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ARTICLE

Effect of Ultrasonic Vibration Coupled with Direct Current on Wetting Behavior

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Abstract: The effect of ultrasonic vibration coupled with direct current on the wetting behavior of Sn liquid solder on Cu substrate at 270 °C was studied by the wetting balance technique. The wetting balance curves were measured. Results show that the ultrasonic vibration coupled with direct current method significantly improves the wettability of Sn liquid solder. With increasing the ultrasonic vibration power and direct current, the maximum equilibrium wetting force is increased. According to the microstructure observations of interface during wetting process, the ultrasonic vibration coupled with direct current method can increase the dissolution of Cu substrate in the molten Sn solder and promote the precipitation of the intermetallic compounds at interface. The sound pressure distribution inside the molten Sn solder was simulated by a finite element software. The maximum sound pressure occurs at the front end of the ultrasonic probe. During the ultrasonic vibration coupled with direct current process, the chemical driving force caused by the precipitation of intermetallic compounds at interface and the strong convection inside the molten Sn solder jointly promote the movement of the triple-phase line, thereby improving the wettability of Sn solder.

Key words: ultrasonic vibration; direct current; wettability; intermetallic compound; driving force

The wettability of molten metal on a solid substrate has a significant influence on the material processing and fabrication[1-3], and it is determined by the temperature, alloying elements, surface oxides, and substrate surface roughness[4]. The common methods to improve the wettability include the temperature change, addition of alloying elements, and coating on the substrate surface. Besides, the introduction of an external field also has a significant effect on the wettability.

The ultrasonic vibration can change the surface state of the liquid/solid system and promote the wetting behavior of the liquid on the solid substrate[5]. Urai et al[6] studied the wetting behavior of water on polytetrafluoroethylene after in situ ultrasonic vibration and found that the wetting angle is decreased during the ultrasonic vibration process and increased slightly after the ultrasonic vibration. Lin et al[7] found that the wettability is improved significantly after ultrasonic vibration. Guo[8] and Benhassine[9] et al studied the effect of ultrasonic vibration on wettability by molecular dynamics. Xu et al[10] studied the wetting behavior of molten Sn solder on bulk metallic glasses, and found that the ultrasonic waves can accelerate the diffusion of the interface atoms and promote the interface reactions, thereby improving the wettability of Sn solder.

When the direct current is applied to the solid/liquid interface, the electromigration, Peltier effect, and convection can be generated inside the solder, which significantly influences the mass transfer during the material processing and the wettability. Chen[11], Chen[12], and Wang[13] et al studied the solid/solid interface reaction of Sn/Ni and Sn/Ag under the direct current and found that the current density and electromigration are the main influence factors for wettability. Zhao et al[14,15] studied the dissolution of solid Ni in liquid Al under the direct current, and found that the dissolution activation energy is decreased after applying the direct current. Xu[16] and Gu[17] et al investigated the wetting behavior of molten solder on the solid substrate under direct current, and found that the polarity of the direct current has little effect

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on the wettability.

Although the effect of ultrasonic vibration and direct current on wettability was individually studied, the combined effect of ultrasonic vibration and direct current on wettability is barely researched. Therefore, the wetting balance method was used in this research to study the wetting behavior of molten Sn solder on Cu substrate under the ultrasonic vibration coupled with direct current. Furthermore, the influence mechanism of ultrasonic vibration coupled with direct current on the wettability of the reaction system was discussed.

1 Experiment

The pure Sn particles and the pure Cu sheet with dimension of 25 mm×8 mm×0.3 mm were selected as the solder and the substrate, respectively. The wetting balance tests were conducted on a solderability tester (SAT-5100, Rhesca, Japan) coupled with a data acquisition software. A hand-held ultrasonic transmitter with a vibration frequency of 28 kHz and an ultrasonic power of 300~700 W was used. A direct current power supply (ZHAOXIN, KXN-3010D, China) with a maximum output current of 10 A was used. The positive electrode was connected to the graphite crucible, and the negative electrode was connected to the Cu substrate. The Sn particles were placed in the graphite crucible with the dimension of 50 mm×40 mm×20 mm and heated by a heating platform. A K-type thermocouple was immobilized in liquid Sn to monitor the real-time temperature, and the temperature error was within ±2 °C. The ultrasonic probe of titanium alloy with a diameter of 10 mm was fixed perpendicularly to the molten solder with a depth of 5 mm. The prefabricated Cu sheet was coated with rosin flux and installed on the holder. The distance from the center line of the Cu sheet to the edge of the vibration probe was 5 mm. The schematic diagram of the wetting balance test apparatus is shown in Fig.1.

