Study on the shear strength degradation of ACA joints induced by different hygrothermal aging conditions

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**Abstract**

The effect of the different hygrothermal conditions on the shear strength degradation of ACA joints and the difference among them are studied in this paper using experiments and numerical simulations. First of all, several hygrothermal aging tests with different accelerating levels are carried out to explore the hygrothermal environment induced degradation paths of interfacial strength of ACA joints. It is found that the shear strength degradation paths differ from each other when the ACA joints are tested under different hygrothermal conditions, while they are similar to each other. For each case, the shear strength firstly decreases rapidly and then slowly approximates to a fixed value, and the shear strength difference between each other goes to an approximate constant after certain early test hours. After that, to understand the degradation mechanism better, finite element modeling is achieved to study the relationship of moisture concentration in ACA layer and shear strength of the ACA joints, and it is found that moisture saturation and diffusion rate are considered to be the main causes of the shear strength degradation.

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1. Introduction

Anisotropic conductive adhesive (ACA) has been considered as a potential replacement of eutectic solder for flip-chip bonding and has been widely used for packaging technologies in chip on flex (COF), chip on glass (COG), liquid crystal display (LCD), outer lead bonding (OLB) and organic light emitting diode display (OLED) [1–3]. The ACA joining technology has numerous distinct advantages, such as fine-pitch interconnection, higher package density, reduced package size, environmental friendliness, elimination of underfilling, low temperature processing, flexible, simple processing and low cost [4–8]. Nevertheless, although it has been adopted for decades, ACA joining process still has its drawbacks, which greatly limits its wider application. Among them, poor reliability of interfacial jointing strength during servicing is one of the most critical ones, particularly when ACA joints work in a hygrothermal environment [9–11]. Hence, a comprehensive study on the reliability degradation mechanism of ACA joints under hygrothermal condition is very important.

Interfacial shear strength is a key index to characterize the reliability of ACA joints, and the moisture absorption is considered to be the main cause of the shear strength degradation. Some recent publications have studied the interfacial adhesion strength of anisotropic conductive film (ACF) joints under hygrothermal condition. Usually, aging tests of joints were performed in chambers with 85 °C temperature and 85% relative humidity (abbr. 85 °C/85%RH). Lin et al. [12] and Gao et al. [13] investigated the effects of 85 °C/85%RH on epoxy-based ACF joints. The results showed that the shear strengths of ACF joints quickly decreased at first and then slowly with the increase of the aging time. Ferguson and Qu [14] studied the moisture effect on the interfacial fracture toughness of different epoxy-based underfill materials. It was found that the absorbed moisture could damage the interfacial adhesion strength between the underfill and solder mask, and the interfacial fracture toughness values decreased by approximately one-half after exposure to 85 °C/85%RH environment. Luo and Wong [15] studied the influence of temperature and humidity on adhesion strength of underfill materials for flip chip packaging. It was found that the adhesion strength degradation was due to the moisture absorption of underfill. The shear strength decreased from 63 to 34 MPa after 24 aging hours during 85 °C/85%RH testing, and reached an equilibrium value about 10 MPa after 96 aging hours. Saarinen and Frisk [16] investigated the adhesion strength of non-conductive adhesive (NCA) joints after humidity tests. It was found that the shear strength decreased significantly by 36% after humidity tests. All the researches mentioned above showed that the adhesion strength of ACF, NCA and epoxy-based underfill joints decreased significantly under hygrothermal environment, especially in the heavy hygrothermal environment, and the strength degradation was associated with the moisture absorption of the interconnection adhesive materials to some extent. However, the degradation mechanism of interfacial adhesion strength under hygrothermal condition is still not clearly known and needs to be further studied. Usually, accelerating 85 °C/85%RH tests, defined in the well-known JEDEC criteria, are used to obtain the deg-
radiation data to evaluate the joints' reliability. Maybe there is no remarkable influence when evaluating the reliability of joints from a qualitative viewpoint, while it will produce certain error when quantitatively evaluating the Mean-time-to-Failure (MTTF) or other reliability index using the test data obtained under such an accelerating test condition, because the actual working environment of joints is usually different from 85 °C/85%RH. To some extent, it is necessary to study the difference of reliability degradation and occurring mechanism of joints under different hygrothermal aging conditions. However, little researches have been done about the difference of the reliability degradation of joints under different hygrothermal conditions. Hence, it is necessary to make clear the strength degradation mechanisms of the ACA joints under different environmental conditions.

