure 5d and Table 3 that the contents of Ag, In and Sn in white needle phase (named as A region) are higher than the dark bulk phase (named as B region), which indicated that A and B regions were composed of Ag-rich phase and Cu-rich phase, respectively. By comparing the microstructure of Figure 5b–f, it is clearly found that the addition of small amount of Pr into 12AgCuZnSn-2In brazing filler metal could produce a remarkable refinement, the grains size of 12AgCuZnSn-2In-0.15Pr brazing filler metal is only 10 μm–15 μm, which is beneficial for the mechanical properties of the brazing alloys.

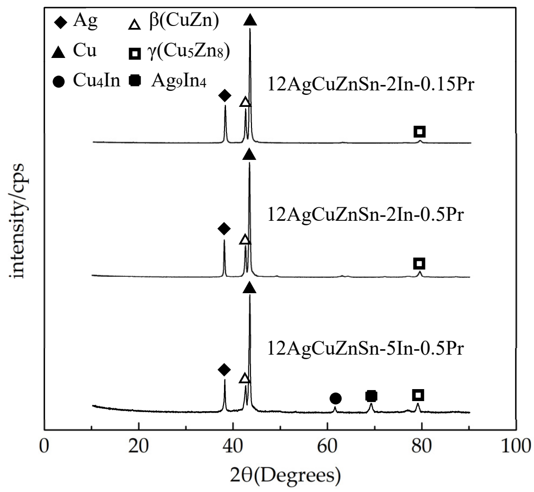

Figure 4. XRD pattern of the novel filler metals.

