Bonding Condition Evaluation using a Melting-Point-Change Type Conductive Paste

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Nowadays advanced board structures are made of two different circuit boards interconnected to each other. As these circuit boards are made of different materials and have to go through different manufacturing processes, the use of a specific conductive paste is needed for a reliable interconnection. To realize this complex structure, we worked on the development of an innovative conductive paste overcoming the weaknesses of the conventional one. It is a melting-point-change type conductive paste in which a large spherical copper (Cu) powder is used as a conductive filler and an alloy of tin (Sn) and bismuth (Bi) is coated on its surface. The conductive paste is expected to stabilize the bonding by tolerating the scatter of the thickness, it also has to maintain a long-term electrical reliability. In this paper, we determined the fundamental composition of the paste to answer these needs. To evaluate the performance of the bonding we studied the influence of the electrical reliability of the paste at its optimum bonding condition, the aggregability of the resin and the bonding strength. Finally, we created a complex structure with the paste to analyze the tolerance of the bonding height and its electrical resistance through a thermal cycle (TC) test.

Key Words: Circuit Boards, Conductive Paste, Melting-point-change, Interconnection Structure
first heating and curing.

In this paper, we established the particle size of the Cu filler, the compound ratio of the resin composition and the bonding process condition (temperature, pressure, time). The electrical resistance stability was measured by a TC test after the bonding. Furthermore, the height and diameter of the paste indispensable for the stability of the bonding were observed. In the circuit connection demonstration experiment, by using the TEG (test element group) board with a daisy chain circuit pattern, the electrical resistance value after the joining was observed, the height tolerance clarified and the resistivity change calculated with a TC test.

2. Experimental procedure

2.1 Evaluation of the bonding condition impact of the electrical resistance

Optimum circuit board bonding conditions must be determined by the process of heating while pressurizing. Experimental conditions were set as follow: two filler particle diameters (32.5/50 (μm), 2 μm Sn-Bi coating), three different contents (89/90/91 (wt%)), three bonding temperatures (150/175/200 (°C)), and three bonding pressures (0.49/1.47/2.45 (MPa)) based on the design of experiments (DOE). The electrical resistance variation was found by applying repeated TC stress on these eighteen combination. The criteria have determined that the change rate of the electrical resistance had to be within a 10% small range when applying a -40°C to +150°C TC stress for 500 times or more.

As regard measuring equipment, I-V characteristics ware acquired with a DC stabilized power supply (Kikusui Electronics PWR 400L) and a strain measuring unit for voltage monitoring (Keyence NR-ST 04) was used in the design of optimum conditions. A temperature cycle chamber (Kusumoto Chemical WINTECH NT1010) was used for the long-term reliability.

2.2 Evaluation of paste bonding strength

It is necessary to ensure the stability of the bonding strength even with a variation in the bonding conditions and the particle size of the spherical Cu powder. Therefore, experiments were conducted to increase the strength of the joint by improving the adhesive component essential for the paste material. This paste material is made of 20 μm to 38 μm Cu particle (2 μm Sn-Bi coating) as conductive filler (content 89wt%), an epoxy resin (bisphenol F type + naphthalene) as additive, and a curing agent (MTHPA : Methyl-tetra-hydro-phthalic Anhydride Acid) as reference. In the experiment, the adhesion strength was changed by adding a thixotropic agent and by changing the curing agent: acid anhydride 13). Table 1 shows the paste sample conditions. A paste was applied to a 110 mm² bonding surface of the Cu test piece, for 90 min under 2.45 MPa and 150°C and subjected it to a tensile test.

As regard bonding strength measurements, a tensile tester (Instron INSTRON 5581) was used and the cross section observation was made by using a metallurgical microscope (Nikon OPTIPHOTO200).

2.3 Evaluation of the paste compression amount

This experiment was conducted to clarify the relationship between the bonding pressure and the compression amount of the paste in order to obtain an ideal condition to realize a stable height after the paste bonding. The paste applied on the Si wafer (20×20 mm) was sandwiched with another Si wafer (20×20 mm), and pressed from 0.02 to 2.89 MPa, while being heated continuously until the curing temperature exceeded 180°C. The change in height and in the applied diameter was measured though the variation of the load, and the pressure conditions to attain an ideal connection were confirmed.