2. Shear strength degradation experiments of ACA joints

In order to study the effects of hygrothermal conditions on the shear strength of ACA joints, more than 1000 ACA joints samples are prepared in our research, which are divided into three groups for later hygrothermal aging tests. Each ACA joint sample is a real frequency identification (RFID) inlay, composed of three components, namely silicon (Si) chip, ACA layer and flexible substrate (Al/PET), as shown in Fig. 1.

The dimension of the silicon (Si) chip is 1.14 mm long and 1 mm wide, with four 65 μm long rectangular bumps. The ACA used in this study is Type DELO MONOPOX AC265 thermostable conductive adhesive supplied by DELO Inc. It is a composite consisting of micro-sized spherical metallic conductive particles that are uniformly mixed in the adhesive matrix, and the adhesive matrix contains thermo-set epoxy resin and other additives. The substrate used in this study is 50 μm thick, with 20 μm Al pad on it. The specifications of the components and bonding process are summarized in Table 1.

<table>
<thead>
<tr>
<th>Description</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conductive particle</td>
<td>Ni/Au</td>
</tr>
<tr>
<td>Particle size (μm)</td>
<td>3</td>
</tr>
<tr>
<td>Silicon chip size (mm)</td>
<td>1 × 1.14</td>
</tr>
<tr>
<td>Flexible substrate</td>
<td>Al/PET</td>
</tr>
<tr>
<td>Bonding temperature (°C)</td>
<td>180</td>
</tr>
<tr>
<td>Bonding time (s)</td>
<td>10</td>
</tr>
<tr>
<td>Bonding force (N)</td>
<td>3.5</td>
</tr>
</tbody>
</table>

Because it is impossible to measure the shear strength of such larger numbers of ACA joint using an in situ method, the shear strength of the ACA joint is measured using an offline method, most commonly used by researchers. Usually, after the samples are moved out of the hygrothermal chamber, the samples surface will form a layer of moisture which can contaminate the test apparatus. So it is inappropriate to do shear strength test immediately. In this study, the samples are placed on a large perforated stainless steel tray for moisture evaporation at room temperature condition for 30 min after they are removed from the chamber, when the samples are cooled to room temperature and the surface moisture has evaporated. Then, the shear strength measurements are carried out as soon as possible. The shear strength test principle is based on the instructions of CONDOR 70-3 Multifunctional bond tester and the solder ball shear test standard [17], as shown in Fig. 2c. A rigid clamping device is used to fix one side of the substrate, and the vacuum plate is used to absorb the bottom of the sample. The shearing speed is 100 μm/s and the height of the shear blade above the substrate is 50 μm. The blade is pushed horizontally from one side. The maximum shear force for each joint, which finally separates the chip from the substrate, is recorded, as shown in Fig. 2d.

3. Analysis of experiment results

After hygrothermal aging tests, the average shear force and its corresponding standard deviation of the 40 joints are calculated for each hygrothermal condition. Shear strength is calculated from the average shear force divided by the joint's bonding area. The results of average shear forces, standard deviations and shear strengths for three different hygrothermal aging tests are listed in Table 2, and the relationship of shear strength versus aging time are graphically shown in Fig. 3.