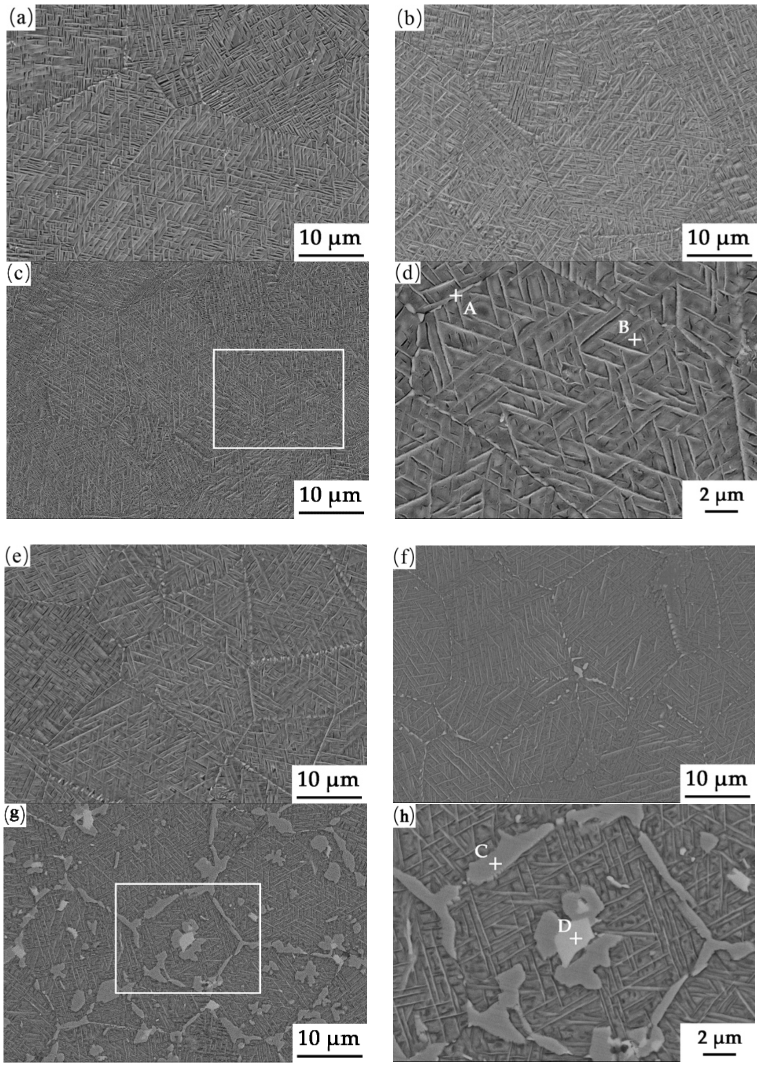

Figure 5. SEM images of the novel low-silver filler metals as-extrude: (**a**) 12AgCuZnSn-1In-0.1Pr, (**b**) 12AgCuZnSn-2In-0.1Pr, (**c**) 12AgCuZnSn-2In-0.15Pr, (**d**) 12AgCuZnSn-2In-0.15Pr (high magnification SEM image of area marked with white square in Figure 5c), (**e**) 12AgCuZnSn-2In-0.3Pr, (**f**) 12AgCuZnSn-2In-0.5Pr, (**g**) 12AgCuZnSn-5In-0.5Pr, (**h**) 12AgCuZnSn-5In-0.5Pr (high magnification SEM image of area marked with white square in Figure 5g).

Table 3. EDS analysis of the points indicated in Figure 5 (at.%).

Points	Ag	Cu	Zn	Sn	In	Pr
A	29.32	35.73	29.84	2.49	2.54	0.08
B	4.85	59.77	34.60	0.41	0.32	0.05
C	13.01	44.99	31.83	2.13	8.02	0.02
D	7.86	30.62	5.56	31.48	0.92	23.56

However, as the In and Pr content in the brazing alloys increases to 5 wt.% and 0.5 wt.%, respectively, some grey phases (named as C region) and bright phases (named as D region) were formed in the matrix. Compared to other areas of SEM images of the novel low-silver filler metals, the results of EDS shown in Table 3 and the XRD patterns shown in Figure 4 indicated that C region was a mixed phase rich in Cu_4In and Ag_9In_4 intermetallic compounds, and D region was a Pr-rich phase. Riani et al. [25] had studied the ternary system of Pr-Cu-Sn, which show that the new Pr-rich phase should be the $Pr_3Cu_4Sn_4$. Figure 6 shows the elements mappings of 12AgCuZnSn-5In-0.5Pr brazing filler metal in Figure 5h, the result indicated that the elements mapping of Pr and Sn overlaps with D region and the In-rich phase surround the rare-earth phase.

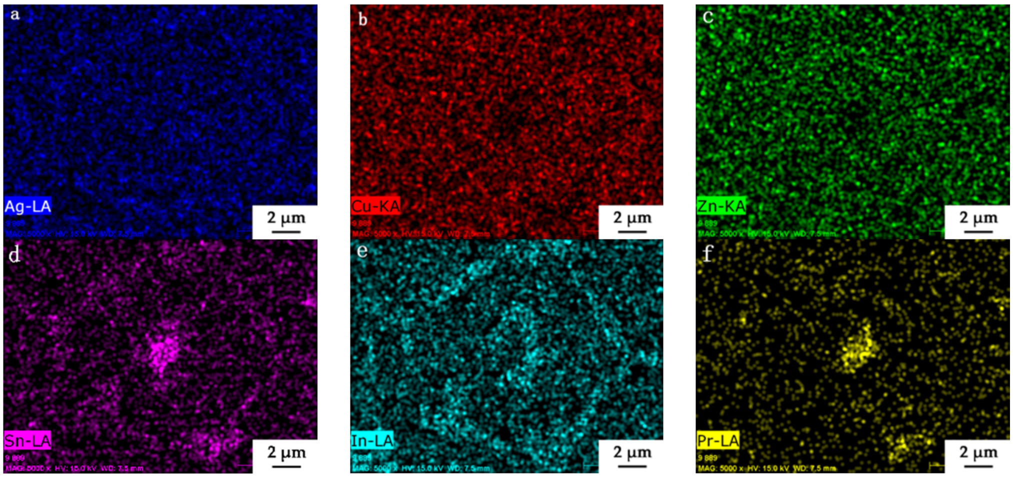

Figure 6. The elements mapping of SEM image of 12AgCuZnSn-5In-0.5Pr brazing filler metal in Figure 5h: (**a**) Ag, (**b**) Cu, (**c**) Zn, (**d**) Sn, (**e**) In, (**f**) Pr.

