

## Article

# Effect of Sn/Ni Addition on the Microstructure and Properties of Flux-Cored Brass Filler Metal and Its Brazed Joints

Wei Liu <sup>1</sup>, Xin Huang <sup>1</sup>, Lijun Ren <sup>1</sup>, Jungang Sun <sup>2</sup> and Yongzhou Bu <sup>2,\*</sup><sup>1</sup> College of Materials Science and Chemistry, China Jiliang University, Hangzhou 310018, China; liuweicjl@cjlu.edu.cn (W.L.); xin.huang@cjlu.edu.cn (X.H.); lijunren@cjlu.edu.cn (L.R.)<sup>2</sup> Hangzhou Huaguang Advanced Welding Materials Co., Ltd., Hangzhou 311112, China; sunjungang@cn-huaguang.com

\* Correspondence: buyongzhou@cn-huaguang.com

## Abstract

In this study, Sn and Ni powders were incorporated into the flux core of brass brazing filler metals, with alloying achieved via in situ synthesis during brazing. The effects of Sn/Ni single and composite additions on the microstructure, melting characteristics, wettability on Q235 steel, and tensile strength of corresponding brazed joints were systematically investigated. Sn addition increased the  $\beta$ -phase fraction, reduced the solidus temperature, and significantly improved wettability—with a maximum unit spreading area of 4.22 mm<sup>2</sup>/mg at 20 wt.% Sn (2.85 times that of the Sn/Ni-free baseline). However, coarse  $\beta$ -phase grains and their transformation to brittle  $\beta'$ -phase at room temperature resulted in no enhancement of joint tensile strength. Ni addition expanded the  $\alpha$ -phase region, refined grains, and induced solid solution strengthening of the  $\alpha$ -phase matrix; joint tensile strength peaked at 501 MPa at 20 wt.% Ni (65% higher than the baseline). Excessive Ni ( $\geq 40$  wt.%) deteriorated wettability due to reduced molten superheat, and 50 wt.% Ni caused  $\alpha$ -phase over-strengthening and embrittlement, leading to a sharp strength drop to 214 MPa. The composite addition of 3 wt.% Sn + 10 wt.% Ni reconstructed the filler metal into a refined, uniform grid-like ( $\alpha + \beta$ ) dual-phase structure without new phase formation, realizing synergistic optimization of wettability and mechanical properties. The co-added sample exhibited optimal performance, with a unit spreading area of 4.51 mm<sup>2</sup>/mg and joint tensile strength of 584 MPa (206% and 93% higher than the baseline, respectively). This improvement was attributed to the coupling of uniform stress dispersion by the grid-like microstructure and dual-element functional complementation (Sn for wettability and  $\beta$ -phase strengthening; Ni for grain refinement and  $\alpha$ -phase strengthening). This work provides a feasible alloying modification strategy for brass flux-cored brazing filler metals, and the revealed microstructure-performance regulation mechanism offers a valuable reference for developing high-performance brass brazing filler metals.



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**Keywords:** brass flux-cored brazing filler metal; Sn/Ni alloying; microstructure; wettability; brazed joint tensile strength

## 1. Introduction

Brass brazing filler metals are the most widely used and highest-volume copper-based brazing materials in industry, owing to their low cost, moderate melting temperature, and excellent wettability [1–3]. Flux-cored brass brazing filler metals represent ideal and key materials for efficient, clean, automated, and intelligent brazing processes, with distinct

advantages including no requirement for external flux, facile wire feeding, and superior operational stability compared with manual brazing [4]. However, they account for only 10–20% of the total brass brazing filler metal market, primarily due to unresolved challenges in product quality control and production costs. Their fabrication involves more demanding technical requirements for flux core cladding, wire drawing, and composition uniformity control than silver-based and aluminum-based flux-cored brazing fillers. Optimizing the manufacturing process to achieve breakthroughs in both cost reduction and performance enhancement thus holds significant theoretical significance and engineering value for advancing the automation and green development of the brazing industry.

The production of flux-cored brazing materials involves multiple plastic deformation processes (e.g., rolling, curling), which impose stringent requirements on the deformability of the brass outer sheath. This technical constraint restricts the selection of alloying elements for the sheath, resulting in an inevitable trade-off between the formability of the outer layer and the brazing performance of traditional brass filler metals—an intractable bottleneck limiting their application in high-end manufacturing [4].

Alloy composition optimization is a mainstream strategy for improving the performance of brazing materials, and extensive studies have focused on this approach for silver-based and aluminum-based brazing fillers [5–8]. For silver-based flux-cored brazing materials, Alan et al. [5] improved wettability, corrosion resistance, and joint strength by adding 2 wt.% Ni to 30Ag fillers; Ma et al. [6,7] realized comprehensive performance enhancement of low-Ag fillers via Ce and Ga addition; Yu [8] developed low-temperature AgCuZnSn flux-cored brazing alloys by adjusting Zn and Sn contents. Nevertheless, these studies focus on silver-based fillers with inherently excellent formability, where only brazing performance requires optimization. For brass brazing fillers, the trade-off between formability and brazing performance is far more prominent: alloying elements that improve the mechanical properties of brass often deteriorate its brazing performance, and vice versa.

In the research field of brazing filler metals, Pb and Cd are excluded for environmental considerations, while Ag, Sn [9], In [10], Ga [11], Ni [12], Cu, and Zn are all commonly used alloying elements. Among these, besides Cu and Zn—the primary elements of the brazing filler metal investigated in this work—Ag, In, and Ga are precious metals, which render Sn and Ni the preferred alloying choices. Sn, in particular, is of great importance for both application and research due to its low melting point, low cost, ability to reduce the solidus and liquidus temperatures of brazing fillers, and improve the fluidity of molten filler metals. Luo et al. [9] systematically summarized and analyzed the effects of Sn on the melting characteristics, wettability, microstructure, and mechanical properties of copper-based brazing filler metals, clarifying the regulatory role of Sn in copper-based brazing systems. Dong [13] noted that Sn exerts a positive effect on the wettability of Cu-Zn fillers and reduces their melting temperature, but the Sn content in industrially manufactured Cu-Zn-Sn fillers must be strictly controlled at a low level. The tensile strength of brazed joints increases gradually with rising Sn content and reaches a maximum at 1.6 wt.% Sn; excessive Sn content leads to the formation of brittle Cu-Sn intermetallic compounds, which distribute within the low-plasticity  $\beta$  phase and cause deterioration in filler metal plasticity, subsequently reducing the tensile strength of brazed joints. As a result, high-Sn-content Cu-Zn fillers suffer from high brittleness that impairs their processing and formability, thus greatly limiting the industrial application of such brass fillers.

Chen et al. [14] systematically investigated the effect of Ni addition on the microstructure of Cu-Zn alloys, revealing that Ni can effectively refine the microstructure and form precipitation-strengthening phases when co-doped with Si. Specifically, the addition of 1.5 wt.% Ni significantly enhanced the tensile strength of the Cu-20Zn alloy, increasing it

from 373.2 MPa to 776.4 MPa, which confirms the remarkable strengthening effect of Ni on Cu-Zn alloy systems.

Previous studies have demonstrated that a suitable alloying strategy can effectively enhance the strength of Cu-Zn alloys. When employed as Cu-Zn brazing fillers, such strengthened alloys can accordingly improve the strength of brazed joints after welding. However, these strengthened Cu-Zn alloys exhibit poor deformability. If used as the outer sheath material, they cannot easily undergo the deformation processes required for the production of brass flux-cored wires. To address this dilemma, an innovative modification strategy has been proposed: adding alloying element powders into the flux core to realize in situ alloying during the brazing process. This method avoids the adverse effects of direct alloying on the formability of the brass sheath. It also allows flexible regulation of the brazed joint composition without altering the conventional preparation process of flux-cored brazing wires. Shi et al. [15] successfully achieved high-content solid solution of B (>0.8 wt.%) in an Ag-Cu-Zn-Sn flux-cored brazing wire alloy via this approach, which effectively refined the grain structure and improved the hardness and tensile strength of the brazed joints. Long [16] prepared metal-particle-reinforced aluminum flux-cored brazing wires by mixing Al-Si/Cu-Si powders with potassium fluoroaluminate flux. The metal powders in the core improved the fluidity and wettability of the molten filler metal. Wang et al. [17] designed a silver-based flux-cored brazing wire with CuSn powder in the core. This enabled the in situ synthesis of high-tin-content AgCuZnSn brazed joints and overcame the formability challenges of high-tin alloys caused by intrinsic brittleness. Pu et al. [18,19] found that the addition of Ga<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> particles significantly enhanced the wettability of Ag<sub>30</sub>CuZnSn flux-cored brazing materials. These studies collectively confirm that incorporating functional powders into the flux core is a feasible technical route for optimizing copper-based flux-cored brazing fillers with strict formability requirements.

