



# Improvement of the bonding between carbon fibers and an epoxy matrix using a simple sizing process with a novolac resin



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## HIGHLIGHTS

- The samples with three different ways of NR addition were fabricated.
- Both interfacial and impact properties were enhanced using NR on fibers.
- Change in fracture mechanism from adhesive failure to cohesive failure was found.

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## ABSTRACT

Novolac resin (NR) sizing modification of the carbon fiber/epoxy composites bonding was investigated by sizing NR on the carbon fibers surface using a conventional sizing process, i.e., immersing fibers tows in a NR solution. Thus, three different types of carbon fibers/epoxy composites were prepared, which NR is introduced in three ways: (1) pure matrix for comparison, (2) adding NR in the matrix, (3) adding NR in the sizing. Interlaminar shear strength (*ILSS*) using short beam shear test and impact strength were investigated. The results showed that both interfacial and impact properties of composites were maximized using the NR sizing on fibers. *ILSS* and impact strength were improved by 22.5% and 56.8%, respectively, compared with the composite without NR. This way introduces no damage to the fibers and has larger advantage than the matrix modification by dispersing NR in epoxy resins. The fracture and surface morphologies of the composite specimens were characterized by scanning electron microscopy (SEM). SEM clearly highlighted a change in the fracture mechanism from a clearly adhesive failure to a cohesive failure.

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## 1. Introduction

The interest in using carbon fiber reinforced polymer composites (CFRPs) are growing due to their outstanding mechanical properties and low density compared with conventional materials. The main usages are aerospace engineering, sporting goods, automotive, shipbuilding, engine, marine and transportation industries [1]. It is now well known that the performance of a fiber reinforced polymer matrix system is controlled by the properties of fiber-matrix interface to a large extent. The properties of interfacial bonding between the fiber and the matrix have an essential effect on the ultimate performance of the composite [2–4]. For example,

a good interfacial bonding ensures efficient load transfer from the matrix to the fiber, which helps reduce stress concentrations and improve overall mechanical properties. Because the properties of an interphase region in the matrix close to the fiber surface are different from the properties of the matrix, resulting in enhancing the structural integrity of the composite and leading to better stress transfer between the matrix and the fibers [5,6].

Resin and fibers modifications are main methods to improve the interfacial bonding. An excellent interfacial bonding can be obtained by adding a new component into matrix [7–9], but it makes curing and manufacturing process complicated. Numerous methods have been studied for fiber surface treatment, such as electrochemical oxidation [10], oxidation in strong acids [11], plasma oxidation [12,13], and ozone oxidation [14]. These treatments on fibers improved the interfacial bonding of the final composites to some extent, but they had drawbacks of high energy

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consumption. Therefore, commercial carbon fibers are often sized, i.e. their surface is covered with a layer of resin [15] or nanoparticles [3]. This method is promising because it can protect the fibers from damage during handling and manufacturing composites. One hand, the sizing generates an interphase, and it can partially dissolve in the matrix. On the other hand, some constituents of the matrix can also diffuse into it. Thus, a concentration gradient of those constituents and consequently a gradient in mechanical properties from matrix to fiber are generated. Moreover, it is known that high matrix cure volume shrinkage (5% or more) on fibers surface has a negative influence on interfacial adhesion [6]. This effect needs to be weakened as well and sizing on fibers can dissolve this problem. The composition and structure of the sizing has a great influence on the level of interfacial adhesion, however, the point has received only limited attention [16]. Therefore, a fundamental understanding and quantitative characterization of the properties of the fiber-matrix interface should be performed widely and it can help design fiber-reinforced polymer composites with improvement of the mechanical properties.

In the past years, the modification of epoxy resin by adding thermoplastics in epoxy matrix has been investigated widely, because there is no reduction in thermal and mechanical properties of the matrix [17,18]. Nevertheless, the adding of thermoplastics in epoxy matrix may result in increasing viscosity and making the manufacture of the composites more difficult. Furthermore, over an upper limit of thermoplastics content often leads to non-uniform distribution throughout the composites [19,20]. Thermoplastics in the fiber sizing eliminate these issues by adding the thermoplastics in the interphase, and the mechanical properties of the composites can be increased by improving the properties of the interphase region. In this paper, a simple coating process was developed, i.e., carbon fibers were immersed into a novolac resin (NR) sizing to coat their surface with a NR layer. The influence of NR sizing on carbon fibers on the interfacial properties was tested by measuring *ILSS*, which was carried out with short beam shear test. A significant improvement of the bonding strength was obtained with optimal conditions. In addition, based on the mechanical properties (interfacial properties and impact properties) and SEM analysis, attempts have been conducted to investigate the modifying mechanisms.

## 2. Experimental

### 2.1. Materials

Epoxy resin TGDDM (tetraglycidyl-4,4'-diaminodiphenylmethane) with an epoxy equivalent weight of 422 used in this study was purchased from Shanghai Research Institute of Synthetic Resins (China). 4,4'-Diaminodiphenylsulfone (4,4'-DDS) as curing agent was obtained from Sinopharm Chemical Reagent (Shanghai, China). A mixing ratio of 3:1 by weight was selected. The thermoplastic NR was provided from Tianjin Chemical factory (China). The chemical structures of these reagents are listed in Fig. 1. Acetone as diluter was purchased from Tianjin Chemical Reagent Co., Ltd., China. The reinforcement materials were continuous polyacrylonitrile-based carbon fibers (3 K) manufactured by Institute of Coal Chemistry, Chinese Academy of Sciences. Their tensile modulus is 211 GPa, tensile strength is 4.05 GPa, elongation at break is 2.07%, and diameter is 7.1  $\mu$ m.

