Numerical simulations of inter-laminar fracture in particle-toughened carbon fiber reinforced composites

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The most predominant failure mechanism in carbon fiber reinforced composites is delamination. In laminated composites dispersed second phase particles have been added to the interlayer region to reduce delamination. In this work a phase field damage model is used to study Mode I crack propagation in particle-toughened interlayers. The simulations show several damage mechanisms observed in experiments **and the processes that produce crack growth**. In particular, failure involves multiple micro-cracks forming ahead of the main crack tip that coalesce and lead to crack branching, deflection and the formation of ligaments. Additionally, increasing the particle stiffness and the **surface energy of the matrix-particle interface** do not lead to an improvement of the interlaminar toughness as the fiber-matrix interface becomes a preferential path for the crack through the nucleation of extended delamination zones ahead of the crack tip.

1 Introduction

Delamination in carbon fiber reinforced polymers (CFRP) is one of the dominating failure mechanisms [1]. In general, microcracks initiate in the resin-rich interlaminar region [2], or at laminate edges [3]; then, the cracks coalesce to form an extended delamination zone. Second phase particles dispersed in the interlaminar region have been identified to suppress or reduce delamination. Experiments show that microcracks interact with particles leading to bridging and a creating a tortuous crack path that helps to increase delamination resistance [4, 5, 6, 7, 8, 9]. Numerical simulations that can predict these mechanisms can be used to advance the design of particle-toughened interlayers by guiding and reducing the number of experiments.

Synchrotron radiation computed tomography (SRCT) and synchrotron radiation computed laminography (SRCL) techniques provide an innovative way to follow crack propagation with resolution of hundreds of nanometers. Borstnar *et al.* [5, 6] use these techniques to conduct in situ observation of Mode I and Mode II fracture of CFRP. In particular, the authors describe the micro scale failure process in interlaminar regions with added second phase particles. The effect of particles with different **mass density** and shape is analyzed but unfortunately, particle and interface properties are not reported in

the papers. Numerical simulations [10] could help to infer the value of some of the unknown mechanical properties of the composites tested by Borstnar *et al.* [5, 6]. The experiments show that the incorporation of second phase particles in the interlayer leads to processes that improve the effective toughness of the composite. Some of these mechanisms are: i) crack trapping, ii) crack deflection, iii) particle-matrix debonding, iv) formation of bridging ligaments, and v) crack path deflection towards the interlaminar region.

Numerical simulations of crack propagation under Mode I are performed with a phase field damage model (PFDM) [11, 12]. A systematic study of the particle stiffness, the **surface energy of the matrix-particle and the fiber-matrix interfaces** and the particle distribution elucidates the influence of these properties on the crack path and the failure mechanisms observed experimentally [5, 6]. The geometries and the particle properties used in the simulations are not extracted from experiments but are a simplification to demonstrate the power of the PFDM approach to predict micro-crack nucleation, bridging and tortuous crack path. For example the simulations show that the crack evolution in Mode I fracture involves multiple micro-cracks forming ahead of the main crack tip that coalesce and lead to crack branching, crack deflection and the formation of ligaments.

The paper is organized as follows, in Section 2, the PFDM is described. In Section 3, the fracture mechanisms in the interlaminar region are investigated by varying the stiffness of the particles as well as the **surface energy of the particle-matrix and the fiber-matrix interfaces**. Section 4 contains the summary of the numerical results and a discussion on design of interlayers with improved properties.

2 Phase Field Damage Model for Brittle Materials

Following Griffith's criterion [13] the energy required to nucleate a crack of area Γ in a brittle material is:

$$W_f[\Gamma] = \int_{\Gamma} G_c d\mathbf{x} \quad (1)$$

where the integral is over the crack area and G_c is the surface energy density needed to create a unit surface crack. This quantity is also referred as the energy release rate.

