Study on factors of interface properties of axial braided C/C composite materials

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In order to study the influencing factors of the tensile properties of axial braided C/C composites, the interfacial shear strength of the fiber rod and matrix was studied and ejecting tests of the fiber rod were carried out. The ejecting test specimens were formed with different thicknesses to obtain the changing rule of the interface shear strength with the thickness of the sample, and the testing method for the interface shear strength of the axial braided C/C composite material. The results show that the recommended thickness of the ejecting test specimens for the interface shear strength of is four times the diameter of the fiber rod. The interface shear strength distribution law of two different batches of materials was obtained through the interface ejecting test. The mesoscopic structure characteristics and pore statistical distribution law of the hole surface after ejecting were analyzed by scanning electron microscopy (SEM), and the mechanism of the difference of the interface shear strength was obtained. The tensile properties of two different batches of materials were obtained by tensile tests. The results show that the tensile properties of the two batches of materials differ greatly. The analysis suggests that the reason for this difference is the differences in interfacial bonding strength between the fiber rod and matrix. The higher the interface shear strength, the better the tensile property of the material will be.

Keywords: *interface properties, axial braided C/C, porosity scale, interface shear strength*

1. Introduction

Carbon-carbon (C/C) composite material, namely, carbon fiber reinforced carbon matrix composite material, is a kind of structural composite material developed in the late 1950s, in which both the reinforcing phase and the matrix phase are composed of pure carbon with special properties [1]. It has a series of excellent properties, such as high specific strength, good thermal stability, abrasion resistance and ablative resistance [2]. In particular, its mechanical properties do not drop but rise with increase in temperature, making it widely used in the field of aviation and aerospace [3]. In the field of aerospace solid rocket motor, throat insert is a key functional component to maintain the predetermined working pressure of the combustion chamber and make the combustion

gas change from subsonic to supersonic to generate thrust [4]. Its working environment is harsh and has an obvious influence on the performance of the solid rocket motor. Failure of the throat insert directly leads to the disintegration of the solid rocket motor [5]. In view of the special position and function of the throat insert, all the well-known manufacturers in the world adopt C/C composite material to prepare the throat insert structure [6], and have made remarkable achievements in the safety assessment of the throat insert structure [7, 8]. C/C composite materials have been applied to the braking system of aircraft, and have played an important role in the braking system of space shuttle, military, and civil aircraft too. Therefore, it is very important to study the mechanical properties of C/C composites and their influencing factors [9].

The interface is the weak link of the composite structure. In C/C composite materials, there are

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two crucial interfaces: one is the interface between a single fiber in the fiber rod (bundle) and the surrounding matrix (microscopic interface); and the other is the interface between the fiber rod (bundle) and the surrounding matrix (mesoscopic interface). The second type of interface will be the focus of this paper [1].

Various testing and analysis methods have been used to analyze, simulate, and calculate the interface properties of C/C composites. It is believed [10] that the toughness of fiber-reinforced ceramic matrix composite is not only controlled by the fiber and the matrix, but also affected by the interface between the two. The strongest interface does not mean the highest toughness, and the relatively weak interface can often improve the slip ability of the fiber when the fiber is broken. Chen Tengfei [11] studied the influence of the interface properties of C/C composite materials, and believed that the interface strength between the rough layer pyrolytic carbon and carbon fiber is higher than that between the smooth layer pyrolytic carbon, while resin carbon and bituminous carbon have higher interface strength due to the existence of chemical bond with carbon fiber. Hatta [12] obtained the interface shear properties of C/C composites under tensile, compression, and shear tests. Zhandarov [13] studied the interface strength testing method of fiber-reinforced composite materials by using the micromechanical testing method, formed the single fiber pull-out and micro-debonding technology, and formed an analytical model to evaluate the interface friction. Kong [14] tested, analyzed, and characterized the interface mechanical properties of fine knitting C/C composites. Meng [15] analyzed the influence mechanism of microstructure on the interface properties of C/C composite through theoretical model and interface ejecting experiment.

In general, the interface is an important structure of C/C composite materials, and there is an inevitable relationship between the interface state and bonding strength and the macroscopic mechanical properties of materials [15]. The reason for the difference in interface properties lies in microscopic defects such as interface cracks, holes, and interface delamination generated during the mold-

ing and processing of C/C materials [16]. After years of exploration by scholars, the experimental techniques used to characterize the fiber rod/matrix interface bonding strength are diversified, and can be divided into in-situ composite test, microscopic composite test, and macroscopic test [17]. However, due to the complexity of specimen preparation and experimental technology and the simplification of microscopic mechanical model, the interface properties measured by various methods differ greatly [18]. Generally speaking, it is a difficult problem to accurately test the interface properties of C/C composites. Therefore, it is a focus of this paper to select an effective and accurate testing method to get the interface properties of axial braided C/C composite materials [19].

