INTERLAMINAR CHARACTERISTICS OF CFRP WITH THERMOPLASTIC PARTICLES

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1 Introduction
Various applications of Fiber Reinforced Plastics (FRP) in primary and secondary load-bearing structures, over the past few decades, have been established as a good substitute for metallic materials. This is because FRP has an excellent specific strength and stiffness. However, mechanical property of FRP in out-of-plane direction was significantly lower than that in in-plane direction. Especially, Low interlaminar strength in laminated structure has been main problem for a long time. To improve this problem, various kinds of investigations have been done.

Effectivity of incorporation of the Thermoplastic (TP) particles into the thermosetting matrix resin of composite laminates on interlaminar shear strength and interlaminar fracture toughness has been reported by many researchers [1]. This technique was applied to UD carbon prepreg that was used in the primary structures of a commercial airliner. By this method, favorable delamination resistance and impact resistance can be obtained and it's possible to control the mechanical properties and long time durability of the composite material. However, effects of various parameters (size, shape, amount, type of resin, surface treatment and so on) of TP particles on interlaminar fracture toughness of composite laminates have not been discussed sufficiently.

In this study, effects of amount, size and shape of TP particles on bending property and interlaminar fracture toughness of woven laminated composites were investigated by using Bending test, Double Cantilever Beam (DCB) and End-Notched Flexure (ENF) tests. In addition, effect of TP particles on fracture mechanism was also estimated by cross-sectional observation and observation for fracture surfaces by using Scanning Electron Microscope (SEM).

2 Materials and specimen preparation
Carbon woven fabric (HONLU TECHNOLOGY CO.LTD) and epoxy resin (jER828; Mitsubishi Chemical Corporation) with hardener (jER cure W; Mitsubishi Chemical Corporation) were used as reinforcement and matrix resin. In this study PA12 (Polyamide 12) thermoplastic particles (Daicel-Evonik Ltd.) were chosen for incorporation into matrix resin. In this study, three kinds of particles with different size and shape were chosen. SEM images of two kinds of TP particle are shown in Fig.1. One is "Potato" shape and the other is spherical shape. The specifications of TP particles for each specimen used in this study are shown in Table1. TP particles of Sample-1 and Sample-2 were prepared for comparing particle size and those of Sample-2 and 3 were for comparing particle shapes.

For fabrication of specimens, first, the resin was heated up at 100°C to decrease the viscosity of liquid resin and stirred at 600rpm using hot stirrer. Particles were preliminarily incorporated into liquid epoxy resin. The incorporation amount of TP particles were 0, 3, 5, 7, 9 wt% for matrix resin. After 24 hours, sufficient dispersion state was obtained. Then, bubbles in the resin were removed in a vacuum for 1 hour, and 12 layers of carbon woven fabrics were stacked with epoxy resin with TP particles impregnated by hand lay-up method. A pressure of 8kPa was applied on the laminates in the oven and cured at 100°C for 2 hours and 175°C for 4 hours. In the ENF and DCB specimen, Polyimide film of 25μm thickness was inserted into the middle.
of layers to introduce precrack. Thickness of specimens was about 2.8 mm.

3 Testing procedures

Dispersion states of TP particles in the specimens were confirmed by cross-sectional observation with optical microscopy. Size of bending specimens, DCB specimens and ENF specimens were 80mm×15mm, 100mm×25mm and 140mm×25mm respectively. Schematic drawing in each specimen is shown in Fig.2(a)(b)(c). The bending test was carried out with test speed of 1mm/min. For measuring mode I interlaminar fracture toughness of the specimens, the Double Cantilever Beam (DCB) testing was performed with testing speed of 1mm/min. Fracture surface after DCB testing was observed by SEM. The end-notched flexure (ENF) test was carried out for measurement of mode II interlaminar fracture toughness. Testing speed was 0.5 mm/min. Fracture surface after ENF testing was also observed by SEM.

4 Result and discussion

4.1 Dispersion states of thermoplastic

Dispersion states of thermoplastic particles in Sample-1, Sample-2 and Sample-3 is shown in Fig.3(a)(b)(c). In Sample-1, TP particles were dispersed uniformly between layers in resin rich region and some of TP particles were in fiber bundles. In Sample-2, TP particles were also dispersed uniformly in resin-rich area as same as Sample-1. In Sample-3, TP particles were not dispersed uniformly in resin-rich area but distributed near fiber bundles.