In this work, Sn and Ni metal powders were mixed with flux powder to prepare flux-cored brass brazing filler metals, with in situ alloying achieved during the brazing of Q235 steel plates. Sn was selected for its low melting point (231.9 °C) and its ability to improve the fluidity and wettability of molten filler metals, as reported by Luo et al. [9] and Dong [13]. Ni was chosen as a strong  $\alpha$ -phase stabilizer in brass, which provides significant grain refinement and solid solution strengthening effects, as reported by Chen et al. [14]. The effects of Sn/Ni single and composite additions on the melting characteristics, wettability, and microstructure of flux-cored brass brazing filler metals were systematically investigated. The tensile strength of the joints brazed on Q235 steel plates with these fillers was also tested. The synergistic regulation mechanism of Sn/Ni co-addition on the microstructure and comprehensive performance of the filler metal was clarified. The optimal addition ratio was determined to realize the simultaneous improvement of wettability and mechanical properties. This study aims to provide a novel alloying modification strategy for copper-based flux-cored brazing fillers and enrich the theoretical basis of microstructure-performance regulation for Cu-Zn-based brazing alloys.

## 2. Materials and Methods

### 2.1. Preparation of Flux-Cored Brass Brazing Filler Metals

The flux-cored brass brazing filler metals were fabricated via the strip rolling method. A brass strip with a Cu content ranging from 57 to 61 wt.% was employed as the outer sheath material. In addition to the primary elements Cu and Zn, the brass strip contained minor additions of Sn, Ni, Mn, and Si. The strip was first rolled into a U-shaped groove, which was then filled with flux powder (blended with Sn/Ni metal powders at designed ratios). The U-shaped strip was subsequently closed to form an elliptical tube and finally drawn for diameter reduction to obtain a flat-round flux-cored brazing wire with a major

axis of 1.9–2.0 mm and a minor axis of 0.6 mm. Sn powder was added to the flux at mass fractions of 0 wt.%, 3 wt.%, 5 wt.%, 7 wt.%, 10 wt.%, 15 wt.% and 20 wt.%, while Ni powder was added at mass fractions of 0 wt.%, 10 wt.%, 20 wt.%, 30 wt.%, 40 wt.% and 50 wt.%, respectively. A total of thirteen samples with different compositions were prepared, including a Sn/Ni-free baseline sample, six Sn-added samples, five Ni-added samples, and a Sn/Ni co-added sample for comprehensive performance testing.

## 2.2. Material Characterization and Performance Testing

For the flux-cored brazing filler metals with different contents of added metal powders, a 5 cm length was cut using pliers. These cut segments were melted into a liquid state using an oxyacetylene flame. After cooling, the surface oxides were ground off with a grinder for chemical composition analysis and metallographic sample preparation. The chemical composition of all samples was analyzed using a JA90-750 photoelectric direct reading spectrometers (Jarrell-Ash Company, now part of Thermo Fisher Scientific, Waltham, MA, USA).

The solidus and liquidus temperatures of the flux-cored brazing filler metals were determined via differential scanning calorimetry (DSC) using a Netzsch STA 404 F3 apparatus (Selb, Germany) under an argon protective atmosphere. The sample mass was controlled at 5–15 mg, and the heating rate was set at 10 °C/min over a temperature range of 200 °C to 1000 °C.

Wettability tests of the brazing filler metals were conducted in accordance with the national standard GB/T 11364-2008 [20] on Q235 steel plates with dimensions of 40 mm × 40 mm × 2 mm. The steel plates were first preheated to 990 °C for 100 s in a muffle furnace, after which 20 mg of the flux-cored brass brazing filler metal was placed on the preheated steel plate surface and reheated to the preset temperature. Following isothermal holding for 10 s, the steel plates were immediately removed and quenched in cold water. The spreading morphologies were scanned, and the spreading areas of the brazing filler metal were measured and calculated using AutoCAD software (version R19.1). Each experimental condition was repeated five times, and the average value was taken as the final test result to ensure the reliability of the data.

Butt joints of Q235 and Q345 steel plates (50 mm × 10 mm × 1 mm) were fabricated via oxyacetylene flame brazing, with acetylene and oxygen flow rates set at 1.4 L/min and 0.9 L/min, respectively. Prior to brazing, the faying surfaces (10 mm × 1 mm) were ground with sandpaper, ultrasonically cleaned with ethanol, and dried in an oven. A 1 mm gap was reserved at the joint to facilitate the flow of the molten filler metal. After brazing, excess filler metal was removed by grinding to ensure the joint thickness was consistent with the base metal. Tensile tests of the brazed joints were performed in accordance with GB/T 11363-2008 [21] at a constant crosshead speed of 3 mm/min using a CMT5105 electronic universal testing machine (Shenzhen Xin Sansi Materials Testing Company, Shenzhen, China). Each experimental condition was repeated at least three times, and the average value was adopted as the final test result to ensure data reliability.

The phase structure of the brazing alloys was identified via X-ray diffraction (XRD). Metallographic samples were prepared from the spreading test specimens and brazed joint specimens, which were subsequently etched with a mixed corrosive solution of FeCl<sub>3</sub>-HCl-H<sub>2</sub>O for 10–15 s. The microstructures of the samples were observed using a Leica DM6M optical microscope (Wetzlar, Germany). High-magnification morphological features and microregional chemical compositions were characterized by a SU8010 scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS) (Hitachi High-Technologies Corporation, Tokyo, Japan).

### 3. Results and Discussion

#### 3.1. Composition of the Flux-Cored Brazing Filler Metal with Sn/Ni Powders

The chemical composition of the flux-cored brass brazing filler metals with varying contents of added metal powders is presented in Table 1. It can be observed that the Sn and Ni powders added to the flux core were effectively dissolved into the brazing alloy matrix during the melting process. The Sn content in the Sn-added samples increased progressively from 1.1 wt.% to 4.02 wt.%, while the Ni content in the Ni-added samples rose gradually from 0.41 wt.% to 10.86 wt.%.

**Table 1.** Chemical composition of the brass flux-cored filler metals.

| No. | Sample                  | Chemical Composition (wt.%) |       |      |       |      |      | X%    |
|-----|-------------------------|-----------------------------|-------|------|-------|------|------|-------|
|     |                         | Cu                          | Zn    | Sn   | Ni    | Mn   | Si   |       |
| ①   | Baseline (0 wt.% Sn/Ni) | 59.01                       | 39.11 | 1.1  | 0.41  | 0.31 | 0.06 | 41.28 |
| ②   | 3Sn                     | 59.14                       | 38.83 | 1.42 | 0.32  | 0.24 | 0.05 | 41.44 |
| ③   | 5Sn                     | 58.89                       | 39.05 | 1.44 | 0.33  | 0.24 | 0.05 | 41.68 |
| ④   | 7Sn                     | 59.05                       | 38.4  | 1.84 | 0.32  | 0.24 | 0.05 | 41.71 |
| ⑤   | 10Sn                    | 59.93                       | 37.65 | 1.71 | 0.32  | 0.24 | 0.05 | 40.76 |
| ⑥   | 15Sn                    | 59.29                       | 37.07 | 3.00 | 0.31  | 0.24 | 0.09 | 42.41 |
| ⑦   | 20Sn                    | 58.37                       | 36.87 | 4.02 | 0.33  | 0.25 | 0.06 | 43.63 |
| ⑧   | 10Ni                    | 59.71                       | 36.73 | 0.92 | 2.12  | 0.41 | 0.11 | 38.20 |
| ⑨   | 20Ni                    | 59.38                       | 34.26 | 1.11 | 4.92  | 0.27 | 0.06 | 33.81 |
| ⑩   | 30Ni                    | 59.36                       | 34.62 | 0.86 | 4.98  | 0.23 | 0.05 | 33.56 |
| ⑪   | 40Ni                    | 57.00                       | 33.66 | 0.88 | 8.29  | 0.22 | 0.05 | 30.00 |
| ⑫   | 50Ni                    | 57.21                       | 30.86 | 0.85 | 10.86 | 0.23 | 0.09 | 24.31 |
| ⑬   | 3 wt.% Sn + 10 wt.% Ni  | 59.75                       | 36.28 | 1.40 | 2.08  | 0.39 | 0.10 | 38.47 |