In this paper, the characterization method of the mesoscopic interface properties of axial braided C/C composites has been obtained through experiments. On this basis, the microscopic differences between two batches of samples with different interface shear properties were studied, and the correlation between the interface microstructure defects and interface properties was established. Then, the tensile properties of two batches of materials were tested, and it was found that they were closely related to the interface properties. The research results indicate the direction for the optimization of material properties.

2. Materials and test methods

2.1. Axial braided C/C composite material

In this paper, axial braided C/C composite material is taken as the research object, and its braiding structure is shown in Figure 1.

In the multiaxial braided carbon fiber-reinforced carbon matrix composite material, the fiber rods by pultrusion molding form an axial reinforcement network and soft carbon fiber bundles are utilized to woven into a pre-woven body. The fiber rods are arranged in a regular triangle in the axial direction, while the fiber bundles pass through them successively from three channels of 0° , 60° and 120° so that increased interlayer is formed; this process is repeated until

Fig. 1. Schematic diagram of braiding form of axial braided C/C composite material. (A) Vertical view, (B) Side view, and (C) Material schematic diagram.

the desired dimension of pre-woven body is completed. Then the pre-woven body is machined by bituminizing, carbonization, densification process, and high-temperature process to manufacture as axial braided C/C composites. The smallest unit of the pre-woven body is symmetric along the axial direction, and the braided thickness is accumulated

along the axial direction, so it is called the axial braided C/C composite material [2].

2.2. Testing methods

Currently, the shear strength of the fiber rod/matrix interface is characterized by the fiber rod ejection method and the pull-out method. In the process of the pull-out method, because the stretching axis is prone to bigotry, it can easily lead to fracture of the fiber rod at the holding site. So the pull-out method is not suitable for testing the interface properties of the fiber rod/matrix. In this paper, the fiber rod ejection method was selected to characterize the interface shear strength of C/C composite material.

The basic principle of the ejection experiment of fiber rod is to apply load along the axial direction of the fiber rod through a rigid pressure head to make it disengage from the matrix as a whole. The load-displacement data are recorded in the ejection process, and then the interfacial shear strength is obtained by means of the corresponding mechanical analysis.

The ejection test system is shown in Figure 2, which mainly includes the micro-loading part and the microscopic observation part. The positioning accuracy of the X and Y displacement platform is $0.1 \mu\text{m}$, the display accuracy of load is 0.25 mN , and the display accuracy of displacement sensor is $0.2 \mu\text{m}$. The test loading speed is 0.2 mm/min , the magnification of the optical microscope is 600 times, and the magnification of the CCD camera can reach 2,000 times.

In the ejection test, it is assumed that the shear stress is uniformly distributed along the fiber rod/matrix interface, then the shear strength of the fiber rod/matrix interface can be calculated by Eq. (1) as follows:

$$\tau_s = \frac{F}{2\pi R_f H} \quad (1)$$

where F is the load value applied when interface debonding occurs, R_f is the radius of the fiber rod, and H is the thickness of the specimen.

From the point of view of the existing interface characterization technology, the interface charac-

Fig. 2. Interface test sample and ejection system. (A) fiber rod ejection system, (B) Sample size.

terization of composites with different braiding parameters and production process is slightly different, and the main difference lies in the determination of the interface test sample. By analyzing the braiding process of axial braided C/C material and combining with the accuracy and range requirements of the interface ejecting system, the designed experimental samples are as shown in Figure 2. H were the fiber rod diameter ($\Phi 1.1$ mm) 1.5 times (1.65 mm), 2 times (2.2 mm), 2.5 (2.75 mm), 3 times (3.3 mm), 3.5 (3.85 mm), 4 times (4.4 mm), 4.5 (4.95 mm), respectively.

In order to reduce the sample damage caused by machining, the sample was machined to the required basic size by wire cutting. The samples were finely ground to meet the requirements of surface smoothness. In particular, the surface of the fiber rod and sample must meet the perpendicularity, otherwise the test results will be a mixture of the shear strength of the interface and the compression performance of the fiber rod.

For the first batch of materials, the fiber rod ejection experiment was carried out according to the thickness of the sample and ejection experiment

method. The number of each thickness of ejecting fiber rod is 200, and the relationship between the obtained sample thickness and the average interface shear strength is as shown in Figure 3.

