RESIDUAL STRESSES AND THERMOPLASTIC ADHESIVE ENVIRONMENTAL CRACKING

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SUMMARY: The work presented here is concerned with thermoplastic-bonded joint durability. Residual stresses in the thermoplastic adhesive layer are generated in thermal bonding process. In application, joints will face various environmental attacks both at the adhesive itself and at joint interfaces. In this study, transparent glass/polycarbonate/glass sandwich samples were used. Adhesive cracking was observed during environmental soaking of the sandwich samples. Furthermore, the adhesive cracking phenomena occurred only when the glass adherend was roughened before bonding. Smooth glass adherend did not cause the adhesive cracking. A finite element model revealed the effects of local adherend surface feature on the residual stress state in the adhesive layer. Residual stresses in the adhesive layer were demonstrated as a primary reason for the cracking.

KEYWORDS: residual stress, durability, adhesion, crack, surface topography

INTRODUCTION

Joint durability has long been one of major concerns in the adhesive bonding area. Most of past research in joint durability has focused on thermosetting adhesives. By combining fatigue load and environmental soaking, recent research has made important progress in many aspects of joint durability [1, 2, 3]. However, thermoplastic adhesive bonding is now finding more and more applications. Bonding with thermoplastic adhesive can be processed very fast [4]. The Thermoplastic bonding can also be incorporated into continuous manufacturing processes such as injection molding [5] and of sandwich structures. A study on the durability of the thermoplastic adhesive joint system is demanded now. Differential material properties between the thermoplastic adhesive and its adherend such as metal or ceramic generate residual stresses at the interface in cooling down the joints. The residual stress development along with the thermoplastic adhesive bonding process was demonstrated in our previous study [6]. The current study is aimed to investigate thermoplastic adhesive joint durability related to residual stresses.

Joint durability is closely related to its application environments such as water, ions in water, solvents, temperature, and loading conditions. Some of them predominantly influence the interface, but some attack the thermoplastic adhesive also. Water has been the most harmful and most commonly encountered environment. Ions generally exist in water such as sodium and chlorine. Al-Momany and Zihlif (1996) studied the effect of Dead Sea water on the mechanical properties of polycarbonate [7]. Effects of solvents were considered in Arnold's study (1995) [8]. Temperature may enhance the degradation of joints and adhesives. Furthermore, environments

could change the failure mode from a predominantly cohesive failure to adhesive failure or vise versa or from one interface to another [9]. Capturing the failure mode in designing joint durability tests is very important in the investigation of the joint failure mechanisms.

For glassy polymers, environmental stress cracking (ESC) is a common cause of failure and its understanding remains incomplete [8]. Many aspects of the polymer such as its chemical compatibility, rate of liquid diffusion, viscoelastic behavior, craze initiation and growth, all play important roles.

In this study on bonding with glassy thermoplastic adhesives, a residual stress related environmental stress cracking phenomena is found in the adhesive layer. Furthermore, the local adherend surface features were found to be the primary cause of the ESC of the adhesive layer. The near interface residual stresses were noticed for the first time.

EXPERIMENTAL

Adherend and Adhesive

A glass/thermoplastic/glass sandwich was chosen as our simulation system. The environmental attacks at the interface or in the thermoplastic can be easily characterized due to the transparency of glass. Glass also has very small thermal expansion compared with thermoplastics and high residual stress will develop in the joints. However, since glass is brittle, high residual stresses at the interface may damage the glass shortly after bonding process. Drory, Thouless, and Evans (1988) also observed a glass cracking phenomena [10]. Therefore, thermoplastics with relatively low modulus such as polycarbonate (elastic modulus, 1.8 GPa) are required to prevent glass cracking. Transparent polycarbonate film (0.254 mm thick) was used for this study.

Two types of glass adherend surface preparations were used in this study, smooth original glass surface (Ra = 0.03 μm) and 60 alumina grit blasted surface (Ra = 4.5 μm).

Environmental Conditions

Five environmental solutions considered in this study: (1) deionized water; (2) deionized water with 0.65% NaCl by weight; (3) deionized water with 0.02% $NaHCO_3$ by weight; (4) deionized water with 0.2% dextrose by wiegh; and (5) ethyl alcohol. Two temperatures, 40 ^{o}C and room (25 ^{o}C), were considered in the study.

Bonding process

The bonding of glass/polycarbonate/glass sandwich was performed in a computer-controlled joining fixture [11]. The processing temperature of polycarbonate is ~280 ^{o}C. A pressure of 206.9 KPa was applied during the bonding process.

