



Contents lists available at ScienceDirect

Ceramics International

journal homepage: [www.elsevier.com/locate/ceramint](http://www.elsevier.com/locate/ceramint)

# A MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub>/SiC anti-oxidation coating for C/C composites prepared at relatively low temperature

Fang-xu Niu<sup>a,b</sup>, Yan-xiang Wang<sup>a,b,\*</sup>, Imran Abbas<sup>b</sup>, Shan-long Fu<sup>b</sup>, Cheng-guo Wang<sup>a,b</sup>

<sup>a</sup> Key Laboratory for Liquid-Solid Structural Evolution and Processing of Materials (Ministry of Education), Shandong University, Jinan 250061, China

<sup>b</sup> Carbon Fiber Engineering Research Center, School of Material Science and Engineering, Shandong University, Jinan 250061, China

## ARTICLE INFO

### Keywords:

Carbon/carbon composite  
Oxidation  
Coating  
Polymer-derived ceramic

## ABSTRACT

In this study, SiC coating for C/C composites was prepared by pack cementation method at 1773 K, and MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub> as an outer coating was successfully fabricated on the SiC coated samples by slurry method at 1273 K. The microstructure and phase composition of the coatings were analyzed. Results showed that a porous β-SiC inner coating and a crack-free MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub> coating are formed. Effect of Si<sub>3</sub>N<sub>4</sub> content on the oxidation resistance of the coated C/C composites at 1773 K in air was also investigated. The weight loss curves revealed that introducing the appropriate proportion of Si<sub>3</sub>N<sub>4</sub> could improve the oxidation resistance of coating. The MoSi<sub>2</sub>-SiOC/SiC coated C/C sample had an accelerated weight loss after oxidation in air for 20 h. However, the coating containing 45% Si<sub>3</sub>N<sub>4</sub> could protect C/C composition from oxidation for 100 h with a minute weight loss of 0.63%.

## 1. Introduction

Carbon/carbon (C/C) composite is a kind of remarkable material used for high temperature applications due to its certain properties such as high specific strength, high specific modulus, good thermal shock resistance and better dimensional stability [1–4]. However, the superior properties of C/C composites will not exist unless the composites are used in inert environment. Researches [5–7] revealed that the rapid oxidation of C/C composites occurs at 773 K in oxidizing environment. The mechanical properties of C/C composites will sharply decrease by 40–50% with a minute increase in weight loss, which severely limits the use of C/C composites.

Coating is a promising technique to protect C/C composites from oxidation at high temperature. MoSi<sub>2</sub> possesses an excellent high-temperature stability and oxidation resistance [8], which can keep performance after oxidation in air for 2000 h at 1923 K. Therefore, MoSi<sub>2</sub> is considered the suitable choice for coating against oxidation. However, high coefficient of thermal expansion (CTE) and accelerated oxidation at 673–773 K ("pest" phenomenon) [9,10] limit its applications for anti-oxidation coating material. Early study [11] indicated that the poor anti-oxidation performance of MoSi<sub>2</sub> at low temperature might be enhanced by addition of 30–50 vol% Si<sub>3</sub>N<sub>4</sub>. In addition, Si<sub>3</sub>N<sub>4</sub> can significantly reduce the CTE gap between the substrate and coating, which can further eliminate the coating cracking caused by thermal cycling.

Pre-ceramic polymer has been widely used in recent years, since the polymer-derived ceramic can be obtained at much lower temperatures compared to other preparation methods [12,13]. High hydrogen silicone oil (H-PSO) is one of the most frequently used precursors to produce SiOC ceramics [14]. Although the pre-ceramic polymer is commonly used to prepare the ceramic composites [15], but its addition often produces void gaps during the coating preparation. Moreover, most coatings with an excellent anti-oxidation protective ability for C/C composites are prepared at high temperature [16–18]. In this study, a new method was proposed to prepare the coating for C/C composites at relatively low temperature. Initially, a porous SiC coating was prepared on C/C composites by pack cementation method at 1773 K. Afterward, a MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub> multi-phase coating on the SiC coated sample was produced at 1273 K by a simple and low-cost slurry method. The H-PSO was used as a dispersant in the slurry process. The preparation, microstructure, phase composition and anti-oxidation performance of the coatings were investigated in detail. Moreover, the concentration effect of Si<sub>3</sub>N<sub>4</sub> on oxidation resistance of the coating at 1773 K in air was also explored.