The original approach of the PFDM is a variational formulation introduced by Francfort and Marigo [14] that was later extended to a phase field numerical algorithm [15, 16, 17, 18]. In this model a phase field variable d defined in the entire domain indicates the amount of damage in the solid, with $d=1$ representing damage and $d=0$ intact status. In the PFDM the energy in Equation 1 is approximated using a diffuse crack topology as

$$\int_V G_c \left(\frac{d^2}{4l_0} + l_0 |\nabla d|^2 \right) d^3x \quad (2)$$

where the integral is over the volume of the solid and the length scale parameter l_0 controls the thickness of the damaged region as visualized in Figure 1. The regularization introduced by l_0 recovers in the limit $l_0 \rightarrow 0$ the sharp crack surface integral in Equation (1) [15, 16, 17, 19].

In other models the fracture onset criterion requires two quantities, the fracture toughness (energy) and the strength (peak stress)[20, 21]. The connection to the PFDM is that the fracture toughness for Mode I is proportional to the critical energy release rate, G_C as

$$G_C = K_{IC} \sqrt{E'} \quad (3)$$

where $E' = E$ for the plane-stress condition, $E' = E/(1-\nu^2)$ for the plane-strain condition, E is the Young's modulus and ν is the Poisson's ratio. On the other hand, a simple calculation [11, 16, 18] shows that the length scale parameter l_0 is related to the peak stress in a 1D homogeneous nucleation example as

$$\sigma_c \propto \sqrt{\frac{E G_c}{l_0}} \quad (4)$$

and therefore, l_0 maybe considered as a material parameter.

Figure 1: (a) Schematic of a sample with a sharp crack. (b) Approximation of the diffusive crack with the phase field $d(x)$.

The phase field damage variable $d(x)$ and the current deformation state are obtained from the solution of coupled partial differential equations that arise from the minimization of the total energy. The steps to arrive to the differential equations are explained in detail in the following paragraphs.

The total energy of the body, W_{total} , being the sum of the elastic energy and the fracture energy, Equation (2), is then given by

$$W_{total}[\varepsilon, \mathbf{d}] = \int_V \left((1 - \mathbf{d})^2 + k \right) \mathbf{d}[\varepsilon] d^3x + \int_V G_c \left(\frac{d^2}{4l_0} + l_0 |\nabla \mathbf{d}|^2 \right) d^3x \quad (5)$$

where the first term takes into account the degradation of the strain energy due to damage, ε is the strain tensor, k is a residual stiffness added to prevent a singular stiffness matrix, and $a[\varepsilon]$ is the elastic energy density. For isotropic materials and small strain the elastic energy density $a\varepsilon$ is calculated as:

$$a[\varepsilon] = \frac{E\nu}{2(1+\nu)(1-2\nu)} \varepsilon_{ii} \varepsilon_{jj} + \frac{E\nu}{2(1+\nu)} \varepsilon_{ij} \varepsilon_{ij} \quad (6)$$

where Einstein's notation on repeated indexes is used. The elastic strain energy rate is:

$$\dot{W}_e[\varepsilon, \dot{\varepsilon}, d, \dot{d}] = \int_V \left(\sigma_{ij} \dot{\varepsilon}_{ij} - 2(1-d)a[\varepsilon] \dot{d} \right) d^3x \quad (7)$$

where the stress tensor is derived from the total energy equation (5) as

$$\sigma_{ij} = \left((1 - \mathbf{d})^2 + k \right) \frac{\partial a[\varepsilon]}{\partial \varepsilon_{ij}} \quad (8)$$

and the energy dissipation due to the crack propagation follows as:

$$\dot{W}_f[d, \dot{d}] = \int_V G_c \left(\frac{d}{2l_0} + 2l_0 \Delta d \right) \dot{d} dx \quad (9)$$

where Δ is the Laplacian operator. The energy dissipation in Equation (9) should be non-negative under the assumption of irreversible damage. This constraint is fulfilled by applying the following two local conditions:

$$\begin{aligned} \frac{d}{2l_0} + 2l_0 \Delta d &\geq 0 \\ \dot{d} &\geq 0 \end{aligned} \quad (10)$$

