Enhanced flexural properties of aramid fiber/epoxy composites by graphene oxide

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Abstract: The reinforcing effect of graphene oxide (GO) in enhancing the flexural strength and flexural modulus of aramid fiber (AF)/epoxy composites were investigated with GO-AFs at a weight fraction of 0.1-0.7%. The flexural strength and flexural modulus of the composite reached 87.16 MPa and 1054.7 MPa, respectively, which were about 21.19% and 40.86% higher than those of the pure epoxy resin, respectively. In addition, the flexural properties and interfacial shear strength (IFSS) of composite reinforced by GO-AFs were much higher than the composites reinforced by AFs due to GO improved the interfacial bonding between the reinforcement material and matrix.

Keywords: graphene oxide; aramid fiber; flexural properties; interfacial bonding

1 Introduction

Among the composites, the flexural properties are affected by the intrinsic flexural properties of the matrix [1]. Epoxy resin as one of the most common polymer matrices, it is utilized extensively in polymer based composites for aerospace applications [2]. However, pure epoxy resins exhibit poor flexural properties, which limits its applications. Fiber-reinforced polymer composites are an emerging class of engineering materials [3]. Previous research reported that the flexural properties of epoxy resin could be improved effectively by carbon fiber and glass fiber etc. [4, 5]. Aramid fiber (AF) is used in aeronautical, automotive applications and construction etc. due to its high modulus and strength, low density, and adaptability [6–8]. However, AF usually possess smooth and chemically inert surface, which results in poor interfacial adhesion between fiber and epoxy matrix [9, 10]. The interfacial shear strength (IFSS) can indicate the degree of interfacial adhesion between fiber and epoxy matrix. Some fiber surface treatment methods have been reported, including acid treatments [11, 12] and chemically grafted [13, 14] which could improve the IFSS of fiber/epoxy composite effectively. Numerous works for fiber’s surface modification, among which the chemical coupling method is one of the important chemical modification methods. The coupling agent is amphiphilic and acts as a chemical bond bridge between fiber and matrix [15]. Xie et al. [16] reported that silane coupling agents with varied silane structures had different effects on the mechanical performance of natural fiber reinforced polymer composites. Shokoohi et al. [17] used silane coupling agents modified glass fiber to improve the interfacial bonding between the fiber and the epoxy matrix. However, those reported methods could only suit for one kind of resin matrix. This is due to the different reagents requirement with different resin matrix, especially for polarity matching [18].

Graphene is the hexagonal honeycomb-like structure with robust honeycomb lattice that it can yield a stable structure [19]. Graphene poses many remarkable properties, including high Young’s modulus (1100 GPa), superior fracture strength (125 GPa), and large surface area (2630 m²/g) [20, 21]. Due to these excellent properties, graphene and its derivatives, in particular graphene oxide (GO) have found interesting applications in nanocomposites [22, 23]. Interestingly, GO has been demonstrated as an ideal reinforcement of fiber-matrix [24]. Research work on polymer nanocomposites was reviewed by Fu et al. [25]. It showed that incorporation of nanoscale fillers, like graphene, with low content into polymers can obtain good enhancements in mechanical and physical properties. Liu et al. [26] directly added GO into epoxy resin, the results showed that
the mechanical properties of GO/epoxy composite were improved effectively. Among them, the maximum flexural modulus and flexural strength reached 2.94 GPa and 130.46 MPa, respectively, which are about 10.11% and 14.67% higher than those of the pure epoxy resin. Pathak et al. [27] found that GO utilization is an effectively approach for improving the flexural properties of carbon fiber reinforced polymer composites. The enhancement can be attributed to the existent of GO, providing fiber with increased roughness and polarity, resulting in strong mechanical interlocking between the fiber and matrix [28]. However, to the best of our knowledge, the enhancement effect of GO modified AF on the flexural properties of epoxy has not been reported yet.

In this work, GO modified the AF surface firstly, then enhanced the epoxy matrix with GO-AFs. Based on the mechanical interlock mechanism [29] mentioned above to systematically examine the role of GO in improving the flexural properties and IFSS of the AF/epoxy composite. Meanwhile, the flexural properties of the composites were also investigated with pure AF for the purpose of comparison.

