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Processing and fracture behavior of a polyethylene-based thermoplastic adhesive and a glass-fiber filled epoxy adhesive

Wenping Zhao, Karthik Ramani*, Barret E. Mueller

Composites and Polymer Processing Laboratory (CPPL), School of Mechanical Engineering, Purdue University, West Lafayette, IN 47907-1288, USA

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Abstract

The processing and fracture performance of a high-density polyethylene (HDPE)-based thermoplastic adhesive and an epoxy-based thermosetting adhesive with a glass fiber additive were studied. Differential scanning calorimetry (DSC) was used to characterize the processing parameters. Double cantilever beam (DCB) fracture testing and butt-joint tensile testing were used to characterize the adhesive joint fracture toughness and strength. Although the thermoplastic adhesive had low strength, it exhibited high DCB fracture toughness because of a large plastic deformation zone in the fracture process. The fiber additive in the thermosetting adhesive stabilized the DCB fracture. Loading speed, adhesive thickness, pre-crack length, and curing temperature were all considered in the study. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Adhesive; Thermoplastic; Fracture; DCB

1. Introduction

Fracture toughness, path, and analysis have recently become effective for characterizing adhesive joint performance. Earlier studies focused on joint fracture of traditional adhesives such as epoxy-based thermosetting adhesives. Bascom and Cottington [1] and Kinloch and Shaw [2] used an elastic–plastic fracture model to interpret the fracture-toughness-related phenomena by incorporating a small plastic deformation zone into the adhesive layer. Adhesive thickness, fracture testing temperature, adherend width, and loading speed were all related to the fracture toughness by Kinloch and Shaw. Truong [3] compared the fracture behavior of unmodified and rubber-modified epoxy adhesive systems. Truong showed that fracture propagated in a brittle, stable manner or a stick/slip unstable manner with the unmodified epoxies, but the fracture shifted to a ductile, stable propagation with the rubber-modified epoxies.

Hot-melt adhesives are very flexible adhesives. Tse et al. [4] used T-peel testing to characterize hot-melt joint strength. They found that viscoelastic properties were significant issues for the hot-melt adhesive and that the joint peel strength was highly dependent on peeling rate.

This study is intended to characterize the fracture performance of high-density polyethylene (HDPE)-based thermoplastic adhesive, Bemis 6343, and epoxy-based thermosetting adhesive, FM 300-2. The Bemis 6343 is not as flexible as the hot-melt adhesive and also not as brittle as the epoxy or rubber-toughened epoxy adhesives. The FM 300-2 has a glass fiber additive in it. Processing, adhesive layer thickness, loading speed, curing temperatures, and pre-crack length are all considered with the adhesives. Double cantilever beam (DCB) specimens with aluminum adherends are used to characterize the fracture performance of the adhesive joint. Butt-joints are used to find the adhesive joint strength.

2. Processing

The two adhesives, Bemis 6343 and FM 300-2, were first studied with differential scanning calorimetry (DSC). The DSC analysis revealed the thermal properties of the two adhesives, including glass transition, melt, and curing. The processing parameters for our DCB and butt-joint specimens were then investigated.

2.1. Differential scanning calorimetry (DSC)

The thermoplastic adhesive, Bemis 6343 from Bemis Associates Inc., Shirley, Massachusetts, was used and

* Corresponding author. Tel.: 765-494-5725; fax: 765-494-0539.
E-mail address: ramani@ecn.purdue.edu (K. Ramani).

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had a melt temperature of ~ 140°C and recrystallization temperature of ~ 100°C. Our DSC analysis indicated that the recrystallization of Bemis 6343 took less than 5 min. Ramani et al. [5] revealed that recrystallization is very important to thermoplastic adhesive joint strength. High crystallinity will result in high joint strength. The design of the cooling process is critical to achieve high-strength thermoplastic adhesive joints.

The thermosetting adhesive film, FM300-2 from Cytec Fiberite Inc., Havre de Grace, MD, had a cure temperature range from 120 to 180°C. Three different curing conditions, curing at 120, 150, and 177°C, were studied with DSC. It took approximately 60 min for FM 300-2 to cure at 120°C, approximately 20 min at 150°C, and approximately 15 min at 177°C. The degree of cross-linking is dependent on both curing temperature and soaking time at the curing temperature. Cross-linking is very important to joint strength development.

2.2. Adhesive joining

The aluminum adherends were grit-blasted with 60 alumina grit and cleaned with soap water, deionized water, and isopropyl alcohol before bonding. The surface roughness of the aluminum was measured at 2.50 μm.