In order to satisfy the conditions in Equation (10), an extended Lagrangian is defined following Miehe *et al.* [17]:

$$L[\varepsilon, \dot{\varepsilon}, d, \dot{d}, \lambda] = \dot{W}_e[\varepsilon, \dot{\varepsilon}, d, \dot{d}] + D[d, \dot{d}, \lambda] \quad (11)$$

where

$$D[d, \dot{d}, \lambda] = \int_V \left(2(1-d)a[\varepsilon] \dot{d} - \lambda Y_{cr} \right) dx \geq 0 \quad (12)$$

is the extended dissipation functional, λ is a Kuhn–Tucker multiplier and Y_{cr} is an inequality constraint function defined as:

$$Y_{cr} = 2(1-d)[\varepsilon] - G_c \left(\frac{d}{2l_0} + 2l_0 \Delta d \right) \leq 0 \quad (13)$$

The variation of the extended Lagrangian with respect to the field variables yields the following Kuhn-Tucker equations

$$\begin{aligned} \sigma_{ij,j} &= 0 \\ \dot{d} &= \lambda \geq 0 \\ Y_{cr} &\leq 0 \\ \dot{d} \cdot Y_{cr} &= 0 \end{aligned} \quad (14)$$

The first equation in (14) is the equilibrium condition. The second equation enforces that the damage can only increase. The last two equations in (14) lead to the governing differential equation of the phase field:

$$\left(\frac{4l_0[\varepsilon]}{G_c} + 1 \right) (1-d) - 4l_0^2 \frac{\partial^2 d}{\partial x_i \partial x_i} = 1 \quad (15)$$

An alternate minimization algorithm [11] is used to find the solution of the damage field and the displacement. This algorithm finds the displacement field and phase field alternatively. At each loading step, the displacement $\mathbf{u}$ is first solved at a fixed phase field d . Then d is solved at fixed $\mathbf{u}$ with the irreversibility condition. The convergence is reached when the change in the variable between two iterative steps is less than certain tolerance. The algorithm steps are listed in Table 3. The simulations are performed using a 3D parallel software¹, MEMOSA, which is based on the cell-centered finite-volume method [11, 22]. The number of cells used in the simulations is 263, 300 with an average cell size of 0.12 μm .

¹ The code is available at <http://c-primed.github.io/fvm/>

- 1) Set $(\mathbf{u}^{(0)}, d^{(0)}) = (\mathbf{u}_{i-1}, d_{i-1})$
- 2) Set $p=1$.
 - (i) Predictor: Solve $\mathbf{u}^{(p)}$ with $d=d^{(p-1)}$
 - (ii) Corrector: $d^{(p)}$ with $\mathbf{u} = \mathbf{u}^{(p)}$ under the constraint $d^{(p)} \geq d_{i-1}$
 - (iii) If $\|d^{(p)} - d^{(p-1)}\| < \delta$ go to 3). Otherwise, $p=p+1$, go to (i).
- 3) Set $(\mathbf{u}_i, d_i) = (\mathbf{u}^{(p)}, d^{(p)})$

Table 1: Alternate minimization algorithm used in the PFDM simulations, loading step i.

3 Simulations of Mode I fracture in the interlayer

PFDM simulations of Mode I fracture are carried out to predict the crack path in the geometry shown in Figure 2. As a simplification and to be able to have a high resolution at the particle-matrix interface a plane strain condition is assumed. An initial $3\mu\text{m}$ crack is located at the center left side of the sample.

Figure 2: Schematic of the sample used in the simulations.

The top and bottom layers are anisotropic lamina with fibers oriented in the x direction. In the simulations homogenized equivalent lamina properties are used for the top and bottom layers, i.e., the fibers are not resolved explicitly. The lamina properties are listed in Table 2 [23].

The interlaminar region is a PMMA matrix with $5\mu\text{m}$ diameter second phase particles. The volume fraction of the particles is 55% and three different particle distributions are used in the simulations. The elastic constants for PMMA used in the simulations are shown in Table 3. The Young's modulus of the particles is varied from 3.9 GPa to

19.5GPa to study the effect of the stiffness of the particles on the crack topology. Unfortunately, particles and interface properties are not reported for the composites tested by Borstnar *et al.* [5, 6], it is only inferred that the mass density of the particles is higher or similar to the mass density of the matrix. Therefore, Young moduli in this range were chosen for the simulations.

