Investigation on the Mechanism of Thermal Cycling Failure in C/SiC Coating Composite Materials

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Abstract

Continuous carbon fiber reinforced silicon carbide (C/SiC) composites possess advantages such as high specific strength and modulus, high damage tolerance, high temperature resistance, corrosion resistance, oxidation resistance, and erosion resistance. They have a wide range of applications in various systems and are of significant importance in the aerospace industry. Understanding the mechanical performance evolution of C/SiC composites under rapid high-temperature cycling conditions is crucial for mastering their temperature fatigue failure mechanisms and predicting their reliability and structural safety.

This paper focuses on the research and improvement of high-temperature structural materials for aircraft in extreme environments, as well as the need to enhance the high-temperature oxidation resistance of matrix materials in the process of coating thermal mismatch reduction. It extracts the significant scientific challenges faced and revolves around the problem challenges The research is focused on the following aspects: the thermal mismatch mechanism of functional coatings on C/SiC ceramic matrix fiber-reinforced composites is not yet clear; the influence of different cooling rates and different numbers of thermal fatigue treatments on the thermal mismatch of functional coatings is not yet clear; how to design targeted thermal expansion coefficients for functional coatings under different operating environments to enhance the durability of the coatings is not yet clear. Based on the refinement of the problems, further attention is given to the testing methods of the studied composite materials and the targeted environmental conditions. The studied materials are C/SiC composites, with a maximum test environment temperature of 1800°C and a minimum of -100°C. The mechanical performance tests mainly include tensile and compressive static forces. The environmental factor superposition method is used for the mechanical performance testing of the materials, continuously superimposing environmental factors to obtain the mechanical performance data of the composite materials under different environmental conditions, and to understand the influence of different environmental factors on the mechanical behavior of thermal protection materials. In order to provide support for subsequent mechanism and failure analysis, microstructure and phase composition detection are performed before and after the experiments, and damage evolution information is monitored online during the experimental process. Based on the results of microstructure observation, in-situ mechanical performance experiments, macroscopic mechanical performance experiments, and macro-micro cross-scale simulation analysis, the mapping relationship between microstructure-in-situ performance-macroscopic performance is determined. Based on microstructure damage observation, phase composition detection, and evolution information online monitoring, the damage evolution process of thermal protection materials is analyzed, and its response mechanism is obtained. Combined with the mechanical behavior curve, a damage evolution model is established. Finally, based on the research results of mechanical behavior under different environmental factor conditions, combined with damage evolution analysis, thermal parameters of the composite materials before and after mechanical performance testing under different environmental conditions are tested, mainly including thermal expansion coefficient, thermal conductivity, and specific heat capacity. For C/SiC composites with antioxidant coatings, a thermal mismatch theoretical model between the coating material and the C/SiC composite material is established based on small deformation elasticity theory, revealing the mechanical behavior response mechanism under rapid temperature change extreme environments, and finally providing a predictive model for the thermal mismatch fracture failure of the coating material.

Modeling and failure mechanism analysis of thermal mismatch in coatings of C/SiC composites under rapid temperature cycling in extreme service environments is a crucial aspect in the development of new aviation science and technology. This is due to the fact that the harsh service environment of C/SiC composites as thermal structural components results in rapid degradation of their oxidation resistance caused by coating material failures such as delamination and fracture due to inconsistent thermal expansion coefficients. Such research provides a theoretical basis and support for the development and improvement of new functional materials.

