Contact resistance and adhesion performance of ACF interconnections to aluminum metallization

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Abstract

Flip chip joining technology using anisotropically conductive films (ACFs) has become an attractive technique for electronic packaging. However, several factors have hindered the wide spread use of this technology. Along with the reliability issue, these factors also include the low availability and high cost of the bumped wafers. This paper introduces the feasibilities of using unbumped die with respect to ACF joints for flip-chip-on-flex (FCOF) assemblies. The unbumped dies contain only bare aluminum pads. Until now the performance of ACF to Al metallization is a controversial issue from the published reports. In this study, two different test vehicles were used to study contact resistance and adhesion performance. Reliability of contact resistance for ACF joints with the unbumped dies was investigated in terms of varying the thickness of the Al pads. Adhesion performance of ACF to the Al metallization was compared with the adhesion performance of ACF to a glass substrate using the same ACF and the same bonding parameters.

FCOF assemblies containing dies with thinner aluminum pads showed lower initial contact resistance and a lower rate of increment during accelerated aging tests. Three factors were considered as the potential causes for the above results: (1) lower concentration of aluminum oxide on the thin Al pad, (2) larger contact area per deformed particle with Au/Ni/Cu electrode for the interconnection of thin Al pad and (3) lower concentration of the defects in the thin Al pad. Contact resistance was found to increase during accelerated testing because of aluminum oxide formation on top of the pads.

Contrary to the usual expectation, adhesion strength of ACF with the Al metallization was increased during 60 °C/95% RH testing. After 500 h of such moisture-soak testing, the adhesion strength becomes 3 times the initial value. The change in chemical state on the aluminum surface is considered to be responsible for higher adhesion strength. It is proposed that oxidation of Al surface due to diffused moisture and the new chemical bond formation at the adhesives/aluminum interface are the key reasons for good adhesion reliability.

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1. Introduction

Anisotropically conductive adhesives films (ACFs) offer the most suitable alternatives to solder. Because they enable ultrafine pitch capability, are lead free, therefore environmental friendly, and require simple processing at low temperatures. Tests by independent researchers and manufacturers [1–3] have demonstrated the excellent reliability of interconnects using ACFs to the noble metallization surfaces. However, for commercial ACF joints bonded to the non-noble surfaces, the contact resistance increases significantly during high temperature and high humidity aging [4,5]. It is commonly accepted that metal oxide formation at the interface of non-noble metal surfaces is responsible for the increases in the contact resistance [4–7]. To retain the low and stable contact resistance, a noble metallurgy,
such as gold-to-gold interconnection, is required. However, due to the high cost of manufacturing Au bumped chips, gold bumping is unattractive to the industry.

Using ACF technology to join chips, which only contain bare aluminum pads, may be a more attractive alternative to the gold-bump interconnection method. It is apparent that this new technology of direct ACF bonding on the aluminum pad will not only satisfy the requirements of advanced electronic packaging for higher density, lighter weight and higher I/O, but can also satisfy the requirements of industry in reducing cost and simplifying processing steps. Although this technology seems to be a breakthrough in fine pitch interconnections, systematic fundamental research work in this topic has not been carried until now. Previous studies so far undertaken on the reliability of ACF joints bonded to aluminum reveal conflicting results [3,8–10]. Some of the results proved that the interconnection of ACF to the aluminum metallurgy performed well [8], while other results demonstrated that the interconnections were unreliable, moreover some of the electrical interconnects even failed after aging tests [9,10].

Adhesion performance of ACFs joints on different substrates has been studied elsewhere with regard to the accelerated aging tests [2,10]. The results showed that degradation of adhesion strength is a very common phenomenon, either for ACF to glass or ACF to flex substrates. There are two potential causes for the adhesion degradation, (1) moisture absorption in the adhesive and (2) delamination introduced by the different degree of swelling of the adhesives and substrates. Adhesion strength of ACFs to chips is reported to be very high in previous literature [11]. It is also crucial to know about the adhesion of the ACF to the conducting metallization. For aluminum metallization, the adhesion reliability is not clear. It is necessary to identify whether the adhesion of ACF to aluminum metallization degrades like other cases or performs in reverse.