## 2. Experimental

### 2.1. Preparation of the MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub>/SiC coating

Small sized substrates (10 mm×10 mm×8 mm) were cut from a

\* Corresponding author at: Key Laboratory for Liquid-Solid Structural Evolution and Processing of Materials (Ministry of Education), Shandong University, Jinan 250061, China.  
E-mail address: [15620980301@163.com](mailto:15620980301@163.com) (Y.-x. Wang).

<http://dx.doi.org/10.1016/j.ceramint.2016.11.151>

Received 14 October 2016; Received in revised form 16 November 2016; Accepted 22 November 2016

Available online xxx

0272-8842/ © 2016 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

bulk piece of 2D C/C composite with density of 1.60 g/cm<sup>3</sup> (Shandong Weiji carbon technology Co. Ltd., Jinan, China). All specimens were hand-polished using 200 and 500 grit SiC papers, and were cleaned ultrasonically with ethanol and dried in an oven at 373 K for 2 h. The SiC inner coating was prepared at 1773 K for 2 h in Ar atmosphere by embedding method and the premixed Si and C were used as embedding powders. The MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub> coating was prepared by slurry method. The MoSi<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> powders were dispersed in H-PSO, with tetramethyl tetra vinyl cyclotetrasiloxane (Vi-D4) as a cross-linking agent, platonic chloride ethanol solution as a catalyst (1# 15 vol% Si<sub>3</sub>N<sub>4</sub>, 85 vol% MoSi<sub>2</sub>; 2# 30 vol% Si<sub>3</sub>N<sub>4</sub>, 70 vol% MoSi<sub>2</sub>; 3# 45 vol% Si<sub>3</sub>N<sub>4</sub>, 55 vol% MoSi<sub>2</sub>; 4# 60 vol% Si<sub>3</sub>N<sub>4</sub>, 40 vol% MoSi<sub>2</sub>). The slurry was mixed ultrasonically in a beaker. Then the as-obtained slurry was brushed directly on the surface of the SiC coated specimens. The samples were treated at 1273 K for 2 h in N<sub>2</sub> atmosphere to form the MoSi<sub>2</sub>-SiOC-Si<sub>3</sub>N<sub>4</sub> outer coating. For comparison, MoSi<sub>2</sub>-SiOC outer coating was prepared by the same slurry method.

## 2.2. Characterization

The coatings were characterized by scanning electron microscopy (SEM, Hitachi SU-70) with energy dispersion spectroscopy (EDS, attached to the SEM), X-ray diffraction (XRD, Rigaku D/max-c, 4°/min) with CuKα radiation (λ=0.154056 nm). In addition, the pyrolysis process of cross-linked H-PSO was analyzed by Fourier transform infrared spectroscopy (FTIR, Bruke Alpha, 4 cm<sup>-1</sup> resolution) and XRD.

To exhibit the effect of coating on flexural properties of C/C composites, three-point bending tests were performed by an electronic universal testing machine to obtain the load-displacement curves. The

tests were carried out with a loading speed of 0.5 mm/min and a support span of 40 mm.

Isothermal oxidation tests were carried out at 1073 K and 1773 K in air to investigate the oxidation behavior of the coated samples. During oxidation tests, the coated samples were kept in a furnace and heated to a given temperatures. After a certain time, the samples were taken out from the furnace and cooled down to room temperature quickly. The mass loss of the samples was measured by an electronic balance with sensitivity of ± 0.1 mg. This oxidation process was repeated several times. Cumulative weight loss of the oxidized samples was calculated and reported as a function of oxidation time. The weight loss (w%) was calculated by Eq. (1) to measure the anti-oxidation property.