From Table 2, it can be seen that the standard deviations of the experimental data are small. Thus, it is reasonable to state that the experimental data are stable and valid. In order to study the shear strength degradation of joints, the decreasing magnitude of the shear strength (AS%) is defined as:
is the initial shear strength of joints and strength decreases markedly from 18.28 to 11.47 MPa and the ally with the aging time going. For 85°C/C/85%RH and 45°C/C/85%RH tests, the shear strength decreases slightly, just from 18.28 to 15.14 MPa, and the ΔS% is 17.2%. The shear strength decreasing path for each group of joints can be approximately divided into two phases. In the first phase, the shear strength decreases more quickly. In the second phase, the shear strength decreases very slowly and gently. For example, in the case of 85°C/C/85%RH test, the shear strength decreases quickly in the initial 96 h, and then nearly stabilizes to a fixed value of about 11.47 MPa. Similarly, for 65°C/C/85%RH and 45°C/C/85%RH tests, the shear strength decreases quickly in the initial period of about 0–144 h and 0–192 h, and then nearly stabilizes to a fixed value of about 13.32 MPa and 15.14 MPa, respectively. Moreover, the shear strength degradation differs from each other when the ACA joints are tested under different conditions, while the difference from each other goes to a constant value after certain early test hours. Hence, it can be concluded with a reasonable confidence that the shear strength of ACA joints follows a similar degradation law when the joints suffer from different hygrothermal conditions in the tested scope, however the shear strength degradation paths differ from each other. It is necessary to take the difference aforementioned into consideration when evaluating the Mean-time-to-Failure (MTTF) or other reliability index of ACA joints using accelerating aging tests.

<table>
<thead>
<tr>
<th>Time (h)</th>
<th>85°C/C/85%RH Average force (k gf)</th>
<th>Standard deviation</th>
<th>Shear strength (MPa)</th>
<th>65°C/C/85%RH Average force (k gf)</th>
<th>Standard deviation</th>
<th>Shear strength (MPa)</th>
<th>45°C/C/85%RH Average force (k gf)</th>
<th>Standard deviation</th>
<th>Shear strength (MPa)</th>
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</thead>
<tbody>
<tr>
<td>0</td>
<td>0.2084</td>
<td>0.05406</td>
<td>18.28</td>
<td>0.2084</td>
<td>0.05496</td>
<td>18.28</td>
<td>0.2084</td>
<td>0.05406</td>
<td>18.28</td>
</tr>
<tr>
<td>48</td>
<td>1.451</td>
<td>0.20408</td>
<td>12.72</td>
<td>1.659</td>
<td>0.15763</td>
<td>14.56</td>
<td>1.780</td>
<td>0.04918</td>
<td>15.61</td>
</tr>
<tr>
<td>96</td>
<td>1.281</td>
<td>0.23232</td>
<td>11.23</td>
<td>1.544</td>
<td>0.15345</td>
<td>13.54</td>
<td>1.789</td>
<td>0.03053</td>
<td>15.71</td>
</tr>
<tr>
<td>144</td>
<td>1.337</td>
<td>0.23836</td>
<td>11.73</td>
<td>1.506</td>
<td>0.08048</td>
<td>13.21</td>
<td>1.774</td>
<td>0.14368</td>
<td>15.56</td>
</tr>
<tr>
<td>192</td>
<td>1.321</td>
<td>0.25531</td>
<td>11.58</td>
<td>1.519</td>
<td>0.21595</td>
<td>13.33</td>
<td>1.729</td>
<td>0.07070</td>
<td>15.17</td>
</tr>
<tr>
<td>240</td>
<td>1.299</td>
<td>0.26338</td>
<td>11.41</td>
<td>1.511</td>
<td>0.12523</td>
<td>13.25</td>
<td>1.728</td>
<td>0.16721</td>
<td>15.16</td>
</tr>
<tr>
<td>288</td>
<td>1.316</td>
<td>0.20492</td>
<td>11.54</td>
<td>1.541</td>
<td>0.07419</td>
<td>13.51</td>
<td>1.727</td>
<td>0.10280</td>
<td>15.15</td>
</tr>
<tr>
<td>336</td>
<td>1.304</td>
<td>0.13529</td>
<td>11.44</td>
<td>1.506</td>
<td>0.11861</td>
<td>13.21</td>
<td>1.721</td>
<td>0.11287</td>
<td>15.11</td>
</tr>
<tr>
<td>384</td>
<td>1.305</td>
<td>0.21081</td>
<td>11.45</td>
<td>1.529</td>
<td>0.15286</td>
<td>13.41</td>
<td>1.731</td>
<td>0.10719</td>
<td>15.18</td>
</tr>
<tr>
<td>408</td>
<td>1.308</td>
<td>0.20320</td>
<td>11.47</td>
<td>1.518</td>
<td>0.10739</td>
<td>13.32</td>
<td>1.726</td>
<td>0.12154</td>
<td>15.14</td>
</tr>
</tbody>
</table>

