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# Research Status and Progress of Lead-Free Solder in Electronic Packaging

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***Abstract:*** Pb in traditional brazing materials Sn-Pb is highly toxic, and leaded brazing materials have been gradually banned in order to minimize the hazards. Research on lead-free solder has made great progress in the past decades, among which SAC alloys are the most representative. SAC has gained wide attention and research because of its relatively good performance, this paper analyzes the performance of SAC alloys with different compositions and alloys doped with different elements, and gives an overview of the latest research on SAC brazing materials in recent years. Due to the problem of the high melting temperature of SAC braze in the field of complex architecture packaging, this paper also classifies and summarizes the low-temperature brazes proposed to solve the high melting temperature of SAC, analyzes the advantages and shortcomings of two kinds of brazes, Sn-based and In-based, and lists the ones that have been previously doped with different elements in different systems, and discusses the possible directions of future research.

***Keywords:*** Lead-free solder, SAC, LTS

## 1. Introduction

Reliable electronic packaging technology is the key to stable data transmission, and the reliability of interconnect materials has always been the focus of attention in the packaging field. Eutectic SnPb brazing alloy is the preferred material in the industry due to its good thermoelectric properties and high reliability. However, due to the toxicity of lead (Pb) element, the application of traditional Sn-Pb binary alloy brazing material is gradually banned. Therefore, the exploration of new lead-free brazing alloys has received extensive attention from researchers around the world. Low-silver solder SnAgCu(SAC)-based brazing material is considered as the most promising brazing material to replace Sn-Pb due to its good performance and low cost, in which it has become a research hotspot due to better market competitiveness. This paper will summarize the research results in the last decade on improving the performance of low-silver SAC brazing materials by adding metals, rare earth elements and nanoparticles, starting from the melting characteristics, microstructure and mechanical properties of brazing materials [1]. On this basis, Sn-Bi and In-based solders are introduced as low-temperature solders (LTS), which can solve the problem of significantly affecting the encapsulation stress due to the high melting temperature of SAC-based solders. Starting from the development of encapsulation materials, this paper analyzes the compositional changes of mainstream solders in the past two decades and the problems they have solved to adapt to the development, and describes the deficiencies of the materials to provide theoretical support for the subsequent research and development of brazing materials.

## 2. SAC System

As interconnect materials, Sn-based brazing materials have been widely used. SAC system brazing material is considered to be the most promising brazing material to replace Sn-Pb due to its relatively good performance. several traditional high silver SAC system solders such as Sn3.8Ag0.7Cu, Sn3.0Ag0.5Cu (SAC305), Sn3.5Ag0.7Cu, etc., have significantly decreased their competitiveness in the market due to the increasing price of silver, and the market competitiveness has decreased. Therefore, low-silver (Ag mass fraction ≤ 1.0%) brazing material has become a research hot spot.

However, when the Ag content decreases, the brazing material shows better ductility, but the melting point increases and the wettability deteriorates, which makes the overall performance decline. Therefore, researchers have added other elements to low silver brazing materials to improve the performance, and a series of research results confirm that the addition of metal elements such as Bi, Ni, Zn, etc. can inhibit the growth of interfacial intermetallic compounds (IMCs), so as to make low-silver brazing materials obtain excellent mechanical properties. In this chapter, we will summarize the research results in the last decade on improving the properties of low-silver brazing materials by adding metals, rare earth elements and nanoparticles, starting from the melting characteristics, microstructure and mechanical properties of brazing materials.

### 2.1. Melting Characteristics

Melting characteristics are an important indicator for evaluating lead-free solders. A representative conventional SAC305 braze has a melting point of approximately 217°C, and an equally representative SAC105 braze has a melting point of 227°C. Reducing the Ag content results in the alloy having a higher melting point. The high melting point results in a higher peak reflow profile of the solder joints, which reduces solderability while elevating the circuit substrate component failure rate. In previous studies, chemical reduction and electrodeposition have been used to prepare SAC nano-braze, and nano-SAC braze can effectively reduce the melting point of low-silver SAC braze, but there is still a big problem in the preparation [2-3]. The mainstream methods to improve the properties of low-temperature brazing materials are alloying and particle strengthening. The melting characteristics of the alloys studied so far are shown in Table 1.

