Integration technologies involving flexible substrates are receiving significant attention owing to the appearance of new products regarding wearable and Internet of Things technologies. There has been a continuous demand from the industry for a reliable bonding method applicable to a low-temperature process and flexible substrates. Up to now, however, an anisotropic conductive film (ACF) has predominantly been used in applications involving flexible substrates; we therefore suggest low-temperature lead-free soldering and bonding processes as a possible alternative for flex-on-flex applications. Test vehicles were designed on polyimide flexible substrates (FPCBs) to measure the contact resistances. Solder bumping was carried out using a solder-on-pad process with Solder Bump Maker based on Sn58Bi for low-temperature applications. In addition, thermocompression bonding of FPCBs was successfully demonstrated within the temperature of 150 °C using a newly developed fluxing underfill material with fluxing and curing capabilities at low temperature. The same FPCBs were bonded using commercially available ACFs in order to compare the joint properties with those of a joint formed using solder and an underfill. Both of the interconnections formed with Sn58Bi and ACF were examined through a contact resistance measurement, an 85 °C and 85% reliability test, and an SEM cross-sectional analysis.

Keywords: Sn58Bi, ACF, Flexible, Low temperature.

I. Introduction

Since the advent of electrically conductive adhesives, the use of solder joints in certain flip-chip assemblies has been avoided owing to a high-temperature process and environmental concerns. Restrictions on the use of lead-bearing solder have driven the electronics industry to seek a reliable replacement [1]. Anisotropic conductive film (ACF) interconnections have drawn significant attention owing to their environmental friendliness through the elimination of lead-containing solder [2]. Moreover, applications involving heat-sensitive devices and flexible substrates have employed ACFs instead of solder for use in low-temperature processes [3]. For these reasons, ACF has played an important role in the electronics packaging industry, encouraging many institutions and manufacturers to investigate and develop ACFs for a wide variety of applications.

However, a shift in the contact resistance during a reliability test and the poor adhesion strength have been pointed out as weaknesses of an ACF [2]. These failures in the bonding process are mostly related with the contact mechanism of the conducting balls and the curing degree of the ACF material. Consequently, the bonding process of a conventional ACF is carried out at a relatively high temperature (around 200 °C) accompanied by a high bonding pressure in order to compensate for the deficiencies in reliability [4].

Unlike an ACF, which enables electrical conduction through mechanical contact between electrodes, solder forms strong intermetallic joints with electrodes [5]. Moreover, the addition of an underfill protects the joints by relieving the thermomechanically induced stresses and promotes overall adhesion strength between flexible substrates. Hence, if the high-temperature process and environmental concerns are the reasons to employ an ACF, bringing down the temperature...
and excluding lead-containing solder from the process would allow a bonding method using solder and an underfill to be readopted.

Therefore, in this paper, we suggest a low-temperature interconnection method using Sn58Bi solder and an underfill suitable for a flex-on-flex structure. All processes including solder bumping and thermocompression bonding were kept to less than or equal to 150 °C for a low-temperature demonstration on a flexible substrate. This study not only focuses on an analysis of the solder joint quality of the newly suggested method, but also investigates the quality of a conventional ACF joint. For a meaningful comparison of the solder and ACF, thermocompression was conducted using similar bonding conditions on the same test vehicles. To examine the electrical and mechanical characteristics of the interconnections of both bonding methods, a 4-point probe measurement of the contact resistance and a cross-sectional SEM analysis of each joint are presented. In addition, an 85 °C, 85% humidity test was conducted on the test vehicles to compare the reliability of the performance.

II. Experiment

1. Materials

The solder paste material used in the bumping process is called Solder Bump Maker (SBM) [6]–[9]. Composed of resin, a deoxidizing agent, and solder powder, the SBM used in this study was optimized for a low-temperature reflow process. A notable difference in the new formulation of the SBM from a previous study is that the SBM used in this study consists of type 7 Sn58Bi solder powder with an increased volume ratio [9]. This formulation is intended to achieve a uniform solder bumping process suitable for FPCB test vehicles, the electrode pitch of which is 250 µm.