In preparation of the adhesive joints, pressure during the polymer's viscous liquid state is critical for promoting adhesive penetration into the micro holes of the adherend surface [6,7]. Thermosetting adhesives are usually low-viscosity liquids at room temperature or even lower temperature. Therefore, applying appropriate pressure is crucial during early stages of the thermosetting adhesive bonding process. Once thermosets cure, the bonding pressure will not change the resulting joint performance. In making our thermoset DCB specimens, we used a pressure of 137.9 kPa. Usually thermoplastic adhesives are viscous between melt and recrystallization. After recrystallization, thermoplastics start to develop strength. Processing pressure is very critical to the joint strength in the molten state and the subsequent cooling stage until glass transition stage of the thermoplastic adhesive. For the Bemis 6343, we applied a pressure of 206.85 kPa in making our DCB specimens.

Fig. 1 shows our typical DCB processing temperature profile with the Bemis 6343 adhesive. For FM 300-2, we used three curing conditions, soaking the joint at 120°C for 90 min, at 150°C for 60 min and at 177°C for 60 min.

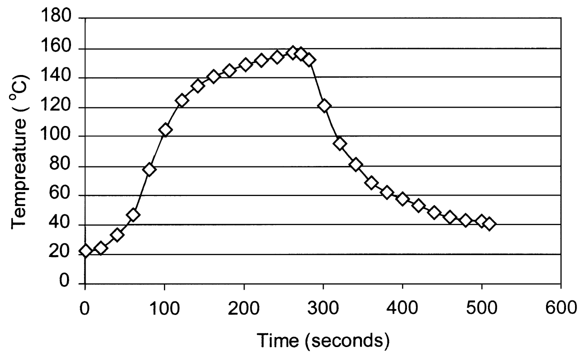

Fig. 1. DCB processing temperature profile for Bemis 6343.

3. Fracture characterization

3.1. Double cantilever beam specimens (DCB)

Double cantilever beam (DCB) and contoured double cantilever beam tests are widely used to characterize the fracture performance of adhesive joints. Much of the earlier work concerned thermosetting adhesives, such as epoxy and rubber-toughened epoxy adhesives [2,8,9]. Joint fracture performance of a thermoplastic adhesive was investigated using DCB tests in this study. The fracture toughness was calculated by [ASTM D 3433 - 93]

$$G_{\mathrm{Ic}} = \frac{4P_{\mathrm{c}}^2(3c^2 + t^2)}{Et^3B^2}, \tag{1}$$

where P_{c} is the load at crack length c (N), c the crack length at load P_{c} (m), E the elastic modulus of aluminum adherend (Pa), t the aluminum adherend thickness (m), and B the aluminum adherend width (m).When $3c^2$ is much larger than t^2, the term $3c^2 + t^2$ can be reduced to $3c^2$. In our DCB testing, the load was observed to reach maximum and then the crack started to propagate. The G_{Ic} was calculated using the maximum load and the corresponding pre-crack length.

Plastic deformation of the aluminum adherend is a concern in the DCB testing. The maximum stress in the length direction of the DCB specimen can be calculated using

$$\sigma = \frac{6P_{\mathrm{c}}c}{Bt^2}. \tag{2}$$

Combining Eqs. (1) and (2), we get the following equation (3) ($3c^2 + t^2$ is taken as $3c^2$):

$$\sigma = \sqrt{\frac{3G_{\mathrm{Ic}}E}{t}}. \tag{3}$$

Therefore, by increasing the adherend thickness, we can avoid plastic deformation of the aluminum adherend. In our DCB testing, we used 9.53-mm-thick aluminum adherends for the loading speed study of Bemis 6343 DCB joints. Other DCB specimens were all 6.35-mm-thick. Plastic deformation of the aluminum adherends was avoided. The width of our DCB specimens was 25.4 mm. The length of the DCB specimens was 250 mm.

3.2. Pre-crack length

Thin Teflon film was placed in between aluminum and the adhesives before bonding to create pre-crack. A series of three different pre-crack lengths was studied with Bemis 6343, 50, 75, 100 mm. A constant loading speed of 2.54 mm/min was used to study the pre-crack effects on the fracture performance of the DCB specimens. Four specimens were tested with each pre-crack length. Results were represented as average ± standard deviation.

3.3. Adhesive thickness

Adhesive thickness was controlled with a steel spacer. For Bemis 6343, a series of four adhesive thicknesses was used: 0.20, 0.30, 0.40, and 0.50 mm. For FM300-2, only one adhesive thickness was used: 0.40 mm. Four specimens were tested with each adhesive thickness for both adhesives. Results were represented as average ± standard deviation.