Table 1: Effect of metal elements on the melting characteristics of SAC solder

| Solder alloys | Solidus (°C) | Liquidus (°C) | Melting range (°C) | Melting points (°C) | Supercooling (°C) |
|---|---|---|---|---|---|
| SAC0507-1Ga | 211.5 | - | - | 225.8 | - |
| SAC103-2Zn | 221.0 | 227.6 | 6.6 | 222.8 | 3.9 |
| SAC103-3Zn | 218.0 | 225.5 | 7.5 | 220.8 | 2.5 |
| SAC105-0.1Fe | 217.4 | 226.4 | 9.0 | - | 23.0 |
| SAC105-0.3Fe | 217.6 | 226.5 | 8.9 | - | 20.3 |
| SAC105-0.5Fe | 217.0 | 226.5 | 9.5 | - | 19.4 |
| SAC105-0.05Fe-1Bi | 217.6 | 230.0 | 12.4 | - | 10.1 |
| SAC105-0.05Fe-2Bi | 214.0 | 229.0 | 15.0 | - | 9.8 |
| SAC0705-3.5Bi-0.05Ni | 206.5 | 215.7 | 9.20 | - | - |
| SAC305 | 203.5 | 227.6 | 24.1 | 217 | 15.0 |

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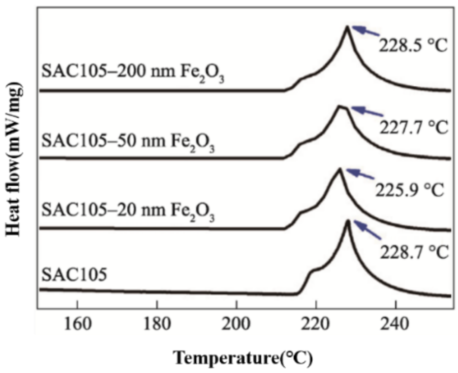
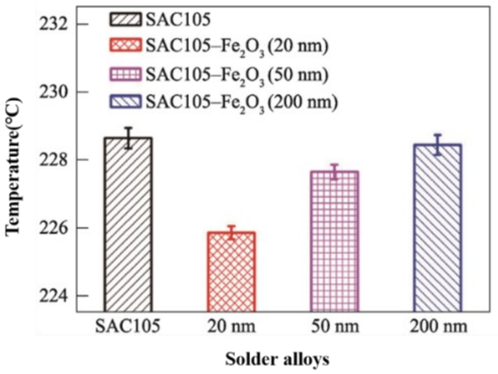
By comparing the results of differential scanning calorimetry (DSC), it was found that in SAC105, with the increase of Bi elemental mass fraction, both the solid and liquid phase lines decreased, but the melting range increased. This view can also be corroborated in the study of Liu et al. In this experiment, Bi and Ni elements were added to Sn0.7Ag0.5Cu (SAC0705) brazing material, and the melting point of the brazing material was significantly decreased, but the melting range was increased, which was mainly caused by the addition of Bi elements [4-5].

In addition, it has been found that the addition of Zn can significantly reduce the melting point of SAC103 brazing material [6]. After the addition of Zn with a mass fraction of 2-3%, the melting point and melting range of SAC system alloys show a decreasing trend, and finally the alloy with 3 wt.% Zn added maintains the solid-phase line temperature around 218.5℃, while the liquid-phase line decreases to 225.5℃ significantly. It can be concluded that the addition of elemental Zn significantly reduces the melting range of the brazing material.

The addition of rare earth elements can also be used to improve the high melting point problem of SAC. Ga is a low melting point metal, and the formation of the alloy can significantly improve the high melting point problem of SAC braze. According to the experimental data, the solid phase line temperature decreases from 217.0℃ to 211.5℃. It can be concluded that the addition of Ga alloy can make up for the shortcomings of the high melting temperature of SAC brazing material [7].