Experimental Results and discussions

The glass/polycarbonate/glass sandwiches were bonded and soaked in different environments at different temperatures. Three samples were used for each experiment. The samples were checked every 12 hours. For *rough glass adherends*, Table 1 summarizes the soaking time at which the

polycarbonate layer started to crack. Figure 1 shows the crack patterns. All the cracks started from the edges of the sandwich samples and then propagated and branched toward the inside of the samples. Adhesion between polycarbonate and glass were lost in the areas with cracks. The polycarbonate cracking and adhesion loss seemed to occur simultaneously. For *smooth glass adherends*, no polycarbonate layer cracking was noticed.

Table 1: Crack initiation time of adhesive (PC) in-between two roughened glass plates in soaking

Soaking environment temperature	Deionized water	Deionized water with 0.65 % NaCl	Deionized water with 0.02% $NaHCO_3$	Deionized water with 0.2% Dextrose	Ethyl Alcohol 190
40 ^oC	No crack was observed	60 hours	36 hours	36 hours	280 hours
25 ^oC	No crack was observed	No crack was observed	280 hours	280 hours	No crack was observed

Our previous studies on the thermoplastic adhesive bonding indicated significant residual stress development in bonding process [6]. However, in this study, the adherend surface topography related polycarbonate environmental crack phenomena required more careful study on the near interface residual stress state in the polycarbonate layer.

Combining with increased temperature (from 25 ^oC to 40 ^oC), ions, Na^+, Cl^-, and HCO_3 in water, dextrose in water, and Ethyl Alcohol enhanced the polycarbonate crack initiation and development. However, the chemical aspects will not be further discussed in this paper.

RESIDUAL STRESSES

Bonding process induced residual stress was first modeled with smooth adherend and then the influence of the local adherend surface features on the residual stress state was modeled with finite element method. In addition, the temperature increase and water absorption were also considered.

Mechanical and Thermal Properties of the Materials

The temperature-dependent mechanical properties of polycarbonate were measured with DMA2980 from TA Instruments (Figure 2). The test was performed at a frequency of 10 Hz. Table 2 shows the other material properties for our modeling.

Incremental Elastic Model

According to the time-temperature superposition study of the polycarbonate [12] and the slow cooling process (~ 1 ^oC/minute), the polycarbonate is viscous above its glass transition temperature. It is reasonable assume that all the residual stresses developed above polycarbonate's glass transition temperature (T_g = 150 ^oC) relax. We used incremental elastic

model validated in our earlier study to evaluate the residual stresses developed in the polycarbonate layer during the cooling process [6].

Table 2: Elastic and thermal properties of materials

	Elestic modulus (GPa)*	Poisson's ratio*	Thermal expansion coefficient (x 10^{-6})	Water absorption (%)*
Polycarbonate	1.8	0.38	67.5	0.35
Glass	68.95	0.2	8.36	-
Steel	190.0	0.30	15.0	-

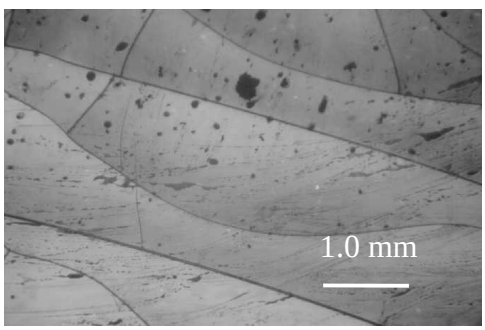

Figure 1: Crack patterns of polycarbonate in-between two roughened glass plates.

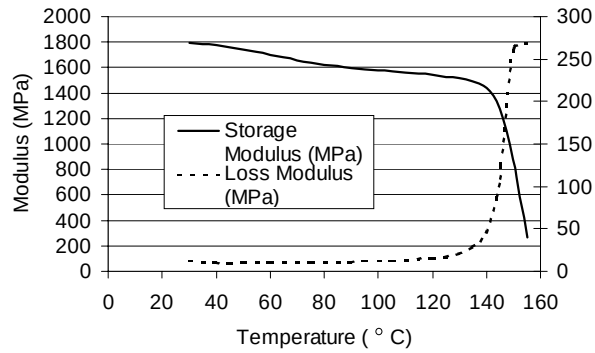

Figure 2: Temperature-dependent mechanical properties of polycarbonate

At a temperature step T_i, the incremental stress $\Delta\sigma_i$ is generated by the incremental thermal strain $\Delta\varepsilon_i$