4.2 Result of three-point bending testing

Relationship between bending strength and incorporation amount of TP particles is shown in Fig.4. Bending strength of Sample-1 and 2 increased with increase in incorporation amount of TP particles up to 7wt% and decreased over 7wt%. In the case of Sample-3, the bending strength increased with increase in incorporation amount of TP particles up to 3wt% and decreased over 3wt%. Compared to Sample-1 and Sample-2 having same potato shape and different particle size, the optimum value of incorporation amount of TP particles for bending strength was same value of 7wt%. And, peak value of Sample-2 was larger than that of Sample-1. From this results, the optimum value of TP particles for bending strength was same when the particle shape was same, and the difference of particle size corresponded to the difference of improvement rate in bending strength. Compared to Sample-2 and Sample-3, maximum bending strength was almost same but the optimum value of incorporation amount of TP particles was different. From this results, the optimum value of TP particles for bending strength was different when the particle shape was different, and the improvement rate in bending strength was almost the same when the particle size was same.

Fracture aspect after bending test in 0wt% and optimum value of each specimen is shown in Fig.5(a)(b)(c)(d). From each photos, transverse crack in 90 degree fiber bundles and tensile fracture of 0° fiber bundle occurs, after that, the main crack progressed to compression side. Further, delamination occurred along with it. Relationship between delamination length and incorporation amount of TP particles is shown in Fig.6. In Sample-1, delamination length decreased with increase in TP particles up to 5wt% and increased over 5wt%. In Sample-2, delamination length decreased with increase in TP particles up to 7wt% and increased over 7wt%. In Sample-3, delamination length decreased with increase in TP particles up to 3 wt% and increased over 3wt%. These results should be correlated to the results of bending strength in Fig.4 because the amount of delamination was less in the samples with better bending properties. The reason the optimum value of Sample-3 was less than Sample-1 and Sample-2 is considered to be the difference in the dispersion states of TP particles. It is considered that the peak appeared in smaller incorporation amount because TP particles distributed near fiber bundles in Sample-3. It is considered that the crack progress is restrained and bending strength increased because fracture toughness could increase with increase in incorporation amount of TP particles. After adding TP particles more than optimum value of incorporation amount of TP particles into resin, bending strength decreased because fracture toughness could decrease with increase in TP particles.
4.3 Result of DCB testing

Initial interlaminar fracture toughness $G_{IC}$ and interlaminar fracture toughness $G_{IR}$ obtained from mode I DCB testing in Sample-1 is illustrated in Fig.7 and Fig.8. From Fig.7, $G_{IC}$ increased with increase in TP particles. From Fig.8, $G_{IR}$ increased with increase in TP particles up to 5 wt% and showed almost the same value in 5 and 7 wt%.

Fracture surface after DCB testing obtained by SEM observation are shown in Fig.9(a)(b). From this observation, there were more amounts of fiber breakages observed at 0 degree fibers and resin on the fracture surface in 7 wt% than that in 0 wt%.

Cross-sectional photos around a tip of crack after DCB testing are shown in Fig.10(a)(b). In 0 wt%, the crack was propagated along interface between fiber and matrix at the interlayer. Meanwhile, in 7wt% the crack was propagated along interlayer or in 0 degree fiber bundles, and also transverse cracks and delamination occurred in and around 90 degree fiber bundles.

From these results, it was clarified that fracture mechanism was changed by addition of TP particles, therefore mode I interlaminar fracture toughness was improved.

4.4 Result of ENF testing

Relationship between interlaminar fracture toughness $G_{IC}$ and incorporation amount of TP particles is shown in Fig.11. In Sample-1, fracture toughness increased with increase in TP particles up to 5wt% and decreased over 5wt%. In Sample-2, fracture toughness increased with increase in TP particles up to 7wt% and decreased over 7wt%. In Sample-3, fracture toughness increased with increase in TP particles up to 3wt% and decreased over 3wt%. Compared to Sample-1 and Sample-2 having same potato shape and different particle size, the optimum value of incorporation amount of TP particles was almost the same value of about 5-7wt%. Peak value of $G_{IC}$ for Sample-2 was larger than that of Sample-1. Compared to Sample-2 and Sample-3 having same particle size and different shape, maximum fracture toughness was almost same but the optimum value of incorporation amount of TP particles decreased in Sample-3.