where $S_0$ is the initial shear strength of joints and $S_{408}$ is the shear strength at 408th hour.

From Fig. 3, it is found that the shear strength decreases gradually with the aging time going. For 85°C/C/85%RH test, the shear strength decreases markedly from 18.28 to 11.47 MPa and the ΔS% is 37.3%. For 65°C/C/85%RH test, the shear strength decreases from 18.28 to 13.32 MPa and the ΔS% is 27.1%. For 45°C/C/85%RH test, the shear strength decreases slightly, just from 18.28 to 15.14 MPa, and the ΔS% is 17.2%. The shear strength decreasing path for each group of joints can be approximately divided into two phases. In the first phase, the shear strength decreases more quickly. In the second phase, the shear strength decreases very slowly and gently. For example, in the case of 85°C/C/85%RH test, the shear strength decreases quickly in the initial 96 h, and then nearly stabilizes to a fixed value of about 11.47 MPa. Similarly, for 65°C/C/85%RH and 45°C/C/85%RH tests, the shear strength decreases quickly in the initial period of about 0–144 h and 0–192 h, and then nearly stabilizes to a fixed value of about 13.32 MPa and 15.14 MPa, respectively. Moreover, the shear strength degradation differs from each other when the ACA joints are tested under different conditions, while the difference from each other goes to a constant value after certain early test hours. Hence, it can be concluded with a reasonable confidence that the shear strength of ACA joints follows a similar degradation law when the joints suffer from different hygrothermal conditions in the tested scope, however the shear strength degradation paths differ from each other. It is necessary to take the difference aforementioned into consideration when evaluating the Mean-time-to-Failure (MTTF) or other reliability index of ACA joints using accelerating aging tests.

4. Degradation mechanism analysis of ACA joints

As mentioned above, it is reasonable to state that the shear strength of ACA joints follows a similar degradation mechanism when suffering different hygrothermal aging conditions aforementioned. However, why they follow similar degradation mechanism and how the hygrothermal condition affects the shear strength of joints is still unclear. The potential relationship between hygrothermal aging conditions and the shear strength degradation should be further studied. In many available literatures [12–16], it has been found that moisture can damage the interfacial adhesion of ACF, NCA or underfill material joints and the adhesion strength degradation is due to the moisture absorption. In this study, owing to the similar material with ACF, NCA and underfill materials, the shear strength degradation of ACA joints is most probably due to the moisture, which diffuses into the adhesive interface of the joints and degrades the adhesive strength of the joints. Therefore, to understand the effect mechanism of hygrothermal aging on the interfacial shear strength, the first thing is to determine the moisture diffusion and the moisture concentration of the ACA joints in different hygrothermal aging conditions.

A computer model is applied to simulate the moisture diffusion at the interface of ACA joints under different hygrothermal conditions in our study. Firstly, the moisture properties of ACA adopted are experimentally determined for the three hygrothermal conditions in our study. Therefore, to understand the effect mechanism of hygrothermal aging on the interfacial shear strength, the first thing is to determine the moisture diffusion and the moisture concentration of the ACA joints in different hygrothermal aging conditions.