Zhao et al. added $Fe_2O_3$ nanoparticles to SAC105 and found that the initial crystallization temperature of the nanobraze could be reduced. Further investigating the effect of $Fe_2O_3$ nanoparticles of different sizes on the alloy, it was found that the addition of $Fe_2O_3$ nanoparticles with a size of 20 nm significantly reduced the initial crystallization temperature of the nanocomposite brazing material (from 228.7℃ to 225.9℃) as shown in Figure 1a, which was due to the higher surface free energy of $Fe_2O_3$ particles, which increased the surface of the nanocomposite brazing material instability and reduced its initial crystallization temperature. In addition, the liquid phase line temperature of the nanocomposite braze was lower than that of the SAC105 braze, with the lowest liquid phase line temperature for the composite braze with the addition of $Fe_2O_3$ of size 20 nm, as shown in Figure 1b.




(a) DSC curves (b) Liquidus melting temperature

Figure 1: Melting parameters of the SAC105-x$Fe_2O_3$ solder (x= 0,20,50,200 nm)

## 2.2. Microstructure

In SAC-system alloy brazing materials, the brazing matrix mainly consists of β-Sn, $Cu_6Sn_5$, and $Ag_3Sn$. In low-silver SAC brazing materials, the number of large lath-like $Ag_3Sn$ IMC in the matrix decreases with the decrease of Ag content, but at the same time, the uneven distribution and coarsening of the grains of β-Sn and $Cu_6Sn_5$ make the mechanical properties and reliability of SAC brazing materials decrease. Taking Sn0.5Ag0.7Cu as an example, Figure 2 shows the Sn0.5Ag0.7Cu brazing matrix microstructure at a cooling rate of 0.3 K/s [8]. It is obvious from the TEM that the

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fibrous $Ag_3Sn$ IMC and slaty $Cu_6Sn_5$ are unevenly distributed in the eutectic region, and these large and brittle $Ag_3Sn$ intermetallic compounds can adversely affect the mechanical properties. Therefore, controlling the mechanical properties of the IMC layer on the alloy as a whole is the key to improving the reliability of low-silver brazing materials.

In the research often through the alloying and particle strengthening methods, refine theβ-Sn, IMC grains in the matrix, change the matrix organization, make the phases uniformly distributed, orderly arrangement, and play the role of fine crystal strengthening. Zn has an obvious modification effect on SAC, easy to replace the Sn atom in $Cu_6Sn_5$ to generate $Cu_6(Sn,Zn)_5$ IMC, but Zn is easily oxidized and should not be added in excess. El-Daly found that the addition of Zn with a mass fraction of 3% to SAC103 can refine the brazing microstructure and improve the alloy strength, without being an alloy that is too strong and leads to poor ductility [6]. In addition, the addition of trace Zn elements will also generate stable $Cu_6(Sn,Zn)_5$ IMC at the interface, compared with $Cu_6Sn_5$ IMC, $Cu_6(Sn,Zn)_5$ IMC has a very small size and more quantity per unit area, and the more dense grain boundaries restrict the diffusion of Cu from the substrate, and at the same time, it has a positive effect on the obstruction of the interfacial $Cu_3Sn$ IMC layer The positive effect on hindering the growth of the interfacial $Cu_3Sn$ IMC layer is also one of the reasons for the improved mechanical properties of the SAC due to the addition of Zn.

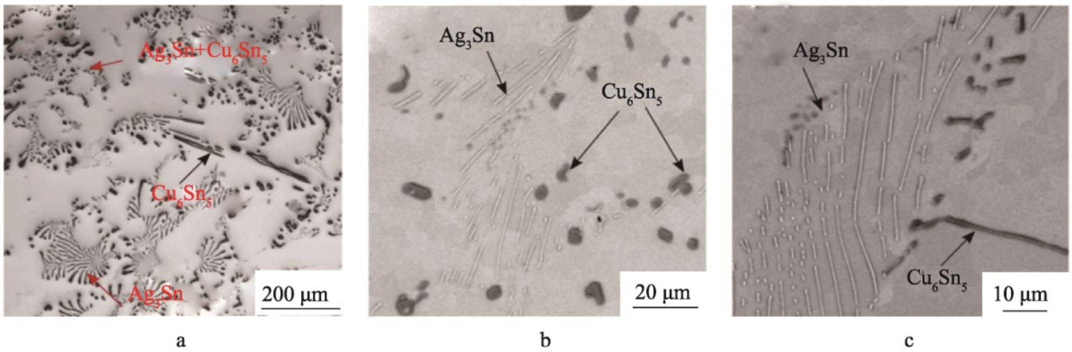


Figure 2: Microstructure of Sn0.5Ag0.7Cu solder at 0.3 K/s cooling rate at different resolutions

