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Measuring fiber–matrix interfacial adhesion by means of a ‘drag-out’ micromechanical test

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Abstract

We have recently developed an experiment to measure the interfacial adhesion in nanotube–polymer composites by ‘dragging-out’ a single nanotube from a polymer matrix using an atomic force microscope tip. To quantify the data, an approximate analysis was used. Here, this ‘drag-out’ configuration is reproduced at a larger scale, namely, using a single flexible fiber (polyethylene) bridging a polymer (epoxy) hole. The data generated from this single fiber drag-out experiment was used as input in a new theoretical model that evaluates the interfacial shear adhesion at the fiber–matrix interface. Comparisons were made between the data generated from the single fiber drag-out and independent pull-out data produced in a classical microbond experiment with the same material system. The drag-out data compare fairly well with the microbond test data, and are found to be of the same order of magnitude as in the literature.

Keywords: B. Adhesion; B. Interface/interphase; C. Micro-mechanics; D. Mechanical testing; Drag-out test

1. Introduction

A relatively complex experimental technique for performing carbon nanotube pullout from a polymer matrix was recently developed in our laboratory [1]. The procedure provides a direct measurement of the shear adhesion of the carbon nanotube/polymer interface for multiwall carbon nanotube (MWNT) specimens. The technique can be briefly summarized as follows. MWNT/epoxy nanocomposites containing holes spanned by MWNTs were prepared, and the MWNTs appeared to be well anchored on either side of the holes. The nanotube–polymer adhesion is then probed by lateral dragging out of the MWNTs from the epoxy matrix using a scanning probe microscope tip. Location of suitable polymer holes and nanotubes and imaging of their subsequent detachment was achieved by transmission electron microscopy. This experiment represents the first attempt to measure the interfacial adhesion in nanocomposites (and was soon followed by a second type of experiment [2,3]). To quantify the experiment in Ref. [1], an approximate analysis was used [1]. Here, this ‘drag-out’ configuration is examined at a larger scale, namely, using a single flexible fiber (polyethylene) bridging a polymer hole. The data generated from this single fiber drag-out experiment serve as input in a new theoretical model that evaluates the interfacial shear adhesion at the fiber–matrix interface. Comparisons are then made between the data generated from the single fiber drag-out and independent pull-out data produced in a classical microbond experiment [4–7] with the same material system.

2. Theory

We start by a brief description of the microbond test. The specimen consists of a matrix droplet of length l_e spread onto a single fiber. A pull-out force P is applied to the fiber to shear it out of the droplet. Assuming that the load is uniformly distributed along the fiber embedded length, the interfacial shear strength (IFSS) is calculated as

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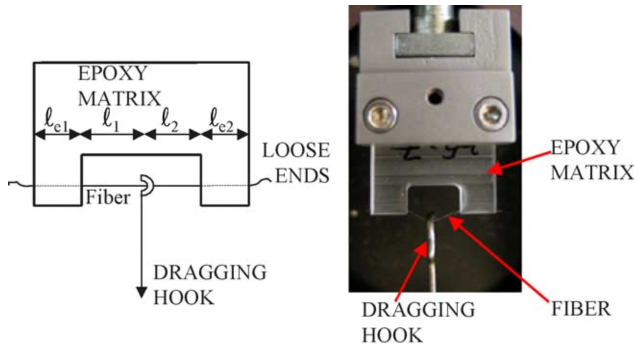

Fig. 1. Schematic diagram of drag out specimen and picture of spectra/epoxy specimen in the tensile holder.

the maximum average interfacial shear stress at failure (τ) using:

$$\tau = \frac{P}{\xi l_e} \tag{1}$$

where ξ is the fiber perimeter [4].