The alloying elements typically modify the phase region of Cu-Zn alloys. The fictitious zinc equivalent (X%) is defined as the amount of zinc substituted by 1 wt.% of alloying elements in the sample, which abides by the zinc equivalent rule and is described as follows [22].

$$X\% = \frac{C_{Zn} + \sum C_i K_i}{C_{Zn} + C_{Cu} \sum C_i K_i} \times 100\%$$

where  $C_{Zn}$  and  $C_{Cu}$  are the actual Zn and Cu contents in brass alloys, respectively; and  $\sum C_i K_i$  is the sum of the products of content  $C_i$  and fictitious zinc equivalent  $K_i$  of all element additions. According to Table 2, the  $K_i$  values of Sn, Ni, Mn, and Si are 2, −1.4, 0.5, and 10, respectively.  $K_i$  data are from Ref. [22], and the fictitious zinc equivalent (X%) of each sample is calculated and listed in Table 1. According to the Cu-Zn binary phase diagram, the maximum solid solubility of Zn in  $\alpha$ -Cu at room temperature is 36 wt.%, so the microstructural type of the samples can be inferred from the zinc equivalent values X% presented in Table 1. The primary phase constituent of the Sn-added samples is the  $\beta$  phase, whereas that of the Ni-added samples is predominantly the  $\alpha$  phase.

**Table 2.** Zinc equivalent  $K_i$ .

| Element | Si | Al | Sn | Mg | Pb | Fe  | Mn  | Ni   |
|---------|----|----|----|----|----|-----|-----|------|
| $K_i$   | 10 | 6  | 2  | 2  | 1  | 0.9 | 0.5 | −1.4 |

### 3.2. Effect of Sn/Ni on the Microstructure and Properties of Brazing Alloy

#### 3.2.1. Microstructural Characteristics of Sn-Added Filler Metal

Metallographic observations (50  $\mu\text{m}$  scale) of brazing filler metals with gradient Sn additions (0~20 wt.%) were conducted, and the results are presented in Figure 1. For samples with Sn addition  $\leq 10$  wt.% (alloy Sn content  $< 2$  wt.%, Figure 1a–e), the microstructure maintained a typical ( $\alpha + \beta$ ) dual-phase structure, where light needle-like  $\alpha$  phases were dispersed in equiaxed  $\beta$  grains. With the increase in Sn content, the grain size of equiaxed  $\beta$  first increased and then decreased, and the volume fraction of  $\alpha$  phase inside  $\beta$  grains decreased gradually. When Sn addition reached 15~20 wt.% (alloy Sn content  $> 3$  wt.%, Figure 1f,g), the  $\alpha$  phase was almost completely eliminated in equiaxed  $\beta$  grains, and the microstructure was dominated by massive  $\beta$  single phase.

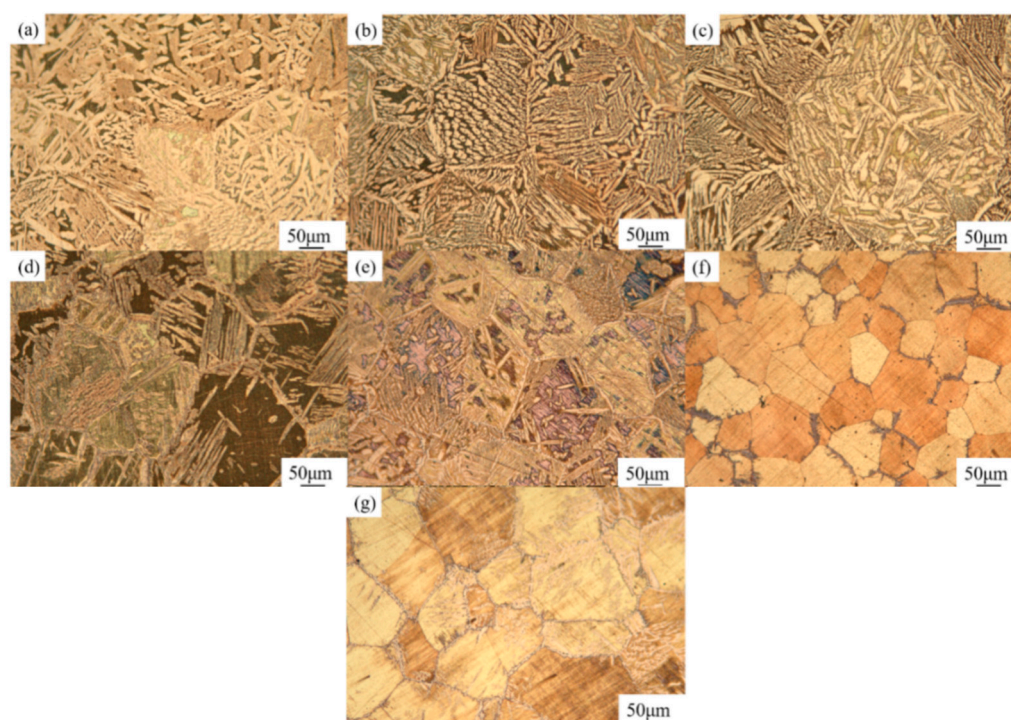


**Figure 1.** Metallographic microstructures of filler metals with different Sn contents: (a) 0 wt.% Sn; (b) 3 wt.% Sn; (c) 5 wt.% Sn; (d) 7 wt.% Sn; (e) 10 wt.% Sn; (f) 15 wt.% Sn; (g) 20 wt.% Sn.

This phase evolution can be reasonably explained by the Cu-Zn-Sn ternary phase diagram (Figure 2). The polythermal longitudinal section shows that the phase transformation temperature between the  $\beta$  single-phase region and ( $\alpha + \beta$ ) dual-phase region at 3 wt.% Sn (point B) is about 100  $^{\circ}\text{C}$  lower than that at 1 wt.% Sn (point A). The decrease in phase transformation temperature makes the alloy tend to form a stable  $\beta$  phase during cooling and solidification, which is the fundamental reason for the gradual increase in  $\beta$  phase volume fraction with Sn addition.

XRD pattern of filler metals (Figure 3) further verified the above microstructural evolution trend: the diffraction peak intensity of  $\alpha$  phase was higher than that of  $\beta$  phase in the Sn-free baseline sample, while the  $\beta$  phase diffraction peak was dominant in the samples with 10 wt.% and 20 wt.% Sn addition, which was a direct reflection of the continuous increase in  $\beta$  phase volume fraction with Sn addition.

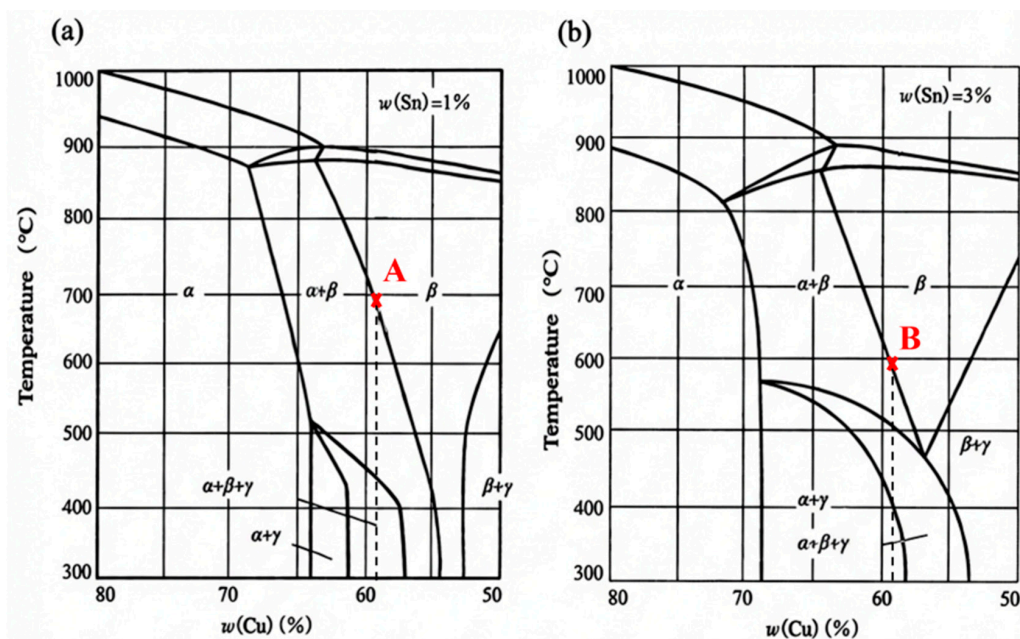


**Figure 2.** Polythermal longitudinal sections of the Cu-Sn-Zn ternary alloy phase diagram [23]: (a) 1 wt.% Sn; (b) 3 wt.% Sn. (Adapted with permission from Jueqi Yu, Practical Alloy Phase Diagram Handbook for Brazing Materials; published by China Machinery Industry Press, 2015).

