AN APPROACH TO DETERMINING RESIDUAL STRAINS AND MOISTURE DIFFUSION COEFFICIENTS OF CURED ADHESIVES IN ELECTRONIC PACKAGING

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ABSTRACT
Polymeric adhesives are popular in the application to electronic or optoelectronic packaging for die attaching, underfilling or interconnection. Their residual strains or stresses (induced by temperature, moisture, and curing shrinkage) and moisture diffusion coefficients have to be determined and cooperated into the package design for better reliability. The purpose of this study is to propose an approach for quantifying adhesive moisture diffusion coefficients and residual strains due to chemical shrinkage, stress relaxation and temperature- and moisture-loading. This approach feature testing fully-cured adhesive/silicon bi-material plates under thermal and moisture loading using Twyman-Green (T/G) interferometry system plus analyses with Timoshenko’s bi-material theory and finite element method (FEM). Three types of adhesives: paste adhesive, film adhesives A and B have been tested for illustrating the approach. The results suggest that the residual strains for the paste adhesive are only induced by CTE mismatch during thermal loading, rather than other factors, after the cured paste adhesive being cooled down to room temperature. On the other hand, the film adhesive A was found to have the additional residual strain caused by chemical shrinkage plus stress relaxation, besides thermal one, is about $2.26 \times 10^{-3}$, which accounts for 85% of thermal strains, after the bi-material plate being cooled down to room temperature. Through moisture diffusion test, the average of the coefficients of moisture diffusion and saturated hygro-strains of the film adhesive B under $30^\circ\text{C}/85\%\text{RH}$ are obtained to be $1.09 \times 10^{-6}$ mm²/s and $1.51 \times 10^{-3}$, respectively. From the aforementioned results, it has been demonstrated that this method with a combination of experimental data and analytical tools can be able to determine the residual strains and moisture diffusion coefficients of the cured film or paste adhesives.

INTRODUCTION
Polymeric adhesives are popular in the application to electronic or optoelectronic packaging. For example, non-conductive paste adhesives are applied as die attachment in conventional packaging or underfilling in flip-chip packaging, and anisotropic conductive film (ACF) or non-conductive film (NCF) adhesives are used for conducting electronic current or holding the connection between the bumps and the electrode pads in chip-on-glass (COG) or chip-on-board (COB) packaging. In case of COG packaging, the adhesive residual strains (or stresses) caused by adhesive chemical (curing) shrinkage, stress relaxation, thermal and moisture loading, might result in the package reliability issues, such as excessive package warpage, interface delamination, and electrical resistance increasing at interconnection[1-6], shown in Fig. 1. In addition to the need of elastic modulus, Tg, and thermal coefficient of expansion (CTE) for these cured adhesives, the related residual strains or stresses (induced by temperature, moisture, and curing shrinkage) and moisture diffusion coefficient have to be determined and analyzed for better designing the joints or bump joints. In the literature, a micro-tester was set-up to measure force response of a droplet of liquid adhesive during curing in order to determine curing shrinkage stresses of non-conductive adhesive [7]. Thermal mechanical analyzer (TMA) associated with dynamic mechanical analyzer (DMA) was used to measure curing
shrinkage stresses of the ACFs [8]. A combination of TMA and thermo-gravimetric analyzer (TGA) approaches was used to the coefficients of moisture diffusion and the hygro-strains of the adhesives [4, 5, 9].

The objective of this paper is to propose an approach for quantifying adhesive moisture diffusion coefficients and residual strains due to chemical shrinkage, stress relaxation and temperature- and moisture-loading. Unlike the approaches in the literature, these coefficients and strains of the adhesives will be determined by an innovative approach in combination of experimental, theoretical and numerical methods: out-of-plane deformations of the bi-material NCF/silicon specimens during moisture absorption will be measured Twyman-Green interferometry, and based on these deformation data, Timoshenko’s bi-material theory and a finite element analysis will be employed for calculating the coefficient of moisture diffusion and the residual strains of the adhesives.

