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Preparation and hydrothermal corrosion behavior of C_f/SiCN and C_f/SiHfBCN ceramic matrix composites

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Abstract

C_f/SiCN and C_f/SiHfBCN-based ceramic matrix composites (CMC) were fabricated using the precursor infiltration and pyrolysis technique (PIP). Their behavior in subcritical hydrothermal conditions was investigated at 150–250 °C using exposition times of 48, 96 and 240 h and shown to rely on active corrosion.

The data of the mass loss as a function of the corrosion time and temperature at S/V of 0.075 were used to rationalize the corrosion kinetics of the CMCs. Both materials have been shown to exhibit excellent stability in subcritical hydrothermal conditions. The corrosion rates of C_f/SiHfBCN were lower than those determined for C_f/SiCN; furthermore, SEM investigation indicates that spallation occurred in the C_f/SiCN samples; whereas the ceramic matrix was still attached on the individual carbon fibers in C_f/SiHfBCN. The results indicate that the incorporation of Hf and B into SiCN matrix leads to a significant improvement of its hydrothermal corrosion performance.

Keywords: Ceramic matrix composites (CMCs); Nanocomposites; Hydrothermal corrosion; Single-source precursors

1. Introduction

Carbon fibers reinforced ceramic matrix composites (CMCs) are promising candidates for the production of advanced high performance structures which have been developed for applications in various fields like aerospace, energy, and the automobile industry,[1] due to their outstanding properties (e.g., high fracture toughness,[2] stability in aggressive chemical environments, high-temperature resistance, lightweight,[3] etc.). They can be fabricated using different processing routes, such as chemical vapor infiltration (CVI),[4,5] polymer infiltration pyrolysis (PIP, also called liquid polymer infiltration – LPI), as well as liquid silicon infiltration (LSI, also called melt infiltration), each of them displaying advantages and drawbacks. For instance, CVI offer high mechanical strength and strain capability of the obtained CMCs, however the CVI preparation of CMCs is time consuming and expensive. Comparatively, PIP/LPI methods can benefit from the low costs; furthermore, the PIP technique allows for fabricating complex-shaped, large-scale components. Various CMC systems were prepared using the PIP method, depending on the preceramic polymer used for infiltration. Thus, C_f/SiC CMCs can be obtained from poly (organosilanes) or poly(organocarbosilanes),[6] C_f/SiCN are prepared upon using poly(organosilazanes) for the PIP process[7]; whereas poly(organoborocarbosilazanes) are used for preparing C_f/SiBCN.[8]

CMCs are candidates for various applications at high temperatures and in hostile environments. However, carbon fibers reinforced CMCs suffer at high temperatures from oxidation issues.[9] Lamoroux et al. devoted initial and intensive studies on the oxidation of carbon fiber reinforced SiC composites.[10,11] In order to improve their oxidation resistance, it has been suggested that for instance a multi-layered matrix design might be a suitable solution.

Furthermore, when exposed to combustion environments, the long term durability of CMCs requires the use of environmental barrier coatings (EBCs) to prevent severe degradation reactions of the CMCs with the combustion products (e.g., steam or salts).

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Silicon-based matrix materials (such as SiC) are prone to strong decomposition in combustion environments, as $Si(OH)_4$ is generated and released from the reaction between the silica-based passivation layer and water vapor. SiC-based CMCs coated with barium strontium aluminosilicate (BSAS)[12] or with other oxide-based materials having low vapor pressure at high temperature (e.g., ZrO_2 HfO_2, rare earth silicates)[13,14] have been shown to effectively protect the CMCs from hot corrosion.

The hot corrosion behavior of CMCs was investigated within the context of their use in turbine engines. Thus, Cheng et al.[15–17] described the high-temperature degradation mechanisms of C_f/SiC composites in complex salt-vapor environments (i.e., $O_2/H_2O/Na_2SO_4$ or Ar/Na_2SO_4). However, few studies concerning the corrosion behavior of ceramic fibers or CMCs under hydrothermal conditions have been reported[18,19] and consequently only scarce information is present in the literature with respect to their behavior.

The purpose of the present study is to demonstrate the possibility of fabrication of C_f/SiCN and C_f/SiHfBCN CMCs via the PIP technique as well as to assess and discuss for the first time their corrosion behavior under subcritical hydrothermal conditions. A comparison between the corrosion behavior of C_f/SiCN and C_f/SiHfBCN CMCs is discussed in detail, indicating that the incorporation of Hf and B into SiCN has a significant effect on the performance under hydrothermal conditions.