Key Words: Thermal Mismatch; Functional Coatings; Failure Mechanism; Ultra-high Temperature

1. Introduction

C/SiC composite materials have become candidate materials in hypersonic aircraft thermal protection systems, aerospace industry, and weapon systems due to their excellent properties such as high strength, high modulus, high temperature resistance, corrosion resistance, and oxidation resistance. Through carbon fiber toughening, carbon fiber reinforced silicon carbide (C/SiC) ceramic matrix composites have advantages such as high strength, high modulus, high temperature resistance, corrosion resistance, and oxidation resistance, making them the preferred choice for high temperature structural materials. With the continuous increase in Mach number of hypersonic aircraft, the service environment, including aerothermal/thermal loads, becomes more severe and complex, resulting in the generation of a large amount of thermal stress inside the materials, leading to complex changes in material behavior. The influence of thermal stress on the structure becomes significant, and it is necessary to study the thermo-mechanical properties of C/SiC composites in order to effectively apply these thermal stresses to structural analysis. Composite materials are essentially anisotropic and inhomogeneous, so they cannot be simply modeled as isotropic materials. The micro/meso structure of C/SiC composites, which are fiber-woven composites, is more complex and highly non-uniform. The microstructure of woven composites determines their properties, so it is necessary to conduct multi-scale analysis to study their properties. Currently, the research on the thermo-mechanical properties of fiber-woven composites mainly focuses on three levels: micro, meso, and macro. In the past few decades, scholars have developed various micro-meso models to predict the mechanical and thermal properties of woven composites. In recent years, researchers both domestically and internationally have conducted extensive studies on the high-temperature tensile, bending, and shear properties of SiC-based composites, achieving a series of outstanding results [1-3]. Meanwhile, many scholars have conducted targeted research on the oxidation problem of C/SiC composites in high-temperature oxidizing environments and have proposed a series of oxidation-resistant coatings to enhance the oxidation resistance of C/SiC composites [4-8]. However, there is little research on the mechanical failure behavior of C/SiC composites with oxidation-resistant coatings under extreme temperature cycling conditions. Currently, there is a lack of clear understanding of the environmental control factors and material performance control factors for material damage and failure, and various established simplified simulation methods have certain limitations. Establishing reliable and effective experimental simulation testing methods, as well as scientifically analyzing experimental results, involves some scientific issues, such as how to establish a simulation testing device in accordance with the principle of "similarity theory"? Can a progressive approximation method be used to establish a simulated experimental testing system? How to accelerate the material's damage evolution to shorten the experimental cycle? How to obtain online information about the material's damage failure process? How to decouple the experimental results of coupled environmental factors to obtain the material's damage failure mechanisms and controlling factors? These are all theoretical and technical issues that need to be addressed in the simulation testing of material service environments.

Among various materials, C/SiC composite material is the only high-temperature composite material available for temperatures up to 2800°C, making it widely used in defense and civilian fields such as aviation and aerospace. However, C/SiC composite materials are not oxidation resistant. Subsequently, oxidation-resistant coatings were developed, but the mismatch in thermal expansion coefficients between the C/SiC composite material and the coating leads to a shortened cyclic life. SiC ceramics can resist oxidation in hypersonic environments up to 1600°C, but the mismatch in thermal expansion also leads to crack formation in the matrix, resulting in direct oxidation of the carbon fiber reinforcement and a shortened cyclic life. Oxidation-resistant coatings typically have a higher thermal expansion coefficient. Due to the mismatch in thermal expansion coefficients between the coating material and the matrix, thermal strain occurs at the interface between the coating material and the contact interface, leading to uneven thermal contraction and lattice distortion, hindering dislocation motion, and causing cracks and decreased strength or even delamination in the coating. This exposes the C/SiC composite material directly to oxygen, leading to oxidative failure and damage. The equivalent thermal expansion coefficient of a composite material can be approximated as the thermal deformation of the material under a unit temperature gradient. Therefore, predicting the equivalent stiffness of the material is necessary before calculating the equivalent thermal expansion coefficient. Based on the hexahedral unit cell model of the micromechanical finite element model, after applying periodic conditions to the cell surface, the expression of the stiffness matrix C can be obtained by applying six sets of different displacement boundary conditions and solving the relationship between stress and strain. When there is a temperature gradient in the free-state unit cell structure, thermal strain is generated, and the constitutive relationship of the material needs to incorporate the contribution of thermal strain to obtain the equivalent thermal expansion coefficient. Ishikawa et al [9]. developed a scheme based on serial-parallel method to predict the thermo-mechanical properties of three-dimensional woven composites, but they only analyzed single-layer warp yarn organization with rectangular cross-section. Due to the limitations of the model, it cannot be applied to three-dimensional models. Cui Chunli et al[10]. predicted the mechanical properties of fiber-reinforced composites at the microscale based on the generalized self-consistent method. Li Diansen et al[11]. predicted the mechanical properties of three-dimensional four-directional woven composites by treating fiber bundles with the same orientation in composite materials as unidirectional composites based on the microscale single-cell model. Zhang Chunchun et al[12]. obtained the relationship between transverse isotropic matrix and internal stress of reinforced fibers based on the two-phase concentric cylinder model, and further predicted the mechanical properties of transversely isotropic fiber-reinforced composites. Naik et al[13]. developed an analytical model to predict the thermo-mechanical properties of three-dimensional thickness orthogonal interlock and interlayer orthogonal interlock woven composites at the micro and meso scales. Ai et al[14]. conducted experimental and numerical studies and used a multi-scale modeling method to predict the thermal expansion coefficient of fiber-woven composites in the temperature range of 300-2500 K. Ullah et al[15]. proposed a multi-scale computational framework and analyzed the homogenized mechanical properties of three-dimensional woven composites using finite element analysis of representative volume elements (RVEs). Siddgonde et al[16]. developed a finite element RVE-based thermo-mechanical analysis method and studied the thermo-mechanical properties of three-dimensional orthogonal interlock woven composites and angle interlock woven composites using multi-scale modeling techniques and periodic boundary conditions.