Prior to experiments, the surfaces of the Cu sheets were mechanically polished to obtain surfaces with roughness (R_a) of 100 nm, which was measured by a stylus-type surface roughness measuring instrument (Laisida, TR200, China) to ensure that this aspect was uniform for all the specimens. The specimens were ultrasonically cleaned in acetone for 3 min. Finally, they were rinsed with distilled water and blow-dried. The experiment temperature, ultrasonic power, ultrasonic vibration action duration, current intensity, and current action duration were 270 °C, 300~500 W, 4 s, 1~3 A, and 40 s, respectively.

At the beginning of the wetting balance experiment, the prefabricated Cu sheet was fixed on the wetting balance tester, the pure Sn particles were loaded into a graphite crucible, and then the crucible was placed on a heating platform. The temperature was kept constant once the solder temperature reached the designed temperature. Then the direct current was applied and the Cu sheet was moved downward to the molten Sn solder at a speed of 3 mm/s until the immersion depth reached 5 mm. Then the ultrasonic vibration was applied immediately. After the ultrasonic vibration coupled with direct current process was conducted for 40 s, the Cu sheet was

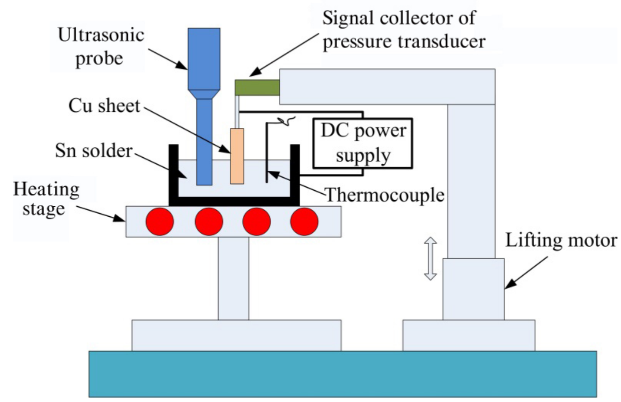

Fig.1 Schematic diagram of wetting balance test device (DC: direct current)

moved upward at the speed of 3 mm/s until the Cu sheet and Sn solder were completely separated. The schematic diagram of the wetting balance test process and the corresponding standard wetting balance curve are shown in Fig.2. The pink rectangular represents the Cu sheet. To reduce the test error, three readings of each parameter were taken and their average value of each parameter was used.

After the wetting balance tests, the Cu sheet was inlaid with a conductive epoxy resin, and then mechanically polished along the cross-sectional direction. The cross-section microstructures were observed by scanning electron microscope (SEM, Quanta FEG450, FEI, USA) coupled with an energy-dispersive spectrometer (EDS). Image-Pro Plus software was used to measure the thickness of intermetallic compounds (IMCs) at the interface.

2 Results

2.1 Effect of ultrasonic vibration coupled with direct current process on wetting behavior of Sn solder on Cu substrate

Fig.3 shows the wetting balance curves of the molten Sn on

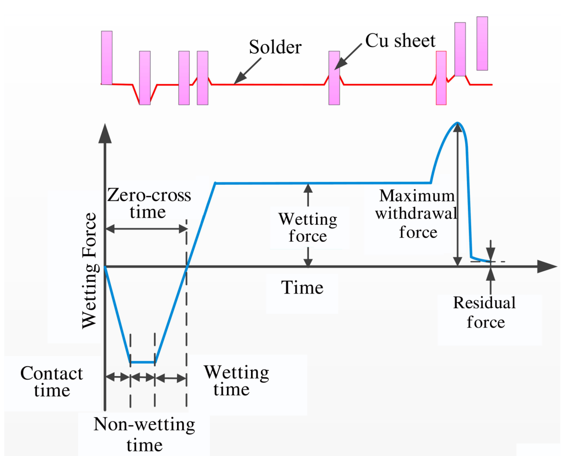

Fig.2 Schematic diagram of wetting balance process and corresponding standard wetting balance curve

