Microstructure evolution and mechanical properties of Cu/Sn/Ag TLP-bonded joint during thermal aging

Huakai Shao\textsuperscript{a},* , Aiping Wu\textsuperscript{a,b,c} , Yudian Bao\textsuperscript{a} , Yue Zhao\textsuperscript{a,c} , Guisheng Zou\textsuperscript{a,c} , Lei Liu\textsuperscript{a,b}

\textsuperscript{a} Department of Mechanical Engineering, Tsinghua University, Beijing 100084, China
\textsuperscript{b} State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China
\textsuperscript{c} Key Laboratory for Advanced Materials Processing Technology, Ministry of Education, Beijing 100084, China

\textbf{ABSTRACT}

Low-temperature transient liquid phase (TLP) bonding is a promising joining technology for high-temperature electronic devices packaging. However, the effect of thermal aging on the joint microstructure and performance has been rarely studied so far. In this paper, the relationship between microstructural evolution and mechanical property was developed for the Cu/Sn/Ag TLP-bonded joint during 350 °C aging. Experimental results show that an interfacial reaction occurs between the intermetallic compounds (IMCs) of Cu\textsubscript{3}Sn and Ag\textsubscript{3}Sn, which induces that a great amount of Cu\textsubscript{4}Sn\textsubscript{11} phase or wedges embed into the Ag-Sn phase layer. The reaction process is controlled by a volume diffusion of Cu elements from the Cu substrate, and capable of promoting the transformation from Ag\textsubscript{3}Sn into Ag(Sn) solid solution. Cu\textsubscript{4}Sn\textsubscript{11} phase can be formed at the expense of Cu\textsubscript{3}Sn and Cu, of which transformation process is completely achieved after aging for 480 h, and the Cu(Sn) solid solution intimately contacts with the opposite Ag(Sn) solid solution in many areas after aging for 960 h. The joint mechanical property initially declines due to the formation of Cu\textsubscript{4}Sn\textsubscript{11} phase and Cu-Sn IMCs grain coarsening, and then increases because of the complete depletion of Cu\textsubscript{3}Sn and substantial precipitation of Cu(Sn) solid solution. After aging for nearly 1000 h, the joint still maintains a high shear strength of > 40 MPa, indicating an excellent thermal reliability.

\section{1. Introduction}

Nowadays, the development of high-temperature semiconductor devices is becoming increasingly important in electronics industry, which can be attributed to the trend of electronic devices being towards high power, miniaturization and high integration, and the demands of harsh environments in new automotive, aerospace, deep oil and gas drilling and energy industries, such as high temperature, large voltage, moisture and corrosion. Therefore, the huge thermal effect is very crucial for the long-term reliability of packages. The interconnection between the chip and the ceramic substrate provides a path for both electrical and thermal conduction in the power module, but the conventional solder joints cannot meet the requirements for high-temperature applications. In recent years, many researches focus on the exploration of new packaging techniques and reliable Pb-free high-temperature solders \cite{1–4}, where low-temperature transient liquid phase (TLP) bonding has been confirmed as a potential candidate.

In the TLP bonding process, a full intermetallic compounds (IMCs) joint can be obtained at low temperature through the diffusion reaction between high-melting substrate (e.g. Cu, Ag) and low-melting interlayer (e.g. Sn, In). Since the IMCs have much higher remelting temperature than the initial solder, the TLP-bonded joint generally possesses an excellent heat-resistant property. Presently, many systems have been proposed for high-temperature packaging, such as Cu-Sn and Ag-Sn systems \cite{5–7}, and the studies focus mainly on the growth behavior of IMCs \cite{8,9}, joint microstructure evolution \cite{5,10}, voids formation and elimination \cite{11,12}, ductility improvement of IMC layer \cite{7,13}, and shortening of bonding time \cite{6,14,15}. However, it has been rarely reported on the joint thermal reliability, which is very crucial for power electronics applications. Thermal aging and thermal cycling are two typical methods for evaluating reliability and lifetime of interconnections \cite{16–18}. In the available publications, the relationship between microstructural evolution and mechanical property has not been established for different systems during thermal aging. For example, Au-Sn TLP-bonded joint has been confirmed to exhibit superior high-temperature performance, its shear strength is still remarkably high (70 MPa) after aging at 250 °C for 3 months \cite{19}, but the detailed microstructural evolution and mechanical property variation were not...
as shown in Fig. 2, and the Cu3Sn/Ag3Sn interface presents as a planar and Ag3Sn was investigated deeply.

