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# THE INTERLAMINAR FRACTURE TOUGHNESS OF CARBON/EPOXY LAMINATES INTERLEAVED WITH POLYAMIDE PARTICLE LAYERS

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**Keywords:** Fracture toughness, Polyamide particles, Interleaving particles, Interface

## Abstract

Toughening epoxy based composite materials using thermoplastic particles is a cost-effective approach to improve their poor delamination resistance. In this paper, the interlaminar toughening effect of different thermoplastic microparticles such as polyamide-6 and polyamide-12 particles for a carbon fibre/epoxy composite was investigated. The particles with a range of different areal weights were directly deposited on the prepreg surface to toughen the interlaminar region of the cured laminate samples. From the scanning electron microscope (SEM) observations, it was found that the effects of the three thermoplastic particles on the interlaminar fracture behaviour were different. It was also found that the particle areal weight affected the Mode-I fracture toughness differently depending on the particle material. In some cases, no toughening effect was observed.

## 1. Introduction

High performance composites usually use thermoset resin as matrix material due to their high modulus and strength and low processing cost. However, one of the drawbacks of thermoset matrix is their low fracture toughness, which results in poor delamination resistance of the composites. This brings the concern on damage tolerance and durability of the in-service composite structures. Therefore, the matrix material is usually required to be toughened without sacrificing the other mechanical properties such as strength, modulus and glass transition temperature. Modifying the matrix with microscale polymer particles such as polyamide (PA) [1], polyethersulphone (PES), polyetherimide (PEI), polysulfone (PSF) [2] in composites is common to improve their toughness extrinsically [3]. Such particles reduce the stress intensity at the crack tip by crack pinning, crack deflection, particle-matrix debonding, particle bridging and microcracking, which results in a higher fracture toughness [4].

However, there is a challenge that mixing fine particles into a liquid resin can lead to high resin viscosity [5], which significantly affects impregnation quality in composite manufacturing processes. One of the solutions is to interleave layers of particle tougheners between plies in a laminate [1,6] rather than direct impregnating the laminate with a highly viscous toughened resin. This can create a high concentration of toughening materials in the interlaminar regions. Such a particle interleaving concept has also been applied with other thermoplastic material formats such as thin film [7], non-woven veils [8] and short fibres [9].

Since thermoplastics have much higher ductility compared to rubbers, they can absorb larger amounts of fracture energy by deformation. However, in order to effectively use such ductility in toughening mechanisms, strong particle-matrix adhesion is essential. The interfacial bonding strength can be affected by the processing temperature. Kim *et al.* [10] investigated the effect of temperature on

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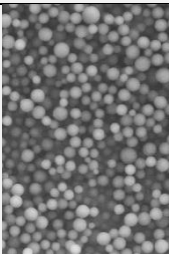
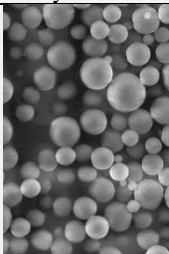
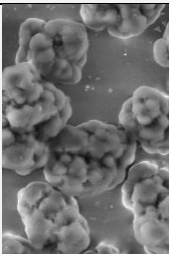
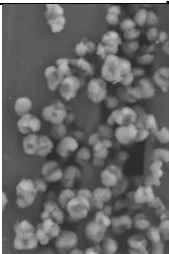
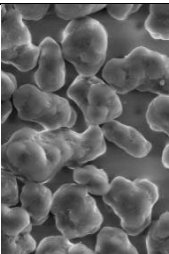
epoxy/polyamide-6 resin blends. They reported that the fracture toughness was affected by the temperature when mixing the polyamide particles and resin, because the partial melting of the polyamide-6 contributed to the interfacial adhesion with epoxy. Additionally, the bonding strength could be influenced by the surface morphology of particles due to the mechanical interlocking effect. A rougher particle surface could result in higher surface area, which was investigated experimentally. In this research, two types of polyamide particles with various of sizes and surface morphologies were used as the interlaminar toughener of a carbon fibre/epoxy composite. Different amount of particles were directly deposited onto the prepreg surface before curing. The Mode-I fracture toughness of the compomposite was measured using double cantilever beam (DCB). Fracture mechanisms were discussed based on the DCB test results and the scanning electron microscope (SEM) images of the fractured surface.