In Mahdavifard et al. Fe and Bi were added to SAC105 braze as shown in Figure 3. In Figure 3(a), $Cu_6Sn_5$ and $Ag_3Sn$ were distributed in the β-Sn dendrites and interdendritic regions. When the mass fraction of Bi reaches 2%, the $Cu_6Sn_5$ grains are significantly refined, as shown in Figure 3(c). This is due to the fact that the addition of elemental Bi degraded the eutectic region to a chain arrangement thereby decreasing the $Cu_6Sn_5$ content and increasing the β-Sn content in the solder. In Figure 3(b), it can be found that with the addition of 0.05 wt.% of elemental Fe, a massive $FeSn_2$ phase distributed at grain boundaries appears, and with the increase of elemental Bi, the $FeSn_2$ aggregates into an elliptical shape. In Huang's study, it was shown that the solid solution limit of Bi element in Sn-Ag based brazing material is 4 wt.% at room temperature, and only when the content of Bi exceeds the solid solution limit, the supersaturated Bi precipitates out as pure Bi phase, which is the reason why Bi did not appear in alloying brazing material in Figure 3 [7].

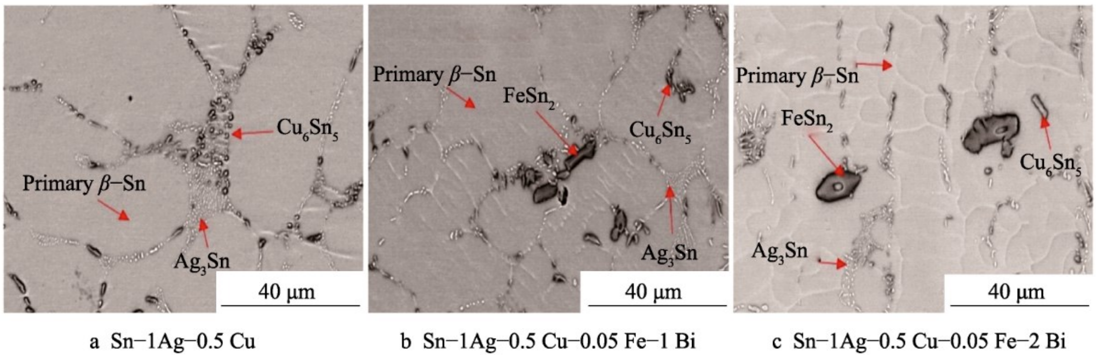


Figure 3: FESEM images of SAC105, SAC105-Fe-1Bi and SAC105-Fe-2Bi solders

## 2.3. Mechanical Property

Good mechanical properties of solder joints are the key to guarantee reliability. In comparison with the conventional SAC305, the low silver brazing material SAC105 has a consequent decrease in the Sn-Ag phase ($Ag_3Sn$) grain content and size in the matrix due to the decrease in Ag content, and the needle-like $Ag_3Sn$ IMC plays an important role in the overall mechanical properties of the solder joints. In a study by Cheng et al, the ultimate tensile strengths of three SAC-based alloys were compared for SAC305, SAC105, and SAC0305 at room temperature (25℃) at different strain rates [9]. The ultimate tensile strengths of SAC105 and SAC0305 are less than SAC305 at different strain rates, which indicates that the mechanical properties of SAC-based brazing alloys weaken with decreasing Ag content.