$$w\% = \frac{m_0 - m_1}{m_0} \times 100\% \quad (1)$$

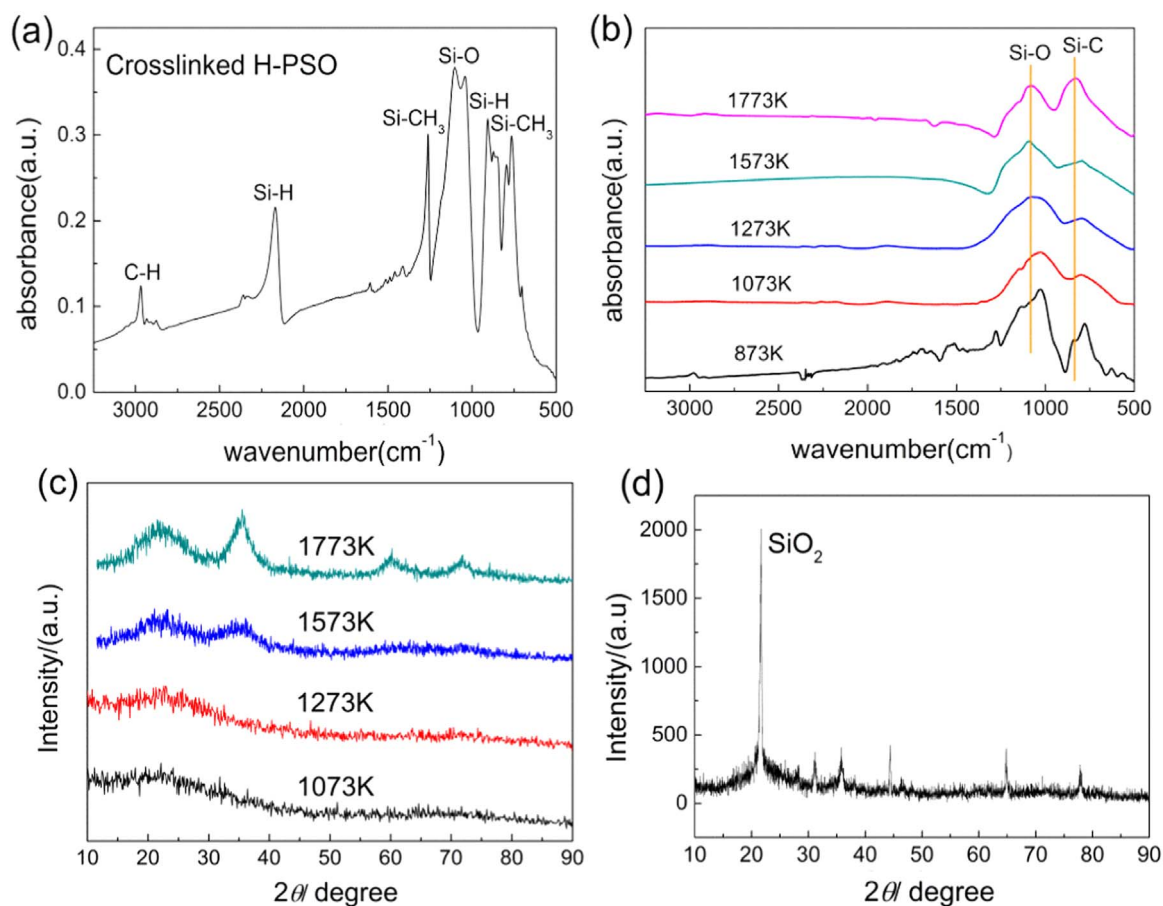
where  $m_0$  and  $m_1$  are the weight of samples before and after oxidation, respectively.

## 3. Results and discussion

### 3.1. Pyrolysis process of cross-linked H-PSO

H-PSO is composed of the basic structural unit Si-O and lots of pendant groups Si-H, Si-CH<sub>3</sub>, but the Si-H groups can give good reaction activity to H-PSO. In order to define the molecular structure of the cross-linked H-PSO after pyrolysis, the pyrolysis products of cross-linked H-PSO at different temperatures were characterized by XRD and FTIR.

Fig. 1(a) and (b) illustrate FTIR spectrum of the cross-linked H-PSO and the pyrolysis products at different temperatures, respectively.



**Fig. 1.** (a) FTIR spectrum of the cross-linked H-PSO; (b) FTIR spectrum of the pyrolysis products of cross-linked H-PSO at different temperatures; (c) XRD of the pyrolysis products of cross-linked H-PSO at different temperatures; (d) XRD of the SiOC ceramics after oxidation at 1773 K.

Download English Version:

<https://daneshyari.com/en/article/5439007>

Download Persian Version:

<https://daneshyari.com/article/5439007>

[Daneshyari.com](https://daneshyari.com)