Fig. 3. The relationship between specimen thickness and interface shear strength.

With the increase in sample thickness, the interface shear strength can be divided into three stages. In the range of 1.65–3.85 mm, the shear strength of the interface increases with the thickness. Within the thickness range of 3.85–4.4 mm, the interface shear strength tends to be stable. When the thickness is greater than 4.4 mm, the interface shear strength tends to increase. The analysis shows that when the sample is thin (less than 3.85 mm), the inter-face damage caused by machining is large, and the measured interface strength is less than the real strength. When the sample is thicker (more than 4.4 mm), the measured strength not only includes the shear strength of the interface but also includes the transverse compression of the fiber rod, because the perpendicularity of the fiber rod axis and the surface of the sample is difficult to be guaranteed. Therefore, 4.4 mm thickness (4 times the diameter of the fiber rod) is appropriate for testing the interface shear strength of axial braided C/C composites.

3. Testing results and analysis

3.1. Results of interface property tests

In order to compare the interface shear strength of the two batches of materials, according to the interface test sample size determined in Section 2, the

interface tests were carried out for the two batches of axial braided C/C composite materials with certain macroscopic properties differences. The interface shear strength of the two batches of materials was measured as shown in Figure 4. These two batches of materials are produced by the same supplier with the same production process. However, due to the fluctuation of the production process, the properties of the two batches of materials are different.

A total of 50 ejection tests were carried out for each batch of material, while 40 ejection tests were carried out for each batch of material. The mean interface shear strength and standard deviation of material 1 were 7.72 and 0.50 MPa, respectively. The mean interface shear strength and standard deviation of material 2 were 9.23 and 0.53 MPa, respectively.

Fig. 4. Normal distribution of interface shear strength. (A) material 1, (B) material 2.

3.2. Difference analysis of interface defects in micro view

In order to find the reasons for the difference in shear strength of interface between the two batches of materials, the end surface morphology of the hole after the ejecting tests was observed and studied, as shown in Figure 5. After the ejecting test of material 2, the interface was greatly destroyed. The microscopic images show that the interface is lamellar structure and oriented along the fiber rod axis, indicating that the interface bonding strength is higher. The interface of material 1 is smoother than that of material 2.

Fig. 5. End face characteristics of matrix hole wall after ejecting fiber rods. (A) Surface morphology of hole wall of material 1, (B) Surface morphology of hole wall of material 2.

The internal characteristics of the wall of hole after the fiber rod ejected are shown in Figure 6. The pore wall of material 2 is rough, and the substrate surface has fiber attachment, which shows better interfacial bonding performance. The wall of hole of material 1 is smoother, and no fiber filament is attached to the surface. This interface difference is related to the surface roughness of the fiber rod.

The morphology of the ejecting fiber rod and its fiber filament are shown in Figure 7. The fiber surface of the fiber rod of material 2 is rough and attached matrix carbon, which is due to the formation

Fig. 6. Wall of hole after ejecting of fiber rod. (A) wall of hole after ejecting of fiber rod of material 1, (B) wall of hole after ejecting of fiber rod of material 2.

of matrix carbon "pinning" at the defect site of the fiber, resulting in local strong interface. The fiber surface of the fiber rod of material 1 is relatively smoother, with a small amount of matrix carbon attachment, and there are grooves parallel to the axial direction, forming a separation.

Through micro-CT combined with scanning electron microscopy (SEM), pores at different positions of the interface were measured, as shown in Figure 8. Two hundred observational tests were carried out to obtain the probability density of pores.

The obtained pore scale data were analyzed, and the pore shape was assumed to be a circular coin shape (the delamination thickness and diameter ratio were determined as 10 according to the previous micro-CT image), and the diameter scale was subject to lognormal distribution, as shown in Figure 9.

$$N(R') = \frac{1}{R'\sqrt{2\pi\sigma}} \exp \left\{ - \left[\frac{\ln(R'/R)}{\sqrt{2\sigma}} \right]^2 \right\} \quad (2)$$

In the formula, R' is the diameter of the pore and R is the average diameter of pores. The R value of material 1 is $138.87 \mu\text{m}$, and that of material 2 is $85.18 \mu\text{m}$. σ is the standard deviation of logarithm

Fig. 7. The end face characteristics of the fiber rod and the apparent characteristics of its fiber filament. (A) End face characteristics of the fiber rod of material 1, (B) Apparent fiber properties of material 1 fiber rod, (C) End face characteristics of the fiber rod of material 2, and (D) Apparent fiber properties of material 2 fiber rod.

