

A Numerical Model to Simulate the Pullout of Carbon Fibre with Radially Grown Carbon Nanotubes

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Abstract A recent development of carbon nanotubes (CNTs) based structural composite materials is to grow CNTs radially on micro-fibres and to use these hybrid fibres to develop superior 3D composites. Due to the extremely high tensile strength and stiffness of CNTs and the increased interfacial areas, it is expected that the fracture toughness of the new composites will be increased substantially. To evaluate the bridging effect of this new hybrid fibre, a finite element model was developed to simulate the pullout of a carbon fibre with radially grown carbon nanotubes. The physical multi-length scale problem is treated at the single macroscopic scale by using equivalent spring elements to simulate the bridging effect from CNTs. The nonlinear properties of the spring elements are obtained from the finite element simulation of a single CNT pullout, where the CNT is simulated by using membrane elements. The bonding and debonding behaviours between the carbon fibre-matrix interface and CNT-matrix interface are described by cohesive laws. The cohesive law for the carbon fibre-matrix interface was calibrated from macroscopic single fibre pullout experimental curves. The cohesive law for the CNT-matrix interface was calibrated from published experimental data on single CNT pullout. The numerical results indicate that the effect of growth CNTs on the carbon fibre is significant. The pullout force of the hybrid fibre has a significant increase at the elastic stage before interfacial debonding, which leads to a higher specific pullout energy.

Keywords carbon nanotube, composite, pullout, cohesive law, finite element

1. Introduction

Carbon fibre, which contains at least 92% carbon [1], has been used in broad fields such as aircraft, marine, energy, biomedical and recreational applications due to its high specific stiffness, specific strength, and low thermal expansion characteristics. Carbon nanotubes (CNTs) are the finest and strongest fibres with tube diameter on the nanoscale and lengths from micro to millimetres (up to 18mm) [2]. They are considered as the new generation of reinforcing phase in fabricating nanocomposite materials. Adding 1wt% of CNTs to matrix material, the stiffness of the composite can increase up to 42% [3]. A recent development of carbon nanotubes (CNTs) based structural composite materials is to grow CNTs radially on micro-fibres and to use these hybrid fibres to develop superior 3D composites, carbon nanotube (CNT)/carbon fibre (CF) hybrid composites, which take the advantages of carbon fibres and carbon nanotubes. The traditional fibres only provide the in-plane (x and y directions in Cartesian coordinates) reinforcement. The delamination always occurs in these traditional laminated composites since there is no reinforcement in the direction of the coordinate z (through-thickness direction) to resist the crack initiation and propagation. Since the CNTs align in multi-directions in the matrix, they can provide the reinforcement to the matrix in different directions as 3D reinforcement.

Due to the extremely high tensile strength and stiffness of CNTs and the increased interface areas, it is expected that the fracture toughness of the new composites will be increased substantially. In recent years, some researchers have grown CNTs on the surfaces of carbon fibres to improve the interfacial strength between the fibres and matrix [4-8]. A single fibre pullout test is one of the most widely used techniques to quantify the interfacial strength. In the past few years, only few researches have been carried out to understand the mechanical performance of CNTs reinforced composites using the single fibre pullout test. For example, Qian, et al. [9] examined the interfacial shear strength of CNT-grafted fibres using both the single fibre pullout and push out test. The

experiment results of the CNT-grafted fibres pullout test showed that the interfacial shear strength have a significant increase (from 75 to 118MPa).

Due to the extremely small size, the role of using this hybrid fibre in composite fracture is still unclear. Thus it is essential to use theoretical and numerical analysis to examine the pullout of this new hybrid fibre from the polymer matrix. Several numerical studies have been carried out on the CNT pullout, but no numerical study on CNT/CF hybrid fibre pullout has been reported. Kulkarni et al. [10] and Nie et al. [11] developed two similar multiscale models to evaluate the effect of interfacial strength on the elastic modulus of CNT/CF fibre reinforced polymer composites. In their simulation, they firstly modelled a nanocomposite formed by a single CNT embedded in the epoxy matrix (CNT/matrix) and numerically predicted the overall mechanical properties of the nanocomposite. The second step is to consider the nanocomposite as an equivalent matrix and use it to form a single carbon fibre nanoreinforced laminated composite.