![Adhesive Residual Strains](image)

**Adhesive Residual Strains**
- Chemical
- Thermal
- Moisture
- Stress relaxation, etc.

![Issues](image)

**Issues**
- Package warpage (Mura)
- Delamination
- Electrical resistance

**EXPERIMENTAL MEASUREMENT**

Twyman-Green (T/G) interferometry is one of full-field optical interference methods which can provide an out-of-plane deformation measurement of the specular surface of the specimen [10]. The interfering fringe pattern represents displacement field governed by

\[ W(x, y) = \frac{\lambda}{2} N(x, y), \quad (1) \]

where the \( W(x, y) \) is the out-of-plane displacement on the x-y plane, the \( \lambda \) the wavelength of the laser beam, and \( N(x, y) \) the fringe order. Since the \( \lambda \) in the study is 632 nm, one fringe order difference represents a displacement of 0.316 \( \mu \)m. That is, the sensitivity of the system is 0.316 \( \mu \)m per fringe. The deformations of bi-material plates will be measured by this system for determining residual strains and moisture diffusion coefficients of the adhesives. And its curvature \( (K) \) can be calculated, based on the assumption of relatively small and spherically bending deformation, by using

\[ K = \frac{1}{\rho} = \frac{2W}{x^2}, \quad (2) \]

where the \( \rho \) is a radius of the curvature of the specimen, and the \( x \) is a pre-defined distance. For residue strain study, the size of test bi-material plates are \( 6 \times 6 \times 0.132 \) (die 0.1/adhesive 0.032) mm for paste-adhesive specimens and \( 4 \times 4 \times 0.113 \) (die 0.075/adhesive 0.038) mm for film-adhesive-A specimens. The curing conditions are at 150°C/0.5 hour for paste-adhesive specimens, and 150°C/2 hours for film-adhesive-A specimens. Note that the dummy silicon dies used in the bi-material specimens are almost perfectly flat below 0.15 \( \mu \)m in the area size of \( 4 \times 4 \) mm. The film adhesive B/silicon bi-material plates with size of about \( 4 \times 4 \times 0.105 \) (die 0.065/adhesive 0.040) mm were fabricated for moisture absorption study. After its full cure the out-of-plane deformation of the bi-material plate was immediately measured by T/G system. Then the specimens were put into a humidity chamber under the condition of 30°C/85%RH for absorbing the moisture. In every five minutes of moisture absorption, the T/G system was applied to it again to document the deformation. The curvatures of the specimen can be obtained using equations (1) and (2) associated with the T/G data before and after moisture absorption.

**THEORETICAL AND NUMERICAL ANALYSES**

**Theory of Moisture Diffusion and Induced Strain**

The moisture diffusion theory of Fick’s Law [11] can be described as follows:

\[ \frac{\partial C}{\partial t} = D \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right), \quad (3) \]

where the \( C \) is moisture concentration, the \( t \) is time and the \( D \) is the coefficient of moisture diffusion. The one-dimensional equation can be written as

\[ \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}, \quad (4) \]

A schematic of the one-dimensional diffusion is shown in Fig. 2, in which the saturated moisture concentration, \( C_s \), is on the top and bottom surfaces of the 2\( h \)-thick dry plate. During the moisture diffusion process, at beginning the moisture concentration is zero inside of the dry plate and \( C_s \) is on its top and bottom surfaces, and then increases with time until the concentration reaches \( C_s \) in the entire plate. Mathematic formulations can be expressed by

**Initial and final conditions** :

\[ C(x, 0) = 0; \quad C(x, \infty) = C_s, \quad (5) \]

**Boundary condition** :

\[ C(-h, t) = C(h, t) = C_s, \quad (6) \]

The closed-form solution to the one-dimensional diffusion problem can be obtained as

\[ \frac{C}{C_s} = 1 - \frac{4}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} \exp(-At) \cos(Bx), \quad (7) \]

where \( A = \frac{D(2n+1)^2 \pi^2}{4h^2} \) and \( B = \frac{(2n+1)\pi}{2h} \).
The moisture-induced stress (strain) is hygro-stress (strain) due to material swelling expansion after the moisture absorption. This hygro-strain ($\varepsilon_M$) can be expressed in terms of coefficient of moisture expansion ($\beta$) and moisture concentration ($C$) as

$$\varepsilon_M = \beta C, \quad (8)$$

**Timoshenko’s Bi-material Theory**

The saturated moisture-induced expansion or residual strains can be determined by T/G experiment associated with Timoshenko’s bi-material theory [12, 13]. The solution of induced curvature ($K$) for a thermally-loaded bi-material plate is described by

$$K = \frac{1}{\rho} \frac{\Delta \varepsilon}{t_1 \left( \frac{(1+nm^3)(1+mn)}{3nm(1+m)} + (1 + m) \right)}, \quad (9)$$

where $m = \frac{t_2}{t_1}$; $n = \frac{E_2/(1-v_2)}{E_1/(1-v_1)}$.