The drag out configuration involves a sample that has a free length and two embedded fiber areas. A force is applied at a point on the free length in a direction perpendicular to the fiber, as shown in Fig. 1. The balance of forces for this configuration is shown in Fig. 2. A hook applies a tensile force F at a distance l_1 and l_2 from the left (A) and right (B) edges, respectively. H is the distance perpendicular to the baseline AB. The force causes a tension T_i in the fiber. The subscripts 1 and 2 refer to the left and right sides, respectively. The tension is a vector with a component P_i parallel to the baseline AB and a component R_i perpendicular to AB. The parallel component is equal to the pull out force that acts to debond the fiber from the matrix. If $l_1 \neq l_2$ then $P_1 \neq P_2$ and there is a horizontal balancing force f. A full expression for the force balance is presented in Appendix A, but for the simple case of $l_1 = l_2 = l_{1/2}$ we have $f = 0$ and

$$P_1 = P_2 = \frac{F}{2}\frac{l_{1/2}}{H} \tag{2}$$

$$R_1 = R_2 = \frac{F}{2} \tag{3}$$

Eqs. (2) and (3) provide the vertical and horizontal force components if the applied force F and the geometrical parameters $l_{1/2}$ and H are known.

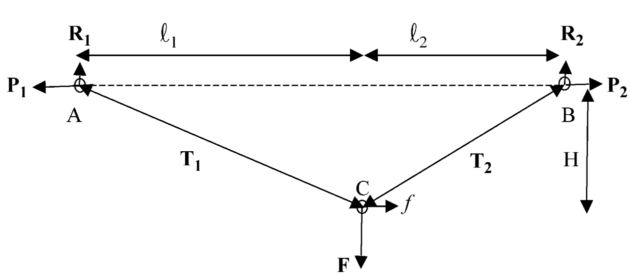

Fig. 2. Force equilibrium for the drag-out configuration.

In the elastic region, governed by Hooke's law, before debonding between the fiber and matrix occurs, the applied force F and the component P parallel to AB are given by (see Appendix A)

$$F = \frac{2AEH}{l_{1/2}}\left(1 - \frac{l_{1/2}}{\sqrt{H^2 + l_{1/2}^2}}\right) \tag{4}$$

$$P = AE\left(1 - \frac{l_{1/2}}{\sqrt{H^2 + l_{1/2}^2}}\right) \tag{5}$$

where A is the fiber cross-section and E is the fiber Young modulus. For small values of $H/l_{1/2}$ these equations can be expanded in a Taylor series:

$$F \cong AE\left(\frac{H}{l_{1/2}}\right)^3 + o\left(\frac{H}{l_{1/2}}\right)^5 \tag{6}$$

$$P \cong \frac{AE}{2}\left(\frac{H}{l_{1/2}}\right)^2 + o\left(\frac{H}{l_{1/2}}\right)^4 \tag{7}$$

Taking the same uniform load distribution assumption as in the microbond test we calculate the IFSS (τ) as

$$\tau = \frac{P_D}{\xi l_e} \tag{8}$$

where P_D is the component P recorded at the deviation point from Eq. (5), ξ is the fiber perimeter, and l_e is the embedded length.

3. Experimental

A single ultra high modulus polyethylene (Spectra 1000, Allied Corp.) fiber was embedded into an Epon 815/Jeffamine T-403 epoxy matrix mixed in a 100:42 ratio. Polyethylene fibers were selected for this experiment for two reasons: their relatively poor adhesion to epoxy (which makes pull-out possible) and their high flexibility (which allows drag-out without premature failure). The epoxy matrix was cured in a silicon mold at room temperature for 5 days before testing was performed. The specimens are U-shaped (Fig. 1), with fiber embedded lengths of $l_e = 2$, 4, and 6 mm on each side. The fiber protrudes over a few millimeters outside the specimens, similar to the microbond test. A 1.5 mm diameter steel hook attached to the moving grip of a tensile tester (MiniMat 2000, Rheometric Scientific) is used to drag out the fiber from the U-shaped polymer specimen at the center of the fiber free length (i.e. $l_1 = l_2 = l_{1/2}$). The force applied, as well as the displacement, are recorded by the Minimat device. The typical drag-out rate used was 0.5 mm/min. The fiber geometry was measured using an optical microscope. Note that since the polyethylene fiber cross-sectional shape varies along its length, we used as an approximation a circular shape with an average diameter of 25 μm in the calculations.