Cohesive zone modelling is a commonly used technique to investigate the failure governed by crack or debonding propagation. It establishes the traction-separation relation for the interface and bridges the gap between the stress- and energy-based approaches [12]. Many studies have been carried out on the interfacial behaviours of fibre-reinforced polymer (FRP) and concrete under mode II conditions [13, 14] by using cohesive zone modelling. However, very little attention has been paid to use cohesive zone modelling to numerically simulate the CNT/CF hybrid fibre pullout.

The purpose of this paper is to investigate the single CNT/CF hybrid fibre pullout test through coupling the single carbon fibre pullout and CNT pullout. The physical multi-length scale problem is treated at the single macroscopic scale by using equivalent spring elements to simulate the bridging effect from CNTs. The bonding and debonding behaviours between the carbon fibre-matrix interface and between the CNT-matrix interface are described by cohesive laws. In this paper, a finite element model of CNT/CF hybrid fibre pullout is firstly presented. Secondly the single carbon fibre pullout and the single CNT pullout are studied by using cohesive zone modelling. The two different cohesive laws for the carbon fibre-matrix interface and CNT-matrix interface are calibrated from macroscopic single carbon fibre pullout experimental curves and published experimental data on single CNT pullout, respectively. The single CNT/CF hybrid fibre pullout test is investigated by using a cohesive finite element model in which the two pullout processes at different scales are coupled. The effect of CNTs on debonding force and specific pullout energy are investigated.

2. Numerical Model

CNTs can be radially grown on the surface of carbon fibres using chemical vapour deposition (CVD). Fig. 1 shows Scanning Electron Microscope (SEM) image of carbon fibre after CNT growth [15]. These hybrid fibres are then embedded in an epoxy matrix to produce CNT/CF hybrid fibre reinforced composites. The diameter and interfacial area of the fibre are significantly increased by growth CNTs which can improve the fracture toughness of traditional fibre-matrix composites.

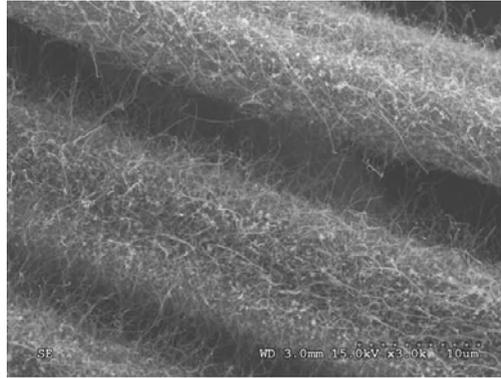


Figure 1. SEM image of a hybrid fiber with CNTs [15]

In a typical single fibre pullout test, there are three stages during the pullout process, including elastic deformation stage before debonding, debonding stage and sliding stage. In the first stage, the fibre and the matrix are well bonded. As the pullout force increases, a crack initiates and propagates along the interface between the fibre and the matrix, leading to a complete debonding, which is the debonding stage. In the last stage, the fibre slides out from the matrix, with friction acting between the two newly formed surfaces. One condition of pullout test is that the length of fibre embedded in the matrix must be less than the critical embedded length for debonding. Otherwise the fibre will break before the debonding occurs.