In a study by El-Daly et al, the addition of different levels of Zn to SAC103 can effectively enhance the tensile strength of the brazing material [6]. The results showed that the addition of 2 wt.% Zn effectively increased the strength of the alloy but at the same time resulted in poor ductility, whereas the alloy possessed both the highest strength and better ductility at an addition of 3 wt.% Zn. This is due to the generation of fine $(Cu,Ag)_5Zn_8$ and fine acicular $Ag_3Sn$, which IMC particles can act as barriers to dislocation movement and thus enhance the strength of the alloy.

The role of element Bi as an excellent additive element to SAC brazing materials has been widely studied. A small amount of Bi can lead to a lower melting point of the brazing material, which improves the mechanical properties of the solder joints due to the solid solution strengthening of the Sn-Bi phase. Excessive amounts of Bi can lead to precipitation of pure Bi phase beyond the solid solution limit, which reduces the reliability of solder joints. Meanwhile, the addition of trace Ni to SAC solder can significantly refine the interface IMC and improve the heat aging resistance of the solder interface, a property that is crucial for the service life of electronic components. Liu et al. added 3.5 wt.% bismuth and 0.05 wt.% Ni to SAC0705 and investigated its mechanical properties. It can be seen from Figure 4 that after aging for 200h, the shear strength of Sn0.7Ag0.5Cu-3.5Bi-0.05Ni is still higher than that of SAC305, which is up to 41.3MPa. Therefore, for SAC0705 brazing material, the addition of 3.5% Bi and 0.15% Ni can obviously improve its mechanical properties, and the mechanism is that the microstructure of the brazing matrix is refined by Ni, while the Bismuth and 0.05wt.% Ni are added to SAC0705 and its mechanical properties are investigated[5]. The mechanism is that the microstructure in the matrix of the brazing material is refined by Ni, and at the same time, Bi element has the role of solid solution strengthening, so it improves the mechanical properties of the brazing material.

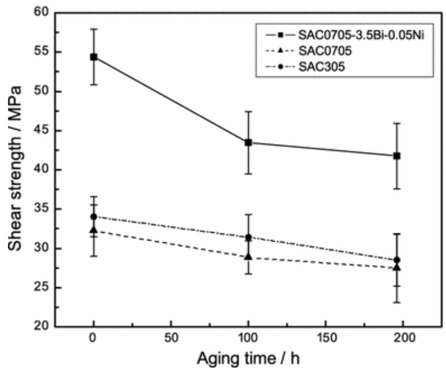


Figure 4: Shear strength of the solder joints before and after HTS aging

The yield strength and ultimate tensile strength of brazing material were substantially increased by adding 0.05 wt.% Fe with 1 wt.% or 2 wt.% Bi to SAC105 brazing material [8]. The yield strength increased from 21.1 MPa to 31.7 MPa and the ultimate tensile strength increased from 27.5 MPa to 40.5 MPa when 1 wt.% Bi was doped, and when the doping concentration reached 2 wt.%, the yield strength increased to 41.8 MPa and the ultimate tensile strength rose to 56 MPa.In addition, the SAC105-0.05Fe$_x$Bi (x =1,2) alloy brazing material has an excellent thermal aging resistance, which is attributed to the reduced rate of IMC coarsening due to the reduction of Sn-Cu or Sn-Ag interactions by Bi[10]. At the same time, Fe atoms in the Sn substrate in the eutectic region significantly reduced the diffusion rate of Ag and reduced the supply of Ag required for the coarsening of Ag3Sn IMC. This conclusion demonstrates that doping Bi and Fe can effectively enhance the mechanical properties of low-Ag SAC system brazing materials.

## 3. Low Temperature Solder (LTS)

Sn-Ag-Cu (SAC) alloys are the most commonly used solder for Sn-Pb solder replacement due to their excellent mechanical properties and reliability. However, their melting range (217℃-222℃) is much higher than that of lead (Pb)-based solders. With advances in microelectronic packaging for aggressive silicon nodes and complex heterogeneous architectures, the high melting temperature of SAC-based solders significantly affects package stress, and multiple high-temperature reflows may damage temperature-sensitive regions. Therefore, research on lead-free low-temperature solder (LTS) is critical. Bi-based and In-based solders have become popular prime candidates for LTS due to their low-temperature alloying capabilities with Sn and Ag.