The objective of this paper is to add to the reliability data for ACF interconnections to aluminum metallization, and to further improve the understanding of electrical and adhesion performance of ACF assemblies. Microstructural analyses were carried out to identify the underlying degradation mechanisms.

2. Experiment

2.1. Sample preparation

Two different test vehicles were used to study contact resistance and adhesion performance. The flip-chip-on-flex (FCOF) assemblies were used to identify electrical performance. Flex substrates of 50 μm thickness with daisy-chained circuits are used in this study. The Au/Ni/Cu electrodes on the flex are about 14 μm high. The dies used in this investigation have the same pattern as introduced in Ref. [5]. Aluminum pads having thickness of 1, 3, 5 μm, were prepared for contact resistance study.

Wafers with a deposited aluminum film were sliced into 30×20 mm pieces and then used as the bottom substrates for the adhesion test vehicles. There were no electrodes on the aluminum metallization layer and the thickness of the aluminum layer is about 1 μm. Au bumps having the height of 16 μm were used to control the gap between top dies and the bottom aluminum (or glass) substrates in the test vehicle for adhesion. Fig. 1(a) and (b) are the schematic diagrams of these two test vehicles.

The ACF used in this study consists of epoxy matrix and dispersed conductive particles. The conductive particles are made up of polymers plated with a thin layer of nickel and gold followed by a thin insulation layer. During the bonding process, the insulation layer will be removed due to the friction of particle with the bump and substrate pad—thus achieves electrical conduction between the chip and the substrate in the Z-direction, while keeping insulation in x–y planes to prohibit short circuit between the adjacent joints. The thickness of ACF is about 35 μm and the particle diameter is about 3.5 μm. Concentration of the conductive particles is
about 3.5 million/mm³. The glass transition temperature \((T_g)\) of the ACF is 130 °C.

2.2. Bonding process

The bonding of die to the flex substrate was carried out using a Toray FC 2000 semi-automatic flip chip bonder. The final bonding conditions were 200 °C at 200 N for 15 s. The samples with aluminum metallization (or glass) substrates were bonded using Karl Suss FCM manual flip chip bonder at 170 °C at 150 MPa for 20 s.

Before each test sample was bonded, all components were cleaned according to a commonly used cleaning process [12]. In addition, aluminum metallized substrates were soaked in 8% ammonia solution for 6–10 s in order to etch surface aluminum oxide as the first cleaning step.

2.3. Reliability test

Two kinds of aging tests, (1) 85 °C/85% relative humidity (RH) high temperature and high humidity condition for 500 h and (2) –50 to 125 °C air-to-air cycling for 600 cycles, were performed to test electrical performance separately. In thermal cycle testing, the dwell time at low and high temperature was 30 min, while the transition time between low and high temperature was 2 min. The contact resistance of FCOF assemblies was measured before the tests and at the different storage time/cycles. The selected 60 °C/95% RH conditions were adapted from an industrial parter to compare the reliability on adhesion between ACF to Al metallization and ACF to glass substrate. The test readout points were selected at 0, 50, 100, 200 and 500 h.

2.4. Contact resistance measurement

The contact resistance of the samples was measured using a four-point probe method as introduced in Ref. [5]. There are 60 daisy-chained bump groups that run parallel to the length of chip in order to measure the contact resistance on each sample. It proved difficult to bond the die containing the very thin pads (1 μm aluminum metallization). Also, because of the co-planarity problem, it was difficult to get a uniform deformation of the conductive particles across the whole length of the long die. Due to this bonding issue for each assembly, just one side of the chip, i.e. total 30 daisy-chained groups had good contact and therefore were measured and calculated for each readout point.

2.5. Die shear test

Die shear test was carried out using an INSTRON MINI-44 universal tensile tester with a cross-head speed of 1 mm/min as depicted in Fig. 2. The displacement of the blade was adjusted deliberately in order to force debonding to occur at the ACF/substrate interface, not at the chip/ACF interface. In shear testing, the load continuously increases after the blade comes in contact with the sample, until the chip with ACF is debonded from the substrate. The maximum force required to completely displace the chip is identified as the shear strength of the sample. Shear strength data is expressed...
as shear force per unit die. Six samples were selected for each readout point.