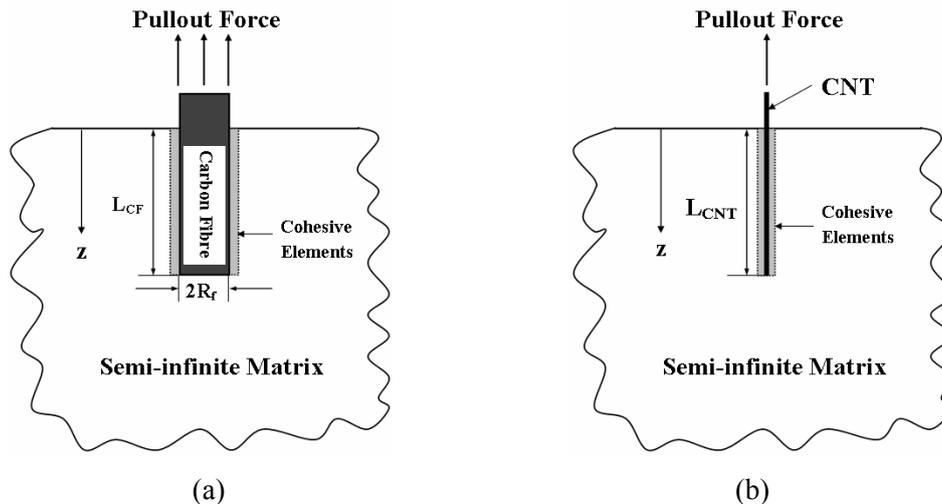


Figure 2. (a) A schematic diagram of a single carbon fibre pullout model; (b) A schematic diagram of a single CNT pullout model

In this study, the CNT/CF hybrid fibre pullout was simulated through coupling of the single carbon fibre pullout and single CNT pullout. In the single carbon fibre pullout, a pullout displacement was applied uniformly on the top surface of the cylindrical fibre embedded in a semi-infinite matrix in the axial direction as shown in Fig. 2(a). The commercial finite element package Abaqus was used to investigate the carbon fibre pullout problem. A two-dimensional axisymmetric model was constructed with a very fine mesh (element type CAX4 and size $1\mu\text{m} \times 1\mu\text{m}$) in the area around the interface between the fibre and the matrix to ensure the accuracy of the numerical results. A small number of four-node, axisymmetric cohesive elements (COHAX4) were used to define the cohesive zone [16]. In the single CNT pullout, another two-dimensional axisymmetric model was developed using a single cylindrical CNT embedded in a semi-infinite matrix. A pullout displacement was also applied on the top of the CNT in the axial direction as shown in Fig. 2(b). L_{CNT} is the total embedded CNT length. Due to its extreme small size (in nanoscale), membrane elements (MAX1)

were used to represent the CNT. The membrane element can be used to represent the thin surface which offers strength in the plane of the element without bending stiffness [17]. The axisymmetric cohesive elements (COHAX4) were also used to define the cohesive zone between the interface of CNT and matrix.

The physical problem of the CNT/CF hybrid fibre pullout was treated as a cylindrical carbon fibre attached with spring elements which perform the bridging effect of CNTs embedded in a semi-infinite matrix, as shown in Fig. 3(a). Spring element is defined as a deformable member which is subjected to tensile or compressive loads in the axial direction. In the pullout simulation, the spring elements are elongated and the spring forces resist the fibre to be pulled out [18, 19]. The parameters involved to define the nonlinear spring elements were obtained based on the numerical results from the single CNT pullout. The radius of the fibre is denoted as R_{CF} and L_{CF} is the total embedded fibre length. As the model developed in carbon fibre pullout [16], a pullout displacement will be applied uniformly on the top surface of the fibre in the axial direction. Since the spring elements are used to simulate the force of CNTs applied on the carbon fibre during the pullout, the debonding of CNT-matrix interface will not be explicitly simulated in the hybrid fibre pullout model. Therefore the debonding only occurs between the carbon fibre and matrix interface, which is assumed to initiate at the carbon fibre-matrix interface and propagate longitudinally along the carbon fibre. It is also assumed that the normal stress along the carbon fibre does not exceed its material ultimate strength, so the carbon fibre will not break before it is pulled out. The plastic behaviour is not considered in this paper.