## 2. Experimental

### 2.1 Materials

The test coupons in this work were made of unidirectional carbon fibre/epoxy prepregs (IM7/8552, Hexcel, US). Six thermoplastic micro particles were used as the toughening materials, which are listed in Table 1. PA12\_d5 and PA12\_d10 particles have a nearly perfect spherical shape and a smooth surface with a small size variation. PA6\_d13 is a polyamide-6 particle with a high surface area due to its surface porosity. Both PA12\_d30 and PA6\_d16 polyamide particles have a less regular shape and some roughness on the surface. No surface treatment was applied to all the particles.

Table 1 Overview of the thermoplastic particles used in this work

Name	PA12_d5	PA12_d10	PA12_d30	PA6_d13	PA6_d16
Material		Polyamide 12		Polyamide 6	
SEM (1000x)					
Size (μm)	~5	~10	25-30	10-16	15-20
Particle shape	Smooth spherical	Smooth spherical	Rough, quasi-spherical	Rough, quasi-spherical	Quasi-spherical
Melting Point (°C)	165-171	165-171	180	210-220	210-220
Supplier	Toray, JP		Goodfellow, UK	Toray, JP	Goodfellow, UK
Tensile Strength	76 MPa			90 MPa	
Elastic modulus	4 GPa			3 GPa	

### 2.2 Specimen preparation

Double cantilever beam specimens were fabricated following the procedure described below. All the polyamide particles were dried at 80°C for 24 hours before use. The particles were spreaded on the middle layer of each sample laminate, and the increased weight was measured to calculate the powder areal amount. A 12 μm PTFE film was used as an insert to create a pre-crack in the interlaminar region.

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Then the laminates were vacuum bagged and cured in autoclave. The curing cycle followed the recommendation from the prepreg supplier; i.e. dwelling at 110°C for 1 hour and curing at 180°C for 2 hours, which led to the cured samples with a thickness of 3.7-3.8 mm.

The cured samples were cut to the dimensions of 20 mm × 160 mm. Piano hinges were bonded (Figure 1A), following the ASTM D5528 test standard. Both side edges of each sample were coated with silver or white paint to visualise the delamination during test.

### 2.3 Double cantilever beam (DCB) test

The test procedure followed the ASTM D5528 standard. A universal material testing machine was used to apply a crack opening load. 1 kN load cell was used to record the force during loading. The cross-head speed was set at 2 mm/min both for initial loading. The initial loading was stopped when the crack propagation length reached 3-5 mm, which provided a sharp crack tip before starting the Mode-I test. The actual DCB test was terminated when the crack propagated up to 30 mm beyond the initial crack tip.

The delamination length was captured using video camera at a sampling rate of 1 Hz (Figure 1B). The cross-head displacement and the applied tensile load during test were recorded at the same time. The modified beam theory (MBT) was used to calculate the Mode-I fracture toughness:

$$G_{IC} = \frac{3P\delta}{2b(a+|\Delta|)} \quad (1)$$

where the  $P$  is the crack opening load,  $\delta$  is the cross-head distance,  $b$  is the specimen width,  $a$  is the delamination length, and  $\Delta$  is a correction factor obtained from the beam compliance.

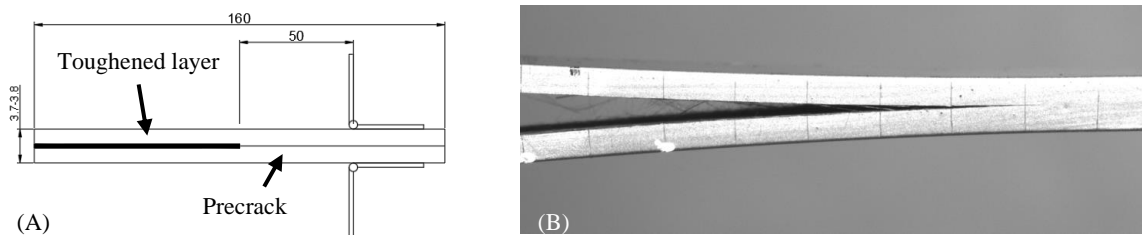


Figure 1 (A) Schematic of the DCB coupon dimensions, (B) crack propagation captured by video camera during the test.

### 2.4 Failure surface analysis

The tested coupons were cut into a few pieces using a composite diamond wheel cutter to investigate the delamination region. The samples were washed in an ultrasonic bath with water to remove the debris from cutting. Fractured surface was observed using a scanning electron microscope (TM3030Plus, Hitachi).

