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Oxidation protection of carbon/carbon composites with a plasma-sprayed ZrB_2–SiC–Si/Yb_2SiO_5/$LaMgAl_{11}O_{19}$ coating during thermal cycling

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Abstract

Carbon/carbon composites were plasma sprayed with a ZrB_2–SiC-Si/Yb_2SiO_5/$LaMgAl_{11}O_{19}$ coating to improve the oxidation resistance at high temperature. Oxidation protection behavior of the coating was investigated by thermal cycling test using a gas flame about 2273 K. Results showed that the weight loss for the completely coated sample was only 0.30% after 10 cycles of heating for 79 min, and it gradually increased to 4.8% until the failure of coating with 24 cycles. The presence of through-cracks and horizontal cracks in the coating due to the durative oxidation of ZrB_2–SiC–Si layer progressively accelerated the oxidation of substrate and failure of coating.

Keywords: Carbon/carbon composites; Coating; Microstructure; Oxidation; Thermal cycling

1. Introduction

Due to the advantages such as low density, high strength-to-weight ratio and retention of mechanical properties at elevated temperatures, carbon fiber reinforced carbon matrix (C_f/C) composites could find specific advanced applications including hot section components for missile engines, exhaust parts for new fighter aircraft, hypersonic vehicle fuselage and wing components, and structures for space defense satellites.[1] These applications often require C_f/C composites to operate in an oxidizing environment. However, C_f/C composites are prone to oxidation in the high-temperature oxidizing environment, which leads to the degradation of mechanical strength and thus significantly limits their available applications.

Oxidation resistant coating is considered an effective approach to protect C_f/C composites against oxidation.[2–4] SiC is one of the promising materials due to good oxidation resistance and excellent compatibility with C_f/C composites. Therefore, the SiC-containing coatings attracted much attention during the past decade.[5–40] To authors' knowledge, the reported attractive SiC-containing coatings for C_f/C composites were listed in Table 1. These coatings could effectively protect C_f/C composites against oxidation according to the negligible weight change after oxidation test. However, these coatings seemed to provide the satisfied protection for C_f/C composites only at temperatures no more than 2000 K in the furnace [see Table 1]. For the new generation of hypersonic weapons and aircrafts, it is desired that the coated C_f/C composites could endure the high-speed flame with temperature about 2273 K or higher. The developed coatings may be inadvisable for C_f/C composites against the high-temperature flame. Therefore, it is necessary to develop

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Table 1
The reported attractive oxidation protective coatings for C_f/C in the past decade.[5–40]