$$\Delta\varepsilon_i = (\alpha_p - \alpha_g)\Delta T_i \tag{1}$$

$$\Delta\sigma_i = -\frac{E_p(T_i)\Delta\varepsilon_i}{(1-\gamma_p)} \tag{2}$$

where α_p : thermal expansion coefficient of the polymer

α_g : thermal expansion coefficient of the glass

$E_p(T_i)$: elastic modulus of the polymer at temperature T_i

γ_p : Poisson's ratio of the polymer

Adding this incremental stress to the stress σ_{i-1} at last temperature step T_{i-1}, the present stress σ_i will be

$$\sigma_i = \sigma_{i-1} + \Delta\sigma_i \tag{3}$$

For the smooth glass adherend, the calculated tensile residual stress with the incremental elastic model is 17.57 MPa in the polycarbonate layer at room temperature, which is about 28% of the tensile strength of polycarbonate.

As the environmental temperature increases to 40°C, the tensile residual stress in the polycarbonate layer will decrease to 15.22 MPa without considering the water absorption of polycarbonate.

GE reported that the water absorption of polycarbonate is 0.35% by weight. Densities of water and polycarbonate are 1.0 g/cm^3 and 1.2 g/cm^3. Therefore the water absorption of polycarbonate is 0.42% by volume (Equation 4). Water absorption induced linear strain is about 0.14% (Equation 5).

$$\frac{dV}{V} = \frac{1}{1 + \frac{1-\alpha_w}{\alpha_w}\frac{\rho_w}{\rho_p}} \tag{4}$$

$$\varepsilon_w = \frac{dl}{l} = \frac{1}{3}\frac{dV}{V} \tag{5}$$

Where α_w : polycarbonate water absorption by weight

ρ_w : density of water

ρ_p : density of polycabonate

ε_w : water absorption induced strain in polycarbonate

Including the water absorption into the linear incremental elastic model, the residual stress is 13.85 MPa at room temperature and 12.00 MPa at 40°C without considering the mechanical property change of the polycarbonate due to water absorption. Our tensile testing of the polycarbonate did not indicate significant mechanical property change due to water absorption.

$$\sigma_{T,w} = \frac{E_T \varepsilon_w}{1-\gamma_p} \tag{6}$$

$$\sigma_T = \sigma_{T,t} - \sigma_{T,w} \tag{7}$$

Where $\sigma_{T,w}$: water absorption induced residual stress at temperature T (either room temperature or 40°C in this study)

σ_T : residual stress at temperature T

$\sigma_{T,t}$: thermal shrinkage induced residual stress at temperature T calculated from the elastic incremental model

Finite Element Modeling

In order to study the rough glass adherend surface related polycarbonate cracking phenomena, a finite element model is developed to for our near-interface residual stress study.

Rough Adherend Surface Features and FEM model

By varying adherend macro-roughness, where surface feature have dimensions in the region of 1 μm or greater, Critchlow and Brewis (1995) studied the influence of the surface macro-roughness on the durability of epoxide-aluminum joints [13]. Critchlow and Brewis immersed their sample in deionized water for a prescribed time before testing. Critchlow and Brewis observed that grit blasting was not necessarily favorable for good joint durability. Keisler and Lataillade (1995) studied the influence of surface roughness on the wettability [14]. Kramer (1983) stated that residual stresses from molding operations were often sufficient to cause

environmental-assisted cracking [15]. Adams and Harris (1987) found that the local geometry, corner in a single lap joint, could influence the joint strength significantly [16]. Rounding the ends of the adherends and filleting the adhesive at the edges could be the effective way to improve the single lap joint strength. In this study, by combining the surface topography analysis with the thermal residual stress modeling, for the first time, the influence of local adherend surface feature on the adhesive cracking was demonstrated.

Figure 3 shows a scanning electron microscope analysis of a joint interface. Various types of joint interface features were noticed. For our model, three types of features, smooth curved, triangular, and rectangular, were used (Figure 4). Based on the symmetric geometry condition, a thermal-stress FEM modeling unit is shown in Figure 5.