## 3. Results and discussion

### 3.1 Fractured surface observation

The SEM images showed all six particles were well impregnated with the resin after curing. However they behaved differently in the Mode-I fracture test, which is illustrated in Figure 2. For the samples toughened with polyamide-12 particles, the crack went through the particles regardless the particle size and surface characteristic (Figure 2A). It can be seen that the particles were ripped off causing plastic

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deformation as shown in Figure 3A and B. However, the main failure mode of the sample toughened with polyamide-6 particles was particle-matrix debonding (Figure 2B). The crack went through the interface between the particles and the matrix.

Since the 8552 resin is amine-cured epoxy [11], it is possible to form strong adhesion with polyamide if the polyamide molecular chains have sufficient mobility or receive enough energy (e.g. at elevated temperature). The possible interactions could be forming covalent bonding during curing between the epoxide groups in the growing epoxy and amide-hydrogen groups in polyamide if the polyamide chains are still reactive, or molecular entanglement at the interface between the polyamide chains and epoxy network (so-called interpenetrating networks, IPNs) [12].

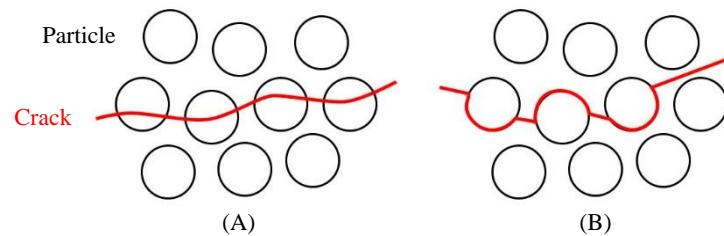


Figure 2 Illustration of the three types of failure behavior observed in this work: (A) particle failure, (B) particle-matrix debonding and

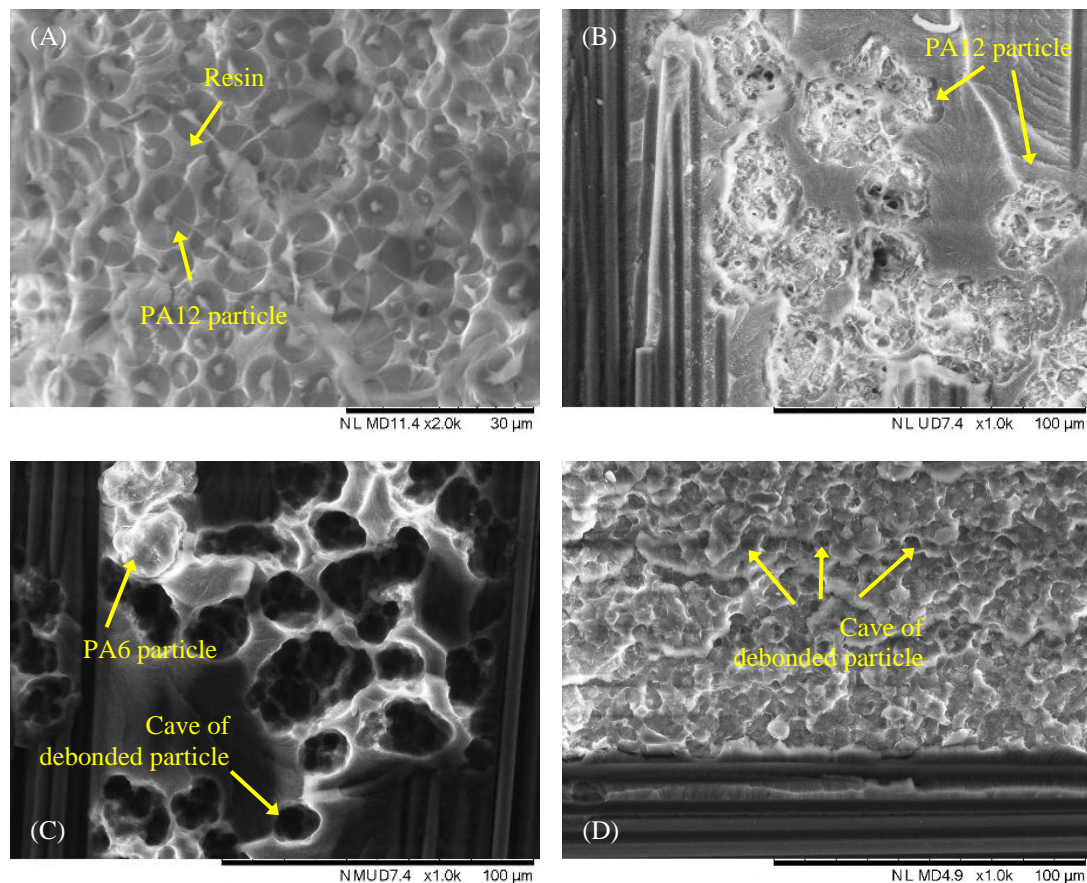


Figure 3 Top-views of fractured surfaces of the specimens with: (A) PA12\_d10, (B) PA12\_d30, (C) PA6\_d16 and (D) PA6\_d13