4.1. Evaluating the moisture properties of ACA

The moisture properties of ACA contain the parameters of diffusion coefficient ($D$, mm²/s) and the saturated moisture concentra-
tion \( (C_{sat}, \text{kg/m}^3) \). Among them \( D \) represents the rate of moisture diffuses through a material and \( C_{sat} \) represents the maximum mass of moisture that can be contained per unit volume \([18,19]\). Typically, a general analytical solution of moisture diffusion, considering one-dimensional case of an infinite plate of thickness \( 2l \), is described by Fick’s second law as follows \([20]\):

\[
\frac{M_t}{M_{sat}} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp \left(-\frac{D(2n+1)^2\pi^2t}{4l^2}\right)
\]

\( (1) \)

\[
C_{sat} = \frac{M_{sat}}{V}
\]

(2)

where \( M_t \) is the weight gain of moisture at time \( t \), \( M_{sat} \) is the saturated moisture weight gain, \( 2l \) is the thickness, and \( V \) is the volume of the specimen.

The method to characterize the moisture properties bases on the JEDEC standard \([21]\). The procedure described in the standards involves exposing a membrane specimen to a specific temperature-humidity condition and monitoring its moisture uptake in weight over time. The procedure to measure the \( D \) and \( C_{sat} \) of ACA in 85 °C/85%RH, 65 °C/85%RH and 45 °C/85%RH conditions is described as follows:

1. Preparing samples at 180 °C for 20 min in a thermal chamber. The sample is a disc shape with a radius of 15 mm and a thickness of 0.5 mm (according to the relation: \( h < 0.05 \pi r \)).
2. Weighing the dry sample and recording as \( W_0(1) \).
3. Placing the samples into the temperature-humidity chamber, and periodically weighing the sample and recording as \( W(t) \).
4. Replacing the sample into the chamber for further absorption until saturate.
5. Plotting the moisture weight gain, i.e. \( M(t) = W(t) - W_0(1) \), verses time. Using the Eq. (1) to calculate the \( D \) and Eq. (2) to calculate the \( C_{sat} \).

The \( M_t \) test data are graphically shown in Fig. 4, and Fick’s curve is used to fit the data. The results of \( D \) and \( C_{sat} \) are listed in the Table 3. Then, the \( D \) and \( C_{sat} \) can be incorporated into the finite element model to depict the moisture behavior for ACA joints under each hygrothermal condition.

4.2. Moisture concentration evaluation of ACA joints

Due to the symmetry of the structure, only one quarter of the joint is simulated for less computing time using Abaqus/Standard version 6.10. A heat-transfer analogous analysis is used to model the moisture diffusion for hygrothermal conditions \([22]\). The nodal output NT11 obtains from the heat-transfer analysis will represent the nodal moisture concentrations. The model uses Heat transfer elements (DC3D4) and consists of 52023 elements and 12023 nodes. The ACA material properties used in the model are given in Table 3. During the finite element simulation, the moisture concentrations in the ACA layer are characterized by a normalized parameter from 0 to 1, namely moisture saturation, and the moisture diffusion simulation is carried out to determine the time of moisture saturation at the ACA layer of joints under different hygrothermal conditions. If saturation is 1 (or 100%), it means the moisture absorbed in ACA is in fully saturated state.

The results of the computer simulation are graphically summarized in Figs. 5 and 6. Fig. 5 shows the moisture distribution in the ACA layer at different aging time for the three types of hygrothermal aging tests. For 85 °C/85%RH, 65 °C/85%RH and 45 °C/85%RH aging tests, the moisture concentration of the ACA is 99.7%, 99.8% and 99.8% respectively after certain early test hours, all of them are near to fully saturated state. Fig. 6 shows the moisture concentration of the ACA layer during hydrothermal aging time.