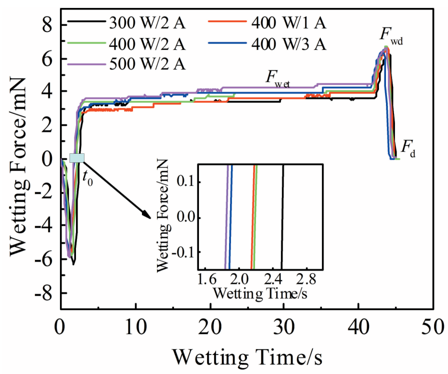

Fig.3 Wetting balance curves of Sn solder on Cu substrate during ultrasonic vibration coupled with direct current process

Cu substrate, and then the zero-cross time t_0, wetting force F_{wet}, maximum withdrawal force F_{wd}, and residual force F_d can be obtained, as listed in Table 1.

It can be seen that the ultrasonic vibration coupled with direct current process has a significant effect on the wettability of the molten Sn solder on Cu substrate. With solely increasing the ultrasonic vibration power from 300 W to 500 W, the wetting force is increased from 3.59 mN to 4.45 mN. With solely increasing the direct current from 1 A to 3 A, the wetting force is increased from 3.96 mN to 4.15 mN. It can be found that during the ultrasonic vibration couple with direct current process, the contribution of ultrasonic vibration to the wettability improvement is larger than that of direct current.

2.2 Effect of ultrasonic vibration coupled with direct current process on interface microstructures of Sn solder on Cu substrate

Fig.4 shows the backscattered electron (BSE) images at the triple-phase regions after ultrasonic vibration coupled with direct current. The scallop-shaped IMC layer consists of Cu_6Sn_5 precipitated on the Sn/Cu interface. Fig. 5 shows the thickness of IMC layers, indicating that the IMC layer becomes thicker as the ultrasonic vibration and direct current increase.

2.3 Sound pressure distribution inside molten Sn solder

Fig. 6 shows the sound pressure distributions inside the molten Sn after ultrasonic vibration coupled with direct current processes with different ultrasonic vibration powers. The sound pressure inside the molten Sn solder is unevenly distributed. The maximum sound pressure occurs at the front end of the ultrasonic probe, which is 9.8×10^6, 1.13×10^7, and 1.28×10^7 Pa with the ultrasonic vibration power of 300, 400, and 500 W, respectively. The farther from the front end of the ultrasonic probe, the lower the sound pressure. It is also found that with increasing the ultrasonic vibration power, the sound pressure inside the molten Sn is generally increased.

3 Discussion

For a reactive wetting system, the final wetting angle depends not only on the surface tension of a liquid metal, but also on the solid/liquid interface reaction. It is reported that the formation of the interface layer has a certain influence on the wetting behavior[18,19]. Therefore, the temperature is fixed in this research and the sufficient amount of molten Sn solder also ensures that it will not be affected by a small amount of Cu substrate. Thus, the effect of surface tension on the wettability of liquid Sn solder is neglected in this research. The dissolution of Cu substrate at the triple-phase regions and the IMC precipitation at interface can influence the final wettability[17]. Therefore, it is important to study the mass transfer behavior of the interface during ultrasonic vibration coupled with direct current process.

3.1 Effect of ultrasonic vibration on mass transfer of interface during ultrasonic vibration coupled with direct current process

When the sound pressure reaches the cavitation threshold (0.3×10^5~0.6×10^5 Pa), the cavitation effect occurs inside the molten solder. The collapse of acoustic cavities may cause the generation of micro-jets and shockwaves, which can form the extremely high temperature and pressure[20,21]. The temperature of the substrate surface caused by the collapse of acoustic cavities can reach 5000 K[22], which is much higher than the melting point of Cu (1728 K) and accelerates the dissolution of Cu substrate into the molten Sn solder. Meanwhile, the microscale high-temperature region formed by the acoustic cavities on the Cu substrate surface can be rapidly quenched by the relatively cold molten solder, thus generating a temperature gradient in the molten solder[23]. It is known that the IMC layer formation at interface hinders the atom diffusion and raises the atomic diffusion barrier, while the formation of temperature gradient weakens the atomic diffusion barrier and increases the Cu diffusion rate in the molten Sn solder. Therefore, the ultrasonic vibration can strengthen the atomic interactions and accelerate the growth of IMCs.