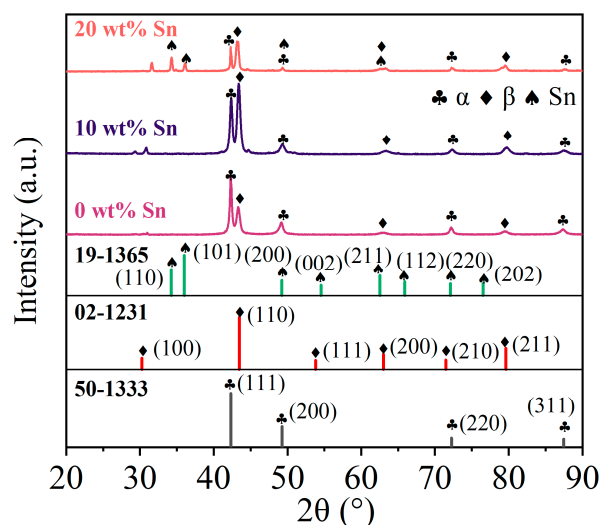


**Figure 3.** XRD pattern of filler metals with different Sn contents.

SEM observations were performed on typical samples (0, 10, 15, 20 wt.% Sn) to characterize the detailed microstructural morphology, as shown in Figure 4 with a scale bar of 20  $\mu\text{m}$ . EDS point analysis (Table 3) was used to identify the phase composition of characteristic regions. The Sn-free baseline sample (Figure 4a) presented an interlaced structure of gray-black  $\alpha$  phase (Region A) and light-colored ( $\alpha + \beta$ ) dual phase (Region B), and the latter showed a distinct lamellar substructure at high magnification. With the increase in Sn content, the  $\alpha$  phase was continuously reduced, the ( $\alpha + \beta$ ) dual phase evolved into a massive  $\beta$  phase matrix (Region D), and a small amount of dendritic  $\alpha$  phase (Region C) was precipitated at the grain boundaries of large-angle oriented  $\beta$  grains. In the 20 wt.% Sn sample (Figure 4d), discrete white secondary precipitated  $\alpha$  phase ( $\alpha\text{II}$ ) was observed in the massive  $\beta$  phase matrix, which is consistent with the microstructural evolution sequence of the alloy in the Cu-Zn binary phase diagram, i.e.,  $L \rightarrow \beta \text{ phase} \rightarrow \alpha + \beta \text{ phase}$ .

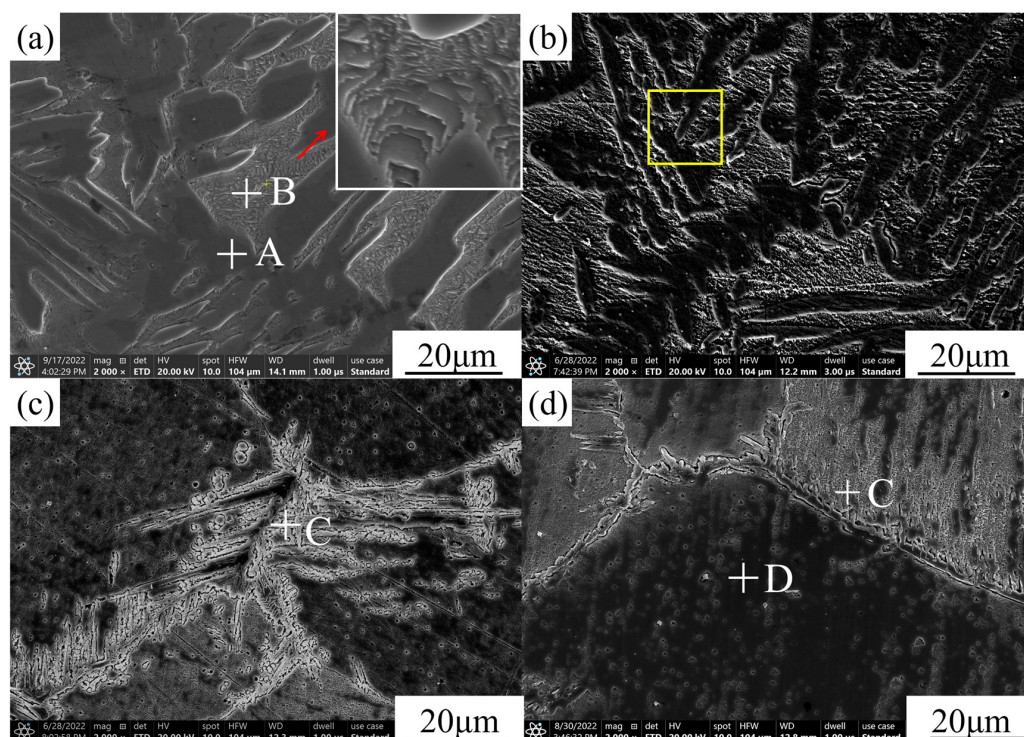


**Figure 4.** SEM images of filler metal with different Sn content: (a) 0 wt.% Sn; (b) 10 wt.% Sn; (c) 15 wt.% Sn; (d) 20 wt.% Sn.

**Table 3.** EDS analysis of characteristic points.

| Point | Mass Fraction |       |      |      | Possible Phase                 |
|-------|---------------|-------|------|------|--------------------------------|
|       | Cu            | Zn    | Ni   | Sn   |                                |
| A     | 63.19         | 35.68 | 0.41 | 0.72 | $\alpha$ -Cu                   |
| B     | 55.90         | 41.32 | 0.39 | 2.38 | $\alpha$ -Cu + $\beta$ (Cu-Zn) |
| C     | 63.60         | 34.50 | 1.90 | -    | $\alpha$ -Cu                   |
| D     | 58.80         | 35.90 | -    | 5.30 | $\beta$ (Cu-Zn)                |

EDS elemental mapping (Figure 5) of the marked region in Figure 4b revealed a distinct enrichment of Sn in the  $\beta$  phase, indicating that Sn has low solubility in the  $\alpha$  phase and high solubility in the  $\beta$  phase in the brass brazing alloy system. During the solidification process, the high-melting-point  $\alpha$ -Cu phase precipitates preferentially; due to its low melting point and limited solubility in the  $\alpha$  phase, Sn atoms are rejected to the solid–liquid interface and gradually segregate at grain boundaries during subsequent solidification, resulting in significant Sn accumulation at grain boundary regions, which further promotes the formation and growth of  $\beta$  phase.

### 3.2.2. Microstructural Characteristics of Ni-Added Filler Metal

Ni addition exhibited a prominent modification effect on the microstructure of brass flux-cored brazing filler metal that was completely different from Sn addition, and the SEM observation results (Figure 6) combined with EDS point analysis (Table 4) showed a clear microstructural evolution law with the increase in Ni addition (0~50 wt.%). The volume fraction of  $\alpha$  phase (Points 1–6 in Figure 6) increased progressively with Ni addition, and at 20 wt.% Ni addition (Figure 6c), equiaxed  $\alpha$  grains formed a continuous and interconnected matrix structure. Meanwhile, the ( $\alpha$  +  $\beta$ ) dual-phase constituent distributed at the grain boundaries of equiaxed  $\alpha$  grains (Points 7–11 in Figure 6) decreased gradually, and finally

evolved into a discrete  $\beta$  single phase at the grain boundaries in the 50 wt.% Ni sample (Figure 6f).

