Review of the test methods of the interface characterization of natural fiber-reinforced polymer composites

Z H Zhu1,3, Y Zhang1, N Zhang1, M Y Hao2 and H W Wu2
1School of Advanced Manufacturing Technology, Guangdong Mechanical & Electrical Polytechnic, Guangzhou, China
2The Key Laboratory of Polymer Processing Engineering, Ministry of Education, South China University of Technology, Guangzhou, China

Email: 184823174@qq.com

Abstract: How to measure the interfacial strength of matrix and reinforcement is one of the key problems in the study of natural fiber reinforced polymer composites. The measurement technologies to study interface strength include destructive methods and non-destructive methods, which principles are introduced in this paper. Different tests can provide theoretical complementarity. Reasonable combination and improvement of interface strength tests can provide strong practical support for the study of interface property.

1. Introduction

Interface is the common boundary of fiber and matrix which plays an important role in fiber reinforced composite. Composite combines matrix with reinforcement by interfacial interactions including chemical bonding, molecular diffusion, mechanical interlock and so on. Therefore, the interface has important influence on the strength, toughness and weather resistance of composite [1].

The existence of interfaces and their important role in the properties of composites have been confirmed by a large number of studies on synthetic fiber reinforced polymer composites. [2-3]. The surface morphology and chemical structure of natural fibers have great heterogeneity and variability and thus the interface between natural fibers and polymer matrix is very complex. [4] Nair et al obtained the high resolution image of elastic modulus distribution in the interface of loycell fiber/PP composite and identified a small transition region that is different from the enhanced phase and the matrix phase for the first time [5]. They also found that there is a significant positive correlation between the thickness of transition region and the macroscopic tensile mechanical properties of composites. The above researches show that although the interface phase scale between plant fiber and thermoplastic polymer may be very small, it has a direct impact on the macroscopic mechanical properties of composite. Therefore, it is necessary to know the methods of measuring and characterizing the interfacial properties of composites to study the relationship between interfacial strength and the properties of composites. The mechanical properties of composites, such as tensile strength, flexural strength and tensile strength, can be determined directly or indirectly to reflect the adhesion of fiber and matrix [6-8]. In addition, there are more direct-measurement technologies to study interface strength which can be divided into two categories according to its principles and the way materials are destroyed: destructive methods and non-destructive methods. Destructive method generally adopts the mechanical test method, which includes single fiber fragmentation test, single-fiber pull-out test, micro-debonding test, squash test, and so on. Non-destructive methods can be classified as chemical, physical, or energy-based, and mainly
include the scanning electron microscopy (SEM), X-ray photoelectron spectroscopy (XPS), atomic force microscope (AFM), and other techniques.

2. Test methods

2.1. Destructive methods

2.1.1. Single fiber fragmentation test

The single fiber fragmentation test is the most widely used method to characterize the interfacial shear strength between fiber and polymer matrix in recent years [9-10].

As shown in figure 1, a fiber is embedded in a dumbbell shaped mold and a casting resin is solidified to obtain the tensile specimen. When the dumbbell specimen is stretched along the axial direction, the fiber in the resin will break because the elongation of the resin is larger than that of the fiber. With the load increasing, the fiber will not break again when reaching the critical length and the shear stress at the interface is not enough to cause the fiber to break. Then, the acoustic emission or the photoelastic technology was used to measure the number of broken fibers and the average fibers break length is within a certain length in the resin. Assuming that the interface shear stress $\tau$ along the fiber length direction is constant, while $\tau$ can be calculated according to the Kelly-Tyson equation [11].

$$\tau = \frac{d_f \sigma_{f(c)}}{2l_c},$$  \hspace{1cm} (1)

where $d_f$ is the diameter of the fiber, $l_c$ is the critical length of fiber, generally, equal to 4.41 mm, $\sigma_{f(c)}$ is the critical fiber strength under $l_c$, which is usually estimated by the Weibull statistical method [12].

![Figure 1. Diagram of fragment length of single fiber.](image1)

2.1.2. Droplet test

Micro-debonding test was firstly proposed by Miller et al. in 1987, which is a vertical embedding of fibers into a very small symmetrical resin drop, as shown in figure 2 [13]. The experiment is similar to pulling out test which is easy to measure the pullout force and bond length or bond area to estimate the interface strength. The experimental model of micro-debonding test generally assumes that the shear stress at the interface is uniformly distributed along the fiber length and thus only the average shear strength at the interface can be calculated. The micro-debonding test can accurately measure the magnitude of the instantaneous debonding force and is suitable for any fiber/matrix study [14-15].

