



A ZrC-SiC/ZrC-LaB₆/ZrC multilayer ablation resistance coating for SiC-coated carbon/carbon composites



Yujun Jia, Hejun Li *, Qiangang Fu *, Jiajia Sun

State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an 710072, PR China

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ABSTRACT

This work proposes a new multilayer coating system containing ZrC-SiC, ZrC-LaB₆, ZrC layer with respectively different functions to protect the carbon/carbon (C/C) composites. The coating system overcame the poor oxidation resistance of ZrC coating and weak anti-scouring performance of modified ZrC coating, which could provide the protection for C/C composites through the whole ablation process. After ablation for 40 s, the liner ablation rate of the multilayer coating was $-2.89 \pm 0.23 \mu\text{m/s}$, decreased by 150.26%, 134.61% and 133.22% relative to the ZrC, ZrC-LaB₆ and ZrC-SiC coating, respectively. The coating showed a weight gain, while all the other coatings lost weight after ablation. The good ablation resistance was mainly ascribed to different functions of these layers in the coating system. The ZrC-SiC layer provided oxidation protection during the initial ablation stage by forming a composite oxide of ZrO₂-SiO₂. In the intermediate stage, the ZrC-LaB₆ layer promoted the formation of a stable scale containing (La_{0.1}Zr_{0.9})O_{1.95}. As the ablation going on, a compact scale containing (La_{0.1}Zr_{0.9})O_{1.95} and La₂Zr₂O₇ was formed, which further protected the composites. During ablation, the ZrC layer provided a foundation for the multilayer coating system against scouring.

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1. Introduction

Carbon/carbon (C/C) composites are ideal materials for applications of high temperature structural components such as nose tips, sharp leading edges and heat shields for reentry vehicles due to their low density, low coefficient of thermal expansion (CTE), high specific strength and retention of mechanical properties [1–5]. However, most of these applications involve oxidizing or ablation environments, where the composites require suitable protection because of the rapid reaction of carbon with oxygen above 500 °C [6–8].

Ultra-high temperature ceramic (UHTC) carbide coating can provide such protection for the composites [9–12]. Among the UHTC carbides, ZrC are attractive coating materials applied in ablation environments owing to its merits of high melting point (3540 °C), relative low density and sufficiently high melting point of the corresponding oxide (around 2700 °C) [13–16]. The merit of monophase ZrC is that it can serve as an anti-scouring layer in the service of the composites. For this kind of coating, it is expected that a dense ZrO₂ layer can form to prevent the oxygen ingress and resist the scouring from the airflows during ablation [17]. However, at temperatures below the melting point of its oxide, ZrC has a poor oxidation resistance [18] and the coating will be subjected to rapid oxygen infiltrating because of the linear rate law of its oxidation [19]. In the applications, there is a process of temperature rise. Only

at an ultra-high temperature, a dense ZrO₂ layer can form. Below this temperature, the oxide scale is still porous. Besides, due to limited thickness of the coating, the formation of such dense scale is always hysteretic or sometimes impossible for the coated composites. In other words, the erosion of the basis material may be prior to forming the dense ZrO₂ scale. These shortages make the ablation resistance of monophase ZrC coating not very exciting.

To solve this problem, most works focus on using additives [20–25]. Addition of silicon-based phases (e.g. SiC, MoSi₂) can significantly improve the oxidation resistance of UHTCs [20–23], since the formed SiO₂ can seal the cracks and pores of the scale below 1800 °C. At higher temperatures, in our previous work [24], incorporation of LaB₆ could significantly improve the oxidation resistance of ZrC-coated C/C composites due to the formation of a dense oxide scale induced by La during ablation. The merits of the above modified coatings are the significant improvement in the oxidation resistance.

However, for the coatings under high heat fluxes containing scouring, owning the satisfactory oxidation resistance alone is insufficient. A strong foundation acting as mechanical support for the oxide scale is also important. Although the oxidation resistance in different temperature range can be improved using the above additives, the melting resistance of the total oxide scale is degraded as the temperature rises to the extremely high temperatures due to the introduction of low-melting-point components [26] such as SiO₂ and La₂O₃ (the oxidation products of the additives, which have melting points lower than ZrO₂). For this reason, the ablation resistance of these modified coatings is also not

* Corresponding authors.

E-mail addresses: lihejun@nwpu.edu.cn (H. Li), fuqiangang@nwpu.edu.cn (Q. Fu).

very exciting under high heat fluxes containing scouring. Thus, from the anti-scouring point of view, it requires an oxide scale with enough-high melting point.

To overcome the poor oxidation resistance of ZrC coating, weak anti-scouring performance of modified ZrC coating and then make the C/C composites adapt to the whole ablation process in the applications, a new multilayer coating system was designed based on the respective merits of pure ZrC coating and different-dopant modified ZrC coating. This coating system contains low temperature SiC-modified ZrC layer, high temperature La-modified ZrC layer, and ultra-high temperature ZrC layer, which was scarcely reported in the open literature.

So far, many coating techniques, such as slurry, chemical vapor deposition (CVD), pack cementation and supersonic atmosphere plasma spraying (SAPS), have been used to prepare coatings for C/C composites. Among these techniques, SAPS is an efficient coating preparing method. The bonding strength of SAPS coating is stronger than CVD and slurry. Although SAPS prepares coatings with weaker bonding strength than pack cementation, it has advantages of depositing almost all ceramic coatings due to the high plasma temperature (10,000 K) and high jet velocities (up to 600 m/s) [27]. Besides, it can precisely control the content of the ceramics, which is much superior to other methods. Therefore, in this study, the multilayer coating system was deposited on the C/C composites using this process. The phase composition, microstructure, ablation properties and behavior of the coatings were investigated.