2.6. Cross-sectional studies and failure analyses

C-SAM (scanning acoustic microscope) scans on different layers of ACF assemblies were carried out using sonix SAM. A high frequency transducer of 230 MHz was used to achieve a high resolution. The microstructures of the cross-sectioned samples were examined using Philips XL 40 FEG scanning electron microscope (SEM). The surfaces of aluminum metallization were investigated using energy dispersive X-ray analysis (EDX) and X-ray photoelectron spectroscopy (XPS). (The monochromatical AlK$_{\alpha}$ excitation line (1486.6 eV) was used in the XPS study.)

3. Results and discussion

3.1. Contact resistance

Contact resistance of the FCOF assemblies was measured to see the variation among the three different thickness (1, 3 and 5 $\mu$m) of Al pads. The contact resistance was found to be relatively low for thin (1 $\mu$m thick) Al pads. The changes of mean contact resistance and standard deviation of FCOF assemblies during 85 $^\circ$C/85% RH conditions for 500 h and during −50 to 125 $^\circ$C thermal cycling for 600 cycles are shown in Figs. 3 and 4. During moisture-soaking tests, a rapid increase in the mean contact resistance is found during first 100 h and then the contact resistance stays in a relatively stable state for the rest of the aging exposure. Samples containing thicker Al pads always show higher contact resistance at each readout point. During thermal cycling test, contact resistance of the samples containing thicker aluminum pads (3, 5 $\mu$m) increases with the number of thermal cycles. Whereas, for the samples containing thinner aluminum pads (1 $\mu$m), a rapid rate of increment of contact resistance was observed at the first 150 cycles, but in the following cycles the contact resistance remained more or less constant. During the accelerated aging tests, no open joints were found for ACF assemblies containing 1 $\mu$m aluminum pads. However, open joints were discovered for the cases of other two thicker aluminum pads and the percentage of open joint was 7–10%. Fig. 5 is the cross-sectional microstructures of ACF assemblies with different aluminum pads after aging tests. After aging exposure, the deformed particles were still trapped between the bumps metallization and the pads, however a few microcracks were observed in ACF joints with thicker aluminum pads, as indicated by arrows in Fig. 5(b) and (c).

It was interesting that FCOF assemblies with thinner aluminum pads had a lower initial contact resistance and a lower rate of increment during aging tests as shown in Figs. 3 and 4. The electrical performance of ACF interconnections is better if large sums free electrons are allowed to flow through the conductive paths, which composed of bumps, trapped particles, and pad metallizations. There are several factors which impede the flow of electrons through the interconnections. It is thought that the main factor that attributes to reducing the number of free electrons that flows through the ACF joints is the surface oxidation of the contact metallizations [5]. Table 1 lists the relative concentration of aluminum metal and aluminum oxide on the original aluminum pads found from high-resolution XPS analyses. The relative concentration of aluminum oxides on the thickest pads is as high as 80 at.%, while for the thinnest aluminum pads the relative concentration is
76.3 at.% The concentration of aluminum oxides on the surface of aluminum pads looks like one potential cause for the difference in the initial contact resistance of FCOF assemblies with bare Al pads. For thicker aluminum pads, higher concentration of aluminum oxide causes to the reduction in the number of aluminum free electrons. Hence, the ACF assemblies with thicker aluminum pads have higher initial contact resistance. The second potential cause may be related to the difference as shown in Fig. 6. Fig. 6 displays the microstructure of ACF joints to aluminum pads before aging tests. Under the same bonding conditions, it looks like that particles tend to en-captured in the thick aluminum pads with little deformation, while have the tendency to flatten with the higher extent of deformation in the thinner aluminum pads. The contact area per particle to Au/Ni/Cu electrode is highest for the ACF joints containing the thinnest aluminum pads. Hence, the contact resistance is the lowest. Another important factor which may be related to the intrinsic property of the sputter deposited Al metallization is the residual stress of the sputtered film. This increases with the thickness of the metallization. Higher residual stress is expected to cause more defects (such as microcracks, voids etc.) in the thick sputter deposited pads [13]. These defects contribute to a higher initial contact resistance.

