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Microstructure evolution, grain morphology variation and mechanical property change of Cu-Sn intermetallic joints subjected to high-temperature aging

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A B S T R A C T

In this work, the aging treatment was carried out on full Cu_3Sn joints at different temperatures for various durations, the microstructure evolution, grain morphology variation and mechanical property change of joints were investigated. The temperature effects on the growth rate and grain morphology of $Cu_{41}Sn_{11}$ phase were found: The complete transformation from Cu_3Sn phase to $Cu_{41}Sn_{11}$ phase in joints needed 300 h at 400 °C, but 4 h at 500 °C. The $Cu_{41}Sn_{11}$ phase formed at 400 °C consisted of columnar grains with an average width of 23.5 μm, while the $Cu_{41}Sn_{11}$ phase formed at 500 °C consisted columnar grains with an average width of 8.5 μm. The average hardness and shear strength values of the $Cu_{41}Sn_{11}$ joints formed at 400 °C were 432.5 HV and 62 MPa, while the joints formed at 500 °C exhibited higher hardness and shear strength (451.5 HV and 65 MPa). Besides, the $Cu_{41}Sn_{11}$ joint was found to exhibit a combination of transgranular and intergranular fracture during shear test.

1. Introduction

Nowadays, some wide band-gap semiconductor devices, such as silicon carbide (SiC) and gallium nitride (GaN) power devices, can demonstrate excellent performance, such as wide band gap width, high saturated electron drift velocity, large critical electric breakdown strength, and high thermal conductivity; in addition, they are stable at temperatures up to 600 °C [1–6]. Therefore, SiC and GaN devices could be widely used in many electronic applications with high operating temperatures, such as those in the down-hole oil and gas industry and in the aircraft, automotive, and space exploration fields [7,8]. For example, the temperature requirement will reach up to 873 K (600 °C) for the deepest geothermal wells for the existing oil or gas wells [9]; the inverters, which control the flow of electric current in the vehicle, consist of power modules with an array of power devices, that will operate at temperature ~873 K (600 °C) [10]; the survey of Venus will make the devices exposed to high temperature up to 480 °C [11]. Therefore, the development of packaging methods that can form high-melting-point joints is extremely crucial to the application of wide band gap semiconductors in power devices.

In recent years, the transient liquid phase (TLP) soldering has been reported as a promising bonding method for producing reliable high-melting-point joints with applications to the power electronics industry operated at elevated temperatures. Cu-Sn TLP method was most widely studied due to the widespread use of Cu in high temperature electronic products, especially on ceramic substrates, and its potential as a device metallization layer [12–15]. A typical TLP soldering process is performed by extending the bonding time until the liquid phase of Sn interlayer is fully consumed and transformed to Cu-Sn intermetallic compounds (IMCs), including Cu_6Sn_5 and Cu_3Sn (melting point: 415 °C and 676 °C) [16–22].

Indeed, the IMCs morphologies, microstructure evolution, and grain orientation distributions of full IMCs joints during different reflow durations have been well investigated [23], many studies also have focused on the reliability of TLP-bonded joints and shown that Cu-Sn system can create more robust bonds with excellent mechanical properties and higher thermal reliabilities than the traditional soldered joints with Sn-based solders [24]. However, all above studies were mainly carried out based on the TLP-bonded joints obtained at process temperatures of no more than 340 °C (e.g., 250, 300 and 340 °C, etc.). Besides, extensive research has been done to study the changes of microstructure and mechanical properties of the Sn-based joints during

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aging or the service period [25]. However, there were limited investigations establishing on the high-temperature (more than 400 °C) reliability, such as microstructure evolution and mechanical property change, of the formed full IMCs joints during aging or the service period, which are crucial for the high temperature performance of the TLP-bonded joint. In this work, the aging treatment was carried out on the formed full Cu_3Sn joints at different temperatures (400 °C and 500 °C) and various durations (1.5 h to 300 h). The microstructural evolution with phase transformation from Cu_3Sn into $Cu_{41}Sn_{11}$ was observed, and the mechanical properties of the full $Cu_{41}Sn_{11}$ joint were also investigated.