4. Results and discussion

Fig. 3 shows a typical experimental result from the drag-out test, and a comparison with the predicted line (Eq. (4) or, equivalently, Eq. (6), using $E = 170$ GPa and $l_{1/2} = 4.5$ mm). Initially, when the applied force F and the deflection H are small, the elongation of the fiber and thereby the experimental force is indeed proportional to the cube of the deflection predicted by Eq. (6). Under isostrain conditions, the difference in elastic behavior between the fiber and the matrix induces a shear stress at the interface. As drag out proceeds, the interfacial shear stress increases until (neglecting bending) the interface fails. Following this, the pull-out force P is governed by friction between the debonded fiber and the matrix. The calculated drag-out force F keeps increasing because of the increasing angle between the fiber and the AB line and assuming no slippage occurred. Fig. 4a–c shows the experimental pull out force P calculated using Eq. (2) compared to the theoretical curve (Eq. (5) or equivalently, Eq. (7)). In the linear-elastic range, the pull-out force increases proportionally to the square of the deflection, as expected from Eq. (7), until fiber debonding occurs. Since the debonding (pull-out) force is smaller than the friction force, the latter continues to increase well after debonding. The increase is larger for longer embedded lengths because the friction area is bigger. For comparison, a microbond test was also performed with the same material system using a fiber embedded in 2 and 4 mm thick epoxy specimens (cured under the same curing conditions), in a direction parallel to the fiber. The interfacial shear adhesion was calculated using Eqs. (8) and (1) for the drag-out and microbond tests, respectively. The results are summarized in Table 1. The interfacial shear adhesion value is about 1.6 MPa for both types of tests. For comparison, Biro at al. [6] reported a value of 8.6 MPa for the IFSS in Spectra/Epon 828 using the microbond test, and Chappel et al. [7] reported a value of 5.7 MPa for the interlaminar shear adhesion using the same system. Thus, the values measured here are of the same order of magnitude as in the literature, albeit lower. This difference may be

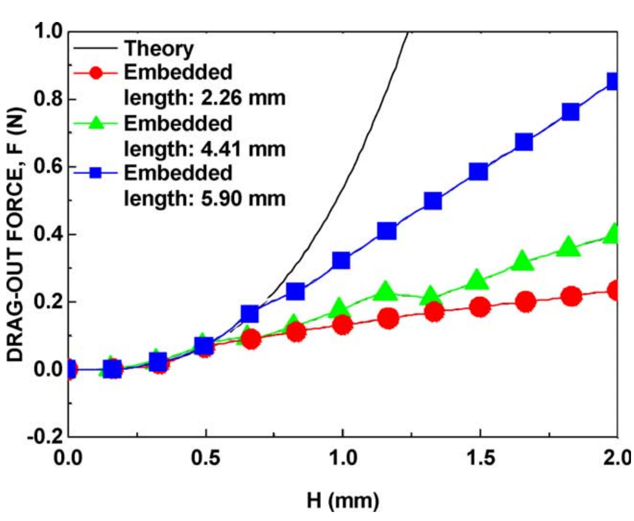

Fig. 3. The drag-out force F as measured with different embedded lengths. The predicted theoretical line (Eq. (4) or, equivalently, Eq. (6)) is shown for comparison.