![Figure 2. Schematic diagram of droplet test.](image2)
2.1.3. Single fiber pull-out test

The single fiber was embedded vertically into the resin matrix and cured, as shown in figure 3. Clamp the resin matrix and continuously apply a very small force to stretch the fiber to pull the fiber out of the resin matrix. Record the fiber debonding moments of force $F_d$, measure the length of the embedded fiber $l$ and fiber diameter $2r$, to calculate the interface strength.

$$
\tau = \frac{F_d}{2\pi rl} \quad (2)
$$

Li studied the Interfacial stress of sisal fiber reinforced high density polyethylene (HDPE) composites using single fiber pull out test [16]. Brahmakumar prepared coconut fiber-reinforced polyethylene composites and studied the effect of natural waxy surface layer of the fiber on fiber/matrix interfacial bonding and strength of composites with a single pull-out test. [17] Although this method is relatively simple, it should be noted that when the fiber-matrix interface bond is strong and the fiber diameter is small, the length of the embedded fiber must be very short. Otherwise, the fiber has broken without pulling out in the process of stretching.

![Figure 3. Diagram of single fiber pull-out test](image)

2.1.4. Extrusion test

Mandell et al. [18] first reported the principle of this method in 1980, as shown in figure 4. The section of the sample should be normal to the fiber axis. Then, the fiber was extruded from the resign matrix with a sharp diamond cone. The debonding force was recorded and the interface shear stress was calculated. An advantage of squash method is that a piece of composites can be directly cut off to conduct experiments and obtain a value close to the actual value and, thus, the entire operation is simple.

![Figure 4. Schematic diagram of the extrusion method.](image)

2.2. Non-destructive methods

2.2.1. scanning electron microscope (SEM). SEM is a method to observe micro-morphology and the basic action of which is that a highly focused electron beam is scanned over the surface of an object and the interaction of the beam with the material excites simultaneously a number of physical processes. The advantages of SEM are as follows. The first is the high magnification up to 200,000-200,000 times. The second is a large depth of field, a large field of vision and a three-dimensional image, which can
directly observe the fine structure of the uneven surface of various samples and thus highlighting how the filler is dispersed or not in the matrix [19]. Therefore, SEM is a very useful scientific research instrument nowadays [20-21].

2.2.2. Atomic force microscope (AFM). A microcantilever is used to sense and amplify the force between the tip and the atom of the sample to get the detection result in atomic force microscope (AFM) test. [22-23] AFM has many advantages over SEM. Unlike electron microscopes, which provide only two-dimensional images, AFM provides true three-dimensional surface maps. Meanwhile, AFM does not require any special treatment of the sample, such as copper plating or carbon plating, which may cause irreversible damage to the sample.

2.2.3. Fourier transform infrared spectrometer (FTIR). Many researchers applied FTIR to analysis the composition of the interface of natural fiber reinforced PLA composites. [24] The light from the source is divided into two beams by a beam-splitter. One beam reaches the moving mirror through transmission, and the other beam reaches the stationary mirror through reflection. The two beams of light respectively go through the stationary mirror and the moving mirror and then return to the beam splitter. The moving mirror moves in a straight line at a constant speed. After the beam splitter rendezvous, the interference light passes through the sample pool. Then, the interference light containing sample information reaches the detector after passing through the sample. Finally, the signal is processed by Fourier transform and finally the FTIR map are obtained.

2.2.4. X-ray photoelectron spectroscopy (XPS). In 1887, Hertz discovered the photoelectric effect and Innes recorded the first human X-ray photoelectron spectroscopy in 1907. The principle of XPS is to use x-rays to radiate a sample and thus the inner electrons or valence electrons of an atom or molecule are excited to shoot out and hence called photoelectrons. Taking the x coordinate of kinetic energy/binding energy of the photoelectron and the relative strength as the y coordinate, photoelectron spectroscopy can be made to obtain the sample information. For example, Hong prepared four kinds of BF and then blended them with polylactic acid (PLA) to prepare PLA/BF composites. FTIR, SEM and XPS were used to characterize and analyze the structure of modified BF and its reinforced PLA-based composites [25].

3. Conclusion
Destructive testing can not only obtain the interfacial bonding strength of composite, but also study the interfacial bonding status and the debonding process of fiber from matrix. The disadvantages of these tests are highly targeted and the practical engineering conditions are complex. Non-destructive tests depend on the test equipment and its accuracy. The real interface morphologies of composites can be described by different devices at different scales, which is helpful to explain the composite interfacial action from the perspective of microphysics. Different tests provide theoretical complementarity. Reasonable combination and improvement of interface strength tests can provide a strong practical support for the study of interface properties.

Acknowledgement
This research was supported by Key Research Platforms and Projects of Universities of Guangdong Province (Grant No. 2018GKTSCX111) and Funding Project for Scientific Research of High-level Talents of Guangdong Mechanical & Electrical Polytechnic (Grant No. Gccrcxm-201903).

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