A fluxing underfill is a bonding adhesive material that is also often called non-conductive paste. FPCB test vehicles with Sn58Bi solder bumps were bonded using a fluxing underfill. As an epoxy-based thermoset, the fluxing underfill material shortens the overall processing steps by combining the functions of a flux and an underfill [10]. Unlike previously developed curing resins [10]–[15] suitable for process temperatures of 180 °C and 250 °C, the material used in this experiment was specially developed and optimized for low-temperature applications (< 150 °C). A detailed chemical and thermal analysis of the curing behaviour of this fluxing underfill can be found in [16].

A commercially available ACF material was used to make the test vehicles for a joint quality comparison. The suggested bonding temperature of this ACF product is 150 °C to 160 °C with a peak duration time of 10 s. The total film thickness is 25 µm with Au/NI-coated conductive particles embedded inside the resin. Table 1 summarizes the specifications of the commercial ACF product, which are excerpted from a technical report provided by the manufacturer.

<table>
<thead>
<tr>
<th>Table 1. Specifications of commercial ACF product.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Item</strong></td>
</tr>
<tr>
<td>Film thickness</td>
</tr>
<tr>
<td>Conductive particle</td>
</tr>
<tr>
<td>Particle size</td>
</tr>
<tr>
<td>Density</td>
</tr>
<tr>
<td>Bonding temperature</td>
</tr>
</tbody>
</table>

A commercially available ACF material was used to make the test vehicles for a joint quality comparison. The suggested bonding temperature of this ACF product is 150 °C to 160 °C with a peak duration time of 10 s. The total film thickness is 25 µm with Au/Ni-coated conductive particles embedded inside the resin. Table 1 summarizes the specifications of the commercial ACF product, which are excerpted from a technical report provided by the manufacturer.
Fig. 2. Schematic diagram of 4-point probe measurement circuitry.

Fig. 3. Process flow diagram of solder-on-pad process: (a) sample preparation, (b) SBM deposition, (c) SBM printing, (d) guide removal, (e) reflow, and (f) residue cleaning.

2. Test Vehicle Design

Figure 1 shows the top and bottom of the FPCB test vehicles designed for the experiments. On top of a polyimide substrate, electrodes and pads are patterned using dimensions relevant to those of display applications. Additional specifications of the test vehicle are presented in Table 2. The UBM structure of the electrode, which consists of Cu/Ni/Au with thicknesses of 11µm, 5µm, and 0.3 µm, respectively, was fabricated using an electroless nickel immersion gold process. Furthermore, daisy chains were formed such that the contact resistance measurements could be monitored. A schematic diagram of the 4-point probe measurement circuitry is shown in Fig. 2.

3. Solder-on-Pad Process

For the preparation of the bumped test vehicles, a solder bumping process called solder-on-pad is employed [7], [17]–[19]. Similar to a stencil printing method, the solder-on-pad process prints the SBM on top of the electrodes. Instead of using stencil masks for the paste printing, the solder-on-pad process employs simple guides to define the printing area. Figure 3 illustrates the overall solder-on-pad process. First, the FPCB test vehicle is prepared with a guide attached to it. Second, the guide is removed after the SBM is printed on top of the FPCB. Third, the test vehicle is reflowed with a peak temperature of less than or equal to 150 °C. Finally, the remainder of the SBM that did not participate in forming the solder bump during the reflow process is cleaned away with a solvent.

The reflow process is carried out using an SMT scope (SK-5000, Sanyo Seiko). Figure 4 shows the reflow profile set for the bumping process. The reflow conditions, as well as the SBM formulation, affect the outcome of the bumping process. The process conditions presented in this study basically follow the optimizing principles suggested in [18]. The resulting heights of the solder bumps after completion of the solder-on-pad process are measured using an Olympus STM6, a 3D optical microscope with ±0.1-µm resolution in the z-direction.

4. Thermocompression Bonding

The thermocompression bonding of the test vehicles is carried out using an M9 Laurier flip chip bonder. Figure 5 shows a process flow diagram of the bonding process using the fluxing underfill. As shown in Fig. 5(b), the underfill is dispensed on the bottom of the FPCB with solder bumps. Then, the top of the FPCB is aligned and bonded according to the bonding conditions, as shown in Figs. 5(c) through 5(d). Finally, the bonded test vehicle is post cured in an oven at 130 °C for 1 h.