For various time durations, and the interfacial reaction between Cu3Sn and aging were not discussed. In the present work, the Cu/Sn/Ag TLP-bonded joint was subjected to high-temperature storage test at 350 °C aging test in air, where the storage temperature is 350 °C, and the aging study was performed with shear rate of 2 mm/min and height of 1 mm to measure the shear strength. At least five specimens were employed to determine average shear strength under one condition. Some typical fractures were analyzed by using the SEM and EDS technologies to determine the joint fracture behaviors.

2. Experimental Procedures

Fig. 1 shows the specific information (shape and dimension) of the substrates for microstructure characterization and shear test, in which the Ag metallization layer is prepared by means of electroplating process. In detail, pure Cu sheets or rods were coated with 2 μm-thick Ni layer followed by 25 μm-thick Ag layer. Prior to bonding process, the Ag-plated and Cu substrates were ultrasonically cleaned in 5% HNO3 solution for 10 min, subsequently rinsed with deionized water and anhydrous alcohol, and finally dried. A commercially pure Sn foil with 10 μm thickness was adopted as the solder interlayer. After cleaned in acetone, the pure Sn foil was sandwiched between the two substrates that were coated with a small amount of rosin flux.

The joining experiment was conducted at a temperature of 300 °C for 420 min in air, in which the heating rate is about 10–15 °C/min. During bonding process, a constant pressure of 0.3 MPa was applied to intimately contact the faying surfaces. The microstructure of the as-bonded joint is composed mainly of Cu3Sn, Ag3Sn, and ζ-phase (Ag4Sn), as shown in Fig. 2, and the Cu3Sn/Ag3Sn interface presents as a planar morphology due to interfacial migration [11]. In order to evaluate the thermal reliability, the as-bonded samples were subjected to thermal aging test in air, where the storage temperature is 350 °C, and the aging time is varied from 24 to 960 h.

The aged samples were mounted in epoxy resin, and then ground by 240, 600, 1000, 1500 and 2000 grade silicon carbide papers, subsequently polished by using diamond polishing agents. The microstructures were characterized by using scanning electron microscopy (SEM) equipped with energy dispersive X-ray spectroscopy (EDS), and electro-probe micro-analyzer (EPMA). In addition, shear test was performed with shear rate of 2 mm/min and height of 1 mm to measure the shear strength. At least five specimens were employed to determine average shear strength under one condition. Some typical fractures were analyzed by using the SEM and EDS technologies to determine the joint fracture behaviors.

3. Results and Discussion

3.1. Microstructural Evolution

Fig. 3 shows the cross-sectional microstructures and elemental linear distributions of the Cu3Sn/Ag3Sn TLP-bonded joints aged at 350 °C for various time durations. After aging for 24 h (Fig. 3a), Ag3Sn compounds transformed totally into Ag(Sn) solid solution, identified by the EDS analysis (Table 1: point A1), and the Cu-Sn IMCs layer contained not only Cu3Sn but also Cu41Sn11 according to the Sn content in the Table 1 (point A2), which is lower than that of Cu3Sn phase. The formation mechanism of the Cu41Sn11 phase will be discussed according to the joint fracture surfaces. In fact, the Sn linear distribution of the Ag-Sn phase did not change during thermal aging from 24 h to 960 h, which demonstrates that no Ag-Sn IMCs remained in the joint. More importantly, the interface between Ag(Sn) solid solution and Cu-Sn IMCs became much rougher in comparison to the initially planar Cu3Sn/Ag3Sn interface (Fig. 2), and a great amount of Cu3Sn particles or wedges embedded into the Ag(Sn) solid solution, indicating that an interfacial reaction occurred between the Ag-Sn and Cu3Sn layers. Adjacent to the Cu/Ag(Sn) interface, it was observed that many Ni-Sn intermetallic particles were formed within the Ag(Sn) solid solution (yellow arrowed), and their distributions were very similar to those in the Ag/Sn/TLP-bonded joint during thermal aging [6].

After aging for 120 h (Fig. 3b), there was no significant change for the microstructure on the side of Ag-Sn phase, but the Cu3Sn particles within the Ag(Sn) solid solution became smaller compared with the 24 h-aged joint, in addition, a thin layer of Cu3Sn solid solution was precipitated between the Cu substrate and Cu-Sn IMCs layer, in consistent with the Cu and Sn linear distributions. After aging for 480 h (Fig. 3c), the Cu(Sn) solid solution continued to grow at the expense of Cu and Cu-Sn IMCs, and the residual Cu3Sn converted entirely into Cu41Sn11 in terms of the decreased Sn content in the elemental linear distributions in comparison to the previous joints. Meanwhile, the interface between Ag(Sn) solid solution and Cu41Sn11 became planar again, similar to the initial Cu3Sn/Ag3Sn interface (Fig. 2), and a few larger Cu3Sn particles were formed within the Ag(Sn) solid solution.