2. Experimental

This study utilizes the high-temperature adhesive produced by Beijing Huicheng Ruixin Co., Ltd. as the coating for the composite material. The coating preparation is conducted using the slurry coating method. The C/SiC composite material specimens for tensile testing are provided by Beijing Composite Material Manufacturing Research Institute, and the preparation process involves 2D plain weave and the PIP process. The ultra-high temperature testing equipment used in this study is provided by Shenzhen Wan Test Equipment Co., Ltd., specifically the 1T universal testing machine.

The microscopic characterization equipment employed is the Zeiss EVO18 instrument. The following figure 1 is the schematic illustration of coated C/SiC composites fabrication procedure

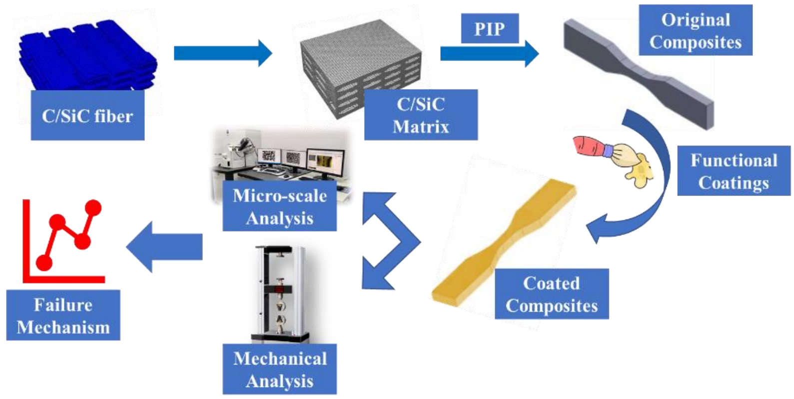


Figure 1 Schematic illustration of coated C/SiC composites fabrication procedure

3. Results and Discussion

3.1 Microstructural characteristics

The failure modes of sample under load can be observed from the diagram, which mainly include fiber bridging, oxidation and thinning fracture of fiber bundle, fiber bundle pull-out, debonding at the fiber bundle-matrix interface, and crack deflection. Based on the above analysis, the fracture mechanism of C/SiC composite material at high temperature can be summarized as shown in the diagram. Without the protection of SiC matrix, carbon fibers at the cut edge of the sample's external layer react with oxygen at high temperature, turning into gas and forming oxidation channels. At the same time, the initial crack on the sample surface propagates from the matrix to the carbon fibers under the action of stress, continuously oxidizing and eroding the carbon fibers, leading to gradual thinning and eventual fracture. In addition, microcracks