2. Experimental Procedures

A sandwich structure of the Cu/Sn/Cu system was utilized in this study to fabricate full Cu_3Sn IMCs joints. The solder interlayer is made by a single pure Sn foil with a thickness of 50 μm. The parent metal substrates are made of two Cu plates with a thickness of 0.5 mm. The Sn foil and the Cu plates were cut into the dimension of 5 mm × 5 mm. After alignment, the sandwich structure was bonded under 300 °C with the reflow time of 36 h. The bonding force applied on these samples during soldering process was 1 N. These conditions produced a uniform interconnect layer of the ε-Cu_3Sn phase in the joints. Following, the formed joint samples were subject to further aging treatment under different temperatures and various durations. In one set, the joint samples were heated at 400 °C for 100, 200 and 300 h. Another set of joint samples was heated at 500 °C for 1, 1.5, 3 and 4 h.

For microstructural observation, the joint samples were mounted in epoxy resin and then manually ground by 600, 800, 1200, 2500 and 3000 grade silicon carbide papers and metallographically polished by 2.5, 1 and 0.05 μm polishing agents. The cross-sectional microstructures and consisting intermetallic phases of the joints were characterized by optical microscopy (OM), X-Ray diffraction (XRD) and transmission electron microscopy (TEM). Componential analysis of intermetallic phases in joints was carried out using an energy dispersive spectrometer (EDS) with the operation voltage of 15 kV and minimum spot size of 1 μm. The specimen for TEM investigation was extracted from the center of joint samples. The Cu substrate was first removed by mechanical grind using 800 grade silicon carbide papers, the extracted specimen was then cut to 3 mm in diameter using an ultrasonic disk cutter, further ground using a dimple grinder and accomplished by ion milling using a precision ion polishing system. The grain morphology of the IMCs was observed by electron backscattering diffraction (EBSD). EBSD testing was carried out at the center of the samples with the accelerating voltage of 20 kV, working distance of 15 mm and step size of 0.2 μm. The mechanical properties of IMCs joints were evaluated by micro-hardness and micro-shear testing. Vickers micro-hardness tests were carried out on the cross-sections of the joints under a 50 g load and with a holding time of 10 s. The micro-shear test was carried out at a constant speed of 100 μm/s at room temperature, and the schematic diagram of the micro-shear test is demonstrated in Fig. 8a. After shear testing, the fracture surfaces were investigated via secondary electron imaging mode in scanning electron microscope (SEM) with the accelerating voltage of 20 kV and working distance of 15 mm.

3. Results and Discussion

3.1. Characteristics of the Produced Full Cu_3Sn Joint Samples

Fig.1a presents the cross-sectional OM image of Cu/Sn/Cu sample bonded at 300 °C for 36 h. EDS analysis revealed that a uniform interconnect layer of the Cu_3Sn (Cu: 77.57 at% and Sn: 22.43 at%) phase, without any other phases, in the joint was produced. Besides, based on the X-ray diffraction analysis (Fig. 4a), the super-lattice ε-Cu_3Sn phase with an orthorhombic structure (a = 0.5529(8) nm, b = 4.775(6) nm and c = 0.4323(2) nm) [26] was confirmed. Fig. 1b shows the grain morphology of Cu_3Sn phase. Different grains presented different gray scales with clear grain boundaries between them. The main features of Cu_3Sn grains presented columnar crystals with the width of 1–4 μm, growing in units of clusters along directions perpendicular to Cu substrates. As Cu_3Sn grains grew from opposite sides, when they contacted each other, the grain growth stopped, the Cu_3Sn boundary lines formed in the center of the joint. It can also be found that a lot of Cu_3Sn grains exhibited fine equiaxed crystals and appeared at the junction sites between columnar Cu_3Sn grains and Cu substrates, and around the Cu_3Sn boundary lines in the center of the joint. This was because Sn could diffuse from Cu_3Sn layers towards Cu/Cu_3Sn interfaces through the Cu_3Sn grain boundaries, which could provide the channels for the Sn diffusion. When diffused Sn contacted Cu from substrate, new Cu_3Sn grains nucleation formed at the junction sites by reaction of $Sn + 3Cu \rightarrow Cu_3Sn$. In addition, when Cu_3Sn grains from the opposite sides just contacted, remaining Cu_6Sn_5 spread residual gaps in the contact areas. Cu atoms from substrates diffusing through Cu_3Sn layers reacted with the remaining Cu_6Sn_5, forming new Cu_3Sn grains by reaction of $Cu_6Sn_5 + 9Cu \rightarrow 5Cu_3Sn$. However, these Cu_3Sn grains could not grow up freely as a result of the constraints of surrounding columnar Cu_3Sn grains, so these Cu_3Sn grains showed equiaxed in shape [27]. Moreover, the Kirkendall voids were found in Cu_3Sn layer.

