Fibre–matrix adhesion and its relationship to composite mechanical properties

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Two major areas of enquiry exist in the field of fibre–matrix adhesion in composite materials. One is the fundamental role that fibre–matrix adhesion plays on composite mechanical properties. The other is what is the “best” method used to measure fibre–matrix adhesion in composite materials. Results of an attempt to provide an experimental foundation for both areas are reported here. A well-characterized experimental system consisting of an epoxy matrix and carbon fibres was selected in which only the fibre surface chemistry was altered to produce three different degrees of adhesion. Embedded single-fibre fragmentation tests were conducted to quantify the level of fibre–matrix adhesion. Observation of the events occurring at the fibre breaks led to the documentation of three distinct failure modes coincident with the three levels of adhesion. The lowest level produced a frictional debonding, the intermediate level produced interfacial crack growth and the highest level produced radial matrix fracture. High fibre volume fraction composites made from the same material were tested for on- and off-axis, as well as fracture, properties. Results indicate that composite results can be explained if both differences in adhesion and failure mode are considered. It will be further demonstrated that fibre–matrix adhesion is an “optimum” condition which has to be selected for the stress state that the interface will experience. The embedded single-fibre fragmentation test is both a valuable measurement tool for quantifying fibre–matrix adhesion as well as the one method which provides fundamental information about the failure mode necessary for understanding the role of adhesion on composite mechanical properties.

1. Background

As composite materials have moved to the forefront of activity in the materials community in the last two decades, there has been an increasing interest in being able to understand the physical and chemical mechanisms responsible for fibre–matrix adhesion as well as the role of fibre–matrix adhesion on composite properties. Early mechanical analyses have shown that when composites are fabricated and the applied load is coincident with the fibre direction, a simple rule of mixtures expression can be used to predict composite properties from the properties of the constituents. The resulting model has limited utility in practice because structural applications for composite materials rarely allow for the applied loads to be in the fibre direction only. There is almost always a component of the structural loads that is in a direction at an angle to the fibre axis. In these situations, fibre–matrix adhesion is important to ensure that a maximum stress level can be maintained across the interface and from fibre to matrix without disruption. At less than optimum levels of adhesion, the load-bearing capacity of the composite has to be reduced to compensate for the reduced adhesion at the interface or the structural component has to be increased in size by increasing ply thickness. Either option is undesirable because it negates a significant portion of the weight and cost advantage of the composite structural component over an equivalent monolithic material.

Most early work on the development of composite materials considered fibre–matrix adhesion to be a necessary condition to ensure good composite properties. The majority of effort was concentrated on increasing fibre–matrix adhesion through the use of surface treatments and coatings. The patent literature on carbon-fibre surface treatments and a large portion of the silane literature on finishes for glass fibres contain descriptions of processes for improving fibre–matrix adhesion.

It is very desirable to have a testing method to measure the adhesion between fibre and matrix which can provide a reproducible, reliable method for investigating and measuring fibre–matrix adhesion. Composite testing of unidirectional composite specimens with loads applied to place the fibre–matrix interface under shear or under a tensile load perpendicular to the fibre axis were commonly used methods. Because the majority of early composite research was
done on glass fibres, these approaches were satisfactory from an engineering sense. However, once different reinforcing fibres (e.g. carbon and boron fibres, polymeric fibres and different glass fibres) were introduced, fabrication of large samples of composites made from experimental runs of new materials tended to be costly.

Several testing methods were developed for measuring fibre-matrix adhesion using single fibres or groups of fibres. The goal was to measure fibre-matrix adhesion in a way that would be a predictor of composite fibre-matrix adhesion, as well as to understand the fundamental physics and chemistry of adhesion itself. Although there has been a wide variety of methods proposed in recent years, most can be reduced to three basic measurement methods. They are fibre pull-out methods, fibre fragmentation methods, and fibre micro-compression methods [1].

In the pull-out experiments, one end of a single fibre is embedded in a block of matrix (Fig. 1). The free end is gripped and an increasing load is applied as the fibre is pulled out of the matrix while the load and displacement are measured. The maximum load, $F$, measured before detachment of the fibre from the matrix is related to the average value of the fibre-matrix shear strength, $\tau$, through the equation.

$$ F = \tau \pi ld $$

where $\pi d$ is the fibre circumference and $l$ is the embedded length. In principle, this is a direct measurement method, but closer inspection shows that the state of stress at the juncture of the fibre and the matrix creates a normal tensile interracial force not encountered in an actual composite. The normal tensile force can act on the interface to reduce the measured shear strength. If the length of fibre embedded in the block of matrix is longer than the critical transfer length for that fibre-matrix combination, the fibre will fracture within the block. This requires the fabrication of thin discs of matrix or very precise control of the depth of the fibre end, because fibre length to diameter ratios of 50–100 are typical. For 10 $\mu$m diameter fibres, this means that the matrix thickness or fibre-end location in a matrix block must not exceed 500 $\mu$m in size [2] or by using half-droplets formed on surfaces [3].

An alternative single-fibre method relies on using the shear stress transfer process to produce fragmentation in a continuous fibre (Fig. 2). In this technique, the fibre is totally encapsulated in a matrix coupon, a tensile load is applied to the coupon, and an interracial shear stress-transfer mechanism is relied upon to transfer the coupon tensile forces to the encapsulated fibre through the interface [4]. As the load is increased on the specimen, shear forces are transmitted to the fibre along the interface. The fibre tensile stress increases to the point where the fracture strength, $\sigma_f$, is exceeded and the fibre breaks inside the matrix. This fragmentation process is repeated as the sample strain is increased producing shorter and shorter fibre fragments within the coupon until the remaining fragment lengths are no longer sufficient in size to produce further fracture through this stress-transfer mechanism. The final fibre fragment length-to-diameter ratio, $l/d$, is measured. A shear-lag analysis is conducted on the fragments in order to calculate an interracial shear strength, $\tau$.

$$ \tau = \left( \frac{\sigma_f d}{2} \right) \left( \frac{d}{l_c} \right) $$

In practice, there is a distribution of critical lengths and Weibull statistics are used to fit the data as shown.

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**Advantages**

- Any reinforcing fibre may be used
- Any matrix may be used
- Direct measurement of load

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**Disadvantages**

- Embedment length is important
- External meniscus is important
- Clamping of the disc is important
- Must be able to grip the fibre mechanically
- State of stress at the exit creates a tensile interracial force
- No information about failure mode
- Interface may not be the same as composite interface
- 1–3 data points per specimen

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**Figure 1** Schematic diagram of the single-fibre pull-out method for measuring fibre-matrix adhesion.