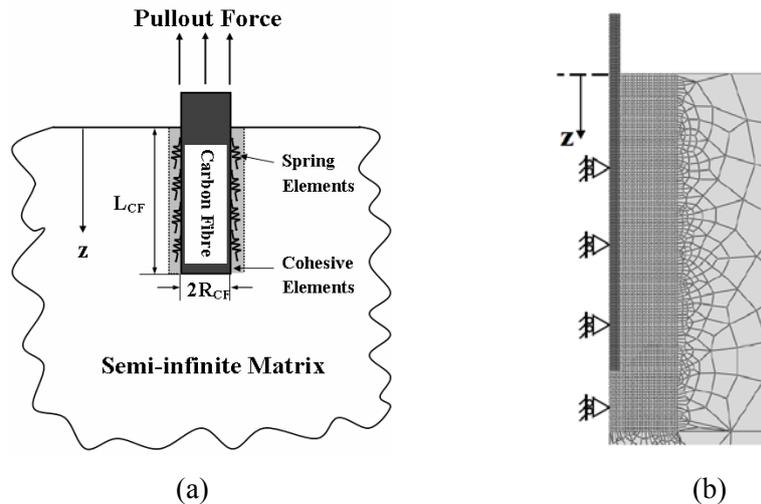


Figure 3. (a) A schematic diagram of the CNT/CF hybrid fibre pullout model using spring element to perform the CNT; (b) Axisymmetric finite element model for a single CNT/CF hybrid fibre pullout with a fine mesh around the interface

Due to the symmetry of this problem, a two-dimensional axisymmetric model was constructed in this finite element analysis. The radius and depth of the matrix are much larger than the dimensions of the fibre in the numerical models so as to simulate a semi-infinite matrix body. The bottom of the model is constrained in both the radial and axial directions. The vertical side along the axisymmetric axis is constrained only in the radial direction. The model contains a total of 5149 four-node quadrilateral elements. A very fine mesh with the smallest elements (CAX4) of $1\mu\text{m} \times 1\mu\text{m}$ were used in the area around the interface as shown in Fig. 3(b). A small number of four-node, axisymmetric cohesive elements (COHAX4) were used to define the cohesive zone. Nonlinear spring elements (SPRING1) were used to model the bridging effect of CNTs in the CNT/CF hybrid fibre pullout. In reality, the CNTs grown on the carbon fibre surface should be

considered as the CNTs embedded in the matrix at an inclination angle of 90° with respect to pullout direction. For this case, the CNT should be considered as a flexible string passing over a frictional pulley, and a snubbing friction model can be derived to relate the pullout force to the fiber inclination angle [20]. Therefore, a frictional pulley model for pullout force of the single inclined CNT was firstly introduced

$$F_{CNT(\Phi)} = F_{CNT(\Phi=0)} e^{f\Phi} \quad (1)$$

where $F_{CNT(\Phi)}$ is the pullout force of inclined CNT with the pullout direction Φ . f is the snubbing friction coefficient which is assumed as 0.1 in this study. Secondly, the pullout force of the single CNT obtained in FE simulation with pulley effect was applied to define the nonlinear properties of the spring elements.

In addition, two cohesive zones were defined between the carbon fibre-matrix interface and CNT-matrix interface in this study. In the single carbon fibre pullout simulation, a bilinear cohesive law was implemented [16]. In the single CNT pullout simulation, the cohesive zone model was adopted from Tvergaard [21] and Chaboche et al. [22]. The interfacial debonding during a fibre pullout test is a pure mode II fracture problem. Therefore the mode II cohesive law [22] can be simplified as

$$T_t = \left(\frac{27u_t^3}{4\delta_t^3} - \frac{27u_t^2}{2\delta_t^2} + \frac{27u_t}{4\delta_t} \right) \tau_{\max} \quad (2)$$

where u_t is the tangential separation and δ_t is the complete tangential separation. τ_{\max} is the interfacial crack initiation stress under shear loading condition. Correlating to energy-based fracture mechanics, the fracture energy G_{IIc} is the area under traction-separation curve.

3. Results and Discussion

3.1 Single Carbon Fibre Pullout Simulation

A comparison study between a finite element simulation and an experimental pullout test was first carried out as shown in Fig. 4 [16]. A $7\mu\text{m}$ diameter HTA-7-6K carbon fibre is embedded in an Epilox epoxy matrix with the embedded length $L_{CF} = 100\mu\text{m}$, which is consistent with the experiment by Bogoeva-Gaceva et al. [23]. The residual thermal stresses were considered on the assumption of $\Delta T = -95^\circ\text{C}$.

