Solid-liquid Diffusion Interconnection Method Based on Cu-In Secondary Micro Nano Layer

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Abstract. A solid-liquid diffusion interconnection method based on Cu-In secondary micro nano layer is proposed. The substrate surfaces of two copper indium secondary micro nano layers with special morphology are contacted with each other, and the contact area is heated to carry out solid-liquid interconnection. The special morphology of copper indium micro nano layer includes copper needle layer and indium micro layer coated on it. During interconnection, liquid indium infiltrates copper needle layer and forms intermetallic compound with good plasticity. The microstructure, intermetallic compounds and shear strength of the interface were analyzed by scanning electron microscope (SEM), transmission electron microscope (TEM) and welding strength tester. The results show that this solid-liquid interconnection method can obtain good interconnection quality and it is of great significance to green packaging.

1. Introduction

The high requirements of electronic products put forward new requirements for high-density interconnection technology. Three dimensional stacking technology is the development direction of high-density interconnection technology, but with the increase of the number of layers, the solder joints become smaller and thinner, and the global interconnection delay increases[1, 2]. Stack interconnection technology has good electrical performance, internal interconnection and high mechanical reliability, which has become an important development trend of 3D interconnection technology[3, 4]. In general, direct copper copper hot pressing interconnection technology requires high temperature and high surface treatment technology such as polishing. This direct copper copper interconnection process requires high bonding pressure, high clean surface, high temperature of 700-1000 °C, vacuum environment, and harsh process conditions limit this technology. Therefore, it is necessary to study a low temperature interconnection technology to reduce process complexity and cost. Zhang et al. used copper cone micro nano layer to increase the interfacial adhesion between copper substrate and epoxy resin [5]. Chen et al. studied the low-temperature interconnection technology between nickel needle micro nano layer and tin solder [6, 7]. Naoya Watanabe et al. reported that copper copper interconnects can be realized by changing the morphology of the two sides of the interconnection couple and complementing the morphology of the two sides[8]. Wang et al. realized the ultrasonic interconnection technology at room temperature by using nickel micron needle powder. However, the interconnection quality and practical application in these studies are not ideal [9].

Copper has a high affinity with oxygen, and it is easy to oxidize under the action of high temperature in the packaging process. In this paper, the copper needle layer and the indium micro layer coated on it were obtained by electrochemical method. After the indium micro layer is coated,
the shape of copper needle layer remains unchanged. The indium micro layer can effectively prevent the copper layer from oxidation. The copper indium micro nano layer has a huge surface area. At the interconnection temperature of 260 °C, indium becomes liquid, and the liquid indium can better infiltrate the copper needle layer. The intermetallic compound Cu2In formed by copper indium is a high-quality phase with good plasticity and greatly improves the interconnection strength.

2. Experiment
Firstly, the copper indium secondary micro nano layer is obtained by electroless plating method; secondly, the surfaces of two substrates (specification: 2cm * 2cm * 0.15mm) with copper indium secondary micro nano layer are contacted with each other to form a contact area; finally, the contact area is heated in air for solid-liquid diffusion interconnection.

The bonding tester with heating plate (Rhesca company, model PTR-1101) is used for interconnection operation, which is as follows: one copper substrate with copper indium secondary micro nano layer is placed on the heating plate, and the other copper substrate with copper indium micro nano layer is placed on the heating plate, so that the copper indium micro nano layers of the two copper substrates are in face-to-face contact, and the interconnection temperature is set at 260 °C. The heating time was 180 min and the heating rate was 10 °C / min. The interconnection process diagram is shown in Figure 1. The whole interconnection process is completed in air. After cooling, the interconnection was measured by push ball test mode.

3. Results and discussion
3.1. Microstructure of Cu-In secondary micro nano array
Copper indium secondary micro nano layer was prepared by electrodeposition on copper substrate and used for subsequent solid-liquid diffusion interconnection research. The copper indium micro nano layer has a secondary structure, the first stage is copper needle layer, and the second stage is indium micron layer, in which the indium micron layer is plated on the surface of copper needle layer; Fig. 2 (a, b) shows the scanning electron microscope images of copper needle micro nano layer with different magnification, the height of copper microneedle is 1-3 μm, the diameter of needle root is 500nm-2 μm, and the copper deposition layer is small, dense and uniform, showing a typical conical array; Fig. 2 (c, d) is the SEM images of different magnification of Cu-In secondary micro nano layer. The thickness of indium micro nano layer is 150 nm-350 nm. With the deposition of indium, the tip of copper needle layer becomes round, but the copper indium secondary micro nano layer still maintains the needle like structure of copper micro nano layer. This array structure has a large surface area and is a typical hydrophobic micro nano structure. On the other hand, the size of the Cu in secondary micro
nano layer is controllable, and the content and type of crystal modifier can be changed during the preparation, and different sizes of micro nano layer can be obtained.

3.2. Microstructure of bonding interface
In order to further observe the insertion state of copper pin and the diffusion process of copper indium in the process of interconnection, field emission scanning electron microscopy (FESEM) was used to characterize (SU8220, Hitachi, The microstructure of Cu-In-Cu interface under different interconnection conditions is shown in Fig. 3. It is found that the protruding copper nanoneedle is firmly connected with the indium layer after interconnection. When the interconnection temperature is 260 °C and the interconnection time is 10 min, there are some hole defects along the interconnection interface, and the microstructure distribution is uneven, and there are many kinds of intermetallic compounds. When the interconnection time is increased to 180 min, there are no holes in the interconnection interface, and the microstructure distribution is relatively uniform, which should be a single intermetallic compound. It can be predicted that when the interconnection time is increased to 180 min, a good interconnection interface can be obtained because on the one hand, the needle-like structure can obtain sufficient mechanical interlock. On the other hand, with the increase of interconnection time, there is enough time for the reaction between copper and indium to obtain a more uniform single intermetallic compound.