induced by stress and heat coupling rapidly initiate and propagate at the interface between fiber and matrix. The presence of fibers causes crack deflection and even detachment of the matrix and fibers at both ends. However, the fibers can still bridge the two ends of the matrix, continue to bear the load, and prevent sudden failure of the specimen. Before cracks and voids inside the material connect with external defects to form oxidation channels, the internal fibers will not be oxidized. However, with the increase of internal stress, when the shear force between the fiber and interface reaches the interfacial shear strength, debonding occurs between the fiber and interface, and some carbon fibers start to pull out from the matrix. In a high-temperature oxygen environment, the interface between fiber bundles and matrix also undergoes debonding due to oxidation and force. At this time, the crack propagating to the fiber bundle will deflect as a whole until it reaches the next fracture interface, resulting in partial fiber bundle pull-out along with the matrix, forming a serrated fracture surface. In addition, the transverse fiber bundles do not play a major load-bearing role under axial force. These fiber bundles directly detach from the matrix as the crack extends and experience tearing under the traction of the matrix and fiber bundles. In the actual process, the various damage and failure modes mentioned above intertwine with each other, forming catastrophic main cracks in various directions. These main cracks rapidly expand and intersect, ultimately leading to material fracture failure.

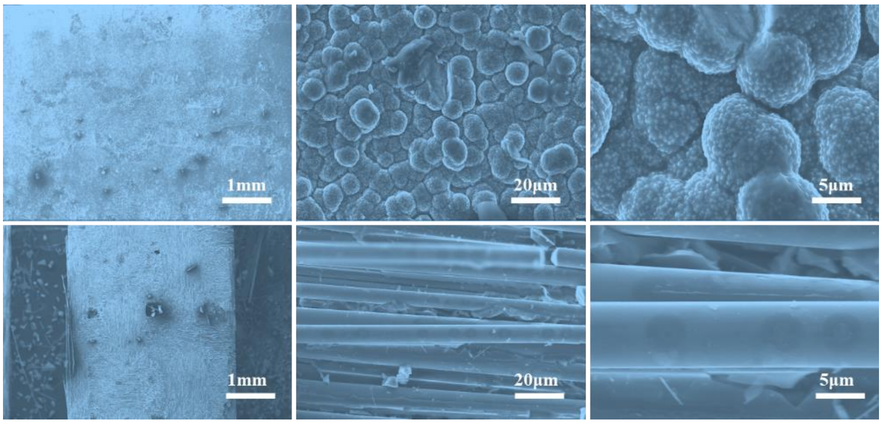


Figure 2 Microstructural characterization of C/SiC composites

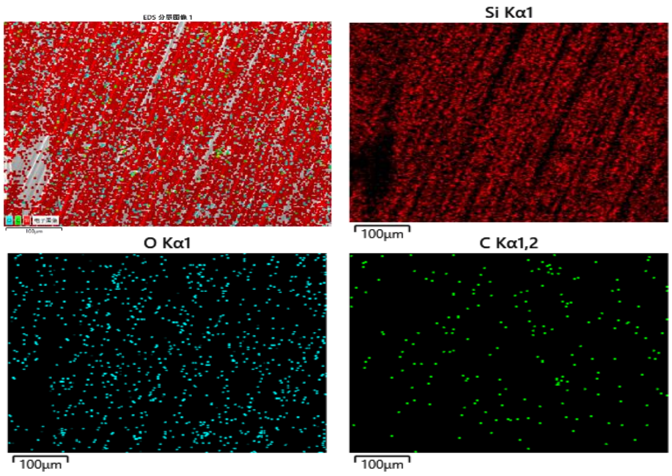
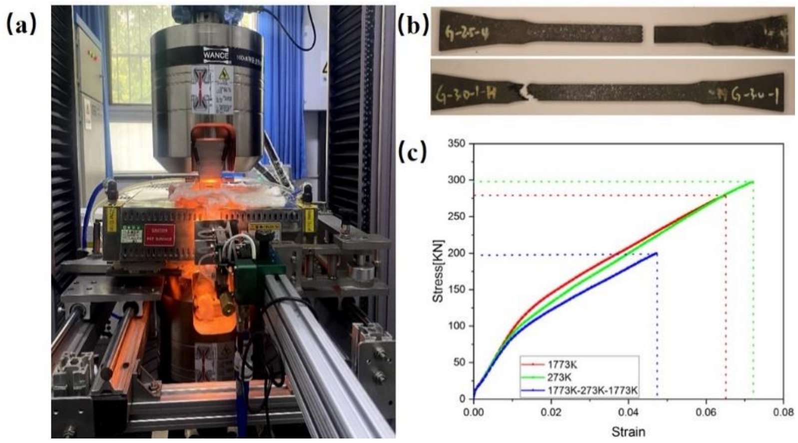