### 3.1. Sn-Bi

Sn-Bi system alloys have become the most widely used LTS alloys due to their low melting temperature, good compatibility and high reliability, etc. Sn-Bi undergoes a eutectic reaction at 138℃, $L \rightarrow \beta\text{Sn} + \text{Bi}$ [11]. The reliability of Sn-Bi is discussed in depth in this section.

Based on Wang et al. it was found that the addition of Bi to Sn not only changed the melting point of the solder, but also the microstructure of the solder, the interfacial behavior between the solder and Cu, and the joint strength [12]. Five Sn-Bi alloy brazes (Sn-5Bi, Sn-15Bi, Sn-30Bi, Sn-45Bi, Sn-58Bi) in the range of 5-58 wt.% were selected in this study. It is found that the IMC layer thickness increases with the increase of Bi content when the Bi content is less than 45 wt.%, while after 45 wt.%, a continuous Bi rich layer is formed close to the interface, and the thickness of the IMC layer starts to decrease, which suggests that the segregation of Bi at the interface inhibits the transformation of Cu6Sn5 to Cu3Sn, and that the continuous Bi rich layer partially blocks the Cu-Sn interdiffusion.

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Among Myung's studies, the addition of a small amount of Ag to the Sn-Bi system was found to thicken the IMC layer with increasing aging time, as shown in Figure 5. With the increase of aging time up to 500 h, the Sn-rich and Bi phases undergo coarsening and the thickness of the IMC layer of the undoped Sn-58Bi increases rapidly [13]. While the bond strength and fracture energy are higher after 0.4-1 wt.% Ag doping, the formed Ag3Sn IMC also consumes the Sn in the solder matrix, which prevents Cu from sufficiently reacting with Sn, thus effectively controlling the increase of the IMC layer thickness after thermal aging. Figure 6 shows the effects of different aging times on the bond strength and fracture energy. The bond strength and fracture energy both decrease with the increase of aging time, but the addition of Ag still makes the solder joints keep high strength. All the above conclusions lead to the conclusion that Ag doping improves the mechanical properties of Sn-Bi system LTS solder joints after thermal aging.

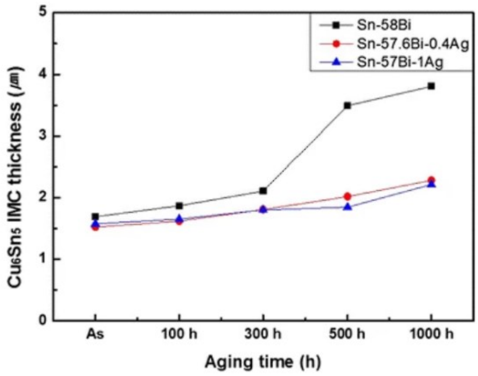


Figure 5: Dependence of $Cu_6Sn_5$ IMC thickness on thermal aging time

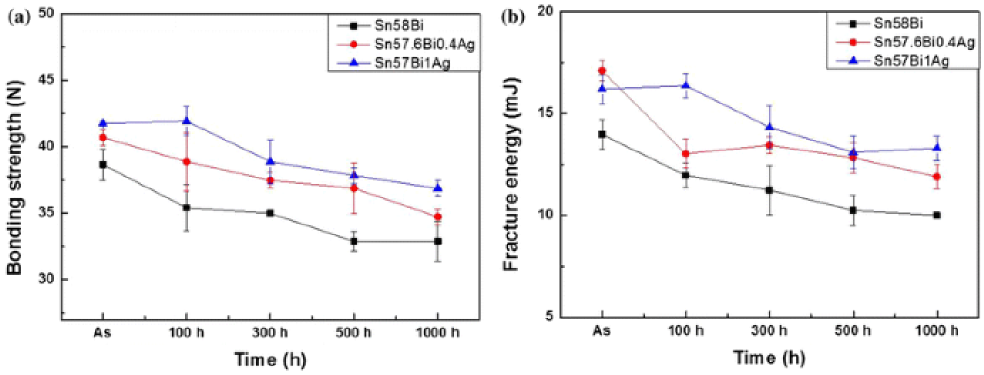


Figure 6: Dependence of (a) bonding strength and (b) fracture energy of Sn-Bi, Sn-Bi-Ag solders on thermal aging time