From Fig. 4, it can be seen that the simulated pullout curve agrees overall very well with the experimental curve from Bogoeva-Gaceva et al. [23] with these fitted parameters $\tau_{\max(CF)} = 45\text{MPa}$, separation displacement of crack initiation $\delta_{d(CF)} = 21.86\mu\text{m}$, and the complete separation displacement $\delta_{t(CF)} = 25\mu\text{m}$. These calibrated parameters of cohesive zone are used to simulate CNT/CF hybrid fibre pullout in the following section.

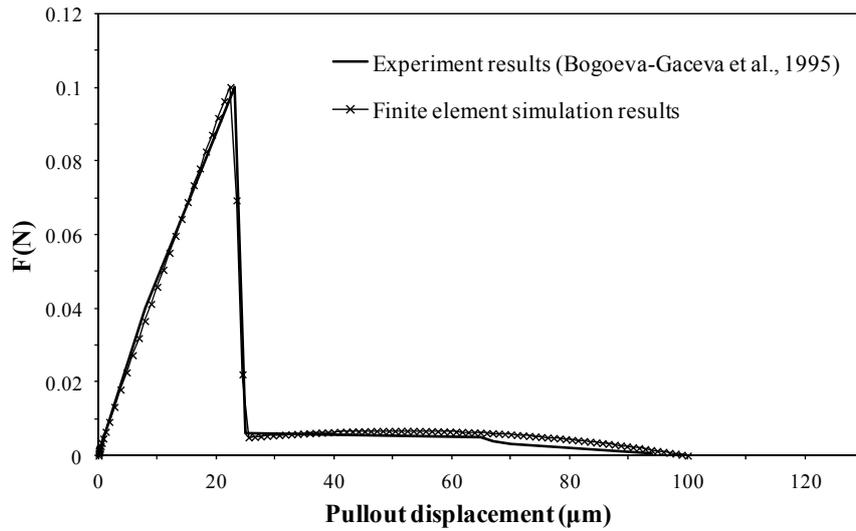


Figure 4. Comparison between experiment and FE simulation for single carbon fibre pullout curve

3.2. Single CNT Pullout Simulation and Validation

Similarly, another comparison study between a finite element simulation and an experimental single CNT pullout test was carried out. In this study, a 24nm diameter multiwalled-carbon nanotube (MWNT) is embedded in an epoxy matrix with the embedded length $L_{CNT} = 2.6\mu\text{m}$, which is consistent with the experiment by Cooper et al. [24].

Fig. 5 shows the simulated single CNT pullout curve. The numerical results were compared with the experimental results as shown in Table 1. W_{CNT} is the total CNT pullout energy, which is the total area under the pullout force-displacement curve. It can be seen that the numerical results agrees overall very well with the experimental results listed by Cooper et al. [24] with these fitted parameters $\tau_{\max(CNT)} = 36\text{MPa}$, $\delta_{d(CNT)} = 140\text{nm}$, $\delta_{t(CNT)} = 410\text{nm}$ which are used to define the nonlinear properties of spring elements in the following section.