As regard measurement equipment, a LSI replacer (Shibuya Kogyo DB200) for pressurized heating machine, a metallurgical microscope (Nikon OPTIPHOTO200) and X-ray line inspection apparatus (Shimadzu SMX-160V) were used.

2.4 Tolerance evaluation of the bonding height

A TEG board was used for the actual construction of the composite structure. Fig. 2 shows a schematic view of the TEG board where (a) is the image view after connection and (b) is the schematic view of the bonding pad layout. By connecting

| Table 1 | Paste conditions for bonding strength examination. |
|---------|--------------------------------------------------|
| Paste type | Filler diameter (μm) | Content (wt%) | Epoxy resin | Curing agent | Additive |
| 1 | 20-38 | 89 | Bisphenol F type + naphthalene | MTHPA * | - |
| 2 | 20-38 | 89 | Bisphenol F type + naphthalene | MTHPA * | Thixotropic agent |
| 3 | 20-38 | 89 | Bisphenol F type + naphthalene | Tepore-based acid anhydride | - |

* Methyl-tetra-hydro-phthalic Anhydride Acid

Fig. 2 TEG board for interconnection structure.

(a) Interconnection structure image after bonding. (b) TEG patterns on board surface for measurement of bonding height and electric resistance.
the daughter board (thickness: 4.2 mm) to the mother board (thickness: 5.7 mm), the electrical resistance of the connection part was measured to establish whether it was good or bad, and the tolerance of the optimum bonding height was determined by observing the cross section of the structure. The bonding process consisted in pre-curing the bonding sheet on the mother board and bonding the daughter board after the paste is filled. The pre-curing conditions were set to 27 sec vacuum, 0.1 MPa and 47°C for 10 sec. The condition for the board bonding were: 2.26 MPa, 170°C for 90 min. Regarding the verification of the long-term reliability, the change rate of the electrical resistance was observed through a TC test (−65°C to +150°C, 500 cycles).

As regard measurement equipment, a metallurgical microscope (Nikon OPTIPHOTO200), a resistance tester (Yokogawa Electric Digital Multimeter 7561/7562) and a TC cycle chamber (Kusumoto Chemical WINTECH NT1010) were used.

3. Results and discussion

3.1 Electrical resistance change by the bonding condition

In order to set optimum conductive paste bonding conditions the distribution of whole composition was studied. Fig. 3 shows the L18 orthogonal table in DOE, and the plot results of the average changing rate of the electrical resistance. This change rate reveals the difference between the electrical resistance value obtained from eighteen different combinations before and after the thermal cycle experiment. By comparing the two diameters, we can observe a lower change rate for the 32.5 μm diameter group combination. In this results, the lowest change rate appeared at the Cu diameter 32.5 μm and curing temperature 150°C combination group. The change rate for this combination was near 0%. From this result, each S/N ratio of control factors were clear and the optimum condition presumed. A series of conditions estimated to be optimal for the bonding process and another one for the comparison are shown in Table 2.

Fig. 4 shows the result of the TC re-test was carried out under these conditions. Fig. 4(a) shows the results of I-V characteristics obtained by studying the relation between the applied current (I) and the voltage (V) changing according to the current (I). In the parameter condition for comparison, after the TC test, the voltage (V) difference is ~11.6% at 2 A. However, when setting the optimum condition parameter, there was no change in the I-V characteristics after the TC test, and a 0.4% ideal rate change was confirmed. Fig. 4(b) shows a bonding cross sectioning of the stable I-V characteristics. This shows that because the contact area and density is increased with the reduction of the particles diameter, an effective intermetallic compound (IMC) is formed between the Cu core and Sn-Bi alloying coating. We can observe this on Fig. 4(b). The Sn-Bi eutectic temperature (137°C), the curing peak temperature (142°C) and the curing reaction of the bonding sheet (from 130°C) and their timing are set according to

![Fig. 3](image-url) Rate of change results of electrical resistance, and experimental factors based on the L18 orthogonal table in the design of experiments (DOE).

![Table 2](image-url) Estimated optimum bonding condition by a S/N ratio.

