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ARTICLE

Research Progress on Intermetallic Compounds in Copper-Aluminum Brazed Joints

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Abstract: The intermetallic compounds (IMCs) in the copper-aluminum brazed joints during formation and application were discussed through reviewing some recent research on brazing copper and aluminum. The review indicates that it is difficult to avoid the formation and growth of the IMCs, which depends on the mutual diffusion between Cu substrate and Al substrate as well as substrates and filler metals. Thermodynamics and kinetics are critical for the nucleation and growth of the IMCs, respectively. Besides, defects (voids, cavities and cracks) in the joint mainly result from the formation and growth of the brittle IMCs because they always result in stress concentration as the source of cracks and accelerate the excessive consumption of the diffused atoms to form voids and cavities. Properties of copper-aluminum joints are severely deteriorated when the thickness of the IMCs exceeds 2~5 μm. Finally, numerous factors (melting point, thermal conductivity, joint design, heat input and chemical composition) strongly impact the formation and growth of the IMCs through changing the mutual diffusion process. Moreover, these factors also have distinct effects on the defects. At present, some efficient methods used to control the IMCs in the copper-aluminum joints are heat input controlling, optimization of joint design and the addition of the third element into filler metals.

Key words: intermetallic compounds; copper; aluminum; dissimilar brazing; factors impacting the Cu-Al joint; atomic diffusion

Copper-aluminum connectors are increasingly used in automotive, marine, electrical and electronic industries due to their exceptional performances of high thermal and electrical conductivities and excellent mechanical properties. Replacing Cu with cheaper and light-weight Al has distinct benefits to reduce the manufacturing costs and save resources [1-6]. In addition, with the promotion of electric vehicles, high-performance batteries are desired to provide enough power for them. Battery packs assembled by many pouch cells can meet this desire. Hence, copper-aluminum joints are one of the key components of the assembly process since pouch cells own a couple of flat connection terminals, which are made of copper and aluminum [7-14]. However, it is of great difficulties to fabricate a fine Cu-Al joint because Cu and Al are significantly different in several aspects of thermal conductivity, linear expansion and density. What's worse, Cu and Al can easily react to form the intermetallic compounds (IMCs) with properties of high hardness, brittleness and resistance [15-17]. The existence of the IMCs in the Cu/Al joint not only results in stress concentration, which is one of the main causes for the formation of cracks in the Cu-Al joint, but also increases the connection resistance, improving the operating costs and shortening the service life of joints[1, 9, 18].

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A lot of studies have been done to investigate the formation and growth of the IMCs and to find methods for the inhibition of them. Phase composition of copper-aluminum vacuum-brazed joints was studied[19,20]. Flash welding is firstly used to join copper to aluminum [21], and then other welding technologies are employed, such as diffusion welding, explosive welding, friction welding, friction stir welding, laser beam welding and ultrasonic welding[3, 4, 14, 22-28]. Although using these welding technologies (except laser beam welding) can make relatively satisfactory Cu-Al joints, they are still not suitable for complex parts and mass production. Compared with those welding technologies, brazing technology is a kind of efficient joining method that is more suitable for complex parts and mass production [1]. The formation and growth of the IMCs during brazing and application are inevitable. So, the formation and growth of the IMCs are still the focus of Cu-Al brazing in the further research.

In this research, some precious studies in recent years on the IMCs in the copper-aluminum brazed joints were reviewed systematically. The formation and growth of the IMCs in the Cu-Al joint during formation and application were discussed. Furthermore, the defects in the Cu-Al joint and factors influencing the defects were summarized, and the relationship between the property of the Cu-Al joints and the IMCs was discussed. Finally, the main causes impacting the change of IMCs were summarized, efficient methods controlling them were listed, and factors influencing the IMCs and defects were explained according to the diffusion mechanism.