The **surface energies** of PMMA, the particles and the lamina are set to $G_c=480\text{Jm}^{-2}$ [12] and $l_0 = 0.3\mu\text{m}$. The interface between the lamina and the interlayer should be interpreted as the effective **interface** between the fibers and the matrix in this model. The surface energy of the fiber-matrix interface $G_c f-m$ and the **surface** energy of the particle-matrix interface $G_c p-m$ are in a range 120Jm^{-2} to 480Jm^{-2} in different simulations. The thickness of the interfaces where the surface energy is set to $G_c f-m$ or $G_c p-m$ is $0.7\mu\text{m}$ (6 cells).

$E_l \quad x \quad [\text{GPa}]$	151
$E_l \quad y \quad [\text{GPa}]$	7.99
$\nu_l \quad xy$	0.32
$\mu_l \quad xy \text{ GPa}$	4.33

Table 2: Homogenized **equivalent lamina** material properties.

$E_m \quad [\text{GPa}]$	2.48
ν_m	0.33

Table 3: **Elastic properties of PMMA** used in the simulations.

In the simulations the crack propagates in the x direction, see Figure 2. In order to control the crack growth rate, the following displacements are applied in the boundary of the sample [24]

$$\begin{aligned}
 u_x &= \frac{K_I}{2\mu} \sqrt{\frac{r(x,y,z,t)}{2\pi}} (\phi - \cos\theta) \cos \frac{\theta}{2} \\
 u_y &= \frac{K_I}{2\mu} \sqrt{\frac{r(x,y,z,t)}{2\pi}} (\phi - \cos\theta) \sin \frac{\theta}{2}
 \end{aligned} \tag{16}$$

where KI is the stress intensity factor for Mode I, $\phi=3-4\nu$ for plane strain condition, and r, θ are the coordinates for a polar system with origin at the crack tip. The distance from the crack tip is a function of the macroscopic crack speed, v , and a pseudo time t :

$$r = \sqrt{(x - vt - x_o)^2 + (y - y_o)^2}$$

where x_o and y_o are the coordinates of the initial position of the crack tip.

3.1 Effect of the stiffness of the particles

In this section a parametric study of the particle stiffness and surface energy of the particle-matrix interface is performed. The effective surface energy of the fiber-matrix interface is kept constant at $G_c \Gamma = 480 \text{ Jm}^{-2}$.

Figure 3 shows the crack path when the particles and the laminae are perfectly bonded to the matrix, i.e., $G_c \Gamma = 480 \text{ Jm}^{-2}$. When $E_p = 3.9 \text{ GPa}$ the crack advances breaking through the particles. A similar behavior is observed in Mode I fracture tests by Borstnar *et al.*[6] when the mass density of the particles is smaller than the mass density of the matrix. Increasing the Young modulus of the particles leads to debonding at the particle-matrix interfaces but the crack still propagates through some of the particles, as shown in Figure 3(b). When the Young modulus of the particles is increased to $E_p = 19.5 \text{ GPa}$ only particle debonding is observed and the particle bridging ligament mechanism appears, Figure 3(c). It is also observed in this last case that the crack propagates along the interface between the lamina and the interlaminar region. This indicates that the crack resistance induced by the high Young modulus of the particles and the perfect bonding between the particle and the matrix induces delamination in the lamina-interlayer interface.

Figure 3: Simulations of Mode I crack propagation with $G_{c\ p-m} = G_{c\ f-m} = 480\text{ Jm}^{-2}$ (a) $E_p = 3.9\text{ GPa}$, (b) $E_p = 9.8\text{ GPa}$ and (c) $E_p = 19.5\text{ GPa}$. In this figure and the following blue represents damaged materials.