4.3. Analysis and discussion

The experimental results of shear strength and the computer simulation of moisture concentration are shown comparatively in Fig. 7. Obviously, from Fig. 7a, it can be seen that the shear strength of joints decreases rapidly from 0 to 96 h, and the moisture concentration of ACA increases rapidly in the same period. After that, the shear strength of joints keeps nearly unchanged and the moisture concentration reaches saturated. Similar results can be found in the cases of 65 °C/85%RH and 45 °C/85%RH aging tests, as shown in Fig. 7b and c. In Fig. 7b, the moisture concentration increases from 0 to 144 h and the shear strength decreases in this period. In Fig. 7c, the moisture concentration increases from 0 to 192 h and the shear strength decreases in this period. It can be said that, for each hygrothermal test, the shear strength firstly decreases with the increment of the moisture concentration and then nearly
keeps unchanged and equilibrium when the moisture concentration is saturated.

From Fig. 7, it is also found that the shear strength degradation of ACA joints takes about 96 h to reach equilibrium point under 85°C/85%RH condition, about 144 h under 65°C/85%RH condition, and about 192 h under 45°C/85%RH condition. It indicates that the shear strength decreases most quickly in 85°C/85%RH condition and most slowly in 45°C/85%RH condition. As mentioned in above section, the ACA moisture diffusion coefficient (D) of 85°C/85%RH, 65°C/85%RH and 45°C/85%RH condition is 2.01 × 10⁻⁶ mm²/s, 1.35 × 10⁻⁶ mm²/s and 1.02 × 10⁻⁶ mm²/s, respectively. This implies that larger the D is, the larger the degradation rate is.

In addition, for 85°C/85%RH, 65°C/85%RH and 45°C/85%RH tests, the saturated moisture concentration (Csat) is 11.18 kg/m³, 5.46 kg/m³ and 2.99 kg/m³, respectively. This implies that the larger the Csat is, the larger the ΔS% is.

To some extent, it is reasonable to conclude that the shear strength degradation of joints under hygrothermal aging conditions is related with the moisture diffusion coefficient (D) and the saturated moisture concentration (Csat). The affecting mechanisms of hygrothermal conditions on the shear strength can be summarized as that the shear strength decreases dramatically when the moisture is quickly diffusing and approximately keeps unchanged and equilibrium when moisture reaches saturated, and the degradation rate of the shear strength is dependent on the moisture diffusion coefficient (D) of the ACA joints. The magnitude of the shear strength decrease (ΔS%) is dependent on the saturated moisture concentration (Csat) of the ACA joints.

Hence, the moisture properties of the ACA play an important role in determining the mechanical reliability of the ACA joints. The shear strength of the ACA joints is greatly dependent on the moisture diffusion coefficient (D) and saturated moisture concentration (Csat). In order to keep high reliable, a preferred ACA material should have low D and minimum Csat.
5. Conclusions

In this study, the affecting mechanism of hygrothermal conditions on the shear strength of ACA joints and the difference from each other have been studied using experimental and simulation methods. Three groups of different accelerating hygrothermal aging tests, i.e. 85 °C/85%RH, 65 °C/85%RH and 45 °C/85%RH, have been carried out for 408 h respectively to investigate the shear strength degradation laws of joints and their difference. For each hygrothermal test, the shear strength of ACA joints decreases gradually with the hygrothermal aging time, quickly at first and then slowly, and the shear strength degradation path can be approximately divided into two phases. In the first phase, the shear strength decreases obviously with the moisture diffusion. In the second phase, the shear strength nearly keeps unchanged and equilibrium when the moisture is saturated. It can be concluded with a reasonable confidence that:

1. Different hygrothermal conditions have different effects on shear strength of ACA joints. However, the shear strength follows a similar degradation law for different hygrothermal conditions, and the shear strength difference between each other goes to an approximate constant after certain early test hours for the given test conditions.

2. The degradation rate of the shear strength is dependent on the moisture diffusion coefficient (D) of the ACA joints. The larger the D is, the larger the degradation rate is.

3. The decreasing magnitude of the shear strength (ΔS%) is dependent on the saturated moisture concentration (Csat) of the ACA joints. The larger the Csat is, the larger the ΔS% is.

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