On the other hand, Fig. 6 illustrates the bonding process using ACFs. The overall bonding process using the ACF has a similar sequence to those of the fluxing underfill. However, the only difference is the underfill dispensing step from Fig. 5(b),
which is replaced with the ACF lamination step, as shown in Fig. 6(b).

Table 3 summarizes the thermocompression bonding of conditions 1 and 2 used for the fluxing underfill and ACF, respectively. The bonding parameters are established based on the conditions recommended by the ACF manufacturer. Because both the ACF and fluxing underfill are specially formulated for low-temperature applications, the peak bonding temperature is set to 150 °C with a holding time of 10 s. However, the conditions differ in terms of the bonding pressure because the pressure generally required for an ACF is higher than that of the solder interconnections owing to its innate contact mechanism.

5. Reliability Test

The test vehicles bonded with two different methods went...
III. Results and Discussion

1. Solder-on-Pad Process

As a result of the solder bumping process, a uniform array of Sn58Bi solder bumps was formed on the FPCB test vehicles. The measurements of each bump height are shown in Fig. 7, where the red line indicates the average value. An average height of 14.7 µm within the range of ± 3 µm was obtained. A magnified photograph of the test vehicles before and after the solder-on-pad process is shown in Fig. 8.

2. Thermocompression Bonding

After the thermocompression bonding process of the Sn58Bi and ACF test vehicles, the contact resistances of each test vehicle were measured using a 4-point probe method. As illustrated in Fig. 2, the 4-point probe method measures the electrical resistance of a joint by detecting a voltage drop across the top and bottom electrodes. The average contact resistances of five samples for each method were 2.7 mΩ and 416.5 mΩ with standard deviations of 0.002 and 0.139 for Sn58Bi and ACF, respectively. The contact resistance of the ACF is approximately 150-times higher than that of the Sn58Bi solder. This substantial difference in the contact resistance can be explained through a cross-sectional image of the two bonding mechanisms.

As shown in Fig. 9, there is a noticeable difference in the contact area of the two cross-section images. The contact area in this case refers to an area that forms an electrical path from bottom to top electrodes. As clearly shown in Fig. 9(a), there is a conducting ball trapped between the electrodes, and the remainder of the gap is filled with epoxy. This deformed conducting particle, which has a diameter of around 10 µm, creates a relatively small electrical path by making contact with the top and bottom electrodes. On the other hand, the Sn58Bi solder makes contact with the entire area of the electrodes, forming a large electrical interconnection. Owing to this
narrow contact area of the ACF, which also indicates a low current density, the measured contact resistance of the ACF is significantly higher than that of Sn58Bi.

As illustrated in Fig. 9(b), the height and uniformity of solder bumps formed using the solder-on-pad process did not cause any bonding defects, such as an electrical shortage or opening, after the thermocompression bonding.

3. Reliability Test

The contact resistance ($R_{\text{contact}}$) of the test vehicles was periodically measured during 1,000 h of the 85 °C, 85% humidity reliability test. Three out of five of the ACF samples were unable to be measured because the contact was lost (open failure) after 1,000 h. On the other hand, none of the five samples using Sn58Bi showed an opening failure even after 1,000 h of testing. The $R_{\text{contact}}$ measurements before and after the reliability test are summarized in Table 4. In these measurements, the samples that showed an opening failure were excluded.

To monitor the shift in contact resistance during the reliability test, the average contact resistance measurements were normalized with respect to the initial measurements, as shown in (1).

$$\frac{R}{R_{\text{initial}}} = \frac{\text{average } R_{\text{contact}}}{\text{average of initial } R_{\text{contact}}}.$$  

Table 4. Contact resistance measurements before and after the 85 °C/85% reliability test.