Table 1 Wetting balance test parameters and related results of Sn solder on Cu substrate during ultrasonic vibration coupled with direct current

Ultrasonic vibration power/W	Direct current/A	t_0/s	F_{wet}/mN	F_{wd}/mN	F_d/mN
300	2	2.48	3.59	6.33	0.035
400	1	2.10	3.96	6.70	0.013
400	2	2.19	4.06	6.72	0.010
400	3	1.82	4.15	6.53	0.050
500	2	1.81	4.45	6.57	0.070

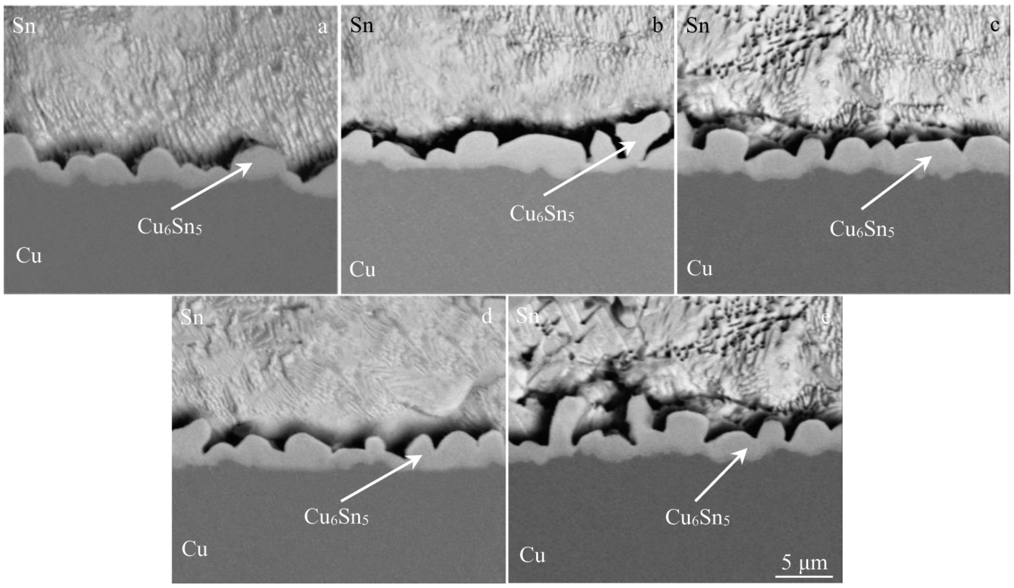

Fig.4 BSE microstructures of triple-phase regions at interface of specimens after different ultrasonic vibration coupled with direct current processes: (a) 300 W/2 A; (b) 400 W/2 A; (c) 500 W/2 A; (d) 400 W/1 A; (e) 400 W/3 A

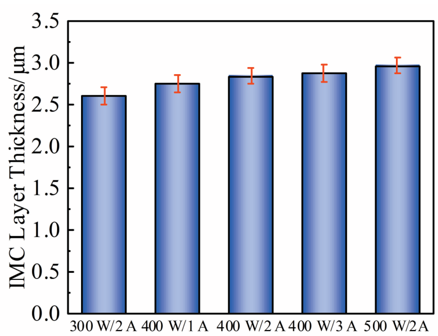

Fig.5 Thickness of IMC layers in triple-phase region of specimens after different ultrasonic vibration coupled with direct current processes

3.2 Effect of direct current on mass transfer of interface during ultrasonic vibration coupled with direct current process