1 Formation and Growth of IMCs

The IMCs formation occurs in the Cu-Al joint during brazing, which is similar to other joining processes. From the Cu-Al phase diagram, IMCs $CuAl_2$, CuAl, Cu_3Al_2, Cu_4Al_3 and Cu_9Al_4 are likely to form in the joint in the temperature range from 150 °C to 500 °C, as shown in Fig.1 and Fig.2. The primary characteristics and properties of four common IMCs are shown in Table 1[7]. Solchenbach et al jointed copper and aluminum using laser brazing without filler metal and flux. They employed a laser beam with a circular spatial power modulation system to irradiate the aluminum surface. The experimental parameters were: repetition frequency of laser beam f=500 Hz, amplitudes of circular beam movement a=0.25 mm, overlap between two irradiated cycles n=0.75, feed rate in x-direction v=55.6 mm/s, peak power P_P=385 W, modulation time t_M=32 μs, modulation period T_M =10 kHz. Research results show that IMCs form and consist of Cu_9Al_4 and $CuAl_2$ phases, as shown in Table 2[9]. Moreover, Solchenbach et al found that the IMCs are converted from Cu_9Al_4 layer and $CuAl_2$ layer to CuAl (or Cu_4Al_3) layer, Cu_9Al_4 layer and $CuAl_2$ layer after annealed at 400~500 °C for 120 h, implying the generation of the new IMC after heat treatment. The total thickness of the IMCs layer increases without the formation of new phases after annealing at 200~300 °C for 120 h, suggesting that the original IMCs grow during brazing due to the heat treatment[26]. Pascal et al used the same method to braze aluminum to copper. The experimental parameters were: repetition frequency of laser beam f=500 Hz, feed rate in x-direction v=50 mm/s, peak power P_P=400 W, modulation time varied from 24 μs to 48 μs in 3 μs steps, modulation period T_M=18 kHz. Results indicate that the Cu/Al interface consists of $CuAl_2$, CuAl, Cu_4Al_3 and Cu_9Al_4 [29].

There are three views to explain the phenomenon. The first one is the theory of formation energy, that is, the phase with the smallest formation energy form first, raised by Chen et al [30]. Hence $CuAl_2$ is the initial phase and then CuAl and Cu_9Al_4 appear simultaneously. The second one is the theory of negative effective formation energy proposed by Gueydan et al[31], who thought that the phase with the largest negative effective formation heat forms as the original phase. Consequently, $CuAl_2$ and Cu_9Al_4 form first. Obviously, these two explanations are incompatible. Recently, Wang et al[21] give a new opinion, that is, thermodynamics and kinetics are critical for the nucleation and growth of the IMCs, respectively. The mutual diffusion between Cu and Al, which cause the formation of the IMCs, is mainly from Cu to Al since the atomic radius of Cu is smaller than that of Al. Phase diagram and diffusion law can be used to predict the formation of the IMCs. Hence $CuAl_2$ and Cu_9Al_4 form first because of proper temperature and great interface concentration gradient between filler metals and Al substrate and Cu substrate, and then CuAl forms, earlier than Cu_3Al_2 and Cu_4Al_3 for the same reason.

The phase composition is strongly changed because of the using of filler metals when brazing. For instance, disordered IMC bulks are presented and consist of $CuAl_2$ and $CuZn_3$ in the Cu/filler metal interface when brazing with Zn-Al alloys [1]. Besides, we can observe $CuZn_3$, $CuAl_2$ and Cu-Ce IMCs in the middle of the joint brazed with Zn-22Al-xCe filler metals. The $CuAl_2$ phase is in a shape of bulky strip [2]. The element's electronegativity is primarily responsible for it. Cu-Ce IMCs are the easiest to form since the electronegativity difference

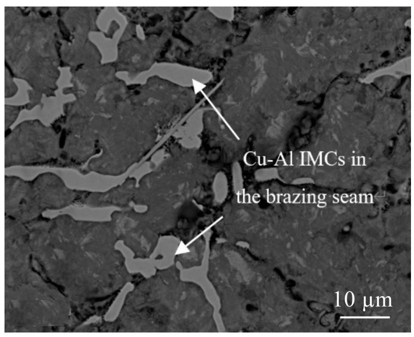

Fig.1 SEM image of Cu-Al IMCs in the brazing seam

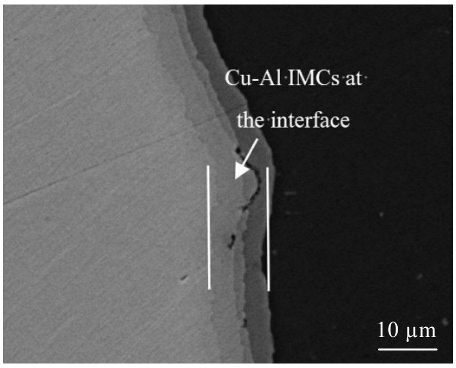

Fig.2 SEM image of Cu-Al IMCs at the interface [21]

Table 1 Characteristics of 4 common IMCs in Al-Cu binary system [7]

Phase	Chemical composition	Crystal structure	Atoms per unit cell
γ_2	Cu_9Al_4	body-centered cubic	36 Cu, 16 Al
ς_2	Cu_4Al_3	Monoclinic	12 Cu, 9 Al
η_2	CuAl	Body-centered orthorhombic	10 Cu, 10 Al
θ	$CuAl_2$	Body-centered tetragonal	4 Cu, 8 Al

between copper and germanium is relatively higher than that between copper and aluminum. Hence Cu reacts with Ce to form Cu-Ce IMCs.