Figure 3 EDS mappings of element distribution corresponding to Si, O and C, respectively.

3.2 Tensile strength experiment under extreme changing temperature atmosphere with oxygen

This article presents the experimental testing of the tensile properties of test specimens using a laboratory-designed and developed ultra-high temperature mechanical performance testing system. The testing mainly includes the tensile performance at 1500 degrees Celsius and the tensile performance after a rapid cooling single-cycle from 1500 degrees Celsius to room temperature. The test results reveal that compared to the tensile performance testing in pure room temperature oxygen environment, the tensile performance of the test specimens under ultra-high temperature is slightly decreased due to the influence of high temperature oxidation. Furthermore, the specimens that have undergone a single extreme temperature cycling exhibit a significant decline in performance.



Therefore, developing functional coatings that can withstand rapid temperature fatigue is one of the urgent issues to be addressed

Figure 4 (a) Mechanical test facility with ultra-high temperature environment; (b) Typical failure results of C/SiC composite; (c) Test results of samples under different circumstances

3.3 Analysis of failure mechanism

Based on the TexGen software, a modeling and simulation of the habitual model of woven composite materials was conducted. In the preliminary stage, the fiber bundle and matrix parameters of C/SiC composite materials were used for modeling calculations, and the calculated tensile modulus of the single cell unit had an error lower than 10%.

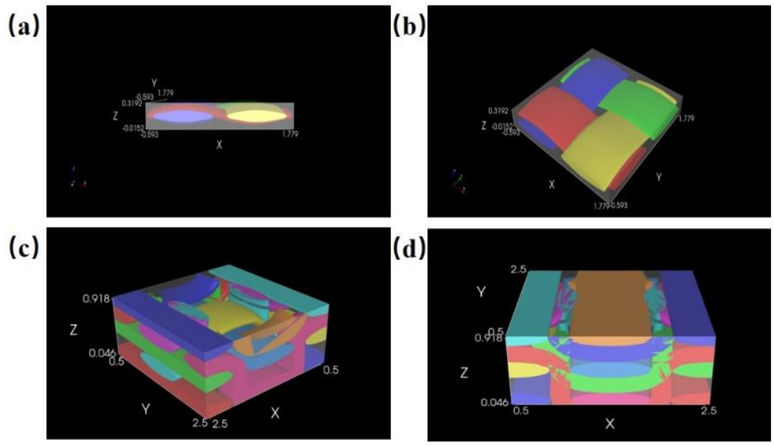


Figure 5 (a)-(b) Model of 2D C/SiC composites; (c)-(a) Model of 2.5D C/SiC composites