<table>
<thead>
<tr>
<th></th>
<th>Sn58Bi</th>
<th></th>
<th>ACF</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Average $R_{\text{contact}}$</td>
<td>Open failure</td>
<td>Average $R_{\text{contact}}$</td>
<td>Open failure</td>
</tr>
<tr>
<td>0 h</td>
<td>2.7 mΩ</td>
<td>0/5</td>
<td>289.0 mΩ</td>
<td>3/5</td>
</tr>
<tr>
<td>1,000 h</td>
<td>3.5 mΩ</td>
<td></td>
<td>1,523.4 mΩ</td>
<td></td>
</tr>
</tbody>
</table>

The normalized contact resistance measurements, denoted by $R/R_{\text{initial}}$, are shown in Fig. 10. The further the $R/R_{\text{initial}}$ value deviates from 1, the larger the shift in the contact resistance value. The blue line with rounded dots represents the normalized contact resistance of the Sn58Bi joints. Although there was a slight shift in the blue line during the initial 100 h or so, the $R/R_{\text{initial}}$ value remained stable throughout the rest of the environmental test. On the other hand, the red line with squared dots, which represents the $R/R_{\text{initial}}$ value of the ACF joints, showed a remarkable leap from the beginning, after which the value displayed an irregular tendency throughout the measurement period. In addition, three out of five of the test vehicles of the ACF joints failed to give a contact resistance value that indicates an electrical opening failure after 1,000 h of the 85 °C, 85% test.

The difference in these reliability performances results from the difference in the connection mechanism of the two bonding methods. The ACF bonding mechanism relies heavily on the mechanical contact of the conducting particles. Thus, this connection can be easily degraded when a swelling of the epoxy resin, caused by high temperature and humidity, occurs [20]. On the other hand, the Sn58Bi solder joint forms a strong intermetallic compound with the top and bottom electrodes.

Fig. 10. Contact resistance measurements during the 85 °C/85% reliability test.

Unlike the simple contact of the conductive particles from the ACF, the intermetallic compound formed during the thermocompression process is a relatively strong metallic bonding, which prevents the electrodes from becoming easily

Fig. 11. Cross-sectional SEM images of ACF joints before and after the reliability test at (a) 0 h and (b) 1,000 h.
detached. Furthermore, the underfill that fills the gap between the joints supports the interconnection by relieving the stress induced by the temperature and humidity. Therefore, given similar bonding and reliability conditions, the environmental test results show that a joint of Sn58Bi using a fluxing underfill is superior to that of an ACF in terms of the electrical stability and mechanical durability.

The idea behind the connection mechanisms stated above can be verified by analyzing the cross-sectional SEM images. Figure 11 shows the cross-section of ACF joints before and after the reliability test. Conductive balls are trapped between the top and bottom electrodes. However, the trapped balls differ in their amount of compression, which is exemplified through the height of the balls, as shown in Figs. 11(a) and 11(b). As the ACF joint goes through a long period of storage under high humidity and temperature, the epoxy material, which fills up most of the gap between the electrodes, expands. Consequently, conducting balls gradually lose their contact with the electrodes, causing a shift in the contact resistance, or even an open circuit failure.

Figure 12 shows cross-sectional SEM images of Sn58Bi solder joints before and after the reliability test. No cracks or delamination of the solder was observed in either of the test vehicles. A noticeable difference between the test vehicles in Figs. 12(a) and 12(b) is the morphology of the Sn58Bi solder and interfacial IMC, which changed over time during the reliability test. A phase coarsening of the Sn- (dark contrast) and Bi-rich (light contrast) regions could be observed because the test vehicles were affected by the high temperature and humidity. Moreover, magnified SEM images of the interfacial IMCs before and after the reliability test are shown in Fig. 13. The scallop-like morphology of the interfacial IMC layer in Fig. 13(a) is regarded as Ni₃Sn₄, which is formed after the thermocompression bonding process [21]. After continuous exposure to the 85 °C, 85% environmental testing, a growth in the thickness of the IMC layer could be observed. The newly formed IMC layer that appeared after the test is considered to be an alloy composed of Au-Ni-Sn-Bi. The formation and growth of such IMCs before and after the reliability test are in accordance with other published observations [21], [22]. In short, from an investigation of the interfacial IMCs, it was shown that the connection mechanism of Sn58Bi solder is not a mere mechanical contact, but an intermetallic bonding that remains relatively stable over time under high humidity and temperature.

IV. Conclusion

In this study, a low-temperature bonding of a flex-on-flex structure was demonstrated. Using a newly formulated SBM with Sn58Bi solder and a fluxing underfill, the solder bumping and thermocompression bonding processes can be conducted at a temperature of less than or equal to 150 °C. The FPCB test vehicles were made of polyimide films with daisy chain electrodes formed on top to verify the bonding capability and reliability.