Apart from the formation and growth of the IMCs in the Cu-Al joint during brazing, studying the changeover of the IMCs in the joint in service is equally important. Copper-aluminum joints are widely used in electrical and electronic industries due to their perfect electrical and thermal conductivities. In addition, to provide high energy for electric vehicles, it is also a dispensable part to fabricate battery pack [9]. It is likely that the formation of new IMCs and the growth of the original IMCs occur due to the passing of electrical current [21, 22, 29].

According to Solchenbach et al's study of influences of aging on Cu-Al joints, electrical aging has greater effects on the growth of the IMCs than thermal aging, because the atomic diffusion promoted by electric current is severer than by chemical potential. Besides, the growth rate of the IMCs increases by 6.5 times when electrical current direction is changed from Al→Cu to the opposite direction [29]. Wang et al recently studied the growth of the IMCs layer in the Cu-Al joint through investigating the growth kinetics of the total IMCs layer, the $CuAl_2$ layer, the Cu_9Al_4 layer and the Cu_4Al_3 layer[21, 22]. The continuous growth of the IMCs occurs with increasing the heating time. Besides, the experimental results show that the growth rate of the total IMCs layer conforms the parabolic time law, which is in great agreement with the conclusions of friction welding and cold roll welding[23, 32, 33]. Wang et al's view can be used to explain this phenomenon. The atomic diffusion rate is accelerated by heat input and electric current, causing phase growth and formation of new IMCs and phases with better kinetics conditions grow faster than other phases.

Table 2 EDS point spectrum analysis results (at%) [9]

Point	Al	Cu	XRD analysis
1	3.6	96.4	
2	49.0	51.0	Cu_9Al_4
3	72.0	28.0	$CuAl_2$
4	81.8	18.2	

From the studies mentioned above, it will come to the conclusion that the formation and growth of the IMCs are likely to occur in the Cu-Al joint during brazing and application. Thermodynamics and kinetics are critical for the nucleation and growth of the IMCs, respectively.

2 Formation of Voids, Cavities and Cracks

The generation of defects (voids, cavities and cracks) which deteriorate the properties of Cu-Al joints cannot be ignored, as shown in Fig.3 [18, 26]. Generally, defects are started in the IMCs. For instance, Xia et al examined the crack in the joint brazed by vacuum brazing with Al-Si filler metals. Examination results indicate that cracks are started on the $CuAl_2$ layer, and then extended to Cu_3Al_2 layer and brazing seam [18]. Zhou et al found that all Cu-Al joints produced by laser brazing are fractured along the $Cu_9Al_4/Cu_{3.2}Al_{4.2}Zn_{0.7}$ interface and $CuZn/Cu_{3.2}Al_{4.2}Zn_{0.7}$ interface, and extended toward the brazing seam [34, 35]. Solchenbach et al's experiment results implied that when the joint was annealed at 500 °C for more than 24 h, cracks are initiated at the interface between $CuAl_2$ IMC layer and Cu layer and grow toward the middle during the heating process[29]. Cavities and voids can be observed in Solchenbach et al's research due to inappropriate heat input [9]. Voids appear in the IMC after electrical aging for 24 h [26]. According to Wang et al's study, the Kirkendall voids form in the IMCs after induction-brazed at 560 °C, and then discontinuous fracture appears with increasing the amount of voids during heat treatment [21, 22].

For the generation of the cracks, Xia et al thought that the stress concentration is always initiated on the brittle IMCs. The IMCs have different thermal expansion coefficient compared with their adjacent phases. The greater the temperature change, the severer the stress concentration generated on the brittle IMCs. And then, the cracks form [18]. The Solchenbach et al's study also came to the same conclusion [26].