3.4. Microstructure of the Brazing Joints

The typical microstructure of copper/304 stainless steel joints with different contents of In and Pr is shown in Figure 7, and the thickness of the brazing seam is slightly different due to the test error and the thermal deformation of the specimens. As we can see from the general view in Figure 7a–f, the successful joint of copper to steel was achieved using flame brazing method, and the microstructure was homogeneous, and no apparent defects were observed in the joint. Moreover, the size of the grey bulk phases is significantly refined when the In and Pr contents are 2 wt.% and 0.15 wt.%, respectively; the main reason may be the enrichment of trace Pr elements in the grain boundary region, hindering the growth of the grey bulk phases in the brazing seam. However, as shown in Figure 7d–f, when the Pr element content in the brazing alloy continued to increase, the refinement of the microstructure of the brazing alloys by Pr element began to weaken; meanwhile, some bright phases were formed in the brazing seam.

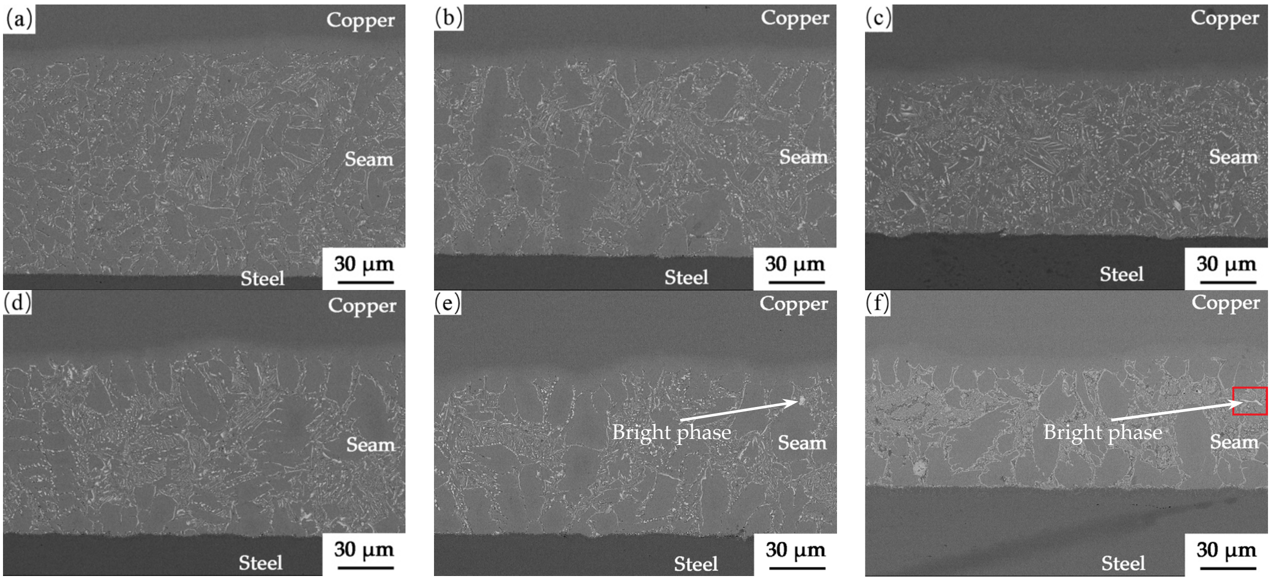

Figure 7. Microstructure of the copper/304 stainless steel joints using different low-silver filler metals: (**a**) 12AgCuZnSn-1In-0.1Pr, (**b**) 12AgCuZnSn-2In-0.1Pr, (**c**) 12AgCuZnSn-2In-0.15Pr, (**d**) 12AgCuZnSn-2In-0.3Pr, (**e**) 12AgCuZnSn-2In-0.5Pr, (**f**) 12AgCuZnSn-5In-0.5Pr.

In order to clarify the microstructure of brazing seam, high magnification SEM micrograph of interfacial microstructures of 12AgCuZnSn-5In-0.5Pr brazing seam and the elements mappings were carried out under certain conditions, and the test results are shown in Figure 8. The microstructure of brazing seam was characterized by three different phases: grey bulk phase (named as A region), bright phase (named as B region) and greyish white phase (named as C region). According to the results of EDS shown in Table 4, the A and C regions are mainly composed of Ag, Cu and Zn, and combining the Ag-Cu-Zn isothermal section at 350 °C, it was inferred that grey bulk phase was a mixed phase of Cu-based solid solution and β-CuZn; greyish white phase was a mixed phase of Ag-based solid solution and Cu-based solid solution; similar results were reported by Cao [26] when the intermetallic compounds formation between brass/steel and AgCuZnSn filler metal was investigated. Notably, the elements mapping of Pr and Sn overlaps with B region; combined with the EDS analysis results, we can conclude that the bright phase should be the $Pr_3Cu_4Sn_4$, and the rare-earth phase is generally brittle, which may worsen the mechanical properties of brazing joints [27].