The presented results might of high relevance within the context of the development of CMCs suitable for petrochemical applications, which are characterized by moderate temperatures (several hundreds of °C) and high hydrothermal pressure.[20]

2. Experimental procedure

2.1. Preparation of ceramic matrix composites (CMC)

A 2D carbon fabric (T300, manufactured by Toray) was used for the preparation of the carbon-fiber-reinforced CMCs. Polysilazane (HTT1800, AZ-EM, Wiesbaden, Germany) and a polysilazane-based SiHfBCN precursor (which was prepared as described in[21]) were considered as preceramic precursors for the PIP fabrication of the composites. The carbon fabric was cut into rectangular pieces with dimension of 44 mm × 16 mm. After performing the infiltration with the preceramic precursor, 16 infiltrated pieces of carbon preform were stacked onto each other within a Teflon mold (Fig. 1a) in a hand lay-up process in order to provide the fiber preform with ca. 3.5 mm thickness. After a curing step, the composites were pyrolyzed in a tube furnace (1100 °C, Ar) to furnish C_f/SiCN and C_f/SiHfBCN-based CMCs (Fig. 1b). The obtained samples were re-infiltrated and pyrolyzed up to 7 times. The specimens were cut and machined into rectangular coupons with dimensions of 44 mm × 3 mm × 3 mm. In order to push down the open porosity, the specimens were re-infiltrated with HTT1800 or SiHfBCN precursor by using one more infiltration prior to the hydrothermal corrosion experiments.

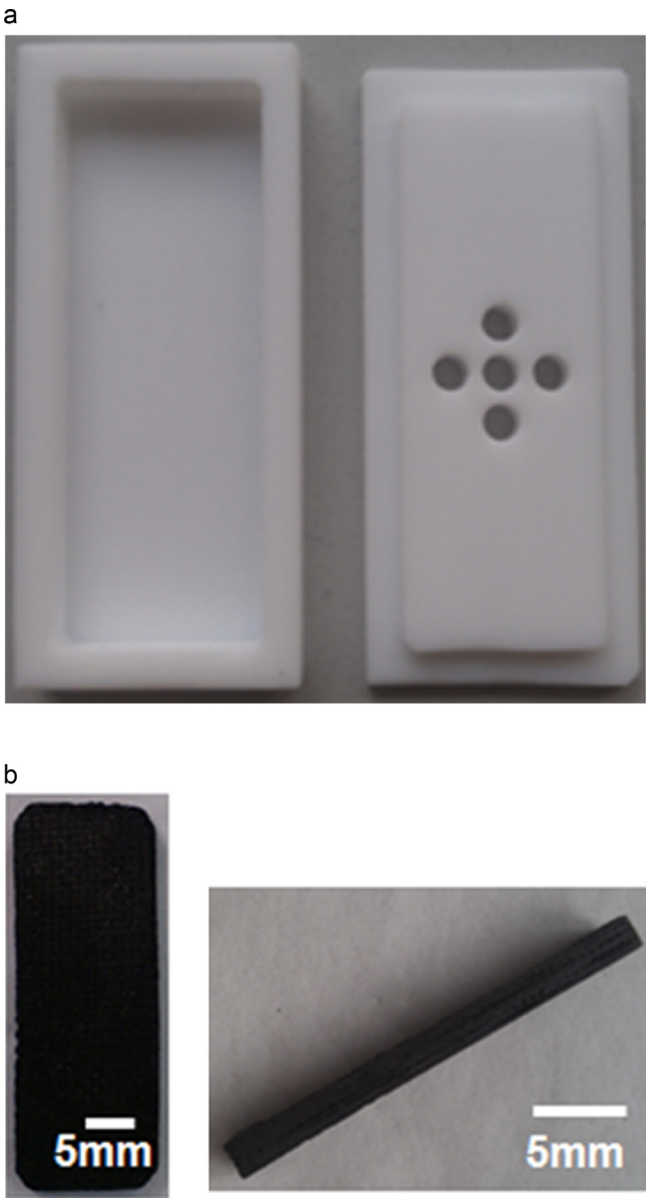

Fig. 1. Photographs of the Teflon mold used for the hand lay-up process (a) and of the prepared C_f/SiHfBCN CMCs (b).

2.2. Hydrothermal corrosion experiments

The hydrothermal corrosion experiments were carried out in closed steel autoclaves with Teflon inlet at temperatures of 150, 200 and 250 °C for 48, 96 and 240 h. The hydrothermal pressure within the autoclaves at the mentioned temperatures was 0.5, 1.5

Table 1
S/V ratios of the hydrothermal experiments performed with the CMCs.

Sample dimensions W × T × L (mm × mm × mm)	Sample Surface (cm^2)	S/V ratio
3*3*20	2.5 ± 0.1	0.4
3*3*8.5	1.2 ± 0.1	0.18
3*3*2.5	0.45 ± 0.05	0.075