Atomic diffusion process is crucial to the formation of the voids and cavities. Experimental results indicated that low heat input can cause the formation of the voids because of incomplete diffusion joining, and high heat input can result in the generation of the voids and cavities due to excessive

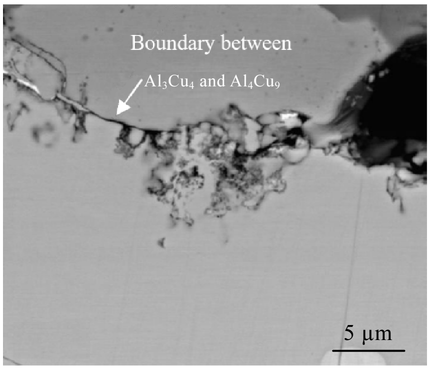

Fig.3 SEM image of cracks in the Cu-Al joint [26]

diffusion [7-9, 21]. Both the formation and transformation of IMCs are beneficial for the generation of the voids and cavities [21, 30]. Because it can promote the atomic diffusion process, and thereby increase the consumption of the diffused atoms. High consumption of diffused atoms results in the voids and cavities.

From the above sections, the formation of cracks needs brittle phase as a source of cracks, which can result in stress concentration. Besides, temperature and temperature change rate provide stress to generate cracks. Voids and cavities appear through atomic diffusion. IMCs accelerate the formation of voids and cavities.

3 Relationship Between Properties of Cu-Al Joints and IMCs

In the above sections, we have mentioned that the formation and growth of the intermetallic compounds are hazardous to the properties of Cu-Al joints. Thus, it is of great necessities to investigate the relationship between properties of the joint and the IMCs.

A notable increasing of the hardness happens in the Cu-Al joint due to the IMCs. It can be seen that the microhardness is 5150 MPa near the Cu side (consisting mainly of $CuAl_2$ phase) while the microhardness reaches more than 1000 MPa in the middle in the brazing seam (consisting of $CuAl_2$ and (Al, Zn)-rich phase) [1, 18]. From Xia et al's experiment, it can be obtained that the transition area between Cu side and the filler metal consists of a Cu_3Al_2 layer (7200 MPa) and $CuAl_2$ layer (5100 MPa) [18, 36].

The above studies show that the IMCs can make the microhardness much higher than that of base metal (below 500 MPa). Meanwhile, the IMCs will make a significant decline in the properties of shear strength, tensile strength and toughness when the thickness of the IMCs exceeds 2~5 μm. For instance, experiment of Solchenbach et al indicated that the IMCs make the shear strength reduce[8]. Pascal et al's investigation showed that the existence of the IMCs decreases the toughness [30]. According to Wang et al's study, the tensile strength is reduced to about 22 MPa [21, 22]. However, some studies show that the existence of the IMC can modify the strength. Ji Feng et al found that a homogeneous distribution of IMCs may increase the shear strength [1], and K. Shinozaki and K. Koyama also proved it. But it still decreases due to the further growth of the IMCs [37].

Moreover, several researchers also suggested that the connection resistance of the copper-aluminum brazed joints increases when the IMCs exceed 2~5 μm. Solchenbach et al's examination indicated that the resistance significantly increases due to the thick IMCs layer [9]. Thereafter, according to Pascal et al's research, the resistance of laser-brazed joints significantly increases due to the IMCs, but is lower than that of the unwelded joint [30].

From these results mentioned above, the IMCs formation and growth significantly damage the comprehensive properties of the Cu-Al joints.

4 Factors Impacting the Formation and Growth of IMCs

The generation of massive IMCs is the primary cause influencing the properties of the Cu-Al joints. The comprehensive properties of the Cu-Al joints deteriorate when the thickness of the IMCs layer exceeds 2~5 μm [8]. Besides, the IMCs are critical for the formation of defects. Factors, influencing the IMCs, generally have an obvious effect on the defects. Hence it is significant to study the factors influencing the formation and growth of the IMCs. Then those factors will be made a good use to control the IMCs formation and growth.