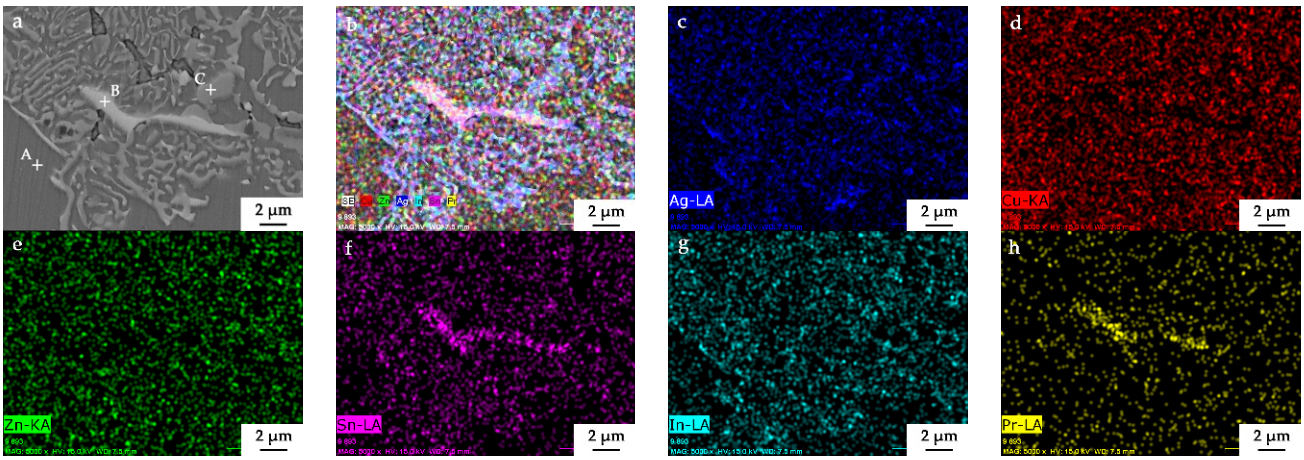

Figure 8. SEM image and corresponding the elements mapping. (**a**) high magnification SEM image of area marked with red square in Figure 7f, (**b**) the elements mapping, (**c**) Ag, (**d**) Cu, (**e**) Zn, (**f**) Sn, (**g**) In, (**h**) Pr.

Table 4. EDS analysis results of the points indicated in Figure 8 (at.%).

Points	Ag	Cu	Zn	Sn	In	Pr
A	6.73	59.56	32.57	0.62	0.47	0.05
B	9.10	28.85	6.86	29.60	0.92	24.67
C	40.24	29.63	20.66	2.54	6.85	0.08

3.5. Mechanical Properties of Brazed Joints

The shear strength measurements of copper/304 stainless steel and 304 stainless steel/304 stainless steel lap joints using the novel 12AgCuZnSn-*x*In-*y*Pr were performed at room temperature with a constant loading rate of 5 mm/min. Notably, the fracture occurred on the copper substrate of the copper/304 stainless steel brazed specimens in all cases after shear tests, which indicated that the copper/steel joints with excellent mechanical properties were achieved.

It is well known that the strength of the 304 stainless steel plate is higher than that of the copper plate, as expected, the fracture occurs in the joints of the steel/steel brazed specimens in all cases, the results of shear strength are shown in Figure 9, and the corresponding fracture morphologies of the brazing seam are shown in Figure 10. As we can see from the Figure 9, the peak shear strength of 427 MPa is acquired using 12AgCuZnSn-2In-0.15Pr for brazing, which increases by 26.7% compared to that of previously studied 12AgCuZnSn filler metals [24], and the corresponding fracture morphology in Figure 10c exhibits typical ductile characteristic with obvious dimples.