3.3. Bonding mechanism
Fig. 4 is the TEM diagram of the interconnection interface when the interconnection temperature is 260 °C and the interconnection time is 180 min. Fig. 4 (a) shows the low magnification image of the copper nanoneedle inserted into the indium layer. It is observed that the interconnection interface is relatively compact with almost no holes. The selected area electron diffraction patterns on both sides of the connecting line correspond to [100] of indium region and [111] of copper region respectively; Fig. 4 (b) (c) shows high-resolution images of both sides of Valley b and c, both cu2in and cu7in3 can be identified, lattice fringes of Cu and In can be identified by measuring lattice spacing, and the transfer region is shown in the middle of (b) (c) image, and there is an amorphous region with a width of 40nm in (b) image. There is a 20nm wide amorphous region in the (c) image, indicating the existence of atomic level interconnection. On the one hand, it may be due to the wedge effect of copper cone structure; on the other hand, the joint density and actual contact time are not evenly distributed along the interconnection interface. The 4 (c) region shows the early bonding stage, while the 4 (b) region interconnects quickly. It can be concluded that in the solid-liquid diffusion interconnection, the copper needle layer structure and indium layer form a physical interlocking
interface. On the other hand, due to the good wettability of indium, it can form intermetallic compounds with copper at the interface, and the interface bonding is dense and the interconnection quality is good.

3.4. Influence of interconnection conditions on shear strength

The parameters affecting the quality of solid-liquid diffusion interconnection include interconnection temperature, interconnection time, etc. Fig. 5 describes the shear strength trend of copper indium copper interconnection interface under different conditions. It is found that the average shear strength of copper indium copper interconnection interface is 6.2 mpa, 6.8 mpa and 11.5 MPa at 260 °C for 10min, 90min and 180min. With the extension of interconnection time, the shear strength increases. This can be attributed to the fact that indium is liquid at 260 °C and has good wettability. The intermetallic compound cu2in has good thermal stability and mechanical properties, and its shear effect is comparable to that of reflow soldering process. The results show that the average shear strength of the interface is 16.2 MPa, 15.3 MPa and 15 MPa at 350 °C for 10 min, 90 min and 180 min. with the increase of interconnection time, the shear strength decreases. According to the Cu in phase diagram [10], intermetallic compounds have a tendency to transform from cu2in to cu7in3, which reduces the shear strength of the interconnection interface. Obviously, satisfactory bonding strength can be obtained at lower interconnection temperature. Compared with the traditional reflow soldering process temperature as high as 400 °C, this kind of solid-liquid diffusion interconnection technology based on Cu in secondary micro nano morphology needs lower temperature, which greatly reduces the adverse effect of chip overheating in high-density laminated packaging due to high temperature. It is expected to replace reflow soldering and become a new interconnection method in high-density 3D packaging.

3.5. Effect of heat treatment on shear strength

In order to further analyze the effect of heat treatment time on the interconnection strength, the samples with interconnection temperature of 260 °C and interconnection time of 180 min were heat treated at 180 °C. The bonding interface of samples with heat treatment time of 10 h and 100 h were analyzed by XRD. Fig. 6 shows the XRD pattern of interconnection interface with heat treatment time of 10 h and 100 h. Comparing the XRD patterns of interconnection interface, when the heat treatment time is 10h, the diffraction peak intensity of Cu and cu2in is relatively strong; when the heat treatment time is 100h, the diffraction peak intensity of cu7in3 is relatively strong, and only the bonding composed of cu7in3 can be obtained by longer heat treatment. After the heat treatment aging, the shear stress of the interconnection interface is tested. The effect of heat treatment aging on the shear strength is shown in Fig. 7 (interconnection time is 180min). It can be found from Figure 7 that the shear strength of the interconnection interface with low shear strength before heat treatment increases significantly after heat treatment, and the increase becomes slow after 10 h; while the shear strength of the interconnection interface with higher shear strength before heat treatment increases after heat
treatment, but the increase is slow, and the shear strength tends to decrease after 10 h. The fracture surface was studied after ball pushing shear test. The morphology of shear section of Cu in Cu interconnection interface under different heat treatment time is shown in Fig. 8. Fig. 8 (a) after heat treatment at 180 ℃ for 100 h, the shear surface is relatively uniform and flat, mainly cleavage fracture. With the crack expansion, secondary cleavage and local tearing occur on the main cleavage surface, and the indium containing section cu7in3 is found at the cross section. Figure 8 (b) after heat treatment at 180 ℃ for 10 h, the tongue like fracture surface shows that the interconnection interface has certain plasticity and toughness, and the indium containing cross section cu2in is found. It can be inferred that the intermetallic compound cu2in with low shear strength is fully embedded in the solid-liquid phase after short heat treatment, and the resulting intermetallic compound cu2in has excellent mechanical properties. With the increase of heat treatment time, the intermetallic compound cu7in3 obtained by long-time heat treatment is brittle and reduces the quality of interconnection. Heat treatment experiments show that the two-stage Cu in micro nano layer interconnection technology can obtain better interconnection strength after short-time heat treatment.

4. Summary
In a word, a solid-liquid diffusion interconnection method is studied in this paper. High quality interconnection can be achieved by two-stage Cu in micro nano layer at 260 ℃ compared with reflow process at 400 ℃. The needle like structure of copper needle layer has good mechanical interlocking force and huge diffusion surface. At the same time, indium has good wettability. The intermetallic compound cu2in formed under the condition of low interconnection temperature is a high-quality phase with good mechanical properties. A good interconnection interface can be obtained by short-time heat treatment. This liquid-solid diffusion interconnection method conforms to the development trend of green packaging.

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