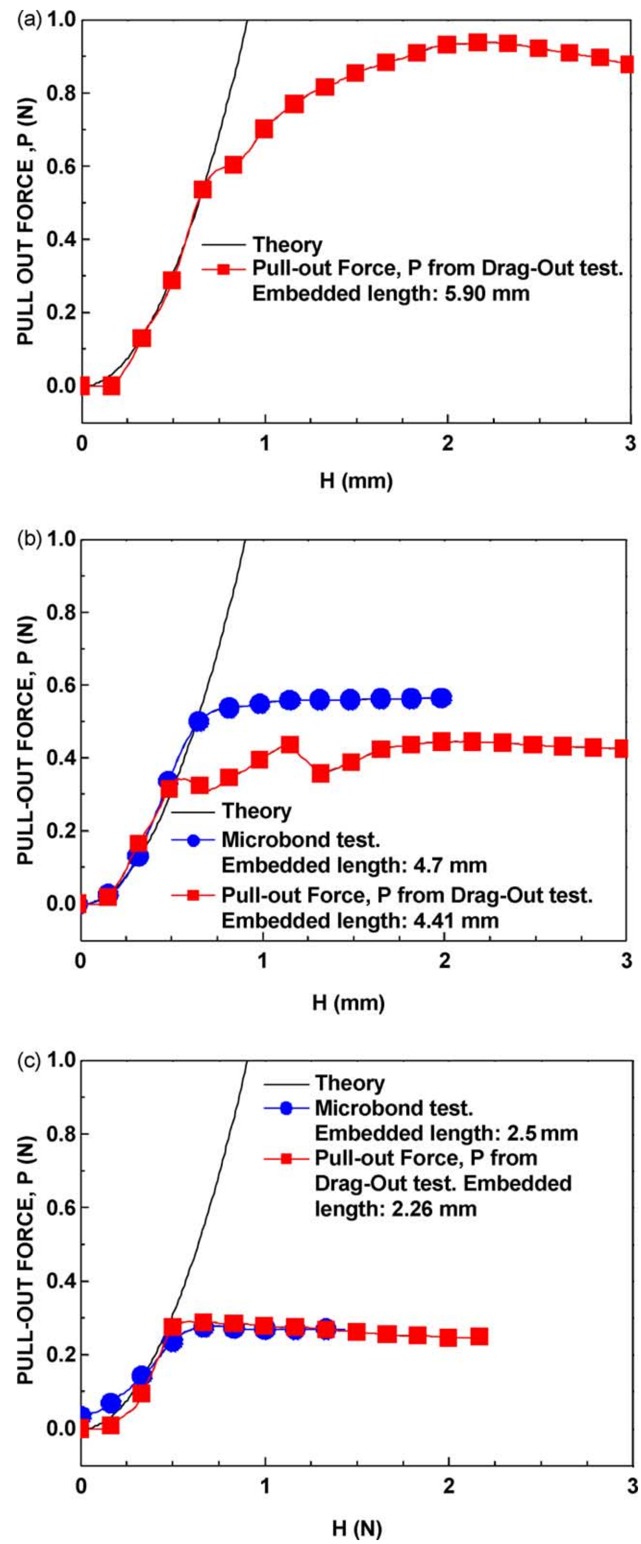

Fig. 4. The horizontal force P as measured for different embedded length (a) 5.9 mm (b) 4.41 mm (c) 2.26, in comparison with Eq. (5) and with traditional microbond test.

Table 1
Summary of IFSS results for epoxy/spectra system

Embedded length (mm)	Drag-out			Microbond		
	No. of samples	Average IFSS (MPa)	Standard deviation (MPa)	No. of samples	Average IFSS (MPa)	Standard deviation (MPa)
2.1–2.5	5	1.57	0.25	3	1.5	0.30
4.1–4.5	4	1.58	0.60	2	1.49	0.22
5.9–6.1	5	1.68	0.48			
Average		1.61	0.46		1.5	0.24

attributed to the different test methods, curing agents, curing cycles and epoxy resins.

5. Conclusions

We have demonstrated a new method for measuring the IFSS in composite systems in which either interfacial bonding is relatively weak or the embedded length is relatively short. The drag-out test can be used with flexible fibers in situations where the fibers have both ends embedded in the matrix. The data presented here compare fairly well with microbond test data. The analysis presented can therefore be used in cases where a drag out configuration is necessary such as in the nanotube–polymer adhesion test used in our previous work using a scanning probe microscope tip [1].