Some studies suggested that an appropriate use of filler metals is beneficial to the properties of the Cu-Al joints. For instance, Feng et al's study showed that brazing with Zn-Al filler metals can reduce the growth rate of the IMCs and thereby enhance the comprehensive properties of the Cu-Al joints [1]. The reduction of the IMCs occurs because filler metals severely react with Cu side. Furthermore, Ji Feng et al examined the effects of Zn-22Al-*x*Ti filler metals on the growth of the IMCs layer in the Cu-Al joints, and found that adding a little element Ti into Zn-22Al filler metals can effectively reduce the thickness of the IMCs layer since the generation of the $TiAl_3$ particles hinders the diffusion of Cu, Al and Zn, which in turn restricts the growth of the IMCs [38]. Finally, Ji Feng et al substituted Zn-Al-*x*Ce for Zn-Al filler metals, and as a result, it can come to the conclusion that the addition of a trace amount of element Ge decreases the thickness of IMC layer due to the formation of the Cu-Ge IMCs, which inhibits the reaction of the Cu and Al [2]. According to Pstruś et al's experiment, obvious restriction of the IMCs growth occurs when brazing with Zn-Al filler metal containing element Cu or element Ag and carrying high pressure on joints [39]. Furthermore, the quality of filler metals is very important. Several studies show that the microstructure and properties of filler metals can be distinctly changed by heat treatment, the third element addition and hot and humid environment[40-42], which may impact copper-aluminum joints. Long et al successfully improved the strength of stainless steel joint by a new filler metal, that is, in-situ-synthesis brazing alloy [43] . The above research implies that filler metals play an important role in the IMCs formation and growth.

Heat input is considered a vital factor influencing the IMCs. For instance, Solchenbach et al investigated the correlation between heat input and the thickness of the IMCs and found that the IMCs rapidly grow due to greater heat input [7] . High heat input can result in an increasing growth of the IMCs because greater amounts of Cu are melted [7, 8] . The same phenomenon was observed in Solchenbach et al' study [9] . Recently, it can be seen in Wang et al's study that great heat input results in a great growth of the IMCs, thereby increasing the resistance [21, 22] . However, Solchenbach et al successfully controlled the heat input by a new laser brazing technology with power modulation system and decreased the thickness of the IMCs layer to 3.2 μm [8] . Thereafter, Pascal et al got the similar result when using this new brazing technology [30] . Zhou et al investigated the influence of laser brazing speed and laser spot offset on the Cu-Al joints. Results indicated that perfect and defect-free joints can be produced by the brazing speed of 0.4~0.6 m/min and laser spot offset of −0.6~0 mm due to proper heat input [34, 35] . Hence the heat input has a significant influence on the IMCs and we can use it to control the IMCs.

According to Solchenbach et al's study, both small amplitudes of circular beam movement and great overlaps between two irradiated cycles can cause a significant growth of the IMCs, resulting in a reduction of shear strength[8] . Besides, the number of brazing seams and their distance strongly impact the connection resistance of the joint because of the formation of the current vortex, which accelerates the growth of the IMCs and reduces the service life of the Cu-Al joints. Then they employed two seams with a large distance to avoid the appearance of the current vertex and thereby significantly minimize the thickness of the IMCs[9] . The above research shows that joint design is important to decrease the growth of the IMCs. Besides, Solchenbach et al thought that the polarity of the electric current is significant for the effect of electrical aging on the growth of the IMCs layer in the joint. Because the growth of the IMCs will be strongly improved while the force direction resulted from chemical potential is in coincidence with electric current (the same force direction enhanced the atomic diffusion rate)[26] .

The above studies are in great coincidence with the results of Pual et al who gave a detailed discussion for joining processes such as friction welding, laser welding and cold roll welding [44] . Numerous factors have critical influences on the formation and growth of the IMCs during brazing, such as physical characteristics (melting point, thermal conductivity), brazing parameters (joint design, heat input) and the addition of the third element. In addition, these factors affect the mechanical properties, microstructures and resistance to corrosion and oxidation of the Cu-Al joints, which thereby impact the service properties. So, these factors should be taken into account when brazing copper to aluminum to make a reliable and stable Cu-Al joint.

5 Atomic Diffusion Mechanism of Formation and Growth of IMCs and Defects

For the formation of the IMCs and defects, the high affinity and unequal mutual diffusion between Cu and Al ($D_{\text{Cu-Al}}$=9.2×10^{-21} m^2/s, $D_{\text{Al-Cu}}$=3.4×10^{-21} m^2/s, T=110 °C) are the main reasons[2, 32, 45] . The formation and growth of the IMCs occur mainly through atomic diffusion between base metal and base metal as well as base metals and filler metals [29] . Hence it can come to the conclusion that atomic diffusion is crucial to the formation and growth of IMCs and defects. The diffusion coefficient is the vital parameter of the atomic diffusion process. The diffusion coefficient D can be described as[46]:

$$D=D_0\exp(-Q/RT) \quad (1)$$

$$D_0=1/6fa^2vz\, c_{\text{v}}^{\text{imp}} \exp(\Delta S_{\text{m}}/R) \quad (2)$$

where D_0 is diffusion coefficient; T is Kelvin temperature; f is correlation coefficients (f_{fcc}=f_{hcp}=0.78, f_{bcc}=0.72); v is the atomic vibration frequency; R is the Boltzmann constant; z is number of near positions of the diffusion atom; a is interplanar crystal spacing; $c_{\text{v}}^{\text{imp}}$ is extrinsic vacancy concentration; Q is diffusion activation energy; ΔS_{m} is diffusion activation entropy.