3.4. Loading speed

The loading speed effects on the DCB fracture toughness were also studied. For Bemis 6343, the loading speeds were 2.54, 25.4, 127.0, 254.0, 381.0, and 508.0 mm/min. For FM300-2, the loading speeds were 2.54 and 508.0 mm/min. Four specimens were tested at each loading speed for both adhesives. Results were represented as average ± standard deviation.

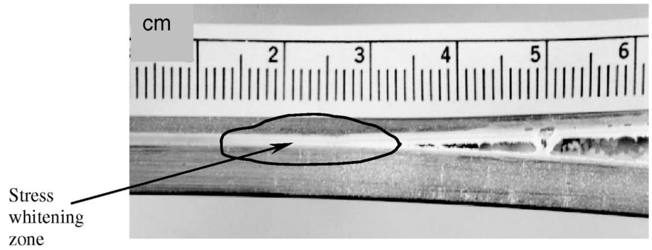

Fig. 2. Double cantilever beam joint testing with thermoplastic adhesive (Bemis 6343).

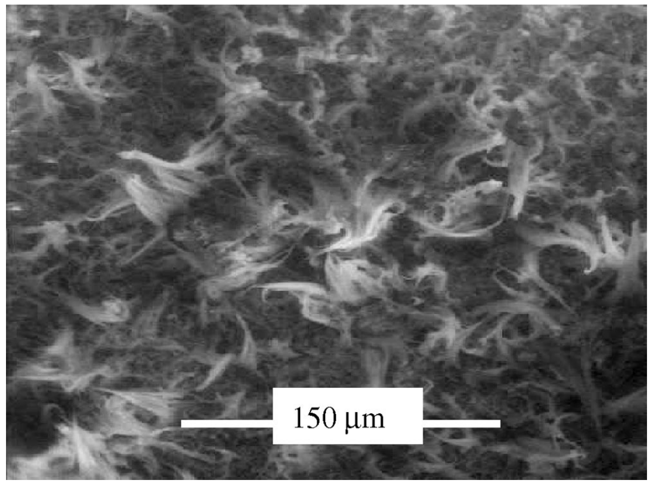

Fig. 3. ESEM analysis of DCB fracture topography (Bemis 6343).

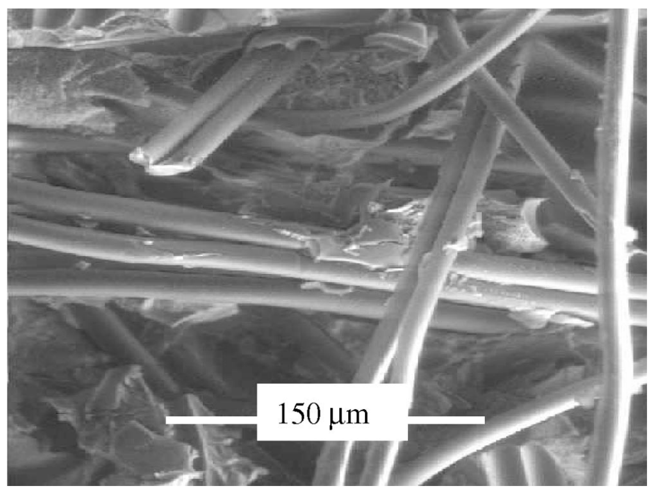

Fig. 4. ESEM analysis of DCB fracture topography (FM 300-2, cured at 120°C).

4. Results and discussion

By analyzing the DCB testing results, we demonstrated that the adhesive plastic deformation significantly influenced the adhesive joint fracture process. Environmental scanning electron microscope (ESEM) analysis of the fracture topography revealed the curing temperature effects on the FM 300-2 DCB fracture performance. The effects of pre-crack length, adhesive thickness, and loading speed on DCB fracture were discussed next.

4.1. Adhesive plastic deformation

The adhesive plastic deformation was clearly seen in testing the thermoplastic Bemis 6343 bonded DCB specimens. However, little adhesive plastic deformation was found in the thermosetting FM 300-2 bonded DCB specimens. A long plastic stress-whitening zone (about 20 mm) was shown in Fig. 2 for Bemis 6343. ESEM analysis of the fracture surfaces also clearly demonstrated more plastic deformation with Bemis 6343 than with FM 300-2 (see Figs. 3–5). Furthermore, FM 300-2 cured at 177°C was more brittle than the FM 300-2 cured at 120°C and, correspondingly, allowed less fiber stretching in the DCB testing (Figs. 4 and 5). A stable crack propagation during the FM 300-2 DCB testing was observed as the result of the fiber entanglement.

Our butt-joint testing indicated the strength of the Bemis 6343 was much lower than the FM 300-2 (Fig. 6).