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Appendix A

In the dragging-out configuration described in Fig. 2, it is assumed that: (a) a steady state exists (slow test rate), (b) no interfacial debonding is initially present between the fiber and the matrix, (c) fiber rigidity in flexure is negligible and a hinge is formed in points A, B and C, and (d) no additional friction is created due to forces R_1 and R_2 since for small values of the displacement H, the force components R_1 and R_2 are close to zero. Under these assumptions, the following force balance equations along and perpendicular of the AB line are obtained:

$$P_1 = P_2 + f \tag{A1}$$

$$F = R_1 + R_2 \tag{A2}$$

From the torque balance we get:

$$R_1 l_1 = R_2 l_2 + fH \tag{A3}$$

Inserting Eq. (A2) in Eq. (A3) gives:

$$R_1 l_1 = (F - R_1) l_2 + fH \tag{A4}$$

Thus, rearranging:

$$R_1 = \frac{F l_2 + fH}{(l_1 + l_2)} \tag{A5}$$

$$R_2 = \frac{F l_1 - fH}{(l_1 + l_2)} \tag{A6}$$

The ratio between P and R is equal to the ratio between l and H:

$$\frac{P_1}{R_1} = \frac{l_1}{H} \tag{A7}$$

and

$$\frac{P_2}{R_2} = \frac{l_2}{H} \tag{A8}$$

Isolating P in Eqs. (A7) and (A8) and inserting the expressions for R from Eqs. (A5) and (A6) gives:

$$P_1 = \frac{F l_2 + fH}{(l_1 + l_2)} \frac{l_1}{H} \tag{A9}$$

and

$$P_2 = \frac{F l_1 - fH}{(l_1 + l_2)} \frac{l_2}{H} \tag{A10}$$

If $l_1 = l_2 = l_{1/2}$, then $f = 0$. Eqs. (A1), (A7) and (A8) become:

$$R_1 = R_2 = \frac{F}{2} \tag{A11}$$

$$P_1 = P_2 = \frac{F}{2} \frac{l_{1/2}}{H} \tag{A12}$$

The tension T in the fiber is:

$$T_1 = \sqrt{P_1^2 + R_1^2} \tag{A13}$$

$$T_2 = \sqrt{P_2^2 + R_2^2} \tag{A14}$$

Assuming that no interfacial debonding is present under a small applied force F, we have:

$$\sqrt{H^2 + l_1^2} = l_1\left(1 + \frac{T_1}{AE}\right) \tag{A15}$$

$$\sqrt{H^2 + l_2^2} = l_2\left(1 + \frac{T_2}{AE}\right) \tag{A16}$$

The right-hand sides arise from the geometry, and the left side from Hooke's law. A is the fiber cross section and E is its Young's modulus. Inserting Eqs. (A5), (A6), (A9), (A10), (A13) and (A14) into Eqs. (A15) and (A16) provides two equations that can be solved numerically to obtain F and f as a function of H. If $l_1 = l_2 = l_{1/2}$, the equations can be solved analytically as follows:

$$\sqrt{H^2 + l_{1/2}^2} = l_{1/2}\left(1 + \frac{\sqrt{P_1^2 + R_1^2}}{AE}\right)$$

$$\sqrt{H^2 + l_{1/2}^2} = l_{1/2}\left(1 + \frac{\sqrt{R_1^2\left(\frac{l_{1/2}}{H}\right)^2 + R_1^2}}{AE}\right)$$

$$\sqrt{H^2 + l_{1/2}^2} = l_{1/2}\left(1 + \frac{F}{2AE}\sqrt{\left(\frac{l_{1/2}}{H}\right)^2 + 1}\right)$$

$$F = \frac{2AE}{l_{1/2}}\left(\frac{\sqrt{H^2 + l_{1/2}^2} - l_{1/2}}{\sqrt{\frac{H^2 + l_{1/2}^2}{H^2}}}\right)$$

$$F = \frac{2AEH}{l_{1/2}}\left(1 - \frac{l_{1/2}}{\sqrt{H^2 + l_{1/2}^2}}\right) \tag{A17}$$

$$P = \frac{F}{2}\frac{l_{1/2}}{H} = AE\left(1 - \frac{l_{1/2}}{\sqrt{H^2 + l_{1/2}^2}}\right) \tag{A18}$$

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