2 Experimental

2.1 Materials

Graphite powders were purchased from Qingdao Jinrilai Shimo Co., Ltd. China. Epoxy resin (WSR615) based on bisphenol-A with an epoxy value of 0.51-0.53 was obtained from Haining Hailong Chemical Co., Ltd. China. The curing agent (Hexahydro-4-methylene phthalic anhydride) was purchased from Zhejiang Alpharm Chemical Technology Co., Ltd. China. Chopped aramid fibers (Kevlar 1414, the diameter of 12 µm) were supplied by Haining Anjie Composite Materials Co., Ltd. China. Acetone, sodium nitrate (NaNO3) and potassium permanganate (KMnSO4) were purchased from Hangzhou Xiaoshan Chemical Reagent Factory and used as received. Concentrated sulfuric acid and hydrochloric acid was obtained from Hangzhou Shuanglin Chemical Reagent Factory.

2.2 Preparation of GO and GO-AFs

Graphite oxide was prepared by oxidizing graphite powders in an oxidation system including NaNO3, concentrated sulfuric acid and KMnO4 in an improved Hummer’s method [30]. And the reaction went through three stages (low temperature intercalation, medium temperature oxidation and high temperature stripping). The obtained graphite oxide was then dispersed in distilled water to form a suspension, then the suspension was treated by ultrasonic technique (100W) for 0.5h to exfoliate the graphite oxide to GO.

GO suspension was used to modify the chopped fibers. For the absorption of GO on the fiber surface, pre-dried chopped AFs were soaked fully with 150 ml GO (0.2 mg/ml) suspension at room temperature and then placed in an ultrasound machine for 1.5h to prepare GO-AFs reinforcement. In the case, chopped AFs were firstly immersed in acetone via ultrasonic treatment for 0.5h to remove the sizing agents and impurities attached on the fiber surface. After the treatments, all the obtained AFs were continuous washed and filtered with enough distilled water for at least 3 times to ensure the complete remove of the remaining solvent, following by drying in a oven at 100±2°C for 6h.

2.3 Preparation of GO-AFs/epoxy composites

GO-AF composites according to the content of 0.1 wt%, 0.3 wt%, 0.5 wt% and 0.7 wt% with 100 g epoxy resin, respectively. All the composites were prepared by an internal mixer (XSS-300, China) at room temperature and a rotor speed of 60 rpm for 716 s. For comparison, pure chopped AFs and epoxy resin were also mixed in the same ratios and experimental conditions. The standard dumbbell-shaped samples used for flexural testing were prepared by cast and curing molding at room temperature. There were post-cure and polishing operations for the sample lastly. A hybrid technology simulation diagram was shown in Figure 1.
2.4 Characterization

The phase purity of the graphite, graphite oxide and GO were characterized by X-ray diffraction (XRD) on a X-ray diffractometer with Cu Ka radiation (λ = 1.5418 Å). And the surface functional groups of GO, AF and GO-AFs were characterized by FTIR. Water contact angle of the fiber surface was obtained by contact angle measuring instrument. When a drop of water (2 µL) firstly encountered the fiber, the photograph was taken and measured the angle as the water contact angle of the fiber surface.

IFSS of fiber/epoxy composite was characterized by single fiber pull-out test (Figure 2). The specimens were tested at a crosshead speed of 10 mm/min and retained 10–15 valid data per set of tests. The flexural specimens were prepared according to the recommendation of ASTM D790-02 [31]. The flexural properties were measured by three-point bending test [32] with a 50 KN load cell with a crosshead speed of 2.0 mm/min, span is 40 mm. Five specimens prepared for each composite were tested for flexural properties. The fracture surfaces of the specimens after testing and pure AF, GO and GO-AFs were also examined by SEM (S-4800, Japan). Before the examination, the fracture surfaces were cleaned using alcohol and then coated with a thin evaporated gold layer to improve conductivity.

3 Results and discussion

3.1 GO analyses

The XRD patterns of the graphite, graphite oxide and GO are shown in Figure 3. The characteristic diffraction peak of the (002) graphite plane is obtained at 2θ = 26.5°, while a relatively low peaks are obtained for graphite oxide and GO at about 2θ = 11.4°. According to the Bragg’s law (nλ = 2d sinθ), the calculated value of interlayer spacing of natural graphite is 3.37 Å, it is about half spacing of GO (d = 7.79 Å). The larger spacing of GO suggested that graphite is mostly oxidized, and various oxygen based functional groups are formed onto the surface [33]. The peak intensity of GO is lower than graphite oxide which demonstrating the disorder structure of GO [34].

