



Qualitative analysis of hafnium diboride based ultra high temperature ceramics under oxyacetylene torch testing at temperatures above 2100 °C

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Abstract

Oxidation tests were carried out on HfB₂-SiC, HfB₂-HfC, HfB₂-WC-SiC, and HfB₂-WSi₂ ceramics using an oxyacetylene torch. The samples were oxidized between 2100 and 2300 °C. From cross-sectional images, scale non-adherence was noted as a limiting factor in oxidation resistance. The sample with the best scale adherence was HfB₂-WSi₂. Factors involving scale non-adherence such as vapor pressure, coefficient of thermal expansion mismatch and phase transformations were considered. In comparing the scale adherence of the samples it was hypothesized that vapor pressure buildup is the principal contributing factor in the scale adherence differences observed among the tested samples. However, the coefficient of thermal expansion mismatch and HfO₂ phase transformation cannot be neglected as contributing factors to scale non-adherence in all samples. © 2013 Elsevier Ltd. All rights reserved.

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1. Introduction

Transition metal borides and carbides with melting temperatures exceeding 2700 °C are commonly referred to as ultra high temperature ceramics (UHTCs) and have been studied as primary candidates for extreme environment thermal protection systems such as those found at the sharp leading edges of hypersonic vehicles.^{1,2} Most commonly explored are the ZrB₂-SiC and HfB₂-SiC (MeB₂-SiC) systems with and without various additives. Most oxidation resistance testing of UHTCs has involved either resistive-element furnace heating or arc jet heating. Over the past decade the cost and limited availability of arc jet testing and the temperature and heating rate limitations of furnace heating have led many laboratories to develop new testing methods in order to probe higher temperatures. The first widely reported test, direct electrical resistance, developed

primarily under Halloran,^{3,4} provided insight into the volatile nature of the oxidation products of ZrB₂-SiC at temperatures up to 2100 °C. Observations of mixing between the ZrO₂ and SiO₂ led to a better understanding of the dynamic characteristics occurring at testing temperature. Using this technique, oxidation resistance and mechanical strength retention comparisons between samples with different chemistries can easily and rapidly be examined.^{5,6} The main drawback of this method, the internal heating, has limited its widespread use. Laser testing of UHTC materials has been utilized by researchers to reach temperatures from benign to beyond the melting points of the UHTC materials.^{7,8} Laser heating technology is versatile and more economical than arc jet testing, yet is also not widely available. As such, several laboratories have developed oxyacetylene torch testing as a rapid screening tool for UHTC materials at temperatures up to 3400 °C.^{9–13} The oxyacetylene torch test is naturally ablative owing to the high velocity gas flow associated with the flame. The oxidation characteristics of the oxyacetylene torch can be controlled by the acetylene to oxygen ratio, while at a given ratio temperature is controlled by the distance to the flame. The torch has been used as a standard test method

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Table 1
Sample composition and sintering parameters with sample ID.

Sample ID	Composition	Sintering temp., hold time, pressure
HS	HfB ₂ –20vol.%SiC	2100 °C, 25 min, 32 MPa
HSW	HfB ₂ –20vol.%SiC–4vol.%WC	2100 °C, 15 min, 32 MPa
HH	HfB ₂ –4vol.%HfC	2100 °C, 50 min, 32 MPa
HW	HfB ₂ –4vol.%WSi ₂	1940 °C, 15 min, 32 MPa

for oxyacetylene ablation testing of thermal insulation materials (ASTM E285-08).