When a direct current is applied to the molten solder, the electromigration, Peltier effect, and convection may occur inside the solder. The Peltier effect is not severe due to the small Peltier coefficient of the metal system[24]. Thus, the influence factors of mass transfer only include the electromigration and Joule heating. The diffusion of Cu atoms and the IMC precipitation at the interface lead to the difference in the internal resistivity of the solder, thereby generating a temperature gradient and concentration gradient inside the molten solder, which enhances the interaction of interface atoms and promotes the precipitation of IMC layer generated by the interface reaction. Although the current densities of 12~36 A/cm^2 in this research are lower than the electromigration threshold, the resistivity of Cu_6Sn_5 is 17.5 μΩ·cm[25], which is higher than that of Sn and Cu. Therefore, the gathering effect of the charge at a scallop-shaped groove may cause a high current density, achieving the threshold value and then triggering the electromigration. Because the resistivity of IMC is relatively large, more heat can be generated at the interface than inside the Sn solder[26], resulting in the temperature gradient which can cause the Marangoni convection in the molten solder.

3.3 Wetting dynamics of Sn solder on Cu substrate during ultrasonic vibration coupled with direct current process

The schematic diagram of the driving force for movement of the triple-phase region is shown in Fig.7. After introducing a quasi-static balance force, the modified Young's equation can be expressed as follows[27]:

$$F = \sigma_{lv}\cos\theta + \sigma_{sl} - \sigma_{sv} \tag{1}$$

where σ_{lv}, σ_{sl}, and σ_{sv} are the surface tensions at the molten Sn/air, molten Sn/IMC, and Cu/air interfaces, respectively. There is an additional tension F at the triple-phase region, which is balanced by the existing surface forces and can change the equilibrium wetting angle θ. The additional tension F consists of the driving force of interface reaction and convection inside the solder.

On the one hand, the interface reaction can initiate the movement of triple-phase region[28-30]. During the wetting process, the interface reaction can take place, as follows:

$$6Cu + 5Sn = Cu_6Sn_5 \tag{2}$$

According to Ref. [31], the system energy transformation per unit spreading length of the molten Sn solder can be expressed as follows:

$$\frac{dE}{dr} = 2\pi r\left[\frac{\rho_{Cu} l_{Cu}}{6M_{Cu}}\Delta G + \gamma_{lv}^{Sn}\left(\cos\theta_i - \cos\theta\right)\right] \tag{3}$$

where r is the instantaneous radius of spreading object; ρ_{Cu}, M_{Cu}, and l_{Cu} are density, molar mass, and reaction layer

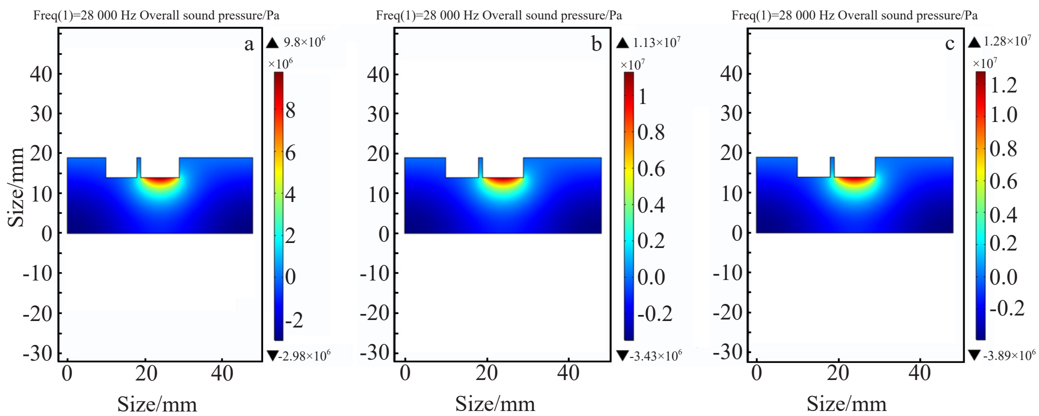

Fig.6 Sound pressure distributions of specimens during ultrasonic vibration coupled with direct current process with different powers: (a) 300 W, (b) 400 W, and (c) 500 W