## 3.2. In-based Systems

As one of the most versatile metals known to man, In has excellent ductility and thermal conductivity, giving it a unique advantage in packaging, In alloy brazes have better mechanical properties for solder joints, and their flip-chip interconnect models show better fatigue life than Sn-Pb in thermal shock experiments. The most widely studied In-based systems are In-Sn and In-Ag, which have better mechanical properties for soft solder joints. These two representative alloys will be discussed in depth in this chapter.

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### 3.2.1. In-Sn

Eutectic In-Sn is widely used in microelectronics due to its low melting point, especially thanks to the excellent bonding effect with metals such as Cu and Ni. In-Sn undergoes eutectic reaction at 118°C:$L \rightarrow \mathrm{Sn(In)} + \beta$, where Sn(In) is the main term and β is the intermediate phase.

In-Sn systems are generally considered to have lower mechanical strength than other Sn-based systems due to the softer nature of the In metal. In addition, In is characterized by excellent ductility and can remain ductile at low temperatures, resulting in In-Sn based solder joints having good fatigue life. On the other hand, the presence of In promotes the formation of $Cu_6(Sn,In)_5$ IMC, which improves the solubility of Cu in the system and enables the formation of strong solder joints with Cu pads even under low temperature soldering conditions. However, the In-Sn system is still deficient in creep resistance, so the study of Sn-In-X alloy brazes is one of the hotspots in the study of LTS nowadays.

### 3.2.2. In-Ag

In-Ag systems are widely used in microelectromechanical systems (MEMS) packaging due to their excellent low-temperature bonding capability and outstanding thermal conductivity. When integrating MEMS, the residual stresses between MEMS multilayer structures due to the process can be effectively solved by using the In-Ag system with a lower bonding temperature. Indium completely melts at 156°C and then reacts with neighboring silver to form AgIn2. At 166°C, AgIn2 decomposes to form molten In and Ag2In, further continuing the reaction of molten In with Ag. The reaction continues until all the molten phases are consumed and the joint solidifies at 180°C [14].

The mechanical properties of In-Ag braze in In-Ag system are similar to those of pure In when the mass fraction of In is > 92%. When In-Ag braze is used for thermal interface materials (TIM), the voiding problem in the braze is critical to its reliability. The voiding problem arises when the preheating temperature is too low to allow sufficient evaporation and removal of the flux. In a study by Otiaba et al. the percentage of voids in TIM decreased with thermal aging time and attributed this phenomenon to high heat flux activity due to prolonged exposure to higher temperatures [15]. In addition, bonding methods without flux have been developed, which is also an effective way to control the In-Ag brazing cavities problem efficiently.

## 4. Conclusion

This paper discusses the melting characteristics, microstructure, mechanical properties and advantages and shortcomings of two types of widely used low-temperature solders of several common Sn-Ag-Cu low-silver lead-free solders, and summarizes the basic information and main characteristics of lead-free alloy brazing materials. The SAC system of lead-free solders has a lot of advantages compared with the Sn-Pb system such as being environmentally friendly, and through the doping of metal elements (Zn, Bi, Fe, etc.), rare earth elements (Ga, etc.), nanoparticles ($Fe_2O_3$, etc.) can have better performance than the solder clamp solder in mechanics, electrical connections, etc. The doping of metallic elements (Zn, Bi, Fe, etc.), rare earth elements (Ga, etc.), and nanoparticles ($Fe_2O_3$, etc.) can provide better mechanical and electrical connection performance than clamp solder. Low-temperature solder brings a major breakthrough in the packaging process and reliability. In the packaging stage, lower reflow temperature means higher reliability and less placement energy consumption. Different alloy formulations can be formulated according to the application or process for greater compatibility.

Although there have been a large number of research results on lead-free solder, most of the research results just stay at the level of academic research, and it is difficult to put into practical applications. Therefore, in the future research and development of lead-free solder in practical application-oriented research will become the focus of the content.

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