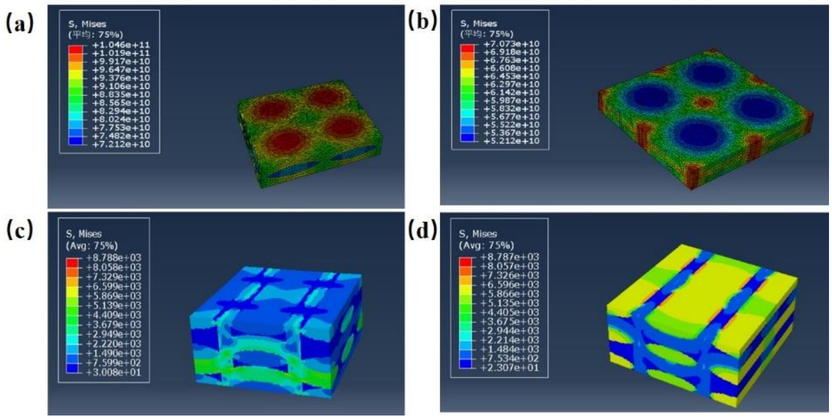


Figure 6 (a)-(b) 2D FEM calculation of C/SiC model; (c)-(d) 2.5D FEM calculation of C/SiC model

The establishment of this microscale model relies on the EasyPBC plugin, which is an ABAQUSCAE plugin used to estimate the homogeneous effective elastic properties of periodic Representative Volume Elements (RVEs). This plugin automatically applies the concept of homogenization of periodic RVEs in the ABAQUS user interface. It allows for the creation of deformable periodic boundary surfaces and provides access to finite element analysis data. The homogenization method of RVE requires the application of uniform strain on the microscale RVE and uses periodic boundary conditions. EasyPBC simplifies the process of applying periodic boundary conditions by automatically generating the required boundary node sets, constraint equations, displacement boundary conditions, and post-processing calculations. The plugin is user-friendly and does not require complex and time-consuming user input or the use of other software. It can be used to analyze RVEs with geometric variations and can be compared with other tools. In the two-dimensional woven single-layer panel, the fiber bundles serve as yarns that are woven to form a two-dimensional preform, which is then stacked and impregnated with resin to form a two-dimensional woven composite material. Compared with the microscale model of a single-layer composite material, the microstructure of a two-dimensional woven composite material is highly non-uniform. The number of fiber bundles and their geometric configuration have a significant impact on its mechanical properties. Therefore, establishing a reasonable microscale model is of greater importance for studying the mechanical properties of two-dimensional woven composite materials. Based on the Key model, a reasonable simulation of the fiber bundle cross-sectional shape and centerline of the reinforcement in a plain weave composite material is conducted to make the fiber bundles smooth, continuous, and have a bending shape close to reality. In a two-dimensional plain weave composite material, the interaction between fiber bundles leads to a non-circular cross-sectional shape of the fiber bundles. Based on previous studies, it is assumed that the deformed cross-sectional shape of the fiber bundles is elliptical, with major and minor axis diameters of a and b, respectively. Considering the repeatability and symmetry of the single cell model, it is assumed that the cross-sectional shape of the fiber bundles remains unchanged, and the warp and weft yarn fiber bundles are arranged uniformly and have the same curvature. The warp and weft yarns use the same material and have the same curvature. Describing the curvature form of the warp and weft yarns is a key aspect in constructing the single cell model of a two-dimensional plain weave composite material.

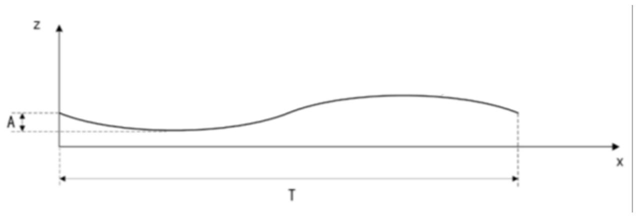


Figure 7 Schematic diagram of bent fiber bundles

$$\hat{x} = A\sin(\frac{2\pi}{T}x) \tag{1}$$

$$T = 2(a+l) \tag{2}$$

$$A = \frac{h-b}{2} \tag{3}$$

$$L = 4\int_{0}^{\frac{T}{4}} \sqrt{1 + A^2 \frac{4\pi^2}{T^2} \cos^2(\frac{2\pi}{T}x)}dx \tag{4}$$

For any curved fiber bundle after bending, its configuration is described by the following sine function, where T represents the period, A represents the amplitude, l is the distance between adjacent fiber bundles, and h is the height of the single cell model.