2. Experimental

2.1. Coating preparation

2D C/C composites with a density of 1.75 g/cm³ were used as substrates. The composites were shaped into cylindrical samples with a diameter and thickness respectively 30 and 10 mm. These samples were cleaned with alcohol and then dried at 80 °C for 12 h for coating.

The above processed samples were coated with a SiC inner coating by pack cementation firstly. The preparing details were reported elsewhere [28].

Four kinds of outer coating were prepared by SAPS: ZrC coating (named ZC), ZrC coating with SiC (named ZS), ZrC coating with LaB₆ (named ZL) and the multilayer coating. ZrC (400 mesh, >98% purity), LaB₆ (325 mesh, 99.5% purity) and SiC (1200 mesh, 99.0% purity) powders were utilized as the starting spraying materials. The composition of ZS and ZL was ZrC + 30 vol.% SiC and ZrC + 20 vol.% LaB₆, respectively. The multilayer coating system was composed of three layers, with the scheme shown in Fig. 1. The composition of the upper layer was ZrC + 30 vol.% SiC, which was the same as that of the corresponding reference coating. This layer was designed to be the first oxygen barrier

layer. Also, the interlayer was composed of ZrC + 20 vol.% LaB₆, which was the second oxygen barrier layer. The inner layer was monophase ZrC.

The powders or composite powders were mixed and roller milled to ensure homogeneous mixing. Before spraying, a process of agglomerating the powders was conducted using a spray dryer to improve their flow ability during SAPS. The powders were mixed with 49 wt.% distilled water and 2 wt.% polymeric binder to form the slurries. These slurries were then forced to pass through the dryer to obtain the final agglomerated powders.

The agglomerated powders were then sprayed on the SiC-coated samples using a SAPS device, which is composed of powder feeder, plasma torch, water-cooling circulator, gas supply, control unit with PC interface and power supply unit. The coatings were prepared according to the parameters: spraying current 380–415 A, spraying voltage 120–150 V, primary gas Ar 74 L/min, carrier gas Ar 10 L/min, second gas H₂ 5–15 L/min, powder feed rate 25 g/min, spraying distance 100 mm and nozzle diameter 5.5 mm. In this study, the total thickness of the multilayer coating was designed to be similar to that in Ref [17,24]. The thickness design for different layers in the system is based on the following consideration:

Thickness of ZS or ZL layer was designed to be between that of the sprayed coatings in Ref [21] and Ref [29] to insure the oxidation protection. The ZC layer should be thicker than the modified layer because the multilayer coating will be tested at high heat flux containing scouring. Thus, the reference coatings of ZC, ZS and ZL were all fabricated by 12 cycles of spray. The outer layer and interlayer of the multilayer coating were respectively formed after 2 cycles of spray. The inner layer of the multilayer coating was fabricated by spraying 8 cycles.

2.2. Ablation test

The coated samples were tested under an oxyacetylene torch with a heat flux of 4.2 MW/m² according to GJB323A-96 [30]. Samples were placed with their surface vertically to the flame. The inner diameter of the nozzle tip was 2.0 mm. The distance between the nozzle tip and the sample surface was held constant at 10 mm. The pressure and flux of O₂ were 0.4 MPa and 0.42 L/s, and those of C₂H₂ were 0.095 MPa and 0.31 L/s, respectively. An infrared thermometer (Raytek MR1SCSF) in two-color mode with an error of ±0.75% was used to measure the sample surface temperature. The detector wavelength pairs of the thermometer were 0.9 and 1.0 μm. The multilayer coating was exposed to the flame for 20 s, 30 s and 40 s. The linear gain rate [31] and mass variation rate [24] were calculated according to the following formula. The ultimate result was the average of three samples.

$$R_l = (l_0 - l_1)/t \quad (1)$$

$$R_m = (m_1 - m_0)/t \quad (2)$$

where R_l and R_m are the linear ablation rate and mass variation rate, respectively; l_0 and m_0 are the sample height and sample mass before ablation, respectively; l_1 is the sample height in the most erosion zone after ablation; m_1 is sample mass after ablation; t is the ablation time.

2.3. Coating characterization

The phase analysis of the coated specimens before and after ablation tests were conducted by X-ray diffraction (XRD, X'Pert Pro MPD) with a Cu K α radiation of wavelength 0.154 nm. The element distribution of the multilayer coating was analyzed by electron probe micro analyzer (EPMA-1720) combined with WDS. The microstructure and chemical composition of the coated specimens were analyzed by scanning electron microscopy (SEM, JSM6460) equipped with energy dispersion spectroscopy (EDS).

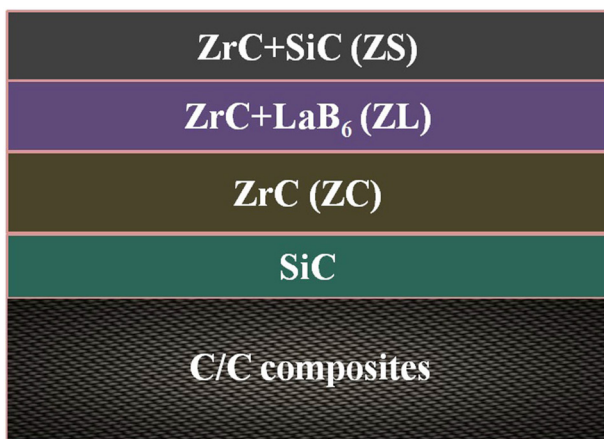


Fig. 1. Designed schematic structure of the multilayer coating system for C/C composites.

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