Eq.(1) and Eq.(2) indicate that the diffusion coefficient depends on six parameters: T, f, α, v, z, $c_{\text{v}}^{\text{imp}}$, which in turn influence the IMCs and defects.

The formation of IMCs is beneficial for promoting the atomic diffusion process, thereby improving the formation of defects. The flux of diffused atoms is[26]

$$J=-D\mathrm{d}c/\mathrm{d}x \quad (3)$$

where dc/dx is atomic concentration gradient along x direction; D is diffusion coefficient.

The formation of IMCs consumes a large quantity of diffused atoms, increasing the atomic concentration gradient (dc/dx). The high atomic concentration gradient in turn promotes the diffusion process and the consumption of atoms, which benefits the formation of the defect. Hence the atomic diffusion and the IMCs formation and growth can promote each other, and defects form during this process.

An appropriate introduction of filler metals has a beneficial effect on decreasing the extrinsic vacancy concentration ($c_{\text{v}}^{\text{imp}}$), effectively restraining the diffusion process and the formation and growth of the IMCs. Then it is not easy to form defects. However, cracks will be formed in large quantities when brazing with unbefitting filler metals. For instance, Xia et al's experiment indicated that Al-Si filler metal can result in many cracks because Si primary (12 000 MPa) crystal replaces the IMCs as the new source of cracks [18]. High heat input can accelerate the diffusion process by changing the temperature (*T*). Meanwhile, the IMCs form and grow, which increases the probability of generation of defects. The brazing seam design has the same effects on the diffusion too. The joint design and electrical current have considerable impact on the number of near positions of the diffusion atom (*z*), thereby changing the diffusion process, the IMCs and defects.

6 Conclusions

Cu and Al easily react to form the IMCs during brazing and application due to the high affinity between them, resulting in additional processes and costs. Thermodynamics and kinetics are critical for the nucleation and growth of the IMCs, respectively. Besides, defects (voids, cavities and cracks) in the joint mainly result from the formation and growth of the IMCs. The properties of copper-aluminum joints deteriorate when the thickness of the IMCs exceeds 2~5 μm. The formation and growth of the IMCs depend on the mutual diffusion between Cu substrate and Al substrate as well as substrates and filler metals. Numerous factors (melting point, thermal conductivity, joint design, heat input and chemical composition) strongly impact the formation and growth of the IMCs through changing the mutual diffusion process. Besides, these factors also have distinct effects on the defects. At present, some efficient methods used to control the IMCs in the copper-aluminum joints are heat input controlling, excellent joint design and the addition of the third element into the filler metal.

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铜/铝异质钎焊连接界面金属间化合物的研究进展

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摘　要：对近年来钎焊铜铝异种材料的相关研究进行回顾，分析了在接头形成与服役时，焊缝中金属间化合物（IMCs）的形成与生长。结果表明，金属间化合物的形成与生长在接头形成与服役过程中是不可避免的。金属间化合物的形成和生长取决于铜铝之间以及与钎料之间的原子相互扩散。金属间化合物的形核和生长必须同时满足热力学与动力学条件。脆硬性金属间化合物容易引起应力集中，且其形成与生长会加剧扩散原子的消耗，因此金属间化合物的形成与生长是导致接头缺陷（如孔洞、空洞和裂纹）的主要原因之一。当界面处金属间化合物层的厚度超过 2~5 μm 时，接头性能会急剧下降。影响金属间化合物生长与扩散和接头缺陷的主要因素有温度，导热性，接头设计，热输入和钎料成分等。以上因素主要通过改变原子扩散过程影响金属间化合物的形成与生长。目前，控制金属间化合物形成与生长的主要方法有控制接头热输入、优化接头设计和在钎料中添加第三元素等。

关键词：金属间化合物；铜；铝；异质材料钎焊；影响因素；原子扩散

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