Fig. 8. Diagram of position of pores on two batches of materials.

of diameter variable. σ of material 1 is 0.55, and σ of material 2 is 0.47. It can be seen from the statistical results that the interfacial pores of material 2 are significantly smaller than those of material 1, which may be the main reason why the interface shear strength of material 2 is higher than that of material 1.

Fig. 9. Probability distribution curve of pores on interface in C/C composite.

4. Interface differences lead to tensile property deviation analysis

These tests were carried out at room temperature using MTS's 858 Mini Bionix helical hydraulic testing machine with a maximum loading force of 25 kN and a loading rate of 0.5 mm/min. The observation surface of the specimen must be sprayed with black-and-white matte paint, and the spots were randomly distributed to facilitate the strain measurement in the test. Tensile strain was measured using ARAMIS noncontact optical full-

field strain measuring system from the German GOM company.

The working principle of the ARAMIS system is to divide the sample surface into small areas. The deformation of the sample is collected in real time in the experiment, the strain of each area is calculated in the post-processing, and the strain information of the sample is obtained. At the heart of the ARAMIS technology is digital image processing and 3D data analysis. The ARAMIS system is ideal for strain testing of highly heterogeneous composites, such as C/C composites.

The axial tensile properties of the two batches of materials were tested. The form and size of the samples are shown in Figure 10. Each batch of material contains eight tensile specimens. The experiment was carried out on the MTS experimental machine with a loading rate of 0.5 mm/min. Using the ARAMIS noncontact optical strain testing method, heterogeneous strains under different tensile loads were obtained, as shown in Figure 11.

Fig. 10. Tensile specimen size.

The observation of the material surface after the test shows that the strain concentration is usually at the interface between the fiber bundle and the matrix, as shown in Figure 12. It may be due to the existence of delamination and porosity at the interface, which leads to the stress concentration under tensile load, thus resulting in the strain concentration.

BE120-10AA(11)-X30 strain gauge was used to obtain strain through YE6261B dynamic data acquisition and analysis system. The typical stress-strain curves of the two batches of materials are shown in Figure 13.

First, it can be seen from the comparison between Figures 11 and 13 that the strain value of the strain concentration areas on the surface of

Fig. 11. Axial tensile strain distribution of axial braided C/C composite material.

Fig. 12. The observation of failure position of specimen after tensile tests.

Fig. 13. Uniaxial tensile stress-strain curve.

the material is significantly higher than the average tensile strain of the whole material during the stretching process, which is basically more than 10 times. These strain concentration areas are mainly the interface junction of the fiber rod (fiber bundle)/matrix. This also indicates that under tensile load, the fiber rod (fiber bundle)/matrix interface will be the weak position of the whole material and the starting point of failure. The interface properties are the main factors that affect the macroscopic properties of materials.

In addition, the tensile nonlinearity of the axial braided C/C composites is obvious, and the curve shows multiple attenuation of stiffness. When the maximum load capacity is reached, the load capacity of the specimen will decrease rapidly. However, due to the transfer effect of the interface on the force, the material still has a certain carrying capacity. The test results show that the tensile strength of materials 1 and 2 is 67.8 and 47.6 MPa, respectively. The two batches of materials have the same braiding process, but although the impregnation, carbonization, and heat treatment processes are similar (but there are some differences between batches), this also leads to differences in the meso interface. It also shows that the difference in interface shear properties results in the difference in tensile properties.

5. Conclusion

In this paper, a characterization method for the interface shear strength of the axial braided C/C composite was obtained. The recommended thickness of the sample of interface shear strength test of the axial braided C/C composite was four times the diameter of the fiber rod.

The mesoscopic structure characteristics of the interface of different batches of materials were obtained in this paper. The shear strength of the fiber rod/matrix interface was related to the pore's scale and roughness of the wall of interface. The smaller the average pore diameter is, the higher the interfacial shear strength. The rougher the wall of hole is, the higher the interfacial shear strength. The rougher the fiber surface is, the higher the shear strength.

The research shows that the axial tensile strength of the axial braided C/C composite material is affected by the interface shear strength. Within a certain range of interface shear strength, materials with large interface strength have high axial tensile strength.

Author contributions

Conceptualization was done by C. W. and W. T.; methodology by C. W.; software and validation by C. W. and W. T.; formal analysis by W. T.; investigation by C. W.; resources by W. T.; data curation by C. W.; writing—original draft preparation by W. T.; writing—review and editing by K. Z.; visualization by K. Z.; supervision by C. W.; project administration by W. T.; and funding acquisition by W. T. All the authors read and agreed to the published version of the manuscript.

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Conflicts of interest

The authors declare no conflict of interest.

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