(a)

![Fig. 4](image-url) Results of TC re-test under the optimum condition. (a) Results of the I-V properties showing the optimum reproducibility of the experimental conditions. (b) Bonding cross sectioning of the stable I-V characteristics.
the curing temperature (150°C). Thus, this timing is interpreted
to be in balance. Thus, the electrical reliability of the paste can
be obtained by maximizing the contact area between the Cu
particles. By appropriately selecting the pressure of the bonding,
the curing temperature and the time, it is possible to obtain a
bonded state that balances appropriately the compounding ratio of
the Cu particles and ensures the electric reliability even after the
thermal cycle.

3.2 Aggregation state and bonding strength of paste
In order to set the optimum process conditions for the
conductive paste, the distribution of the entire composition
including the bonding material was examined. The bonding
strength of the paste material in section 3.1 often varies
depending on the conditions of heating and pressure at the time of
bonding. Therefore, while taking into consideration the bonding
stability of the paste during the heating process, we examined
the improvement of the adhesive component for a steadier
bonding strength. Fig. 5 shows a cross sectioning comparing
the bonding state in which the difference in strength occurs at
the bonding portion of the paste. Fig. 5(a) shows an example of
how Cu particles are dispersed due to poor bonding conditions.
The bonding sheet fluidity associated with the heating during
the structure construction is most likely the reason for this filling
state. Fig. 5(b) is an ideal form with a strong bonding strength.
Cu particles collectively aggregate at the center, and the contact
area to be alloyed is also large. It is necessary for the composition
of the paste material that such agglomeration change stays small
even after a change in the behavior of the bonding sheet at the
time of pressurization and heating. The bonding strength and
the aggregability after the curing was investigated in order to
determine the resin composition to ensure an ideal density to
maximize the bond of the Cu particle. Fig. 6 shows the state of
the test piece after the bonding experiment. The bonding area is
the overlapping inner side of each test pieces. Fig. 6(a) shows
the interface of the paste type 1 with the basic composition as
a reference. The central part of both test pieces is an aggregated
part in which the Cu particles of the paste are cured. In contrast,
Fig. 6(b) is a paste type 2 to which a thixotropic agent is added.
We observed that the aggregation state was clearly improved in
comparison with the paste type 1. In addition, Fig. 6(c) is a paste
type 3 with a different curing agent. Its aggregation state is clearly
inferior to Fig. 6(b). However there is no big difference from the
reference aggregation state of the paste type 1.

Next, the results of the numerical comparison of the bonding
strength are shown in Fig. 7. By visual observation, the state of
aggregation appeared to be a slight different, whereas the strength
difference appear larger. Compared to the reference paste with
a basic composition, the type 2 added thixotropic agent shows a
42% improvement effect while the paste type 3 with the altered
curing agent shows a 49% reduction. This is believed to be due to
the addition of the thixotropic agent improving the printability
when applying the paste, allowing the Cu particles to be easily
bonded by maintaining a high viscosity even at the time of curing
after the coating.

On the other hand, the strength remarkably decreased in the
paste type 3 in which the curing agent was changed. In this
case, the curing agent was also changed to a highly viscous one
in order to suppress the outflow of Cu particles, but expected
effects could not be obtained. This is believed to be due to the
fact that the adhesion force of the resin itself contributes greatly
to the bonding strength, and therefore modified curing agent was
inferior in reactivity to the reference and the adhesive strength of
the resin was also lowered. From this result, it can be interpreted
that the method of increasing the viscosity with the curing

![Fig. 5](image)

**Fig. 5** Cross sectioning of bonding condition cured under 2.45
MPa, 150°C for 90 min. (a) Condition of few filler. (b) Ideal
connected condition.

![Fig. 6](image)

**Fig. 6** Comparison of the aggregation state of paste after tensile test. (a), (b), (c) are paste type 1, 2, 3. (d) Test piece: connected area
is 110 mm².

![Fig. 7](image)

**Fig. 7** Comparison of bonding strength of paste by tensile test.
agent eventually hindered the agglomeration of the Cu particles and further decreased the adhesive strength of the resin itself. Therefore, it can be said that the increasing thixotropy is the most effective approach to increase cohesiveness.