This article simplifies the C/SiC model of functional coatings into numerous vertical base cylinders and extracts one unit for discussion to calculate the stress state caused by thermal mismatch. Only the thermal mismatch between the functional coating and the ceramic layer is considered, and the entire system is considered to cool rapidly from high temperature to room temperature or low temperature. Only the temperature gradient is considered, and the constraints of the base are not considered, nor is the creep behavior and high-temperature oxidation behavior. The equivalent thermal expansion coefficient of the composite material can be approximately regarded as the thermal deformation of the material under a unit temperature gradient. Therefore, the prediction of the equivalent stiffness of the material is required before calculating the equivalent thermal expansion coefficient. Based on the hexahedral unit cell model of the microscale finite element model, after applying the periodic conditions on the cell surface, six sets of different displacement boundary conditions are applied, and the relationship between stress and strain can be obtained by solving the expression of the stiffness matrix C. When there is a temperature gradient in the free state unit cell structure, thermal strain will be generated accordingly, and the constitutive relationship of the material needs to include the contribution of thermal strain to obtain the equivalent thermal expansion coefficient. Based on the steady-state temperature analysis, the coupling equation of a uniform temperature field is used to solve the thermal expansion coefficient of the unit cell. Since the temperature distribution of the unit cell model is uniform, the thermal deformation in three directions of the unit cell can be obtained, and the following equation is used to predict the thermal expansion coefficient components in the corresponding directions of the unit cell:

$$\alpha_{ij} = \frac{\varepsilon_{ij}}{\Delta T} \quad (5)$$

where αij is the thermal expansion coefficient, εij is the strain, and ΔT is the temperature difference. By applying a constant temperature load to the unit cell, the thermal strain in three directions can be obtained. The thermal expansion coefficient components in three directions of the unit cell can be obtained according to the equation.

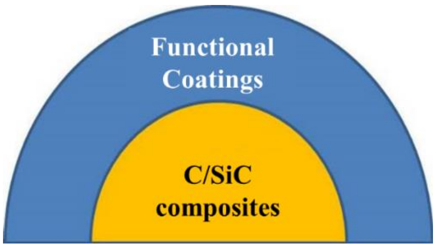


Figure 8 Schematic diagram of the geometric model for thermal mismatch in functional coatings

4. Conclusions

This paper investigates the mechanical properties and microstructural characteristics of C/SiC materials in a 1500℃-oxidizing environment using a self-developed thermo-mechanical-oxygen coupling tensile device and SEM characterization method. Important data regarding the high-temperature tensile strength, fracture morphology, and internal microstructural evolution of C/SiC materials are obtained. The research results show that under the combined action of thermo-mechanical-oxygen environment, the fracture modes of the composite material include fiber bridging, fiber bundle oxidation, pull-out, interface debonding, and crack deflection. These mechanisms collectively lead to the failure of the composite material. Among them, the oxidation of carbon fibers is one of the main failure causes, especially in the absence of SiC matrix protection. Internal cracks and voids do not immediately cause fiber oxidation, but when the internal stress reaches a certain level, they will propagate and lead to debonding and pull-out. The damage and failure of the composite material are the comprehensive result of multiple factors, including mechanical loads. High-temperature oxidation and interface degradation. These factors interact with each other, resulting in complex damage and failure behavior of composite materials under extreme environments. This study deepens the understanding of the damage and failure mechanisms of C/SiC composites in high-temperature oxidizing environments. The research provides a foundation and guidance for evaluating the performance of C/SiC composites in high-temperature applications and improving and optimizing the high-temperature service performance of C/SiC composites in the future.

Acknowledgements

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