The surface energy of the particle-matrix interface is set to $G_{c\ p-m} = 120\text{ Jm}^{-2}$ in the simulations in Figure 4. Under this condition, debonding at the particle-matrix interfaces is the dominant mechanism making the crack path tortuous. In the experiments of Borstnar et. al. [5, 6] the crack is deflected from the fiber-matrix interface towards the interlayer. The same behavior is predicted in the simulations when $E_p = 9.8\text{ GPa}$ and $E_p = 19.5\text{ GPa}$, Figures 4(b) and 4(c). This may be the result of a stronger bonding between the fibers and matrix than the particles and the matrix. Particle bridging ligament is observed in the samples with particles with higher mass density in the experiments by Borstnar et. al. [5, 6] and it is captured in the simulations with $E_p = 19.5\text{ GPa}$, see Figure 4(c).

Figure 4: Simulations of Mode I crack propagation with $G_{c\ p-m} = 120\text{ Jm}^{-2}$ and $G_{c\ f-m} = 480\text{ Jm}^{-2}$. (a) $E_p = 3.9\text{ GPa}$, (b) $E_p = 9.8\text{ GPa}$ and (c) $E_p = 19.5\text{ GPa}$.

The evolution of the crack path with $E_p = 19.5\text{ GPa}$, $G_{c\ p-m} = 120\text{ Jm}^{-2}$ and $G_{c\ f-m} = 480\text{ Jm}^{-2}$ is shown in Figure 5 to examine the processes of particle bridging and crack deflection at the fiber-matrix interface. Figure 5(b) shows micro-crack nucleation ahead of the crack tip as the mechanism that results in particle bridging. As the load is increased the micro-cracks coalesce to form a continuous crack.

Figure 5: Crack path evolution with $E_p=19.5$ GPa, $G_{c\ p-m}=120\text{Jm}^{-2}$ and $G_{c\ f-m}=480\text{Jm}^{-2}$. The load is increased from (a) to (d).

3.2 Effect of particle distribution

The effect of the particle distribution is investigated numerically by introducing a particle-depleted region of length $18\mu\text{m}$ in the samples, see Figure 6. The effective critical fracture energy density between the fibers and the matrix is kept constant at $G_{c\ f-m}=480\text{Jm}^{-2}$, the Young's modulus of the particles is set to $E_p=3.9$ GPa, $E_p=9.8$ GPa and $E_p=19.5$ GPa in different simulations.

Figure 6: Schematic of a sample with a particle-depleted region.

In Figure 7 the propagation of the crack with perfectly bonded particles in the interlaminar region is shown for different particle stiffness. For $E_p=3.9$ GPa and $E_p=9.8$ GPa the crack is confined to the interlayer. In Figure 7(c) a crack nucleates along the fiber-matrix interface ahead of the particle depleted region. In the simulations in Figure 8 the surface energy of the particle-matrix interface is reduced to $G_{c\ p-m}=120\text{Jm}^{-2}$. Particle bridging and crack deflection are observed in the particle rich

regions. In all the configurations shown in Figure 8, micro-cracks nucleate ahead of the particle depleted region. This process is depicted in Figure 9, where crack branching is observed as a consequence of micro-crack nucleation and coalescence ahead of the main crack tip.

Figure 7: Simulations of Mode I crack propagation with a particle-depleted region and $G_{c\,p-m} = G_{c\,f-m} = 480 \text{ Jm}^{-2}$. (a) $E_p = 3.9 \text{ GPa}$, (b) $E_p = 9.8 \text{ GPa}$ and (c) $E_p = 19.5 \text{ GPa}$.

Figure 8: Simulations of Mode I crack propagation with a particle-depleted region, $G_{c\ p-m} = 120\ \text{Jm}^{-2}$ and $G_{c\ f-m} = 480\ \text{Jm}^{-2}$. (a) $E_p = 3.9\ \text{GPa}$, (b) $E_p = 9.8\ \text{GPa}$ and (c) $E_p = 19.5\ \text{GPa}$.

Figure 9: Crack path evolution with a particle-depleted region with $E_p = 19.5\ \text{GPa}$, $G_{c\ p-m} = 120\ \text{Jm}^{-2}$ and $G_{c\ f-m} = 480\ \text{Jm}^{-2}$.