It is commonly accepted that the main cause for the degradation in electrical performance of ACF joints which have undergone aging tests is corrosion and oxidation of the non-noble metals. Aluminum metallization is thought to be very active in forming oxide and hydroxide oxidation when exposed to moisture and high temperature. Aluminum oxide layers accumulate on the surface of aluminum metallization during aging exposure, which then results in the reduction of free electrons, hence a higher contact resistance increases. In some cases, the thickness of the metal oxide layer is sufficient to impede electrons from flowing through the ACF interconnections, resulting in electrical failure [5]. The thicker pads contain more defects, they are more prone to oxidation. This might be one of the reasons why higher contact resistance were found for thicker Al pads during the accelerated testing. In addition, microcracks in ACF joints may attribute to the increase in the contact resistance. However, during the accelerated 85 °C/85% RH test, contact resistance for each sample

### Table 1
List of relative concentration of aluminum and aluminum oxide on the surface of aluminum pads

<table>
<thead>
<tr>
<th>Al-pad (thickness)</th>
<th>Peak</th>
<th>Center</th>
<th>SF</th>
<th>Pk area</th>
<th>FWHM</th>
<th>Tx. function</th>
<th>Norm area</th>
<th>at.%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al-1 μm</td>
<td>Al-oxide</td>
<td>76.5</td>
<td>1.00</td>
<td>995.479</td>
<td>1.436</td>
<td>2836.3</td>
<td>0.00025</td>
<td>76.383</td>
</tr>
<tr>
<td></td>
<td>Al-metal</td>
<td>73.8</td>
<td>1.00</td>
<td>308.079</td>
<td>0.690</td>
<td>2833.5</td>
<td>0.00008</td>
<td>23.617</td>
</tr>
<tr>
<td>Al-3 μm</td>
<td>Al-oxide</td>
<td>76.5</td>
<td>1.00</td>
<td>571.391</td>
<td>1.436</td>
<td>2836.3</td>
<td>0.00014</td>
<td>79.504</td>
</tr>
<tr>
<td></td>
<td>Al-metal</td>
<td>73.8</td>
<td>1.00</td>
<td>147.437</td>
<td>0.690</td>
<td>2833.5</td>
<td>0.00004</td>
<td>20.496</td>
</tr>
<tr>
<td>Al-5 μm</td>
<td>Al-oxide</td>
<td>76.5</td>
<td>1.00</td>
<td>649.048</td>
<td>1.436</td>
<td>2836.3</td>
<td>0.00016</td>
<td>79.968</td>
</tr>
<tr>
<td></td>
<td>Al-metal</td>
<td>73.8</td>
<td>1.00</td>
<td>162.737</td>
<td>0.690</td>
<td>2833.5</td>
<td>0.00004</td>
<td>20.032</td>
</tr>
</tbody>
</table>

Fig. 5. Microstructures of ACF assemblies using bare aluminum pads; (a), (b), and (c) after 85 °C/85% RH tests and (d), (e) and (f) after thermal cycling tests. (a), (d) for 1 μm aluminum pads; (b), (e) for 3 μm aluminum pads; (c), (f) for 1 μm aluminum pads.
did not obviously increased beyond 100 h of moisture soaking. The potential reason for producing such stable contact resistance is attributed as the formation of a stable passive layer of aluminum oxide at the early stage of moisture-soaking tests. The passive layer, composed of Al₂O₃ (aluminum oxide), has the property for pre-venting further oxidation and hence shows a slower rate of increment in contact resistance at these readout points. Whereas, during dry thermal cycling test, we found unstable contact resistance up to 600 cycles for the samples having thicker Al pad. Because of the dry environment, contrary to moisture soak testing, it took a much longer time to form a stable oxide layer.