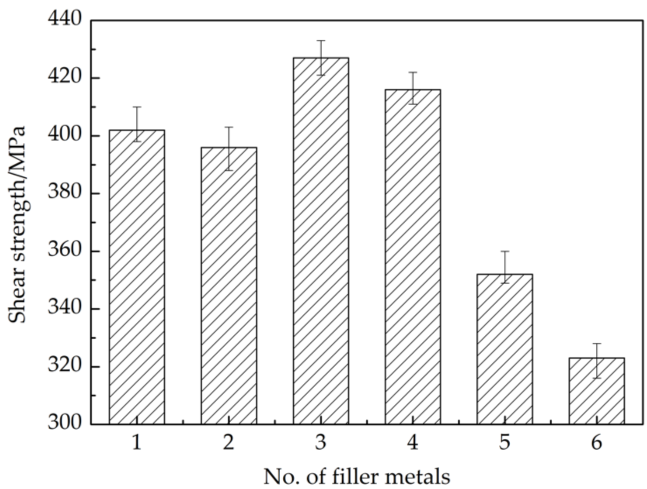

Figure 9. Shear strengths of the steel/steel lap joints using 12AgCuZnSn-*x*In-*y*Pr.

However, the shear strength of brazed joints drops when the content of In and Pr is further increased. As the In and Pr contents of the brazing alloy reach 2 wt.% and 0.5 wt.%, respectively, the shear strength of the brazed joint is reduced to 352 MPa, the corresponding fracture morphology in Figure 10e shows a mixed fracture. The elements mappings of the fracture morphology of the brazing joint of 12AgCuZnSn-2In-0.5Pr filler metal were shown in Figure 11. It is clearly found from Figure 11f that the globular particles are the Pr-rich phase, and rare-earth phases easily become the source of crack initiation and propagation and deteriorate the mechanical properties of brazed joint [28]. In addition, when the content of In and Pr reaches 5 wt.% and 0.5 wt.%, respectively, the shear strength is reduced to 323 MPa, even lower than that of the 12AgCuZnSn brazed joint, and the corresponding fracture morphology in Figure 10f shows a typical brittle fracture. All of the above analyses indicated that the changes in the fracture morphologies are attributed to the microstructure and the shear strength of the brazed joints.

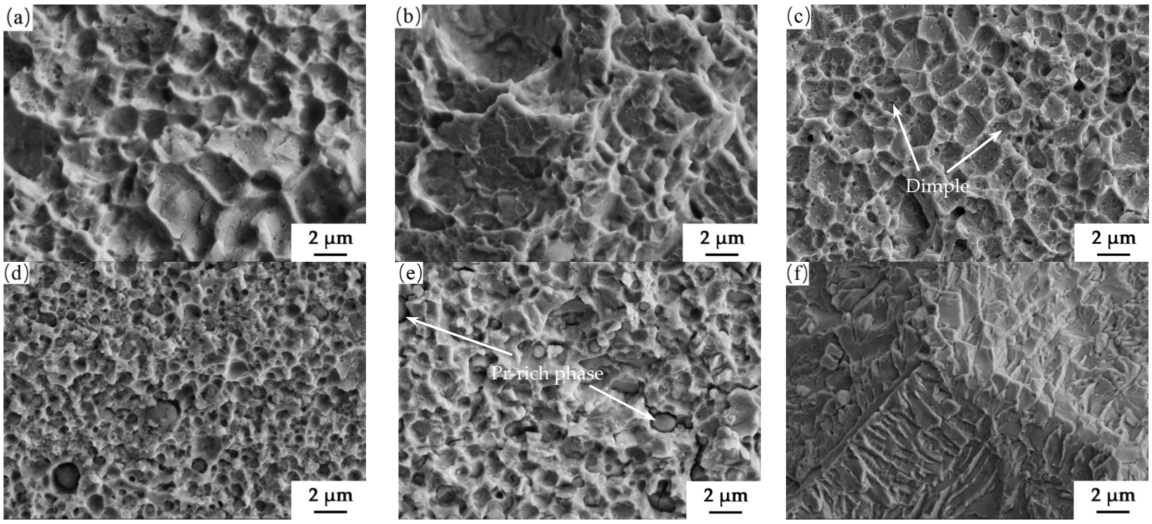

Figure 10. Fracture morphologies of the brazing joints using different low-silver filler metals: (**a**) 12AgCuZnSn-1In-0.1Pr, (**b**) 12AgCuZnSn-2In-0.1Pr, (**c**) 12AgCuZnSn-2In-0.15Pr, (**d**) 12AgCuZnSn-2In-0.3Pr, (**e**) 12AgCuZnSn-2In-0.5Pr, (**f**) 12AgCuZnSn-5In-0.5Pr.