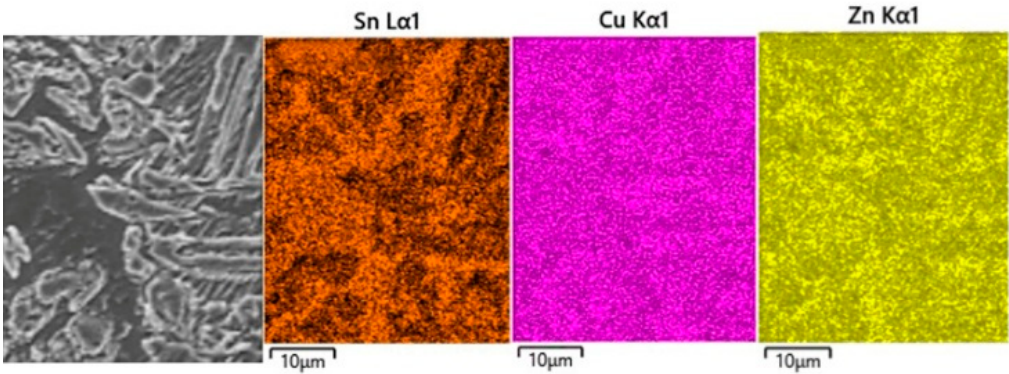


Figure 5. EDS mapping of the marked box in Figure 4b.

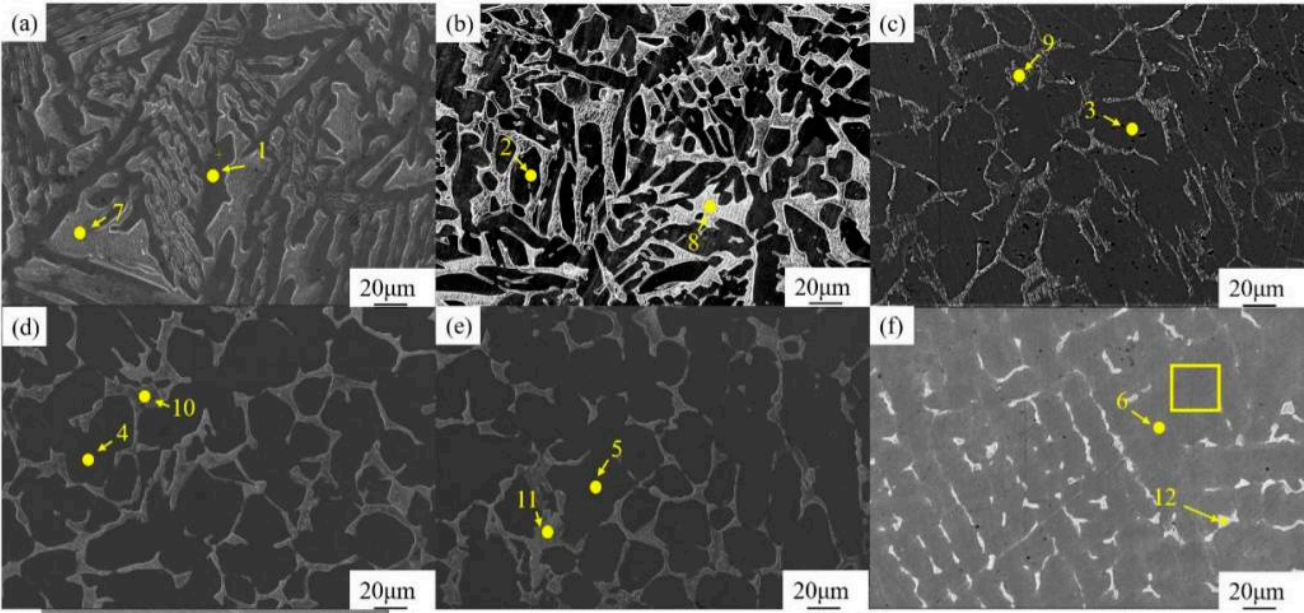


Figure 6. SEM images of filler metal with different Ni content: (a) 0 wt.% Ni; (b) 10 wt.% Ni; (c) 20 wt.% Ni; (d) 30 wt.% Ni; (e) 40 wt.% Ni; (f) 50 wt.% Ni.

Table 4. EDS analysis of characteristic points in Figure 6.

| Point | Mass Fraction (wt.%) |      |      |     | Possible Phases                |
|-------|----------------------|------|------|-----|--------------------------------|
|       | Cu                   | Zn   | Ni   | Sn  |                                |
| 1     | 63.5                 | 36.5 | -    | -   | $\alpha$ -Cu                   |
| 2     | 60.4                 | 35.3 | 4.3  | -   | $\alpha$ -Cu                   |
| 3     | 58.8                 | 35.5 | 5.7  | -   | $\alpha$ -Cu                   |
| 4     | 57.2                 | 34.9 | 7.9  | -   | $\alpha$ -Cu                   |
| 5     | 56.5                 | 32.4 | 11.1 | -   | $\alpha$ -Cu                   |
| 6     | 54.2                 | 33.3 | 12.5 | -   | $\alpha$ -Cu                   |
| 7     | 56.8                 | 41.6 | -    | 1.6 | $\alpha$ -Cu + $\beta$ (Cu-Zn) |
| 8     | 52.9                 | 41.1 | 4.0  | 2.1 | $\alpha$ -Cu + $\beta$ (Cu-Zn) |
| 9     | 53.0                 | 41.9 | 5.2  | -   | $\alpha$ -Cu + $\beta$ (Cu-Zn) |
| 10    | 50.3                 | 40.0 | 6.7  | 3.1 | $\alpha$ -Cu + $\beta$ (Cu-Zn) |
| 11    | 48.3                 | 38.9 | 8.6  | 4.1 | $\alpha$ -Cu + $\beta$ (Cu-Zn) |
| 12    | 49.3                 | 31.4 | 13.1 | 6.2 | $\beta$ (Cu-Zn)                |

The fundamental reason for the expansion of the  $\alpha$  phase region by Ni addition is the change in Zn solubility in the Cu-based matrix: the maximum solubility of Zn in pure Cu is about 36 wt.%, while in the Cu-Ni-Zn cupronickel system, the Zn content can reach 44–73 wt.%. Ni forms a continuous substitutional solid solution with Cu, which enables Zn to form a broader single-phase solid solution region in the Cu-Ni matrix, far exceeding its solubility limit in the pure Cu-Zn system, thus expanding the  $\alpha$  phase region and increasing its volume fraction. In addition, EDS data of Point 12 in Table 4 showed that the  $\beta$  phase in the 50 wt.% Ni sample contained 6.2 wt.% Sn, while the average Sn content in the alloy was only 0.85 wt.%, which confirmed that Sn was completely segregated in the  $\beta$  phase with the increase in Ni content. Due to its low melting point, Sn atoms are enriched at the front of the solid–liquid interface during solidification, which drives the transformation of ( $\alpha + \beta$ ) dual phase into  $\beta$  single phase and forms the oriented discontinuous point-chain microstructure observed in Figure 6f.

XRD pattern of filler metals (Figure 7) of Ni-added samples further confirmed the gradual decrease in  $\beta$  phase volume fraction with increasing Ni content: the  $\beta$  phase diffraction peak disappeared in the 50 wt.% Ni sample, and only  $\alpha$  phase diffraction peaks were detected. Notably, the  $\alpha$  phase diffraction peaks showed a significant overall right shift, which is due to the complete miscibility between Cu and Ni: the atomic radius of Ni (124 pm) is smaller than that of Cu (128 pm), and Ni atoms substitute Cu atoms in the crystal lattice to form a substitutional solid solution, leading to lattice contraction and reduction in interplanar spacing  $d$ . According to Bragg's Law ( $2d\sin\theta = n\lambda$ ), the decrease in  $d$  leads to the increase in diffraction angle  $\theta$  under the condition of fixed wavelength  $\lambda$  and diffraction order  $n$ , thus resulting in the right shift in  $\alpha$  phase diffraction peaks. The Sn-rich boundary phase was not detected in the XRD pattern of filler metals due to its low content (below the 5% detection limit of XRD).