3.2. Microstructure Evolution and Grain Morphology Variation of the Full Cu_3Sn Joints Subjected to 400 °C

Fig. 2a–c show the cross-sectional OM images of full Cu_3Sn joints with additional treatment at 400 °C for various durations. After being aged for 100 h, new phases with scallop shape were formed on the original Cu_3Sn/Cu interfaces. These new phases exhibited a discontinuous and random distribution on the interfaces (Fig. 2a). After the treatment time was increased to 200 h, these new phases grew up obviously and contacted with each other, forming continuous phase layers on both interfacial sides. Beyond that, the thickness of Cu_3Sn layer was decreased with the increase of treatment time (Fig. 2b).With further treatment for 300 h, the whole joint was occupied by the single new phase with the Cu_3Sn phase disappeared (Fig. 2c). In addition, some voids were found near the new phase/Cu interfaces after a period of treatment time, and the number of voids increased visibly with the increase of the amount of new phases (Fig. 2b–c). In order to accurately determine above new phases, the EDS, XRD and TEM analysis were carried out on them. The major elements of different points (Fig. 2b) detected by EDS are listed in Table 1. According to the EDS analysis results and the Cu-Sn binary phase diagram [28] (illustrated in Fig. 3), the new phase formed on the Cu_3Sn/Cu interfaces was confirmed to be δ-$Cu_{41}Sn_{11}$. Besides, according to the X-ray diffraction analysis (Fig. 4b), the δ-$Cu_{41}Sn_{11}$ phase with cubic structure (a = 17.98 nm) [29] was confirmed. Fig. 5 shows the selected area electron diffraction patterns of $Cu_{41}Sn_{11}$ phase from a TEM sample extracted from the center of joint in Fig. 2c. Based on the phase transformation, as illustrated in Fig. 2a–c, and the Cu-Sn binary phase diagram, the δ-$Cu_{41}Sn_{11}$ was formed by the reaction of $11Cu_3Sn + 9Cu \rightarrow Cu_{41}Sn_{11}$ during aging above 350 °C, with consuming the Cu_3Sn phase and the Cu from substrate [30]. When a certain amount of Cu diffused from substrates into Cu_3Sn layers, the voids were subsequently formed in Cu substrates. Accordingly, the process of aging treatment above 350 °C till the full $Cu_{41}Sn_{11}$ joints formed can be described as follows: when the full Cu_3Sn joint is aged for a period of time, the δ-$Cu_{41}Sn_{11}$ is formed by the reaction of $11Cu_3Sn + 9Cu \rightarrow Cu_{41}Sn_{11}$. With further reaction, $Cu_{41}Sn_{11}$ phase grows up and contacts with each other, forming a continuous layer. The thickness of $Cu_{41}Sn_{11}$ layer increases with the increase of treatment time, but with the decrease of the thickness of Cu_3Sn layer. For longer treatment time, the $Cu_{41}Sn_{11}$ layer occupies the whole joint cross-section with the remaining Cu_3Sn layer completely consumed. According to the Cu-Sn phase diagram, δ-$Cu_{41}Sn_{11}$ is a thermodynamically stable phase over the temperature range of 350 to