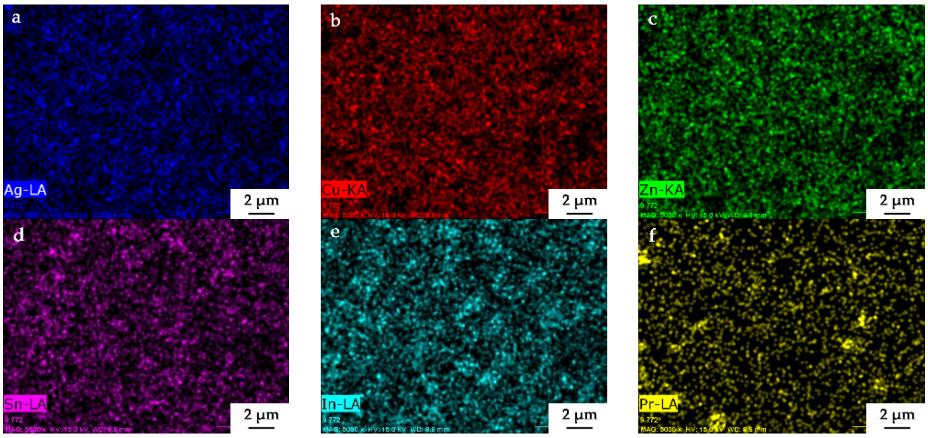

Figure 11. The elements mappings of the fracture morphology of the brazing joint of 12AgCuZnSn-2In-0.5Pr filler metal in Figure 10e: (**a**) Ag, (**b**) Cu, (**c**) Zn, (**d**) Sn, (**e**) In, (**f**) Pr.

4. Conclusions

The effects of In and Pr on the melting temperature, wettability and microstructure of 12AgCuZnSn-*x*In-*y*Pr brazing filler metals were studied. Moreover, the mechanical properties and fracture morphologies of the brazing joints using different low-silver filler metals were investigated, and the following conclusions were obtained:

(1) The decreases in both T_s and T_l are attributed to the addition of In; trace amounts of Pr have little impact on the melting temperature of the low-silver filler metals.
(2) The spreading area of the filler metals on copper and 304 stainless steel substrate reaches the peak when the contents of In and Pr are 2 wt.% and 0.15 wt.%, respectively. Excessive Pr elements will inhibit the wettability of the filler metals.
(3) The microstructure of the filler metal and brazing joints produces a significant refinement when the Pr content is 0.15 wt.%. However, Cu_4In, Ag_9In_4 and bright $Pr_3Cu_4Sn_4$ phase were formed in the 12AgCuZnSn-5In-0.5Pr brazing alloy.
(4) The peak shear strength of steel/steel brazing joint is obtained using 12AgCuZnSn-2In-0.15Pr filler metal, and the corresponding fracture morphology exhibits typical ductile characteristic with obvious dimples. However, some globular Pr-rich particles are found in the fracture morphology when the content of Pr reaches 0.5 wt.%.

Author Contributions: Conceptualization, S.X.; methodology, P.Z.; software, J.W.; validation, J.W.; formal analysis, J.W.; investigation, J.W.; resources, J.W.; data curation, J.W.; writing—original draft preparation, J.W.; writing—review and editing, J.W.; visualization, J.W.; supervision, S.X.; project administration, S.X.; funding acquisition, S.X. All authors have read and agreed to the published version of the manuscript.

Funding: This work was funded by the National Natural Science Foundation of China, Grant No. 51975284, and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD).

Acknowledgments: The authors sincerely acknowledge the financial supports by the National Natural Science Foundation of China, Grant No. 51975284, as well as the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD).

Conflicts of Interest: The authors declare no conflict of interest.

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