3.3 Paste compression amount

By observing the change in height according to the pressure applied to the aggregated paste itself, the relationship between the pressure at which the bonding was stabilized and the crushed amount was confirmed. Fig. 8 shows a cross-sectional photograph (left side) of the crushed state of the paste for each load and an X-ray photograph (right side) showing a change in the coating diameter. The increase in paste diameter shows little change from 0.46 MPa to 1.34 MPa, but from around 1.34 MPa a sharp rise is observed, and the increase rate exceed 80% at 2.84 MPa. Then, at 2.86 MPa, which is the largest load, X-ray is observed on a photograph short circuit between adjacent pastes.

**Table 7**

| Pressure | Paste height | Paste diameter |
|----------|--------------|----------------|
| 0.02 MPa |              |                |
| 0.15 MPa |              |                |
| 0.44 MPa |              |                |
| 1.34 MPa |              |                |
| 2.08 MPa |              |                |
| 2.86 MPa |              |                |

**Fig. 8** Observation result of cross section of bonding height. Pressure was at 0.02 to 2.89 MPa, curing was heated continuously until 180°C is exceeded.

**Fig. 9** Graphic representation of the bonding height load transition and diameter spread ratio. Pressure was at 0.02 to 2.89 MPa, curing was heated continuously until 180°C is exceeded.

From these results, it was found that keeping the load at about 1.5 MPa or less is the most suitable condition to keep a stable paste diameter. In particular, the rapid change in paste application diameter is believed to be due to the melt bonding of the coating metal between the Cu particles progressing as the Cu particles pressurization becomes larger than 1.34 MPa. Cu particles deformation is considered to be due to higher pressure and thereby its volume is extruded. Since the periphery of each paste is in an open state and as the resin becomes highly liquid with the heating, the Cu particles may have been extruded with the synergy effect of the pressure.

The paste height behavior shows that the paste itself acts as a stopper, which means that the stable bonding height can be determined. This means that this conductive paste can be used in a relatively low load in the bonding process of the circuit board. At the same time, this leads to a reduction in the pressure applied to the bonding sheet, which makes the control of the pressurizing and the heating process easier. In this respect, the bonding sheet material can suppress the melting viscosity to a comparatively low level, and if the material having a small repulsive force against the bonding load can be adopted, excessive resin flow can be suppressed, the shape of the paste bond portion can be stabilized, and a highly reliable bonding can be obtained.

3.4 Tolerance of bonding height

**Fig. 10** shows the cross section of the connecting part in the interconnection structure by TEG board. The bonding state of Cu fillers in the paste connecting the upper and lower pad is observed, both cross sections shows a good resistances state.
The difference on the aggregated state of the Cu fillers can be observed and the height variation between the upper and lower pad occurring by the scatter of aggregation was studied. Fig. 11 shows the histogram of the distribution of the height between the 791 bonding pads with a good electrical resistance. From these distributions, we found that the maximum height, the minimum height, the average height and the standard deviation were respectively 75 μm, 25 μm, 54 μm and 10.7 μm. Therefore, for an optimum electrical resistance the bonding height tolerance should fluctuate within a 50 ± 25 μm range.

This result almost matches the 60 μm crushed state of the paste after increasing the load in Fig. 8. When a paste is applied to the actual circuit board, the scatter of board thickness is inevitable. Circuit boards requiring new interconnected structures could become high-multilayer thick boards (4 to 6 mm) and in some cases, the thickness scatter is near ±100 μm. In this case, the board thickness has to be uniform and the paste needs to tolerate the bonding height scatter. This experiment showed that the cohesive force by adding a thixotropic agent increases the bonding strength by 42%. A 1.5 MPa or less load of is the best condition to maintain the paste diameter unchanged.

The bonding height tolerance is at around 50 ± 25 μm and there is only 5% electrical resistance variation after 500 thermal cycles.

4. Conclusions

In this paper, the bonding conditions of composite circuit board structures with a melting-point-change type conductive paste were studied. The results are summarized below:

An Optimal bonding conditions allows a ±10% electrical stability.

An improvement of the cohesive force by adding a thixotropic agent increases the bonding strength by 42%.

A 1.5 MPa or less load of is the best condition to maintain the paste diameter unchanged.

These results show that with this paste, it is now possible to create a bonding in a relatively low load region and at the same time the pressure applied to the bonding sheet can be reduced and the shape of the paste bonding part stabilized.

The bonding height tolerance is at around 50 ± 25 μm and there is only 5% electrical resistance variation after 500 thermal cycles.

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