Functional groups present on the surface of GO were evaluated by FTIR and it is showed in Figure 4. It can be seen that GO is rich in oxygen-based functional groups. For example, the broad bands observed at 3404.35 cm⁻¹ correspond to the O-H stretching [35]. The peaks at 1612.49 cm⁻¹, 1396.46 cm⁻¹ and 1055.98 cm⁻¹ corresponds to the stretching vibration of C=C, C-OH and C-O respectively [27]. At 1217.08 cm⁻¹, 985.62 cm⁻¹ and 844.42 cm⁻¹ are assigned to the stretching vibration of C-O-C [36].
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### 3.2 GO-AFs reinforcement analyses

Figure 5 shows the photos of pure and modified AFs. The untreated pure chopped AFs are yellow in color while GO-AFs exhibit a dark gray color, implies that the surface should be covered by GO. It could also be confirmed from the FTIR spectra of AF and GO-AFs (Figure 6). The FTIR spectroscopy of pure AFs shows the stretching vibration peaks at 3431 cm\(^{-1}\) (O-H and N-H), 1645 cm\(^{-1}\) (C=O), 1508 cm\(^{-1}\) (C=C), 1315 cm\(^{-1}\) (Ph-N) and the bending vibration at 1541 cm\(^{-1}\) (N-N), respectively [37]. For GO-AFs, in addition to these peaks shown above for pure AFs, a new characteristic peak at 1385 cm\(^{-1}\) is seen in the spectrum of GO-AFs, which is attributed to the epoxy group in GO [38], and confirms the modification of GO on AFs. Figure 7 shows the microscope images of AF before and after GO modification. It is obvious that the surface roughness of GO-AFs is larger than that of AF. The surface morphology of AF before and after GO modification can be also observed by SEM as shown in Figure 8. Figure 8A displaces a rather smooth surface of pure AFs. By contrast, the surface of GO-AFs becomes rougher as shown in Figure 8B. The rougher surface of GO-AFs should be attributed to the modification of GO. The GO surface (Figure 8C) has a distinct wrinkle morphology which greatly increases the roughness of the fiber surface, causing it to mechanical meshing with the substrate. The modified AFs are used as a reinforcing material for epoxy resin, which could effectively improve the interfacial bonding strength with the epoxy matrix for GO’s wrinkled surface and functional groups.

Furthermore, we have also conducted ultrasonic washing experiments for GO-AFs. Figure 9 presents the SEM micrographs of GO-AFs ultrasound in distilled water for 10 min (Figure 9A) and 30 min (Figure 9B), respectively. It implies that the stability of GO on AF surface is good.

### 3.3 Flexural properties of GO-AFs/epoxy composites

The flexural properties of the samples were calculated according to the following Eq. (1) \[39\]:

\[
\sigma_f = \frac{3PL}{2bd^2}
\]  

Where \(\sigma_f\) is the flexural strength, \(P\) is the maximum load observed during the test, \(L\) is the support span, \(b\) is the measured specimen width and \(d\) is the measured specimen thickness. The flexural strength and flexural modulus of the specimens are displayed in Figures 10 and 11. The flexural strength of the GO-AFs/epoxy composite reaches the maximum value at the GO-AF content of 0.1 wt%, about 21.19% higher than that of the pure epoxy resin. However, the flexural strength decreases with further increasing the GO-AF content. The flexural strength of the counterpart (AFs/epoxy composites) increases with increasing the AF content (from 0.1 wt% to 0.7 wt%). Furthermore, the flexural strength of AFs/epoxy composites with 0.7 wt% AF is about 13.54% higher than that of pure epoxy. The flexural modulus of epoxy-based composites with different reinforcement materials (pure AF and GO-AFs) from 0.1 wt% to 0.7 wt% are both increasing, reach the maximum at the same content of 0.7 wt%. The flexural modulus of the AFs/epoxy and GO-AFs/epoxy composites reach 817.51 MPa and 1054.7 MPa, respectively. Which are about 9.05% and 40.86% higher than those of the pure epoxy resin, respectively. This is mainly due to the flexural modulus of AF and GO are much higher than that of epoxy resin [21, 40].