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Polyamide-12 has a melting point close to the curing temperature of the 8552 epoxy resin (180 °C), which could meet the aforementioned conditions and resulted in high interfacial bonding between the polyamide-12 and the resin. Therefore, it could be deduced that the deformation and tearing of the polyamide-12 particles contributed large absorption of fracture energy during test. However, the polyamide-6 particles debonded from the epoxy resin (Figure 3C and D), which showed they were not interacted with the growing 8552 epoxy network during curing. This could be their high melting point (above 200 °C) restricted the mobility of the polyamide molecular chains. Additionally, the interfacial bonding was not improved by the high surface roughness of the PA6\_d13 particles.

### 3.2 DCB test results

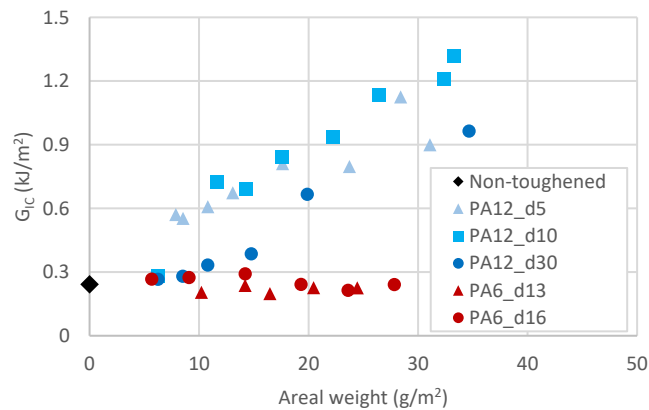


Figure 4 Initiation  $G_{IC}$  for different areal weights of polyamides

Figure 4 shows the initiation  $G_{IC}$  for the areal weight of the particles. The Polyamide-12 and phenoxy particles were more effective to improve the interlaminar fracture toughness than the polyamide-6 particles. PA12\_d5, PA12\_d10 and PA12\_d30 (blue in Figure 4) resulted in a linearly increasing  $G_{IC}$  with the increase of the deposited particle amount. Especially, the  $G_{IC}$  for the sample interleaved with 33.3 g/m<sup>2</sup> of PA12\_d10 (1.32 kJ/m<sup>2</sup>) was approximately 4.5 times higher than the non-toughened sample (0.242 kJ/m<sup>2</sup>). This clearly shows the effect of the strong interfacial bonding between the polyamide-12 and the matrix, which resulted in a large amount of fracture energy was absorbed by the plastic deformation of the particles (Figure 3A and B). The particle size difference between PA12\_d5 and PA12\_d10 did not influence the toughening effect. However, the larger particles PA12\_d30 showed slightly lower  $G_{IC}$  improvement compared to the other two smaller polyamide-12 particles.

For the polyamide-6 particle toughened samples (red in Figure 4), there was no clear correlation between  $G_{IC}$  and the particle areal weight both for PA6\_d13 and PA6\_d16. This was caused by the poor particle-matrix adhesion resulted in interface debonding (Figure 3C and D).

### 4. Conclusions

In this paper, interlaminar toughening of carbon fibre/epoxy composites by direct deposition of thermoplastic micro-particle on the prepreg surface was investigated. Three types of polyamide-12 particles enhanced the  $G_{IC}$  with the increase of the particle amount. Due to the effective interfacial bonding between the polyamide-12 and the epoxy resin, the fracture energy was mainly absorbed by the plastic failure of the particles. A maximum  $G_{IC}$  improvement of 450% was observed in this work.

The two polyamide-6 particles showed no obvious improvement on  $G_{IC}$  with the amount of powder. SEM image revealed that the polyamide-6 particles deboned from epoxy, which indicated no effective interfacial bonding was developed during composite curing. The higher particle surface roughness did not improved the fracture toughness.

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