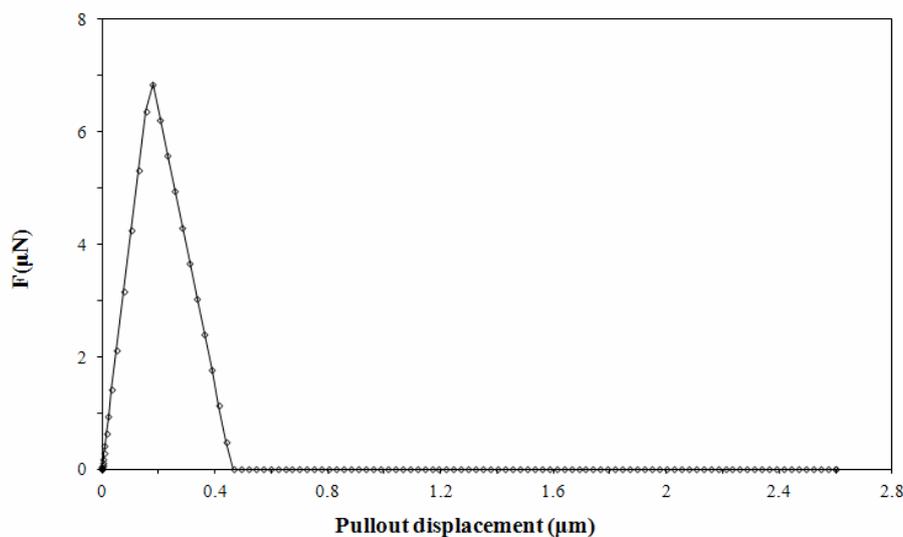


Figure 5. FE simulation for single CNT pullout curve

Table 1. Comparison between experiment and FE simulation for single CNT pullout

	FE simulation	Cooper et al. [24]
Maximum pullout force $F_{\max(CNT)}$ (μN)	6.845	6.8 \pm 1.7
Total CNT pullout energy W_{CNT} (J)	1.63 \times 10 ⁻¹²	1.6 \times 10 ⁻¹²

3.3. CNT/CF Hybrid Fibre Pullout

3.3.1. Estimation of length and number of CNT grown carbon fibre

To simulate a single CNT/CF hybrid fibre pullout, the length and number of CNT radially grown on the single carbon fibre should be identified. In this study, the length and number of CNTs were estimated based on the experimental results obtained by Zhang et al. [25]. In their experiment, carbon nanotubes were successfully grown on the surface of a 7 μm diameter carbon fibre T650 by chemical vapor deposition. The diameter of the CNT/CF hybrid fibre measured was approximately 20 μm . Therefore, the length of CNTs (L_{CNT}) grown on the carbon fibre surface can be estimated as 6.5 μm .

The number of CNTs grown on the carbon fibre surface was calculated based on the CNT population density estimated from the experiment. The CNT population density at the carbon fibre surface was estimated as high as 8 \times 10⁹ tubes/cm² [25]. In this study, it is assumed that CNTs are uniformly grown on the carbon fibre surface. The number of CNTs grown on the carbon fibre surface per unit length can be calculated as 1760 tubes/ μm . It is also assumed that there are 100 layers of CNTs uniformly distributed along the carbon fibre-matrix interface ($L_{CF} = 100\mu\text{m}$ with element size 1 $\mu\text{m} \times 1\mu\text{m}$). Therefore, the total number of CNTs distributed on each layer was estimated as 1760 tubes. In addition, 99 layers of CNTs were used to joint with the carbon fibre in the hybrid fibre simulation which excluded the bottom layer of CNTs.

3.3.2. CNT/CF hybrid fibre pullout force

The maximum pullout force of hybrid fibre ($F_{\max(H)}$), is one of the most important parameters recorded from a pullout test, which is used to calculate the average interfacial strength. Fig. 6 showed the case of CNTs completely pulled out with the single carbon fibre. It can be seen that the magnitude of the maximum pullout force for CNT/CF hybrid fibre ($F_{\max(H)} = 0.39\text{N}$) is much higher than the carbon fibre pullout ($F_{\max(CF)} = 0.1\text{N}$). The maximum pullout force of the hybrid fibre has a significant increase by 290%. The increased pullout force was observed in the early stage of elastic deformation before the interfacial debonding occurred in carbon fibre. An et al. [26] performed an experiment of the CNT/CF hybrid fiber pullout to confirm that the maximum pullout force of hybrid fibre had a significant increase up to 120%. The difference probably is due to the different number of CNTs and carbon fibre embedded length used in this study.

The interfacial shear strength (IFSS), which is traditionally used to evaluate the interfacial bonding, is normally calculated from the load-displacement curve of a fibre pullout test. It can be defined as [27]

$$\tau_s = \frac{F_{\max}}{2\pi R_{CF} L_{CF}} \quad (3)$$

The IFSS increases from about 45.5MPa to 177MPa. This improvement of the interfacial shear strength is due to the increased interfacial area and roughness, which can provide a stronger interfacial bonding between the CNTs, carbon fibre and the matrix. A significant improvement in the IFSS was also observed in the experiment studies [7, 9, 26, 28]. This study indicates that the effect of growth of CNTs on the carbon fibre surface is significant, which can enhance the adhesion between the fibre-matrix and thus improve the delamination resistance of the composite.