For a comparison of the electrical and mechanical characteristics of the interconnection, flex-on-flex test vehicles bonded with a low-temperature ACF were prepared using similar processing conditions. The average contact resistance measured using a 4-point probe method was 2.7 mΩ and
416.5 mΩ for Sn58Bi and ACF joints, respectively. The shift in the contact resistance was monitored during the 85 °C, 85% test. Owing to the different interconnection mechanisms of the two bonding methods, the reliability of the two joints showed a noticeable difference. Cross-sectional analyses of Sn58Bi and an ACF were conducted to identify how the joints changed over time under high humidity and temperature.

In conclusion, from the reliability test and a cross-sectional analysis of the interconnections, the stability and durability of the newly formed low-temperature Sn58Bi joint was verified.

Acknowledgement

This work was partly supported by ETRI, the components and the R&D Convergence Program of MSIP (Ministry of Science, ICT and Future Planning), and ISTK (Korea Research Council for Industrial Science and Technology) of Rep. of Korea, Grant B551179-12-04-00 (Development of an image-based, real-time inspection, and isolation system for hyperfine faults).

References

Haksun Lee received his BS degree in electrical engineering from Purdue University, West Lafayette, IN, USA, in 2010 and his MS degree in electrical engineering from the Georgia Institute of Technology, Atlanta, GA, USA, in 2012. He is currently pursuing his PhD in electrical and computer engineering at Georgia Institute of Technology. From 2012 to 2015, he was a researcher with ETRI, Daejeon, Rep. of Korea, where he was involved in the development of novel interconnections and fine-pitch flip-chip technologies.

Kwang-Seong Choi received his BS degree in material science and engineering in 1995 from Hanyang University, Seoul, Rep. of Korea, and his MS degree in electronic material science and engineering in 1995 and PhD in telecommunication engineering in 2008 from the Korea Advanced Institute of Science and Technology, Daejeon, Rep. of Korea. From 1995 to 2001, he was with Hynix Semiconductor Ltd., Incheon, Rep. of Korea, where he was involved in the development of chip scale packages, PoP, and highspeed electronic packages for DDR, Rambus, and RF devices. Since 2001, he has been with ETRI, where he is currently working as a principal member of the engineering staff. His research interests include the development of 3D modules with through-silicon via, flexible interconnections, and flexible solar cell.

Yong-Sung Eom received his BS degree in aeronautical engineering from Korea Aerospace University, Hwajeon, Rep. of Korea, and his MS in aeronautical engineering from the Department of Aerospace Engineering at the Korea Advance Institute of Science and Technology, Seoul, Rep. of Korea, in 1988 and 1991, respectively. He worked at the Korea Institute of Aeronautical Technology, Korean Air Ltd., Seoul, Rep. of Korea, as a design and process engineer for the composite materials of the MD-11 Aircraft Spoiler from 1991 to 1995. In 1999, he received his PhD in material engineering from the Department of Material Science Engineering at École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland. After returning to Korea, he worked at Hynix Semiconductor Ltd., Icheon, Rep. of Korea, as a packaging engineer for memory devices from 2000 to 2001. Since 2001, he has been with ETRI of Korea, where he has been working as a packaging engineer. His research activities include the development of interconnection materials based on a polymer for the electronic packaging and process design of 3D-IC and MEMS packaging.

Hyun-Cheol Bae received the BS and MS degrees in electrical engineering from Dongguk University, Seoul, Rep. of Korea, in 1999 and 2001, respectively, and his PhD in electrical engineering from Chungnam National University, Daejeon, Rep. of Korea, in 2009. Since 2001, he has been with ETRI, where he is currently working as a senior researcher. His current research interest includes the design and fabrication of integrated passive devices, 3D stacked chip packaging using TSV, and wafer-level packaging for MEMS devices.

Jin Ho Lee received his BS degree in physics from Kyungpook National University, Daegu, Rep. of Korea, in 1980 and his MS degree from Kyungpook National University, Seoul, Rep. of Korea, in 1982. He received his PhD in physics from Kyungpook National University in 1988. Since 1982, he has been with the ETRI, where he has been involved in the development of semiconductor devices and advanced flat panel display devices, such as memory devices, TFTs, flexible devices, and power devices. He is currently the managing director of the IT Components and Materials Industry Technology Research Department, in ETRI.