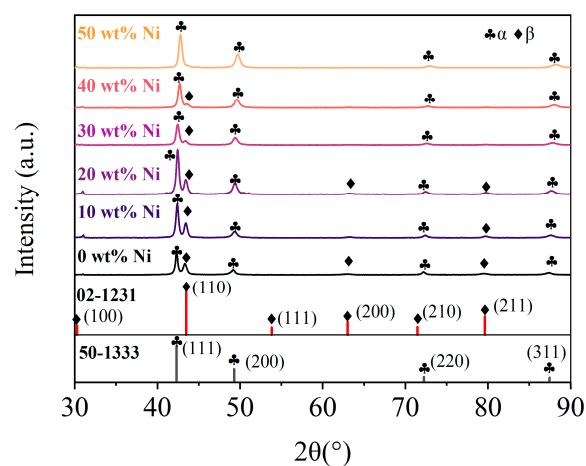


**Figure 7.** XRD pattern of filler metals with different Ni content.

In general, the microstructure of Ni-added samples gradually evolved into a continuous  $\alpha$ -phase matrix with a small amount of ( $\alpha + \beta$ ) dual phase or  $\beta$  single phase distributed discontinuously at the grain boundaries of equiaxed  $\alpha$  grains. Compared with Sn addition, Ni exhibited a significant grain refinement effect, which is an important structural characteristic for improving the mechanical properties of brazing filler metals.

### 3.2.3. Performance of Sn/Ni Added Filler Metal

Figure 8 presents the variation in the solidus temperature and unit spreading area of the brazing filler metal with Sn addition content (0–20 wt.%). The line graph shows a continuous decrease in the solidus temperature of the filler metal with the increase in

Sn addition, while the bar graph reveals a significant and steady improvement in the unit spreading area on Q235 steel plates, indicating a positive correlation between Sn addition and the wettability of the brazing filler metal.

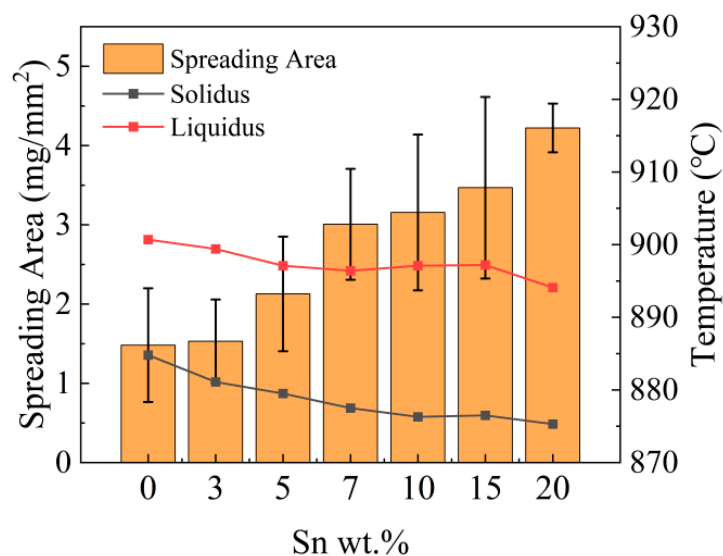


**Figure 8.** Melting temperature range and spreading area of filler metal with different Sn contents.

The enhancement mechanism of Sn on the melting and wettability properties is closely related to its low melting point (231.9 °C) and interdiffusion behavior with the brass matrix. The melting process of the flux-cored brazing filler metal is a dynamic interdiffusion process: the low-melting-point Sn powder added in the flux core melts preferentially during brazing, and the formed molten Sn liquid drives the successive melting of the brass outer sheath through interdiffusion and fusion. This process effectively reduces the solidus temperature of the brass brazing filler metal, induces superheating of the molten metal, and further reduces the viscosity of the molten filler metal while improving its fluidity. The improved fluidity significantly promotes the spreading and wetting behavior of the molten brazing filler metal on the steel plate surface. When the Sn addition reaches 20 wt.%, the unit spreading area of the filler metal reaches the maximum value of 4.22 mm<sup>2</sup>/mg, which is 2.85 times that of the Sn-free baseline sample, realizing a remarkable improvement in wettability.

Figure 9 depicts the changes in the solid–liquid temperature, melting temperature range and unit spreading area of the brazing filler metal with Ni addition content (0~50 wt.%), and the wettability of the Ni-added filler metal exhibits a trend of first increasing and then decreasing, which is quite different from the monotonic improvement of Sn addition. For the Ni-free baseline sample, the solidus temperature is 882 °C, the liquidus temperature is 914.2 °C, and the melting temperature range is 32.2 °C. With the increase in Ni addition, the melting temperature range of the filler metal narrows gradually: when the Ni addition reaches 50 wt.%, the solidus and liquidus temperatures are 888.31 °C and 911.2 °C respectively, and the melting temperature range is reduced to 22.89 °C, a 29% reduction compared with the baseline sample. A narrowed melting temperature range is conducive to reducing the viscosity and improving the fluidity of the molten brazing filler metal, which is beneficial to its spreading and gap-filling performance.

Consistent with the change in the melting temperature range, the unit spreading area of the Ni-added filler metal increases first and peaks at 20 wt.% Ni addition, with a maximum value of 2.46 mm<sup>2</sup>/mg (1.67 times that of the baseline sample). However, when the Ni addition exceeds 20 wt.% and reaches 40 wt.% or above, the unit spreading area drops sharply and is even lower than that of the baseline sample, accompanied by

the phenomenon of incomplete melting of the brazing filler metal. This is due to the high melting point of Ni (1450 °C): low-content Ni addition narrows the melting temperature range and optimizes fluidity, while high-content Ni addition elevates the overall melting temperature of the brazing filler metal. At a fixed brazing temperature, this leads to a significant reduction in the superheat degree of the molten filler metal, resulting in impaired fluidity and poor spreading performance.

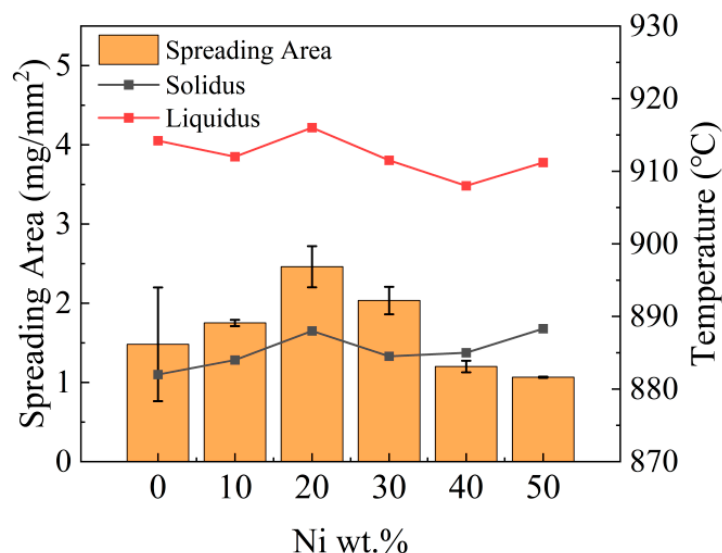


**Figure 9.** Melting temperature range and spreading area of filler metals with different Ni contents.

Figure 10 shows the effects of Sn and Ni addition on the tensile strength of Q235 steel brazed joints, and the two elements exhibit completely different regulation effects on the mechanical properties of the joints. Despite the remarkable improvement in wettability, Sn addition exerts a negligible effect on the tensile strength of the brazed joints—the tensile strength of joints with different Sn addition contents remains at approximately 300 MPa, consistent with the baseline sample. In sharp contrast, Ni addition significantly improves the tensile strength of the brazed joints, and the strength exhibits a trend of first increasing and then decreasing with the increase in Ni addition content.