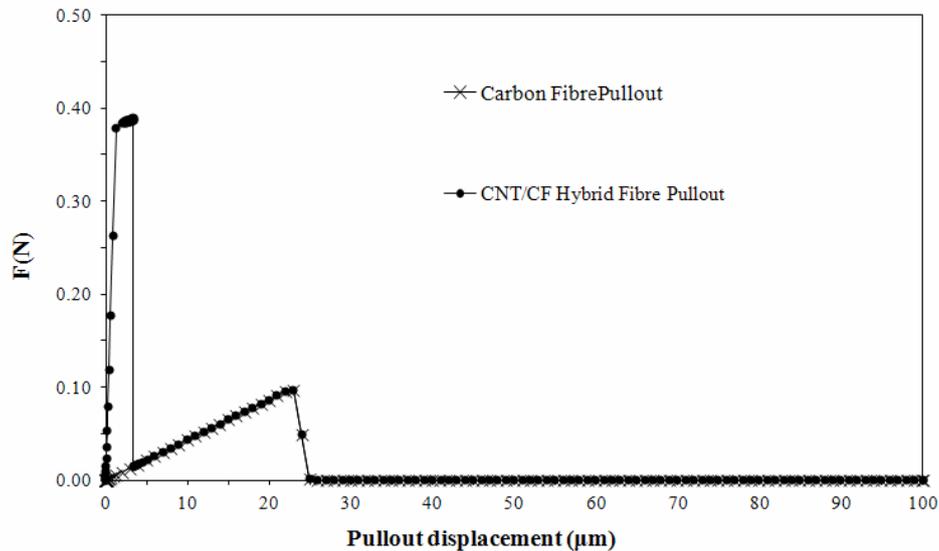


Figure 6. Comparison between the single carbon fibre pullout and the single CNT/CF hybrid fibre completed pullout curve.

3.3.3. Specific pullout energy

The specific pullout energy can be well used to quantify the beneficial effect of growth of CNTs on the bridging resistance of fibres in composites, which is first introduced in this paper. The specific pullout energy ω is defined as

$$\omega = \frac{W}{2\pi R_{CF} L_{CF}} \quad (4)$$

where W is the total fibre pullout energy, which is the total area under the pullout force-displacement curve. Specific pullout energy can be used to quantify the fibre bridging effect in a composite failure analysis. Fibre bridging appears behind a major crack tip in an intralaminar fracture, which enhances the fracture resistance of fibre reinforced composite. According to the calculated results, it can be seen that the influence of growth of CNTs on the specific pullout energy is significant. The magnitude of the specific pullout energy of CNT/CF hybrid fibre ($\omega_H = 1032\text{J/m}^2$) is much higher than the carbon fibre ($\omega_{CF} = 565\text{J/m}^2$). The specific pullout energy of the CNT/CF hybrid fibre increases by 83%. This is due to the large resistance induced by the carbon nanotubes, which requires more energy to pullout the fibre.

4. Conclusion

A finite element model was developed to simulate the pullout of a carbon fibre with radially grown carbon nanotubes. The bonding and debonding behaviours between the carbon fibre-matrix

interface and CNT-matrix interface were described by cohesive laws. Equivalent nonlinear spring elements were used to simulate the force of CNTs applied on the carbon fibre during the pullout. The numerical results of the CNT/CF hybrid fibre pullout from a case study show that the growth of CNTs on the carbon fibre surface has a significant effect on the maximum pullout force, and they increase the resistance in fibre at the elastic deformation stage before interfacial debonding. There also a signification improvement in the interfacial shear strength, which indicates that growth of CNTs can provide a stronger interfacial bonding between the CNTs, carbon fibre and the matrix. The beneficial effect of growth of CNTs on the bridging resistance of fibres in composites can be well quantified by the specific pullout energy. The increased specific pullout energy of the hybrid fibre indicates that growth of CNTs can enhance the fracture resistance of fibre reinforced composites.

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