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Preparation of carbon fiber-reinforced zirconium carbide matrix composites by reactive melt infiltration at relative low temperature

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An attractive way to prepare carbon fiber-reinforced ZrC matrix composites was proposed and confirmed experimentally. The experimental results showed that the resulting composites were fabricated by immersing carbon/carbon preforms in molten Zr_2Cu at 1200 °C for 3 h. ZrC was the main phase, with a content of 42.2 ± 1.3 vol.%. The composites exhibited excellent ablation resistance when undergoing ablation with an oxyacetylene torch, the mass loss rate and linear recession rate being 0.0006 ± 0.0001 g s^{-1} and 0.0003 ± 0.0001 mm s^{-1}, respectively.
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Zirconium carbide (ZrC), which is a member of the ultrahigh-temperature ceramics family, has a number of unique properties, including an extremely high melting point, high strength and hardness, relatively low density, and high retained strength at high temperatures [1–5], which make it attractive for aerospace applications associated with hypersonic flight and rocket propulsion. However, ZrC ceramics are brittle and display little to no plasticity across a broad temperature range [5,6]. It has been proven in earlier works that introducing carbon fibers is an effective way to improve the fracture resistance of ZrC [5–7].

Known as a rapid and low-cost manufacturing process [5,8–14], the reactive melt infiltration (RMI) process has been widely used to produce structural components with complex geometries made of SiC- [14,15], ZrC- [5,11,16] and HfC-matrix [13] composites. Recently, carbon fiber-reinforced zirconium carbide (C_f/ZrC) composites with porous carbon/carbon (C_f/C) preforms and pure Zr metal have been prepared by this method [5,7]. Due to its high melting point (1850 °C) [17], however, the Zr melt reacts strongly with fibers, which is detrimental to the reinforcement effect. Introducing copper to form Zr_2Cu (melting point 1025 °C) can reduce the processing temperature effectively, and can wet and infiltrate well into a porous preform [2,10,18]. Zr_2Cu instead of Zr has been used as an infiltrator to produce W/ZrC composites at temperatures as low as 1200 °C [11,16,19–21]. However, C_f/ZrC composites fabricated by the RMI process using Zr_2Cu as the infiltrator have not yet been reported.

In this study, an attractive way to prepare C_f/ZrC composites by RMI with Zr_2Cu as a reactive infiltrator is proposed for the first time. The microstructure and composition of the composites were investigated. Furthermore, the ablation properties of the composites were also determined using an oxyacetylene torch.

Felts with a fiber volume fraction of about 30 vol.% (T300, with an average diameter of about 7 μm, a tow size of 3 K, a carbon content of 93% (impurities of O + H + N + S ⩽ 7%) and a bulk density of 1.76 g cm^{-3}; Toray, Japan) were prepared by a needle-punching technique with alternatively stacked 0° weftless piles, short-cut-fiber webs and 90° weftless piles.

The preparation of C_f/C preforms included the following stages: first, phenolic resins (flaxen solid, manufactured by Taihang Co. in Xian) were smashed into powder and dissolved into ethanol with the weight ratio of 1:1; next, the carbon felts fixed by modular molds were located in a hermetic container; then the phenolic resins/ethanol solution were infused into the container and overwhelmed the felts when the container pressure was below 10 Pa. After dwelling there for 6 h, the saturated felts were cured at 80 °C for 4 h and then at 150 °C

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for 3 h. Finally, the cured felts were pyrolyzed at 1200 °C for 60 min in N_2 (purity ⩾ 99.999 vol.%, $O_2 + H_2O$ ⩽ 0.001 vol.%) under atmospheric pressure, with a heating rate of 10–15 °C min^{-1}. Four cycles of infiltration and pyrolysis were necessary to obtain the C_f/C preforms with appropriate porosity.

Commercial Zr_2Cu ingots (purity ⩾ 99.2 wt.%, impurities (Hf + Fe + Ti + Si + Mg + Al + Ni) ⩽ 0.8 wt.%; Hunan Rare Earth Metal & Material Institute, Hunan, China) were used as the reactive infiltrant. The ingots were prepared with spongy Zr pieces (purity ⩾ 99.6 wt.%) and copper cylinders (purity ⩾ 99.99 wt.%) by arc melting. To allow for homogenization, each ingot was flipped and remelted three times.

The preparation of C_f/ZrC composites included the following stages: the Zr_2Cu alloy was rapidly heated up to 1200 °C, the reactor keeping the preforms separated from the alloy during heating. After the alloy had melted completely, the porous C_f/C preforms were mechanically driven into the melt and kept there for 3 h, before being separated from the liquid Zr_2Cu bath and left to cool naturally to room temperature. Any excess solidified melt adhering to the sample was removed by a grinding machining with a diamond wheel.