### 2.2. Addition of NR in the matrix and NR sizing on the fibers and production of unidirectional carbon fiber/epoxy composites

- (1) Weighed NR and TGDDM were mixed until the NR particles were uniformly dispersed in the epoxy resin. Subsequently, the mixture was diluted by adding acetone. Then, 4,4'-DDS was added to the solution while stirring. The obtained solution was coated on dry fibers by means of solution dip coating. The prepregs were placed in a vacuum oven to remove the acetone. Prepreg layers were then stacked up and cured at 90 °C for 0.5 h, 150 °C for 1 h, and then postcured at 200 °C for 2 h. During the curing process, the pressure was 0.6 MPa.
- (2) The NR/acetone solution (NR concentration: 2 wt%, 5 wt%, 8 wt% and 11 wt%) was prepared for the sizing treatment on the fibers. The dry fibers were then sized with the NR solution for 5 min. Excess sizing was removed by a

scraper after passing through the sizing bath, then the fibers were dried in a vacuum oven at 70 °C for 30 min. Last, the NR sized fibers was coated by epoxy solution. The prepreg layers were then stacked up and cured as above process.

### 2.3. Characterization

Interlaminar shear strength (*ILSS*) of carbon fibers/epoxy composites was measured on an universal testing machine (AG-X10KNSTD, Shimadzu, Japan), using a three-point short beam bending test method described in ASTM standard D2344 at room temperature. *ILSS* values were calculated according to the following formula:

$$ILSS = \frac{3P_b}{4bh} \quad (1)$$

where  $P_b$  is the maximum compression load at fracture in N,  $b$  is width of specimen in mm, and  $h$  is thickness of specimen in mm. Each reported *ILSS* value was the average of the results from measurement of six specimens. The relative error was estimated to be within 10% based on reproducibility of the data among different specimens.

The fibers multifilament tensile strength test was also carried out by using an universal testing machine (AG-X10KNSTD, Shimadzu, Japan) at a crosshead speed of 2 mm/min, according to GB/T 3362-2005.

A TGA-92 instrument (Separam, France) was used to characterize the NR content coated on the carbon fibers. 5–10 mg specimens from the fibers randomly were proper for TG test. The specimens for TGA were ground into powder, and these specimens were placed in a platinum crucible and heated. A constant nitrogen flow of 45 mL/min was used in order to avoid oxidation of carbon fibers. The temperature from room temperature to 600 °C at a heating rate of 10 °C/min was applied and then kept for 120 min at 600 °C. This heating program is designed to achieve a constant of residue while the temperature is not high enough to cause fibers to damage. The mass content of NR *NR%* was obtained from the TG curves, which were calculated according to the following formula:

$$NR\% = \frac{RRF - RRNF}{100 - RRNF} \quad (2)$$

where *RRF* is the residue rate of the neat fibers, *RRN* is the residue rate of the neat NR, and *RRNF* is the residue rate of NR coated fibers. Three samples were tested based on reproducibility of the data among different specimens.

Nonstandard impact specimens of 60 mm in length, 6 mm in width, and 2 mm in thickness were performed by using the homemade Charpy impact testing equipment [21]. All the test specimens were unnotched.

The fracture surfaces of *ILSS* samples were examined by scanning electron microscopy (SEM, JSM-6360LV) with an excitation voltage of 30 kV. The fracture surfaces were cleaned with alcohol in order to eliminate impurities like dusts before test, and were coated with a thin evaporated layer of gold to improve conductivity.

## 3. Results and discussion

### 3.1. Influence of the NR sizing on the fibers strength and its composites

Multifilament tensile strength of carbon fibers was carried out for studying the effect of NR sizing solution concentration on the fibers strength. The relationship between multifilament tensile strength and NR sizing solution concentration was shown in Fig. 2. As a result, the tensile strength of carbon fibers almost increases constantly until NR sizing solution concentration is up to 5 wt%. However, it remains almost a constant (almost 3.58 GPa) as the concentration is over 5 wt%. It indicates that carbon fibers can be protected effectively when NR solution concentration is up to 5 wt%.

A TGA analysis of the pristine fibers, NR powders and NR coated on fibers are shown in Fig. 3. The residue rate of the neat fibers (*RRF*) is 98.2 wt%, and only 1.8 wt% weight loss of fibers can be occurred. In this temperature range (room temperature to 600 °C), the weight loss is attributed to the possible moisture and carbon fibers show little decomposition. The residue rate of the neat NR (*RRN*) is about 57.9 wt% from NR isothermal curve. In addition, the residue rates of 2 wt%, 5 wt%, 8 wt% and 11 wt% NR on fibers (*RRNF*) are 97.52 wt%, 96.39 wt%, 94.75 wt% and 91.63 wt%, respectively. Thus, the weight content of NR on fibers surface treated with 2 wt% NR, 5 wt%, 8 wt% and 11 wt% NR concentrations is 1.6 wt%, 4.3 wt%, 8.2 wt% and 15.6% from calculated

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