The misfit strain ($\Delta \varepsilon$) can be described as

$$\Delta \varepsilon = \Delta \varepsilon_T + \Delta \varepsilon_M + \Delta \varepsilon_R, \quad (10)$$

where thermal misfit strain $\Delta \varepsilon_T = (\alpha_2 - \alpha_1)(T - T_0)$; moisture misfit strain $\Delta \varepsilon_M = \beta_2 C_2 - \beta_1 C_1$; and residual misfit strain $\Delta \varepsilon_R = \varepsilon_{R2} - \varepsilon_{R1}$. Among the above equations, the $T_0$ is a stress-free (initial) temperature, while the $T$ is final one. The $t_i$, $E_i$, $v_i$, $\alpha_i$, $\beta_i$, $C_i$ and $\varepsilon_{Ri}$ are thickness, elastic modulus, Poisson’s ratio, and coefficient of thermal expansion, coefficient of moisture expansion, moisture content, and residual strain, respectively, for the bottom layer ($i = 1$) and top layer ($i = 2$) of the bi-material plate. The curvatures for the film adhesive B/silicon bi-material plates before and after moisture absorption can be measured by T/G system. Since the silicon cannot absorb moisture (i.e. $\beta C = 0$), the difference of both curvatures can be used to determine the moisture-induced expansion strain of the film adhesive B by the equations (9). Similarly, the residual strain of adhesives can be determined by the same calculation.

**RESULTS AND DISCUSSION**

**Mechanical Properties of Adhesives**

The elastic moduli and CTEs for the film adhesive A materials have been determined from the DMA and TMA tests, respectively, in terms of temperatures. These data can be expressed in Fig. 3 by curves with two constant lines and linear ramp for CTEs and Es with respect to temperatures. Similarly, those data for paste adhesive material, which is provided by material vendor, can be also expressed by the curves, as shown in Fig. 4. The material properties listed on Tables 1 and 2 are used in the theoretical and FEM analyses.

![Fig. 3 Elastic modulus and CTE vs. temperatures for the paste adhesive, obtained from DMA and TMA tests](image)

![Fig. 2 Illustration of one-dimensional diffusion](image)

**Table 1: Mechanical properties of materials**

<table>
<thead>
<tr>
<th>Material</th>
<th>$E$ (GPa)</th>
<th>CTE (ppm/°C)</th>
<th>$\nu$</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>Die (Silicon)</td>
<td>163</td>
<td>2.5</td>
<td>0.28</td>
<td></td>
</tr>
<tr>
<td>Paste Adhesive</td>
<td>$E_1 = 2$</td>
<td>$\alpha_1 = 75$</td>
<td>$\alpha_2 = 200$</td>
<td>0.4</td>
</tr>
<tr>
<td>Film Adhesive-A</td>
<td>by DMA</td>
<td>by TMA</td>
<td>0.4</td>
<td></td>
</tr>
</tbody>
</table>

**Table 2: Material properties for moisture diffusion**

<table>
<thead>
<tr>
<th>Material</th>
<th>$E$ (GPa)</th>
<th>$\nu$</th>
<th>$D$ (mm²/s)</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chip</td>
<td>163</td>
<td>0.28</td>
<td></td>
<td>0</td>
</tr>
<tr>
<td>Film Adhesive-B</td>
<td>$E_1 = 1$</td>
<td>0.4</td>
<td>by experiment &amp; FEM fitting</td>
<td>by experiment &amp; bi-material theory</td>
</tr>
</tbody>
</table>
Residual Strains of Paste Adhesive and Film Adhesive A

Typical results of fringe patterns (warpage) are shown in Fig. 5 for paste adhesive/die bi-material plate under heating from room temperature (29°C) to 110°C. The density of the con-centric fringes decreases with temperature increasing, and almost becomes zero at 110°C. Further through data reduction by equation (2), the resulting curvatures for three specimens are plotted against various temperatures, as shown in Fig. 6 along with theoretical and FEM predictions (which only takes into account adhesive thermal strains). The well consistent results among them indicate that the paste adhesive has only thermal strain, without others (such as residual strains due to chemical shrinkage or stress relaxation) after the specimens cured and right cooled down to room temperature.