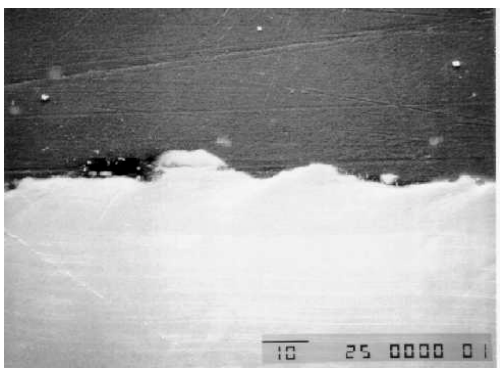

Figure 3: Scanning electron microscope (SEM) analysis of joint interface

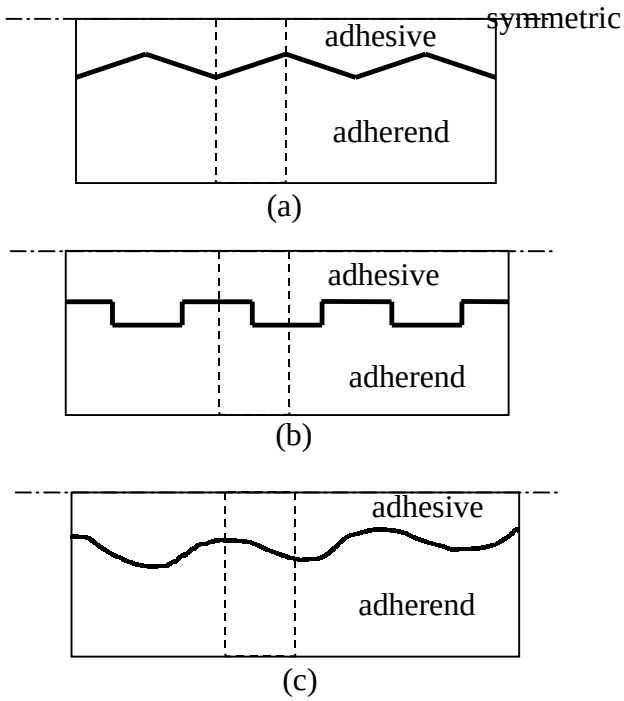

Figure 4: Interface features for modeling, (a) triangular, (b) rectangular, and (c) smooth curved (All the interface corners were rounded in modeling.)

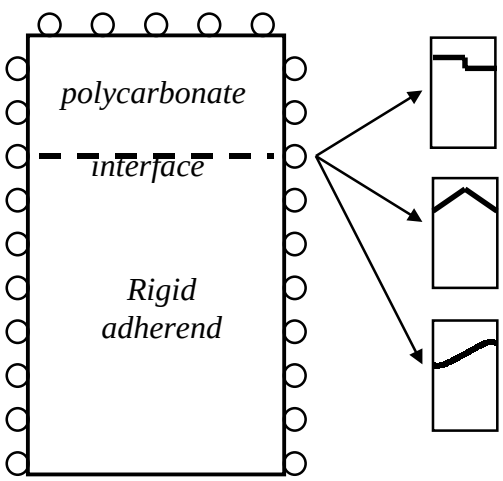

Figure 5: Modeling unit (All the interface corners were rounded in modeling)

A finite element analysis package, ABAQUS 5.6-1, was used in this study. Eight-node biquadratic element type was used. The residual stress was assumed to start at T_g of polycarbonate (150°C). In the model, the polycarbonate expansion induced by water absorption

was subtracted from the thermal shrinkage of the polycarbonate. However, mechancial properties were assumed not to be influenced by the water absorption.

FEM Modeling Results and Discussions

For smooth glass adherend, the residual stress calculated using the thermal-stress FEM model was 11.75MPa in the polycarbonate layer at 40°C, including the water absorption properties of the polycarbonate. The thermal stress predicted by the FEM model was comparable with that of the incremental elastic model.

For rough glass adherend, the stress distribution in polycarbonate was quite different in the valleys and at the peaks of the adherend surface. *In the valleys*, the stress state was tri-axial tensile (Figures 6, 7, and 8). A tri-axial tensile stress state is not good to the interface strength and durability and it may also cause brittle fracture of the adhesive itself [17]. The region with tri-axial tensile stress state could also be a potential interface defect locus. *At the peaks* of the adherend surface feature, the residual stress was compressive in the thickness direction of the sandwich sample (Figures 6, 7 and 8). The compressive stress in the thickness direction might be good for the interface, but large compression could also cause fracture of the adhesive especially when the corners of local surface features are sharp and the residual stresses are in tension in the lateral directions.