Fracture aspect after ENF test in 0wt% and optimum value of each specimen in result of ENF test is shown in Fig.12(a)(b)(c)(d). From Fig.12(a), only the the main crack was observed in 0wt%. From Fig.12(b) and (c) in Sample-2 and Sample-3, Transverse cracks in 90 degree fiber bundles were confirmed around the main crack.

Fracture surface after ENF testing obtained by SEM observation is shown in Fig.13(a)(b). In specimen with potato particles (Fig.13(a)), the striation of resin was observed around TP particles. In the specimen with spherical particles (Fig.13(b)), the striation of resin was observed around TP particles. In addition, spherical particles was elongated.

Consequently, in the improvement of interlaminar fracture toughness, exists of the optimum value exists was clarified in particle size and incorporation amount of TP particles as same as the results of bending test. Compared to Sample-1 and Sample-2 having same potato shape and different particle size, the optimum value of incorporation amount of TP particles for interlaminar fracture toughness $G_{IC}$ was same value of about 7wt%. And, peak value of $G_{IC}$ for Sample-2 was larger than that for Sample-1.

Here, the reasons for existence of optimum value for bending strength and interlaminar fracture toughness were explained below. As shown in Fig.14, when incorporation amount of TP particles is optimum, crack propagation is restrained by TP particles and the crack branch off from around particles. While, when incorporation amount of TP particles is over optimum value, particles were neighboring and crack propagate around particles in a row and is not restrained. In the case of spherical particles, ductility of particles restrain the crack growth. Because of it, it's considered that interlaminar fracture toughness increased with smaller amount of the particles.

5 Conclusion

5.1 Effect of particle size on CFRP

In the case of the same shape with different size of TP particles, the bending strength was increased and delamination length was decreased until the optimum value with increase in TP particles. Interlaminar fracture toughness was also increased with increase in the size of particle. In addition, transverse cracks in 90 degree fiber bundles around the main crack occurred with increase in the size of particle and apparent interlaminar fracture toughness increased.
5.2 Effect of particle shape on CFRP

In the case of the same size with different shape of TP particle, the optimum value of incorporation amount of TP particles for bending strength and interlaminar fracture toughness changed.

References

Fig. 1 Shapes of TP particles

Table 1 Specifications of each specimen

<table>
<thead>
<tr>
<th>Name</th>
<th>Shape</th>
<th>Particle Size [µm]</th>
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<tbody>
<tr>
<td>Sample-1</td>
<td>Potato</td>
<td>8</td>
</tr>
<tr>
<td>Sample-2</td>
<td>Potato</td>
<td>24</td>
</tr>
<tr>
<td>Sample-3</td>
<td>Sphericity</td>
<td>23</td>
</tr>
</tbody>
</table>
Fig. 2 Schematic drawing of each specimen

(a) Bending specimen

(b) DCB specimen

(c) ENF specimen
Fig. 3 Distribution of TP particles

Fig. 4 Relationship between bending strength and incorporation amount of TP particles
Fig. 5 Fracture aspect after bending testing

(a) 0wt%

(b) Sample-1 7wt%

(c) Sample-2 7wt%

(d) Sample-3 3wt%

Fig. 6 Relationship between delamination length and incorporation amount of TP particles

Fig. 6 Relationship between delamination length and incorporation amount of TP particles
Fig. 7 Relationship between $G_{IC}$ and incorporation amount of TP particles.

Fig. 8 Relationship between $G_{IR}$ and incorporation amount of TP particles.

Fig. 9 Fracture aspect after DCB testing
Fig. 10 Aspect of crack growth in the middle of DCB testing

(a) 0 wt%

(b) 7 wt%

Fig. 11 Relationship between interlaminar fracture toughness $G_{IC}$ and incorporation amount of TP particles

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Fig. 11 Relationship between interlaminar fracture toughness $G_{IC}$ and incorporation amount of TP particles
Fig. 12 Fracture aspect after ENF testing
Fig. 13 Fracture surface after ENF testing

(a) Potato shape

(b) Spherical shape

Fig. 14 Schematic drawing of the crack growth