The different effects of Sn and Ni on joint tensile strength are essentially determined by their regulated microstructures. For the Sn-added filler metal, the primary phase is the coarse  $\beta$  phase (average grain size  $> 50 \mu\text{m}$ ), and the  $\beta$  phase is prone to transform into the brittle ordered  $\beta'$  phase at room temperature; the coarse and brittle phase characteristics make the Sn-added filler metal lack plastic toughness, thus failing to improve the tensile strength of the brazed joints. For the Ni-added filler metal, the primary phase is the fine-grained  $\alpha$  phase (average grain size  $20\sim 50 \mu\text{m}$ ), and Ni atoms form a substitutional solid solution with Cu atoms, exerting a distinct solid solution strengthening effect on the  $\alpha$ -phase matrix. The combined effect of grain refinement and solid solution strengthening significantly improves the mechanical properties of the filler metal, leading to a steady increase in joint tensile strength with Ni addition. When the Ni addition reaches 20 wt.%, the joint tensile strength peaks at 501 MPa, a 65% increase compared with the baseline sample (302 MPa).

When the Ni addition is further increased to 50 wt.%, the joint tensile strength plummets sharply to 214 MPa, which is lower than the baseline sample. This is due to the double adverse effect of the microstructure evolution of the 50 wt.% Ni sample: on the one hand, the microstructure evolves into an  $\alpha$ -phase matrix with discrete Sn-enriched  $\beta$ -phase boundary phases, and the Sn-enriched  $\beta$  phase is an inherently hard and brittle constituent that easily causes stress concentration; on the other hand, the Ni content in the

$\alpha$  phase reaches a maximum of 12.5 wt.% at this addition level, and excessive solid solution strengthening induces severe hardening and embrittlement of the  $\alpha$ -phase matrix, resulting in a dramatic reduction in the tensile strength of the brazed joints.

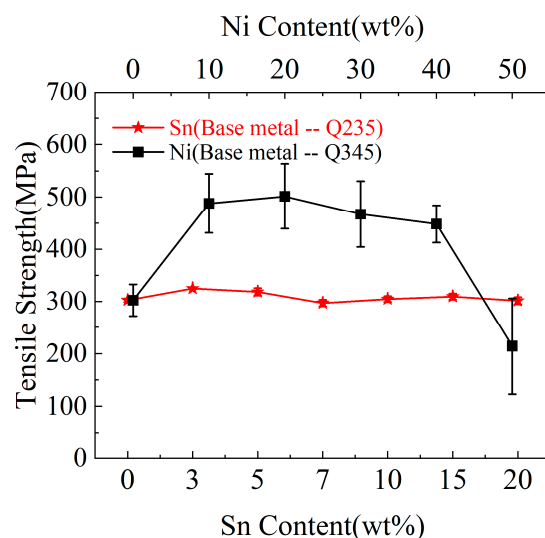


**Figure 10.** Tensile strength of joints with different Sn/Ni contents.

### 3.3. Effect of Sn/Ni Co-Addition on the Microstructure and Properties of Brass Filler Metal

To optimize the comprehensive performance of brass flux-cored brazing filler metals, a composite alloying experiment with 3 wt.% Sn + 10 wt.% Ni (No. ⑬, Table 1) was conducted based on the single-element addition results. The microstructure and performance of the co-added sample were systematically investigated by comparative analysis with the Sn/Ni-free baseline sample (No. ①, Table 1), 3 wt.% Sn single-added sample (No. ②, Table 1) and 10 wt.% Ni single-added sample (No. ⑧, Table 1), with the results presented in Figures 11–13.

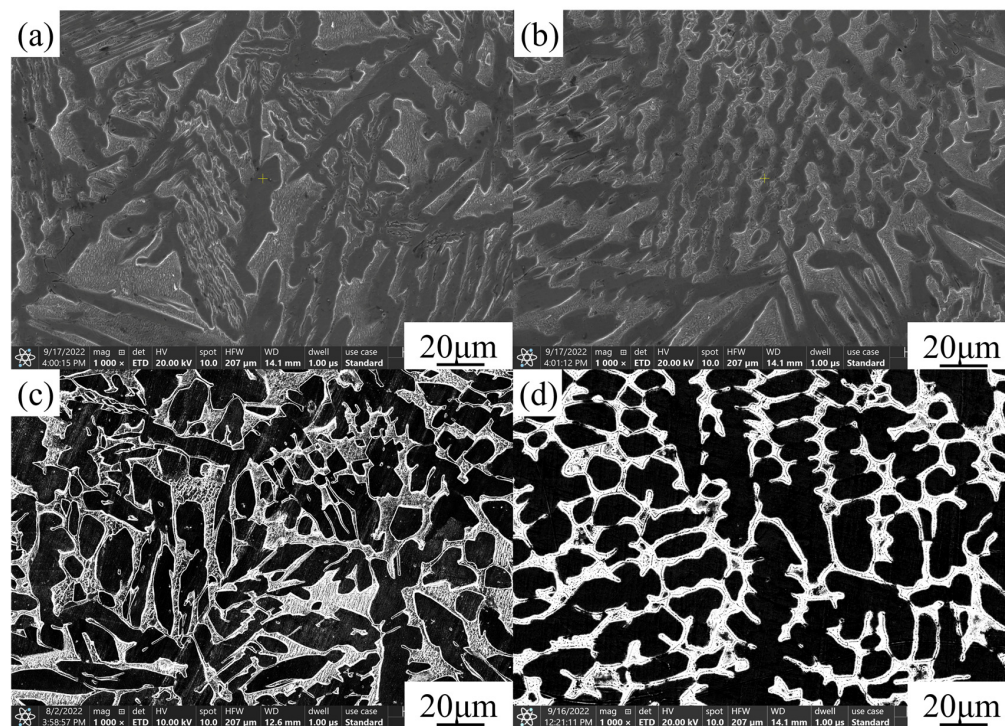


**Figure 11.** Microstructure of (a) Sample ①: baseline; (b) Sample ②: 3 wt.% Sn; (c) Sample ⑧: 10 wt.% Ni; (d) Sample ⑬: 3 wt.% Sn + 10 wt.% Ni.

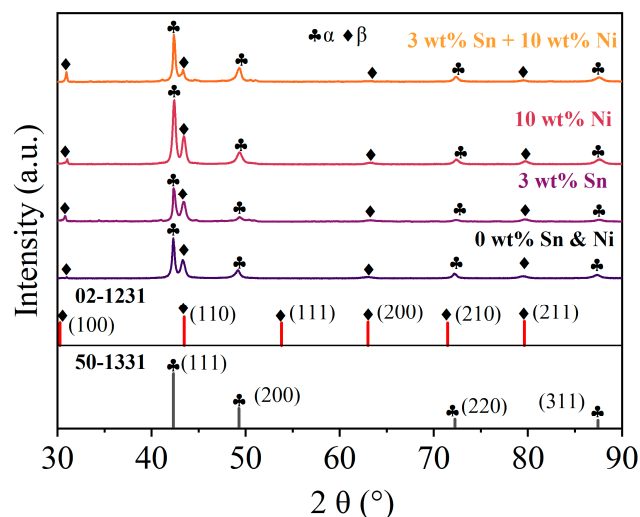


**Figure 12.** XRD pattern of filler metals with different Sn and Ni content: baseline; 3 wt.% Sn; 10 wt.% Ni; 3 wt.% Sn + 10 wt.% Ni.

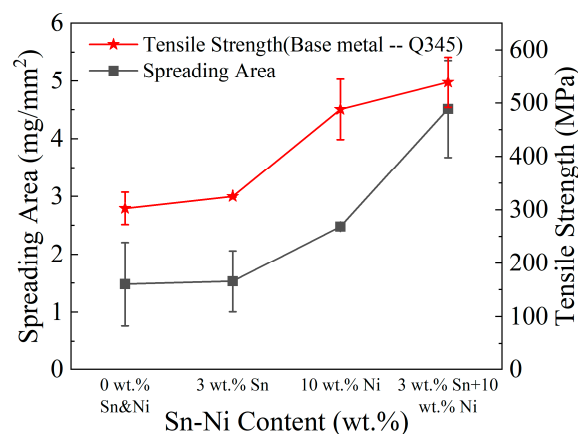


**Figure 13.** Spreading area of filler metals and joint tensile strength with different alloy contents.

### 3.3.1. Microstructural Characteristics of Sn/Ni Co-Added Filler Metal

Figure 11 displays the high-magnification microstructures of the four typical samples (20  $\mu\text{m}$  scale), revealing a distinct microstructural evolution induced by Sn/Ni co-addition compared with single-element modification and the baseline. The baseline sample (①, Figure 11a) exhibits a directional lamellar dendritic microstructure with obvious anisotropy, while the 3 wt.% Sn single-added sample (②, Figure 11b) is dominated by coarse massive  $\beta$ -phase grains with a small amount of  $\alpha$ -phase precipitated at grain boundaries. The 10 wt.% Ni single-added sample (⑧, Figure 11c) forms a continuous  $\alpha$ -phase matrix with discontinuous ( $\alpha + \beta$ ) dual-phase distributed at the grain boundaries of equiaxed  $\alpha$  grains. In contrast, the 3 wt.% Sn + 10 wt.% Ni co-added sample (⑬, Figure 11d) completely eliminates the directional dendritic morphology of the baseline and the coarse phase distribution of single-element addition, forming a uniform grid-like ( $\alpha + \beta$ ) dual-phase microstructure. In this structure, the soft  $\alpha$ -phase matrix is uniformly dispersed in the whole microregion, and part of the  $\alpha$  phase is surrounded by a continuous/semicontinuous ( $\alpha + \beta$ ) dual-phase network, with refined and evenly distributed grains and clear, smooth phase interfaces.