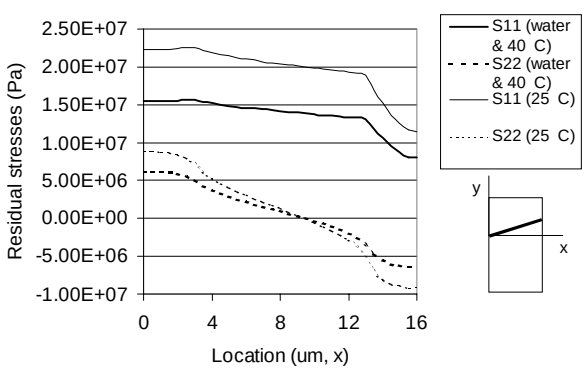

Figure 6: Residual stress distribution along interface (with triangular interface feature, S11 stands for stress in x-direction, S22 stands for stress in y-direction)

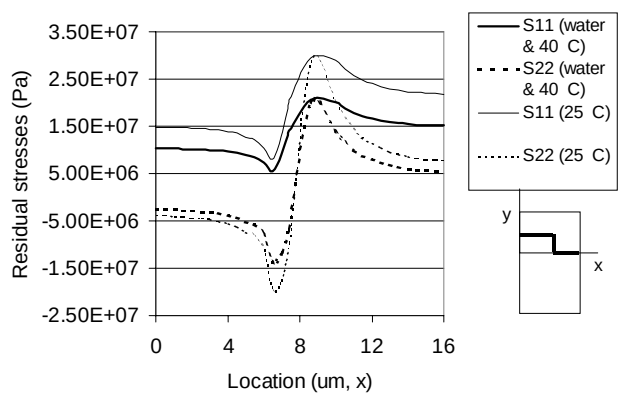

Figure 7: Residual stress distribution along interface (with rectangular interface feature, S11 stands for stress in x-direction, S22 stands for stress in y-direction)

In the model, the valleys and peaks of the adherend surface features were evenly spaced. In reality, surface features can be dominated by peaks ($S_k > 0$) or valleys ($S_k < 0$) (see Figure 9). S_k is a parameter used to describe the symmetry of height distribution of surface features. Surface features can also be nodular structures (Figure 9). Local adherend surface features and their interactions can make the residual stress state very complicated. However, the surface features selected in our model were designed to capture the typical stress state in the adhesive layer.

In the above model, the local mechanical properties of the polymer were not differentiated from the global properties. The material performance at micron level will need to be studied and understood in the future. When dealing with high stress concentration, nonlinear mechanical

performance and creep of the polymers will need to be incorporated into our model. In addition, our model can also be used for joint design with complex substrate shape at a macro level.

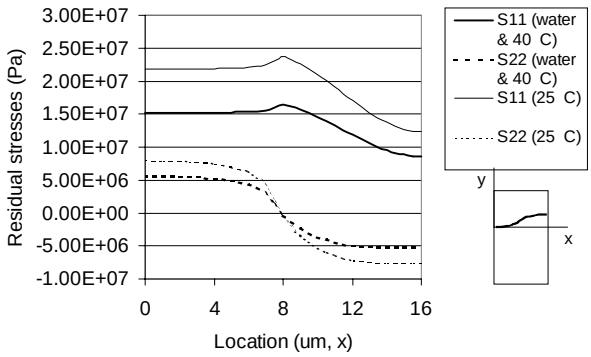

Figure 8: Residual stress distribution along interface (with smooth curved interface feature, S11 stands for stress in x-direction, S22 stands for stress in y-direction)

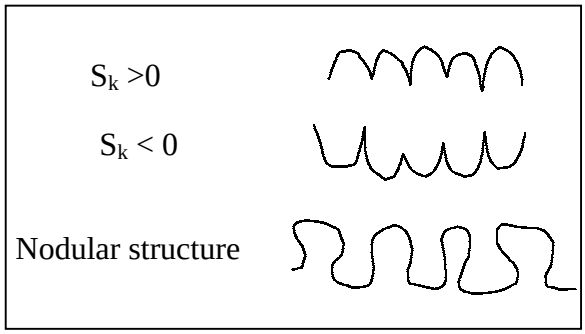

Figure 9: Adherend surface features

CONCLUSIONS

Residual stresses are commonly encountered in adhesive bonding. The residual stress induced environmental thermoplastic adhesive cracking was revealed in this study. A glass/polycarbonate/glass sandwich sample was successfully selected to demonstrate effects of the residual stresses and environments on the joint cracking and durability. It was found that the polycarbonate in-between two roughened glass adherends cracked in environmental soaking of the sandwich sample, however, the polycarbonate in-between two smooth glass adherends did not.

Adherend surface roughness was shown to enhance the adhesive cracking by changing the residual stress distribution in the adhesive. Both temperature and water absorption were incorporated into the incremental elastic model. Three types of adherend surface features were selected based on the SEM analysis of the interface. A FEM model was developed to investigate the local adherend surface feature effects on the residual stress distribution.

The experiments also demonstrated water or solvent, temperature, and ions in water could accelerate the loss of adhesion and adhesive cracking in combination with residual stresses in the thermoplastic adhesive layer.

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