The film adhesive A has also been investigated using the same approach. The typical fringe patterns (warpage) of a dry adhesive A/die bi-material plate under thermal cycling are shown in Fig. 7. Similar to one with the paste adhesive, the fringe patterns of this specimen are con-centric and fringe density decreases with temperature increasing. And the fringe density approaches to zero near 110°C and returns to the original fringe density after the specimen rapidly cooled down to the room temperature. It is worth mentioning that this thermal cycling test had to be completed in a short period of time (about 20 min in heating, and 20 sec in cooling) to prevent the specimen from absorbing moisture. The resulting curvatures of two specimens are plotted against the temperature in Fig. 8 and compared with those from Timoshenko’s solution and FEM results. The curvature gradients (slope) with respect to temperature for these three solutions are almost the same in the temperature region below the Tg, but far off above the Tg. This means that the temperature-induced deformations of the
specimens are well addressed by Timoshenko and FEM solutions at the temperature below the Tg, but not above the Tg (probably due to the time-dependent material properties of the adhesive). However, there is a large discrepancy of the curvature values between experimental and analytic (Timoshenko and FEM) results at the temperature below the Tg, although Timoshenko and FEM results are very consistent. This discrepancy implies that some residual strains, other than thermal strains, of the adhesive have contributed into it. This residual strain would be possibly from the chemical shrinkage plus stress relaxation of the adhesive A during curing above the Tg, and can be determined by putting the difference of the curvature (ΔK) into equation (9). The residual strain is calculated as about 2.26×10⁻³ at the room temperature, which is close to 85% of thermal strains after the cured adhesive A cooled down to room temperature.

**Moisture Diffusion Coefficient and Hygro-Strain of Film Adhesive B**

The coefficients of moisture diffusion and saturated hygro-strains of the film adhesive B at the condition of 30°C/85%RH have been determined with a combined approach of Twyman-Green experiment, bi-material theory and FEM. Typical resulting curvature of a bi-material plate, as shown in Fig. 9, changes with time during the moisture absorption at condition of 30°C/85%RH. It is shown that the bi-material plate with curvature of 1.97×10⁻³ 1/mm right after full cure at 150°C/10mins starts to change its curvature with time increasing, when absorbing the moisture at 30°C/85%RH condition. After 30 min, its curvature reaches to 0.94×10⁻³ 1/mm and becomes almost constant. This curvature difference (Δk = 1.03×10⁻³ 1/mm) was used for calculating saturated hygro-strains of the film adhesive B using equation (9) by assuming moisture saturation at 30 min. The saturated hygro-strain for the film adhesive B at 30°C/85%RH condition is 1.6×10⁻³. Prior to determining the coefficients of moisture diffusion, the transient moisture analysis of a FEM model was validated by comparing with the close-form solution in equation (7). The results of level of moisture saturation along the line P’P (central line through the thickness of the adhesive) was plotted with different times for both solutions. It is shown that both solutions are almost identical. Those curvature data in Fig. 9 can be plotted again with curvature change (curvature difference between at initial and current states) vs. time, and shown in Fig. 11, associated with validated FEM calculation for best fitting and then determining the coefficients of moisture diffusion (D=1.1663×10⁻⁶ mm²/s obtained). Three bi-material test specimens have been analyzed and their moisture diffusion coefficients and saturated hygro-strains of the film adhesive B are shown in Fig.12. The results show that the average moisture diffusion coefficients and saturated hygro-strains of the film adhesive B under 30°C/85%RH are 1.09×10⁻⁶ mm²/s and 1.51×10⁻³, respectively.
CONCLUSIONS

This study aims to propose an approach for quantifying adhesive moisture diffusion coefficients and residual strains due to chemical shrinkage, stress relaxation and temperature- and moisture-loading. This approach involves testing the adhesive/silicon bi-material plates under thermal and moisture loading using Twyman-Green interferometry plus analyses with Timoshenko’s bi-material theory and finite element method. Three types of adhesives including paste adhesive, film adhesives A and B have been tested in this study for illustrating the approach. The results suggest that the residual strains for the paste adhesive are only induced by CTE mismatch during thermal loading, rather than other factors, after the cured paste adhesive being cooled down to room temperature. By contrast, the film adhesive A was found to have the additional residual strain caused by chemical shrinkage plus stress relaxation, besides thermal one, is about $2.26 \times 10^{-3}$, which equals 85% of thermal strains, after the bi-material plate being cooled down to room temperature. Through moisture diffusion test, the average of the moisture diffusion coefficients and saturated hygrostrains of the film adhesive A under 30°C/85%RH are obtained to be $1.09 \times 10^{-6}$ mm²/s and $1.51 \times 10^{-3}$, respectively. Accordingly, this method combining with experimental data and analytical tools has been demonstrated to enable determining the residual strains and moisture diffusion coefficients of the cured film or paste adhesives.

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