3.3 Effect of fiber-matrix bonding

In this section the interplay between the **surface energy** of the particle-matrix and fiber-matrix interfaces studied. To take into account the effect of the particle distribution at the same time, the geometry with a particle-depleted region shown in Figure 6 is used.

Figure 10 shows simulations of Mode I crack propagation with $G_{c\ p-m} = 480\ \text{Jm}^{-2}$ and $G_{c\ f-m} = 120\ \text{Jm}^{-2}$ for different particle stiffness. When the **surface energy** of the particle-matrix interface is higher than the **surface energy** of fiber-matrix interface, cracks nucleate in the lamina-interlayer interface ahead of the initial crack tip. In the simulations shown in Figure 11 the **surface energy** of the particle-matrix interface is reduced to $G_{c\ p-m} = 120\ \text{Jm}^{-2}$. In these simulations the initial crack advances through the particle interfaces in the interlayer but a new crack is nucleated at the lamina-interlayer interface ahead of the particle depleted region. **Crack propagation towards the lamina-interlayer interface is observed in the experiments of Borstnar *et al.* [5, 6] in resin rich regions in agreement with our simulations.** Snapshots of the crack evolution are shown in Figure 12 for $G_{c\ p-m} = 120\ \text{Jm}^{-2}$ and $G_{c\ f-m} = 140\ \text{Jm}^{-2}$. Upon further loading, the crack tip and the crack nucleated ahead of the particle depleted region coalesce to form a continuous crack, see Figures 12(c) and (d).

Figure 13 shows the crack path in simulations with $E_p = 3.9\ \text{GPa}$, $G_{c\ p-m} = 120\ \text{Jm}^{-2}$ and $G_{c\ f-m} = 140\ \text{Jm}^{-2}$, $180\ \text{Jm}^{-2}$ and $240\ \text{Jm}^{-2}$. In Figures 13 (a) and (b) a crack is nucleated at the interlayer-lamina interface. However, when the **surface energy** of the fiber-matrix interface is increased to $240\ \text{Jm}^{-2}$ the crack continues growing confined to the interlayer region. In Figure 14 similar simulations are performed with $E_p = 3.9\ \text{GPa}$ and $G_{c\ p-m} = 120\ \text{Jm}^{-2}$ and $G_{c\ f-m} = 120\ \text{Jm}^{-2}$, $140\ \text{Jm}^{-2}$ and $160\ \text{Jm}^{-2}$. Clearly the propensity of the crack to nucleate at the interlayer-lamina interface depends not only on the **surface energy** of the interfaces but also on the relative stiffness of the particle and the matrix and the distribution of particles. Even more important is the size and distribution of rich resin regions that drive the crack towards the weakest interface and may result in delamination along the fibers.

Figure 10: Simulations of Mode I crack propagation with a particle-depleted region in with $G_c p-m = 480 \text{ Jm}^{-2}$ and $G_c f-m = 120 \text{ Jm}^{-2}$. (a) $E_p = 3.9 \text{ GPa}$, (b) $E_p = 9.8 \text{ GPa}$ and (c) $E_p = 19.5 \text{ GPa}$.

Figure 11: Simulations of Mode I crack propagation with a particle-depleted region with $G_c p-m = 120 \text{ Jm}^{-2}$ and $G_c f-m = 120 \text{ Jm}^{-2}$ (a) $E_p = 3.9 \text{ GPa}$, (b) $E_p = 9.8 \text{ GPa}$ and (c) $E_p = 19.5 \text{ GPa}$.

Figure 12: Crack path evolution in with a particle-depleted region, $E_p = 9.8 \text{ GPa}$, $G_c p-m = 120 \text{ Jm}^{-2}$ and $G_c f-m = 140 \text{ Jm}^{-2}$.

Figure 13: Simulations of Mode I crack propagation with $E_p=3.9 \text{ GPa}$ G_c
 $p-m = 120 \text{ Jm}^{-2}$, (a) $G_c f-m=140 \text{ Jm}^{-2}$, (b) $G_c f-m=180 \text{ Jm}^{-2}$ and (c) G_c
 $f-m=240 \text{ Jm}^{-2}$.