The porosity of the specimen was equal to one minus the quotient of the bulk density to the theoretical density. The bulk density of the specimen was obtained from measurements of its dry weight and external dimensions. The theoretical densities were calculated using rule of mixtures calculations, based on the theoretical densities of 6.63 g cm^{-3} for ZrC, 1.55 g cm^{-3} for the deposited C, 8.96 g cm^{-3} for Cu, 6.49 g cm^{-3} for Zr and 1.76 g cm^{-3} for the T300 fiber. The volume fractions of the solid phases were measured by inductively coupled plasma (ICP) and chemolysis analysis, which were described previously [22]. The density and open porosity of the specimen was measured using the Archimedes method, with distilled water as the immersing medium, after open porosity was saturated under vacuum for 12 h. The density was measured for at least five samples.

In order to analyze the contents of each phase, the resulting composites were crushed into powder by milling machining with a diamond milling cutter and granulated using a 100-mesh sieve, before being immersed in an acid solution (1 part HCl, 1 part HNO_3 and 2 parts distilled water) for 30 min. When tested by reacting with pure Zr, Cu and ZrC, respectively, this solution can dissolve Zr and Cu but not ZrC. After filtering, the fluid underwent ICP analysis to determine the amount of residual Zr and Cu; the solids left on the filter were dried in an oven at 80 °C for 5 h and then kept in muffle furnace at 1200 °C for 5 h, where the solid powders were oxidized thoroughly into ZrO_2. The amount of ZrC was calculated from the content of ZrO_2, and the amount of the residual carbon (containing carbon matrix and fiber) was equal to the weight difference between the drying powders and ZrC. Since the fiber fraction was a known quantity, the content of the residual carbon matrix could be calculated.

The phases were analyzed by X-ray diffraction (XRD) with a Bruker D8 Advance instrument. The microstructures were observed by scanning electronic microscopy (SEM; Quanta-200) and energy-dispersive spectroscopy (EDS). All specimens for SEM/EDS analyses were prepared by incised, ground and polished with a series of diamond pastes to a surface finish of 0.5 μm. Image analysis software package (ImageJ, National Institutes of Health, Bethesda, MD) was used to determine the average diameters of the fibers in the composites. The average diameter was calculated by measuring 30 fibers per image of at least 10 different SEM images.

The ablation properties were tested with an oxyacetylene flame according to the GJB323A-96 Ablation Standard. The ablation tests were conducted in air, and the pressures of oxygen and acetylene were 0.4 and 0.095 MPa, respectively. The surface temperature of the sample was monitored with an optical pyrometer. During the test, the ablation gun, with a gunpoint diameter of 2 mm, was first ignited. When the fluxes of acetylene and oxygen were turned to 1.116 and 1.512 m^3 h^{-1}, respectively, the gun was moved perpendicular to the sample surface. The distance between the tip of the gun and the top of the sample was 10 mm and the exposure time under the torch flame was 30 s. The heat flux was calculated from the heat absorbed by water flowing through a calorimeter, and was found to be 4200 kW m^{-2} (10% error). At least three samples with dimensions of 30 mm × 30 mm × 3.5 mm were examined in each test. The mass and linear ablation rates (R_m and R_l) were defined as follows:

$$R_m = (m_0 - m_1)/t$$

$$R_l = (l_0 - l_1)/t$$

where m_0 and m_1 represent the weight before and after ablation, respectively; l_0 and l_1 represent the thickness before and after ablation, respectively; and t is the ablation time. In detail, l_1 is the average thickness of seven equidistant points along the longest chord of the ablated region with an approximately circular shape. The overall ablation rates of the composites were taken from three specimens on average.

The reaction between carbon and Zr_2Cu melt can be expressed as Reaction (1). Thermodynamics calculations were conducted in the standard state to estimate the direction of this reaction. The thermodynamics data of the Zr_2Cu phase were calculated using Pandat 7.0 software based on the database optimized by Zeng et al. [23]. The thermodynamics data of the other reactant and products are from Chase [24]. The changes in Gibbs' free energy and enthalpy above the melting point of Zr_2Cu are shown in Figure 1, which reveals that

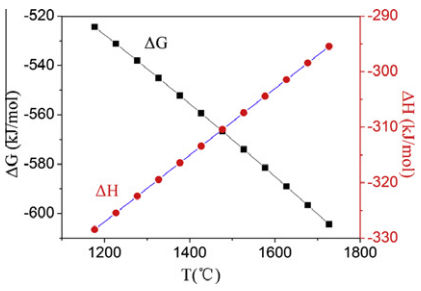

Figure 1. Changes in Gibbs' free energy and enthalpy as a function of temperature above 1200 °C for Reaction (1).

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