3.2. Shear test

Fig. 7 shows the change of adhesion strength of ACF assemblies with aluminum (glass) substrates during high temperature/high humidity (60 °C/95% RH) aging exposure. Shear strength for the Al substrates increased about three times after 500 h aging tests. Whereas, the shear strength of ACF assemblies to glass substrates decreased continuously during aging exposure. The adhesion degradation of adhesives’ joints was investigated in earlier studies and is now commonly attributed as the formation of delaminations and voids, which due to the moisture absorption of the epoxy adhesives [2,12]. However, the result of adhesion strength of ACF to Al metallization is contradictory to the usual expectation. To verify any delaminations or voids in the ACF joints to the Al metallization, C-SAM study was carried out on the samples that had undergone 500 h of moisture-soaking tests. Few voids were found in the chip-to-ACF interface (Fig. 8a). Also, no voids or cracks were observed in the layer of ACF/aluminum metallization as shown in Fig. 8(b). Cross-sectional examinations of the samples had undergone C-SAM investigations were also carried out to reveal interfacial bonding under SEM with high magnification (8000×). Fig. 9 shows a typical SEM image of the section plan perpendicular to the faces of the interface, which underwent the moisture-soaking test for 500 h. The cross-sectional examination also supports the SAM experimental results, displaying that the interface is very strong and not containing any defect. Another finding from the cross-sectional examination is that the original Al metallization increases in thickness by 1.5–2 times, which was only 1 μm thick before the aging test. EDX analysis on this Al metallization reveals the existence of oxygen after moisture soaking. As hydrogen (H) is not detectable by EDX, we have not yet confirmed whether it is aluminum oxide or hydroxide. However, it is certain that Al absorbed moisture and transformed it to aluminum oxide or hydroxide during the moisture-soaking exposure. Al has a strong affinity to oxygen and reacts vigorously with the
water molecules, which were diffused to the ACF during the moisture-soaking exposure. There is a less chance for water molecules to remain, which could contribute to the swelling of the ACF. Whereas, for glass substrates, there is no chance absorbing water molecules through the glass surface. Thus all the moisture absorbed by the ACF, contributes to the swelling of the ACF and the formation of cracks or delaminations at the ACF-to-glass interface.

Al has a strong affinity of to oxygen. Al also reacts with the oxygen atom containing radicals such as COO, C=O, C–O etc. of the epoxy chain. Thus a strong chemical bond between the epoxy of the ACF to the Al metallization is also formed during the aging time. Fig. 10 depicts such chemical bonding of the Al surface to the ACF matrix. Similar adhesion mechanism for Al containing surface to the epoxy was previously described by other investigations [13–16]. Thus shear strength of ACF to Al metallization increases with time during high temperature/high humidity test.

4. Conclusions

Electrical and mechanical performances of ACF interconnections to aluminum metallization were examined and discussed in this study. Flip chip containing bare aluminum pads were used to study contact resistance of the FCOF packages. Thickness of the Al pads was varied from 1 to 5 μm to investigate the effects of the thickness on the electrical resistance. Accelerated aging conditions were used to examine the electrical performance. The results showed that FCOF assemblies of thinner aluminum pads had a lower initial contact resistance and a lower rate of increment during aging test. Three factors were considered as the potential causes for the above results, (1) lower concentration of aluminum oxide on the thin Al pad, (2) larger contact area per deformed particle with Au/Ni/Cu electrode for the interconnection of thin Al pad and (3) lower concentration of the defects in the thin Al pad. During 85 °C/85% RH testing, all FCOF assemblies showed a rapid increase in the mean contact resistance during the first 100 h of testing and then maintained a relatively stable position at the following readout points. The contact resistance of ACF assemblies of thicker aluminum pads increase continuously while thermal cycle increases. Potential causes for such degradation of contact resistance, could be due to the increase of aluminum oxidation, the reduction of aluminum free electrons and the formation of microcracks in ACF joints.

Aluminum metallized substrates, which did not contain electrodes, were used to study the mechanical performance. Adhesion of the ACF assemblies to aluminum metallized substrates demonstrates a better reliability. Contrary to the expectation, adhesion strength at the ACF/aluminum metallization actually increases during high temperature and high humidity exposure. To verify this result, the other tests were conducted...
using glass substrates. It was found that adhesion strength of the glass substrate decreases with the aging (moisture soaking) time for the same ACF bonded at the same bonding parameter. It is proposed that diffused water molecules play in a different way for the glass substrate than for the Al metallization. Al metallization absorbs the water molecules to form aluminum oxide or aluminum hydroxide. Chemical reactions are proposed between oxygen containing radical of epoxy and aluminum that might contribute to the increase in adhesion strength. These aliphatic chains and networks protect the interface from further contact with free water. There were no cracks or voids found at the ACF to Al metallization interface. Hence the adhesion strength increases between ACF matrix and aluminum metallization.

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