Figure 14: Simulations of Mode I crack propagation in $E_p=9.8 \text{ GPa}$ with G_c
 $p-m = 120 \text{ Jm}^{-2}$, (a) $G_c f-m=120 \text{ Jm}^{-2}$, (b) $G_c f-m=140 \text{ Jm}^{-2}$, and (c)
 $G_c f-m=160 \text{ Jm}^{-2}$.

4 Summary

PFDM simulations of Mode I crack propagation are performed to study the failure mechanisms that dominate the crack evolution in a particle toughened interlayer. The elastic constants of the particles, the **surface energies** of the particle-matrix and fiber-matrix interfaces are varied to study their effects. It is important to emphasize that three configurations with different random particle arrangements were performed for each plot reported. In the three simulations the crack paths observed were quantitative similar. The PFDM simulations are able to capture several fracture mechanisms observed during damage of particle-toughened interlayers [5, 6], i) crack trapping, ii) crack deflection, iii) particle-matrix debonding, iv) bridging ligaments, v) crack path deflection towards the lamina and back to the interlaminar region. **The failure processes observed for different sets of material properties are summarized in Table 4.**

The PFDM simulations show that failure in the interlayer is a heterogeneous process with distributed onset of damage ahead of the crack tip. This is in agreement with experiments that reveal distributed damage sites rather than the propagation of a single crack tip [6]. As an example of this behavior, what seems a crack deflecting towards the lamina in Figure 12 is in fact a crack nucleation site ahead of the crack tip that coalesces with the crack upon further loading. Crack branching, shown in Figure 9, is another example that results from distributed initiation sites that coalesce with the advancing crack tip.

The simulations show that the **surface energy** of the particle-matrix and fiber-matrix interfaces need to be included in models to determine the crack path due to the competition between these two types of interfacial failure. In sections 3.2 and 3.3 the fiber and matrix are assumed to be perfectly bonded and therefore, the crack remains confined to the interlayer. An exception is the simulation in with $E_p=19.5$ GPa (Figure 3 (c)) where due to the high stiffness of the particles the crack may propagate to the lamina-interlayer interface.

The particle distribution plays a predominant role as well. The PFDM simulations show that in resin rich regions the crack path may be driven towards the fiber-matrix interface in agreement with experiments. Finally, increasing the particle stiffness and the **surface energy** of the particle-matrix interface does not always lead to an improvement of the composite toughness. When the particles are stiffer than the matrix, the fiber-matrix interface becomes a preferential path for the crack by the nucleation micro-cracks ahead of the crack tip creating extensive delamination zones.

E_p[GPa]	$G_{c\ p-m}$[Jm⁻²]	$G_{c\ f-m}$[Jm⁻²]	failure processes
3.9	480	480	i) crack propagates through particles. Figure 3(a), Figure 7(a)
9.8	480	480	i) crack propagates through particles, ii) crack advances towards the interface with the lamina. Figure 3(b), Figure 7(b)
19.5	480	480	i) particle bridging ligament, ii) crack propagates along the interface between the lamina and the interlaminar region, iii) micro-crack nucleation ahead of the crack tip. Figure 3(c), Figure 7(c).
3.9	120	480	i) particle debonding, ii) micro-crack nucleation ahead of particle depleted regions. Figure 4(a), Figure 8(b).
9.8	120	480	i) particle debonding. ii) micro-crack nucleation ahead of particle depleted regions. Figure 4(b), Figure 8(b).
19.5	120	480	i) particle debonding ii) micro-crack nucleation ahead of the crack tip, iii) crack deflection towards the interlaminar region, iv) crack branching. Figure 4(c), Figure (5), Figure 8(c), Figure 9.
3.9-19.5	120	120	i) particle debonding. ii) micro-crack nucleation ahead of particle depleted regions, iii) delamination along the matrix-fiber interface. Figure 11, Figure 12.

3.9-19.5	480	120	i) delamination along the matrix-fiber interface. Figure (6).
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Table 4: Failure processes observed in the simulations for different sets of material properties.

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