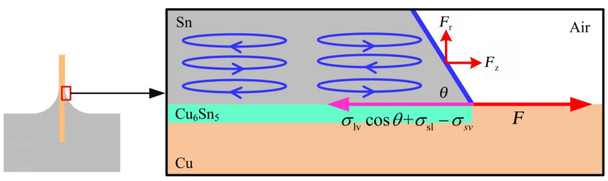

Fig.7 Schematic diagram of forces at triple-phase region

thickness of the Cu substrate, respectively; γ_{lv}^{Sn} is the surface energy of the molten Sn solder (0.56 J/m^2)[32]; θ_i and θ are the dynamic wetting angle and equilibrium wetting angle, respectively; ΔG is the Gibbs free energy change of the Cu_6Sn_5 formation at the interface (ΔG=7747.65−0.371T, with T representing the temperature)[33]. The term $(\rho_{Cu}l_{Cu}/6M_{Cu})\Delta G$ represents the excess free energy generated by the formation of Cu_6Sn_5, which is also the driving force of the interface reaction. The reaction layer thickness l_{Cu} is 2.6, 2.75, 2.84, 2.88, and 2.96 μm and the corresponding driving forces of interface reaction is 483.87, 511.79, 528.54, 535.98, and 550.87 J/m^2 for the specimens under the processing condition of 300 W/2 A, 400 W/1 A, 400 W/2 A, 400 W/3 A, and 500 W/2 A, respectively. The term $\gamma_{lv}^{Sn}(\cos\theta_i-\cos\theta)$ represents the driving force caused by the difference in surface energy (<0.56 J/m^2). Therefore, the driving force of the interface reaction plays an important role. The driving force of interface reaction for the movement of triple-phase region is increased with increasing the ultrasonic vibration power and direct current.

On the other hand, the strong convection inside the Sn solder can also initiate the movement of triple-phase region[34], and it generates a rotating component along the z direction (F_z), which provides a physical driving force for the movement of triple-phase region. Thus, both the micro-jet caused by the ultrasonic vibration and the Marangoni convection caused by the direct current can lead to the enhancement of convection inside the solder. The wettability of the molten Sn solder on the Cu substrate is improved by ultrasonic vibration coupled with direct current process.

4 Conclusions

1) The ultrasonic vibration coupled with direct current process has a significant effect on the wetting behavior of the molten Sn solder on the Cu substrate surface. With increasing the ultrasonic vibration power and direct current, the maximum wetting force is increased. The contribution of ultrasonic vibration to the wettability improvement is larger than that of direct current. Meanwhile, the dissolution of Cu substrate and the precipitation of intermetallic compound (IMC) layer at the interface are promoted by applying the ultrasonic vibration power coupled with direct current process.

2) The precipitation of IMC layer at interface provides a chemical driving force, and the convection inside the molten Sn solder provides a physical driving force for the spreading of molten Sn solder on the Cu substrate. Since the ultrasonic vibration coupled with direct current process can enhance the precipitation of IMC layer and convection, the wettability of Sn solder on Cu substrate is improved.

References

1 Ambrose J C, Nicholas M G, Stoneham A M. *Acta Metallurgica et Materialia*[J], 1992, 40(10): 2483
2 Ambrose J C, Nicholas M G, Stoneham A M. *Acta Metallurgica et Materialia*[J], 1993, 41(8): 2395
3 Yin L, Meschter S J, Singler T J. *Acta Materialia*[J], 2004, 52(10): 2873
4 Kumar G, Prabhu K N. *Advanced Colloid Interface*[J], 2007, 133(2): 61
5 Panteli A, Robson J D, Brough I *et al*. *Materials Science & Engineering A*[J], 2012, 556: 31
6 Urai T, Kamai M, Fujii H. *Journal of Materials Engineering and Performance*[J], 2016, 25(8): 3384
7 Lin Q L, Xing C X, Sui R *et al*. *Experimental Thermal and Fluid Science*[J], 2019, 102: 351
8 Guo W B, Ma K, Wang Q *et al*. *Materials Letters*[J], 2019, 264: 127 118