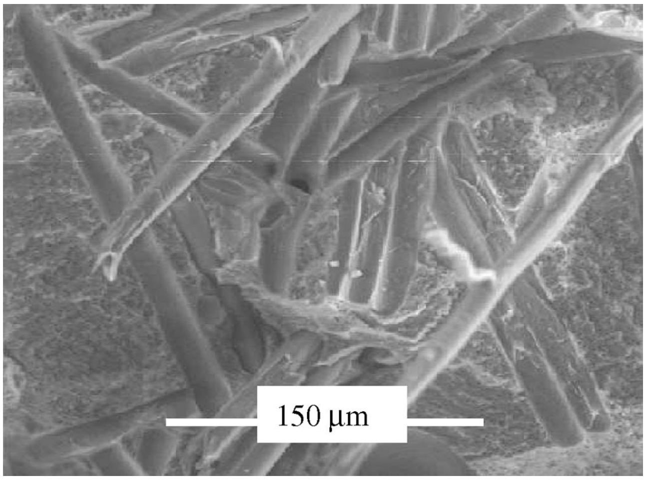

Fig. 5. ESEM analysis of DCB fracture topography (FM 300-2, cured at 177°C).

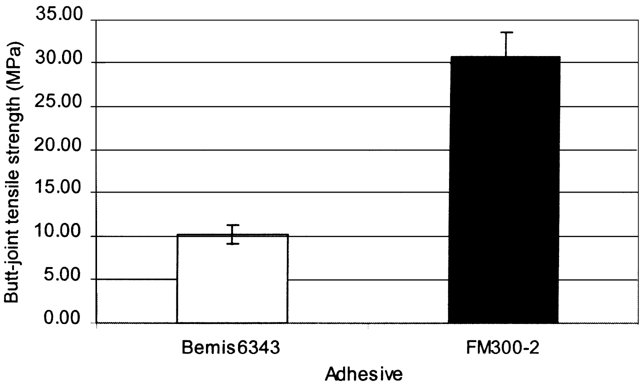

Fig. 6. Butt-joint strength (average ± standard deviation, four specimens for each adhesive, loading speed = 2.54 mm/min).

However, the fracture toughness of Bemis 6343 DCBs was higher than that of the FM 300-2 DCBs (Figs. 6–10). Plastic deformation of the adhesive was considered significant in DCB testing. A similar mechanism can be found in epoxy and rubber-toughened epoxy [2,9].

4.2. Effects of pre-crack length

Because of the long plastic deformation zone, it is difficult to characterize the crack length during crack growth for the Bemis DCB specimens. We made specimens with three different pre-crack lengths to observe crack length effect on DCB fracture toughness. The pre-crack lengths used were 50, 75, and 100 mm. Fig. 7 shows that there are no significant effects of pre-crack length on the fracture toughness with the Bemis 6343 adhesive.

4.3. Effects of adhesive thickness

Based on the fracture micromechanics of adhesive joints [9], it is possible that adhesive thickness could

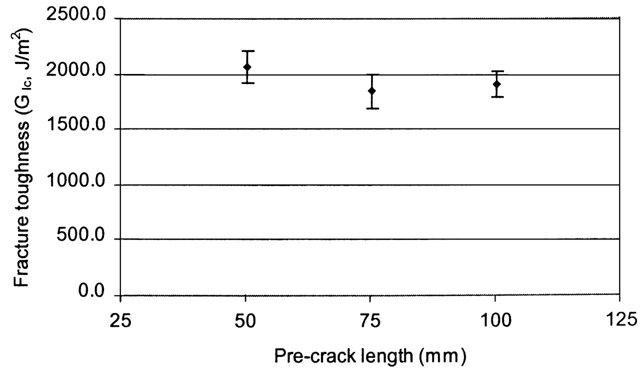

Fig. 7. Effects of pre-crack length on DCB fracture toughness (Bemis 6343, adhesive thickness = 0.4 mm, loading speed = 2.54 mm/min).

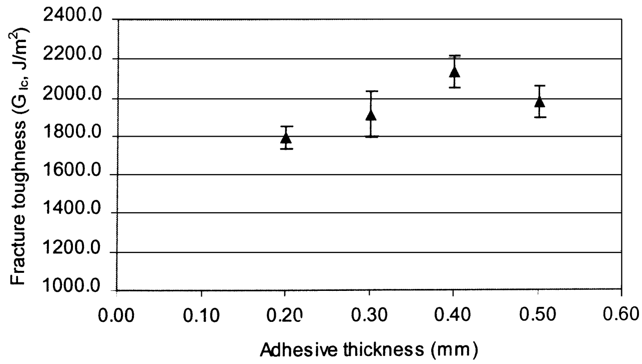

Fig. 8. Effects of adhesive thickness on DCB fracture toughness (Bemis 6343, loading speed = 2.54 mm/min).

influence the fracture toughness for the ductile adhesive bonded system. In our testing, adhesive thickness ranged from 0.2 to 0.5 mm for Bemis 6343, and we did not find significant dependence of fracture toughness on the adhesive thickness (see Fig. 8). An adhesive thinner than 0.2 mm could induce processing defects in the adhesive, especially for grit-blasted adherends. For application, an adhesive thicker than 0.5 mm might not be economical.