Coatings	Oxidation test[a]			Refs.
	Temp (K)	Time (h)	Weight loss	
SiC/yttrium silicate/glass	1773	164	1.65 wt.%	[5]
SiC/yttrium silicate	1773	73	1.93 wt.%	[6]
$SiC/ZrO_2–SiO_2$	1773	10	1.97 wt.%	[7]
$MoSi_2–SiC–Si$	1773	200	1.04 wt.%	[8]
$SiC–B_4C/SiC/SiO_2$	1773	50	<1.3 wt.%	[9]
SiC whisker-toughened $SiC–CrSi_2$	1773	50	0.66 wt.%	[10]
SiC/Cr–Al–Si	1773	197	0.079 wt.%	[11]
SiC/yttrium silicates/borosilicate glass	1873	202	2.87 mg cm^{-2}	[12]
SiC/Si–Mo–W	1773	252	1.56 wt.%	[13]
Si–SiC	1773	166	0.61 wt.%	[14]
SiC_n/SiC	1873	64	1.3 wt.%	[15]
$SiC/SiC–Mo_2Si$	1573	110	1.6 wt.%	[16]
C/SiC/Si–SiC	1873	170	1.64 wt.%	[17]
SiC nanowire-toughened $SiC–MoSi_2–CrSi_2$	1773	155	0.64%	[18]
Yttrium silicate	1873	202	2.87 mg cm^{-2}	[19]
$SiC/MoSi_2$	1773	346	2.49 mg cm^{-2}	[20]
$C/SiC/MoSi_2–Si$	1773	300	1.4 wt.%	[21]
C/SiC/Si–Mo	1873	300	0.65 wt.%	[22]
$MoSi_2–CrSi_2–SiC–Si$	1873	300	0.5 wt.%	[23]
SiC/Si–W–Cr	1773	51	2.26 wt.%	[24]
$SiC/SiC–Si–ZrB_2$	1773	386	−0.08 wt.%	[25]
$SiC/ZrB_2–SiC/SiC$	1773	217	0.56 wt.%	[26]
C/SiC/Mo–Si–Cr	1873	300	0.25 wt.%	[27]
SiC nanowire-toughened $CrSi_2$-SiC-Si	1773	316	1.24 wt.%	[28]
$MoSi_2–CrSi_2$–Si/B-modified SiC	1873	300	0.41 wt.%	[29]
SiC nanowire-toughened Si–Cr	1773	185	−0.79 wt.%	[30]
SiC/SiC–YAG–YSZ	1773	150	−1.77 wt.%	[31]
$SiC–MoSi_2/ZrO_2–MoSi_2$	1773	260	1.31 wt.%	[32]
$SiC/TaB_2–SiC–Si$	1773	300	2.6 mg cm^{-2}	[33]
B_2O_3-modified $MoSi_2–CrSi_2$–Si/B-modified SiC	1873	900	−0.03 wt.%	[34]
$TaC–SiC/TaB_2–TaSi_2–SiC–Si$	1773	330	0.9 wt.%	[35]
SiC/Mullite	1773	322	0.489 mg $(cm^2\ h)^{-1}$	[36]
$SiC/AlPO_4$	1873	100	1.8 wt.%	[37]
$SiC/TaB_2–TaC–SiC$	1773	400	1.43 wt.%	[38]
SiC/LaB_6 modified $MoSi_2$	1773	750	0.108 wt.%	[39]
$SiC–Si/ZrB_2$ modified $SiC–Si/ZrB_2$ modified $MoSi_2–SiC–Si$	1953	50	2.44 wt.%	[40]

[a] Isothermal oxidation test in the furnace.

new coatings to protect C_f/C composites from oxidation under the gas flame with temperature about 2273 K.

Our group designed a novel coating system which consisted of the inner layer of silicate and the top layer of thermal barrier ceramic.[41–43] The heat resisting and thermal stability of silicate layer are improved, thanks to the heat insulation of the top thermal barrier layer. In previous works, $Yb_2SiO_5/LaMgAl_{11}O_{19}$ (YSO/LMA) coating was plasma sprayed on C_f/SiC substrate.[41,42] The coated and uncoated C_f/SiC samples were thermally cycled by the gas flame with temperature about 2273 K. It was found that the weight loss for the sample coated on one-side was 4.1 wt.% after 11 cycles of heating for 85 min, for the uncoated sample it was as high as 20.6 wt.%.[41] It is speculated that the YSO/LMA coating may be effective to protect C_f/C composites at high temperature. The coefficient of thermal expansion (CTE) of C_f/C composites is so low that the mismatch of CTE between the coating and substrate is quite serious. ZrB_2–SiC composites have good compatibility with C_f/C composites[44–46] and may be the promising bond layer between YSO/LMA coating and C_f/C substrate. In this study, the C_f/C composites were plasma sprayed with the YSO/LMA coatings with and without ZrB_2–SiC composites bond layer. Microstructure and thermal cycling behavior of the coated samples were investigated on a burner-rig setting using a gas flame with temperature about 2273 K. It is expected that this study could provide some valuable information for developing the high-temperature oxidation protective coatings for C_f/C composites.

2. Experimental procedure

The raw materials used in this study were the commercial powders of Yb_2O_3, SiO_2, MgO, Al_2O_3, La_2O_3, Zr, Si and B_4C powders. The characteristics of the starting powders with their sources are presented in Table 2. The YSO and LMA powders were synthesized by solid-state reactions according to Eqs. (1) and (2), respectively, and the ZrB_2–SiC-20 wt.%Si (ZSS) composite powders with mole ratio of 2:1 for ZrB_2 to SiC were