9 Benhassine M, Saiz E, Tomsia A P *et al*. *Langmuir: the ACS Journal of Surfaces & Colloids*[J], 2009, 25(19): 11 450

10 Xu Z W, Li Z W, Zhong S J *et al*. *Ultrasonics Sonochemistry*[J], 2018, 48: 207

11 Chen S W, Chen C M, Liu W C. *Journal of Electronic Materials*[J], 1998, 27(11): 1193

12 Chen C M, Chen S W. *Acta Materialia*[J], 2002, 50(9): 2461

13 Wang C H, Kuo C, Chen H *et al*. *Intermetallics*[J], 2011, 19(1): 75

14 Zhao J F, Unuvar C, Anselmi-Tamburini U *et al*. *Acta Materialia*[J], 2007, 55(16): 5592

15 Zhao J F, Unuvar C, Anselmi-Tamburini U *et al*. *Acta Materialia*[J], 2008, 56(8): 1840

16 Xu Q G, Liu X B, Zhang H F. *Transactions of Nonferrous Metals Society of China*[J], 2010, 20(8): 1452

17 Gu Y, Shen P, Yang N N *et al*. *Journal of Alloys and Compounds*[J], 2014, 586: 80

18 Choi W K, Yoon S W, Lee H M. *Materials Transactions*[J], 2001, 42(5): 783

19 Liu C Y, Tu K N. *Journal of Materials Research*[J], 1998, 13(1): 37

20 Ji H J, Chen H, Li M Y. *Ultrasonics Sonochemistry*[J], 2017, 35(A): 61

21 Cherepanov P V, Melnyk I, Andreeva D V. *Ultrasonics Sonochemistry*[J], 2015, 23: 26

22 Suslick K S, Hammerton D A, Cline R E. *Journal of the American Chemical Society*[J], 1986, 108(18): 5641

23 Cherepanov P V, Melnyk I, Skorb E V *et al*. *Green Chemistry*[J], 2015, 17(5): 2745

24 Sheibani H, Liu Y, Sakai S *et al*. *International Journal of Engineering Science*[J], 2003, 41(3-5): 401

25 Frederikse H P R, Fields R J, Feldman A. *Journal of Applied Physics*[J], 1992, 72(7): 2879

26 Sun Xuemin, Yu Weiyuan, Wu Baolei *et al*. *Rare Metal Materials and Engineering*[J], 2020, 49(10): 3425 (in Chinese)

27 Manor O, Dentry M, Friend J R *et al*. *Soft Matter*[J], 2011, 7(18): 7976

28 Wang H Q, Gao F, Ma X *et al*. *Scripta Materialia*[J], 2006, 55(9): 823

29 Lin Q L, Li F X, Wang J B. *Journal of Alloys and Compounds*[J], 2018, 767: 877

30 Wang H Q, Wang F J, Gao F *et al*. *Journal of Alloys and Compounds*[J], 2007, 433(1-2): 302

31 Yost F G, Romig A D. *MRS Online Proceedings Library Archive*[J], 1987, 108: 385

32 Eustathopoulos N, Nicholas M G, Drevet B. *Wettability at High Temperatures*[M]. Oxford: Elsevier, 1999

33 Sobiech M, Krüger C, Welzel U *et al*. *Journal of Materials Research*[J], 2011, 26(12): 1482

34 Kozlova O, Voytovych R, Protsenko P *et al*. *Journal of Materials Science*[J], 2010, 45(8): 2099

超声振动和直流电耦合作用对润湿行为的影响

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摘 要：采用平衡润湿测试法研究了270 ℃时，施加超声振动和直流电耦合作用对液态Sn钎料在Cu基体上润湿行为的影响。通过对润湿平衡曲线的测量发现：超声和直流电耦合作用能显著改善Sn钎料的润湿性，随着超声功率和电流强度的增加，最大平衡润湿力也随之增加。对润湿过程中界面微观结构的观察结果显示，超声和直流电耦合作用增强了Cu基体在熔融Sn钎料中的溶解，促进了界面金属间化合物的析出。利用有限元软件对液态Sn钎料内部的声压分布进行模拟，发现最大声压发生在超声探头的端部。在超声振动和直流电耦合作用下，界面析出金属间化合物时所产生的化学驱动力以及超声和直流电作用引起钎料内部强烈的对流作用共同促进了三相线的移动，从而改善了钎料润湿性。

关键词：超声振动；直流电；润湿性；金属间化合物；驱动力

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