XRD pattern analysis of the four samples (Figure 12) further verifies the phase composition characteristics of the co-added sample. The XRD peaks of the four samples are consistent in position, with only slight differences in peak intensity, indicating that Sn/Ni

single or co-addition does not introduce new phases into the brass matrix. All samples are composed of Cu-based solid solution ( $\alpha$  phase) and Cu-Zn-based solid solution ( $\beta$  phase), and the variation in peak intensity is only the result of the change in the volume fraction of  $\alpha/\beta$  phases caused by alloying element addition. This confirms that the excellent performance of the co-added sample is not derived from the generation of new phases, but from the optimization of microstructural morphology and the synergistic effect of Sn/Ni dual-element solid solution strengthening.

### 3.3.2. Performance of Sn/Ni Co-Added Filler Metal

Figure 13 presents the unit spreading area of the four brazing filler metal samples on Q235 steel plates and the tensile strength of the corresponding brazed joints, clearly demonstrating that Sn/Ni co-addition achieves a simultaneous improvement in the wettability and joint mechanical properties of the brazing filler metal, which is difficult to realize by single-element addition.

The baseline sample exhibits the poorest comprehensive performance, with a unit spreading area of only 1.47 mm<sup>2</sup>/mg and a joint tensile strength of 302 MPa. Single Sn addition (3 wt.%) significantly improves the wettability of the filler metal (unit spreading area increased to 3.82 mm<sup>2</sup>/mg) but has almost no enhancement effect on the joint tensile strength (305 MPa). Single Ni addition (10 wt.%) effectively improves the joint tensile strength (426 MPa), but the promotion of wettability is limited (unit spreading area of 2.15 mm<sup>2</sup>/mg). By contrast, the sample with 3 wt.% Sn + 10 wt.% Ni co-added exhibits the best overall performance among all specimens: its unit spreading area reaches 4.51 mm<sup>2</sup>/mg, which is a 206% increase compared with the baseline sample and 18% higher than the 3 wt.% Sn single-added sample; the average tensile strength of the corresponding brazed joints is 584 MPa, a 93% increase relative to the baseline sample and a 37% increase compared with the 10 wt.% Ni single-added sample.

### 3.3.3. Mechanism of Synergistic Performance Improvement by Sn/Ni Co-Addition

The significant synergistic effect of Sn/Ni co-addition on the comprehensive performance of brass flux-cored brazing filler metals is the result of the coupling of microstructural optimization and dual-element functional complementation, with the core mechanisms as follows:

Grid-like microstructure realizes uniform stress dispersion and crack propagation inhibition: The grid-like ( $\alpha + \beta$ ) dual-phase microstructure formed by co-addition eliminates the structural anisotropy of the baseline sample and the coarse phase distribution of single-element addition. The continuous/semicontinuous ( $\alpha + \beta$ ) dual-phase network forms a “reinforced skeleton” in the  $\alpha$ -phase matrix, which can uniformly disperse the external tensile stress to the whole microregion and avoid local stress concentration. Meanwhile, the refined grain size and numerous phase interfaces significantly increase the crack propagation resistance, and the soft  $\alpha$ -phase matrix can effectively relieve the stress at the phase interface and block the rapid expansion of microcracks.

Dual-element functional complementation realizes the synergy of wettability and strength: Sn element exerts its advantage of improving the wettability of the brazing filler metal—its low melting point (231.9 °C) reduces the melting temperature of the filler metal, improves the fluidity of the molten metal, and ensures the excellent spreading performance of the co-added sample; at the same time, Sn is enriched in the  $\beta$  phase to realize the solid solution strengthening of the hard phase. Ni element refines the grains, expands the  $\alpha$ -phase region, and forms a substitutional solid solution with Cu to realize the solid solution strengthening of the  $\alpha$ -phase matrix, which significantly improves the mechanical properties of the brazing filler metal. The co-addition makes Sn and Ni

exert their respective functional advantages without mutual interference, realizing the simultaneous improvement of wettability and strength.

Synergistic solid solution strengthening of dual elements enhances the overall performance of the matrix: Sn-enriched  $\beta$  phase and Ni-strengthened  $\alpha$  phase form a “soft matrix-hard network” structural system in the co-added sample. The solid solution strengthening effect covers both the  $\alpha$  matrix and the ( $\alpha + \beta$ ) reinforced network, and the grid-like microstructure ensures the uniform distribution of the strengthened phases, avoiding the local brittle hardening caused by the excessive enrichment of a single element, thus realizing the synergistic improvement of the plastic toughness and strength of the brazing filler metal.

In summary, Sn/Ni co-addition helps optimize the microstructure of brass flux-cored brazing filler metals and exerts the functional advantages of the two elements in a synergistic manner, which effectively alleviates the mutual restriction between wettability and mechanical properties in single-element modified brazing filler metals, and significantly improves the comprehensive performance of the brazing filler metal and the corresponding brazed joints.

#### 4. Conclusions

This work studied the effects of Sn/Ni single and composite additions on the microstructure, melting characteristics, wettability and tensile strength of Q235 steel brazed joints with brass flux-cored brazing filler metals, and clarified the corresponding alloying mechanism.

- (1) Sn addition enhanced the wettability of the brazing filler metal (maximum unit spreading area:  $4.22 \text{ mm}^2/\text{mg}$ ,  $2.85 \times$  baseline at 20 wt.%) by lowering its melting temperature and improving molten fluidity, but induced coarse  $\beta$ -phase grains and brittle  $\beta'$ -phase transformation at room temperature, resulting in negligible improvement in the tensile strength of brazed joints.
- (2) Ni addition expanded the  $\alpha$ -phase zone, refined grains and realized solid solution strengthening of the brazing filler metal. Joints' tensile strength peaked at 501 MPa (65% higher than the baseline) at 20 wt.% Ni, and the melting temperature range was narrowed by 29% at 50 wt.% Ni. High Ni addition ( $\geq 40$  wt.%) deteriorated wettability; 50 wt.% Ni caused over-solid solution strengthening and embrittlement of the  $\alpha$ -phase matrix, leading to a sharp decrease in tensile strength to 214 MPa.
- (3) Composite addition of 3 wt.% Sn + 10 wt.% Ni reconstructed the filler metal microstructure into a refined and uniform grid-like ( $\alpha + \beta$ ) dual-phase structure, making up for the defects of single-element addition and achieving synergistic optimization of comprehensive performance. The optimal performance was obtained with the unit spreading area reaching  $4.51 \text{ mm}^2/\text{mg}$  (206% higher than the baseline) and the joint tensile strength up to 584 MPa (93% higher than the baseline).

The synergistic improvement was attributed to the coupling of dual-element functional complementation and microstructural optimization: Sn improved wettability and strengthened the  $\beta$ -phase, while Ni refined grains and strengthened the  $\alpha$ -phase matrix, forming a “soft matrix-hard network” structure. This grid-like ( $\alpha + \beta$ ) structure dispersed stress and hindered crack propagation, realizing the balanced strength and toughness.

This study provides a feasible alloying strategy for high-performance brass flux-cored brazing filler metals, and the revealed microstructure-performance regulation mechanism supports the development of advanced copper-based brazing filler metals.

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W.L. and L.R.; writing—review and editing, W.L., X.H., L.R. and Y.B.; supervision, W.L. and Y.B. All authors have read and agreed to the published version of the manuscript.

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