4.4. Effects of loading speed

It was found that the DCB fracture toughness of the Bemis 6343 joints gradually increased as loading speed increased from 2.5 to 508.0 mm/min (Fig. 9). The loading-speed-dependent fracture toughness could be caused by viscoelastic properties of the thermoplastic adhesive [4].

4.5. Effects of curing temperature and residual stresses

No significant influence of the curing temperature on FM 300-2 DCB fracture toughness was found (Fig. 10). Curing temperature could influence residual stresses and the interaction between the polymer and fiber. For

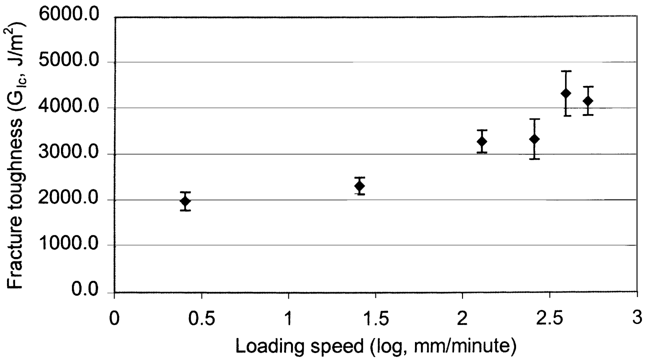

Fig. 9. Effects of loading speed on DCB fracture toughness (Bemis 6343, adhesive thickness = 0.4 mm).

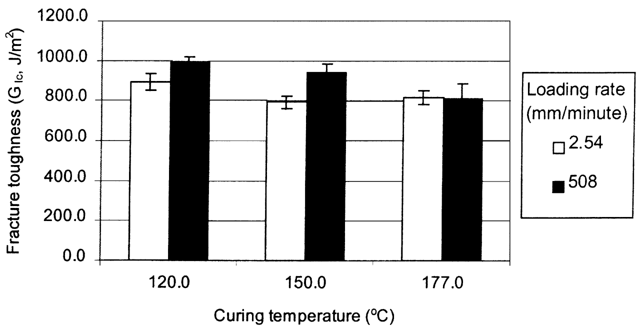

Fig. 10. DCB fracture toughness (FM300-2, adhesive thickness = 0.4 mm).

thermoplastic Bemis 6343, the residual stresses and other factors, such as adhesive thickness, could influence the plastic deformation of the adhesive layer [10]. More study is under way to tailor the multi-factor effects.

5. Conclusions

The processing parameters of the two adhesives, thermoplastic Bemis 6343 and thermosetting FM 300-2, were characterized using differential scanning calorimetry (DSC). The recrystallization of Bemis 6343 took less than 5 min. The curing of FM 300-2 took about 60 min to cure at 120°C, about 20 min at 150°C, and about 15 min at 177°C. Application of processing pressure to both adhesives when they were in viscous liquid states was vital in the bonding process. However, the temperatures of the two adhesives differed greatly when each was in the viscous liquid state.

High DCB fracture toughness (~ 2000 J/m^2) was found in the thermoplastic Bemis 6343 bonded DCB due to the plastic zone effect at the crack tip. As the loading speed of the Bemis 6343 DCB increased from 2.5 to 381.0 mm/min, the fracture toughness increased almost 100%. The viscoelastic properties of the Bemis 6343 could be the reason for the loading-speed-dependent performance of its DCB.

The FM 300-2 showed higher butt-joint strength (~ 30 MPa) than the Bemis 6343 (~ 10 MPa). However, the FM 300-2's DCB fracture toughness was lower than that of the Bemis 6343 DCB. Little adhesive plastic deformation was found in the FM 300-2 DCB testing. The glass fiber additive in the adhesive stabilized the DCB crack propagation.

Adhesive thickness (from 0.20 to 0.50 mm) and pre-crack length (from 50 to 100 mm) were found to have negligible effects on the Bemis 6343 DCB fracture toughness. For FM 300-2, curing temperature and loading speed had little effects on the fracture toughness.

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