It could be further seen from Figure 10 that the flexural strength of GO-AFs/epoxy composites are consistently higher than the AFs/epoxy composites. This is mainly due
Figure 7: Microscope images of (A) AFs and (B) GO-AFs

Figure 8: SEM images of (A) pure chopped AFs; (B) GO-AFs; (C) GO

Figure 9: SEM images of GO-AFs ultrasounded with different time: (A) 10 min. (C) 30 min
to the fact that the flexural strength of the composite material strongly depends on the interfacial adhesion between fiber and matrix [41]. It is expected that the introduction of hydrophilic GO can not only increase the surface polarity of the fiber, but also increase the specific surface area, thus can provide a desirable interfacial adhesion with resin [28, 42]. The surface polarity of fibers is characterized by its water contact angle. As can be seen in Figure 12, the untreated AF has a water contact angle of 56.3°. While the water contact angle of GO-AF decreases to 46.3°, which illustrating the addition of GO indeed improves its surface polarity.

Herein, as showed in Figures 13A and 13B, the bonding between fiber and epoxy resin is weak, this is due to the fact that the chopped AF surface pulled-out on the fracture surface is free of epoxy infiltration. Conversely, in Figure 13C, epoxy matrix is firmly adhered on GO-AF, which demonstrates that there may be some chemical and physical chelation effect between GO and matrix [43, 44]. This good interfacial adhesion allows the material to transfer stress from the epoxy matrix to the fibers when subjected to external forces [45]. The mechanism for the interfacial properties can be further explained as follows: on the one hand, the oxygen-based functional groups of GO interact with groups on the epoxy resin to enhance the interfacial adhesion of the fibers to the resin; furthermore, the wrinkled morphology of GO can form mechanical interlock with epoxy resin [44].

On the other hand, the flexural strength of GO-AFs/epoxy composites decreases with further increasing the GO-AF content (wt% > 0.1 wt%). Compared with AFs/epoxy composite at the AF content of 0.7 wt% (shown in Figure 13B), Figure 13D exhibits that the GO-AF aggregation appears at the content of 0.7 wt%. This can be attributed to the increase of AF’s polarity by GO which increases the probability of GO-AF aggregation. And it results in stress concentration and thus the flexural properties of GO-AFs/epoxy composites declined.

### 3.4 IFSS of GO-AFs/epoxy composites

The IFSS was calculated according to the following Eq. (2) [46]:

\[
\tau = \text{IFSS} = \frac{F}{\pi dl}
\]

Where \( F \) is the maximum pull-out force, \( d \) is the diameter of the monofilament and \( l \) is the embedded length of the AF in the epoxy. Table 1 displays the IFSS of the AFs/epoxy composite and GO-AFs/epoxy composite by single fiber pull-out test. The IFSS of AFs/epoxy composite is 6.20 MPa, while the IFSS of GO-AFs/epoxy composite is 8.29 MPa, which is about 36.8% higher than that of AFs/epoxy composite. This is consistent with previous flexural strength.
Figure 13: SEM images of fracture surface of fibers/epoxy composites with different mass fractions of AF and GO-AFs; (A) 0.1 wt% AF; (B) 0.7 wt% AF; (C) 0.1 wt% GO-AFs; (D) 0.7 wt% GO-AFs

Table 1: IFSS of AF/epoxy composite with AF modified before and after

| Sample     | IFSS (MPa) | Increase of IFSS compared with AF |
|------------|------------|----------------------------------|
| AF/EP      | 6.20       | 0                                |
| GO-AF/EP   | 8.29       | 36.8%                            |

results, which highlights the significant role of GO in enhancing interfacial bonding between AF and epoxy matrix.

4 Conclusions

In summary, although pure aramid fibers can be used to reinforce epoxy resin, significantly improved flexural strength, flexural modulus and IFSS can be obtained by introducing GO to modify aramid fibers.

1. The flexural strength and flexural modulus of the GO-AFs/epoxy composite containing 0.1 wt% GO-AFs are 87.16 MPa and 1054.7 MPa, respectively, which are about 21.19% and 40.86% higher than those of the pure epoxy resin.
2. The flexural strength and flexural modulus of the AFs/epoxy composite containing 0.7 wt% AFs are 81.59 MPa and 816.51 MPa, respectively, which are about 13.54% and 9.05% higher than those of the pure epoxy resin.
3. The IFSS value of GO-AFs/epoxy composite reaches 8.29 MPa, which is higher than that untreated AF by 36.8%.
4. GO is an excellent potential modifier to modify AFs.

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