As UHTCs have been tested using these various techniques with parameters that include higher temperatures (>1800 °C) and high velocity flow,^{14–19} it has become apparent that there is a point at which the protective SiO₂–MeO₂ scale that is formed on MeB₂–SiC begins to fail. Above a critical temperature, the viscosity of glassy SiO₂ will be too low to remain integral to the scale. Under these conditions the SiO₂ will flow out of the pores of the MeO₂ and from the sample surface resulting in a less protective porous outer scale.¹⁹ Oxide scale non-adherence at temperatures in excess of 2000 °C has also been observed during oxidation testing of MeB₂–SiC materials.^{9,20,21} The loss of scale adherence could be a result of many factors including stress induced by the difference in thermal expansion coefficient (CTE) of the MeB₂ and MeO₂-based layers upon heating/cooling, phase transformation of the MeO₂ and its associated volume increase upon cooling or fracture caused by the escape of gaseous by-products of oxidation such as CO, SiO and B₂O₃.

The present paper focuses on evaluating the relative performance of four different compositions among the HfB₂ containing UHTCs, at temperatures above 2100 °C for long duration, using an oxyacetylene torch. A baseline sample of HfB₂–SiC was tested and compared to W-containing samples. W has been found to be a beneficial additive for both ZrB₂ and HfB₂-based UHTCs when samples are tested under furnace heating up to 2000 °C,^{20,22,23} and as such two chemistries including W, HfB₂–WC–SiC and HfB₂–WSi₂, were tested. A qualitative analysis of the differences in behavior is presented.

2. Materials and methods

Commercially available HfB₂ (Materion, 99.9%, –325 mesh), WC (Materion, 99.5%, –325 mesh), WSi₂ (Materion, 99.5%, –325 mesh), Hf (Materion, 99.8%, –325 mesh), C (Materion, 99.5%, –325 mesh) and β–SiC (Materion, 99.9%, 1 μm) were used to prepare four different sample compositions: HfB₂–20vol.%SiC [HS], HfB₂–20vol.%SiC–4vol.%WC [HSW], HfB₂–4vol.%HfC [HH] and HfB₂–4vol.%WSi₂ [HW]. The powder mixtures were ball milled in isopropanol for 24 h with SiC grinding media, dried at room temperature, and subsequently dry milled for 12 h. Typical weight loss of the SiC grinding media after milling was 0.2 mg (0.2 wt.% of the total batch). The powders were sieved through an 80-mesh (177 μm) screen.

Sample composition and sintering conditions are summarized in Table 1. Milled powders were loaded into a 20 mm diameter graphite die to produce a 13 mm thick cylinder. A layer of BN and graphite foil separated the powder from the die with

the powder in contact with the graphite foil. The powder-filled dies were cold pressed at approximately 50 MPa. The powders were sintered using field assisted sintering (FAS: FCT Systeme GmbH, Model HPD 25-1, Rauenstein Germany) at 2100 °C (HSW, HH, HW) or 1940 °C (HW) for 25 min (HS), 15 min (HSW and HW), or 50 min (HH) under a 32 MPa load. The controlled heating and cooling rates were 50 °C/min. The load was applied during heating to 1600 °C and released on cooling to 1000 °C. The graphite foil was removed from the sample by manual grinding, and the faces of the disks were polished to 45 μm for exposure to the torch.

Samples were oxidized using the oxyacetylene torch apparatus developed at Loughborough University.¹³ Samples were held in place using a carbon–carbon foam insert in a water-cooled graphite holder. The surface of the sample was 25 mm from the exit point of the torch. During the test, the temperature of the exposed face was recorded using a 2 color pyrometer (Marathon MR1SCSF, Raytek GmbH, Berlin, Germany) while the back face temperature was measured by a J-type thermocouple inserted through a hole in the graphite holder to contact the back of the sample. Fig. 1 shows a photograph of the test along with the front face heating profile for the samples. The photograph (Fig. 1a) was taken through welding glasses to limit the brightness. An oxygen rich flame was chosen for testing the UHTC samples. The acetylene to oxygen ratio was 1:1.35 with flow rates of 0.8 m³ h^{–1} and 1.1 m³ h^{–1} respectively. The heat flux was measured at 25 mm using the same acetylene to oxygen flow rates and ratio with a water-cooled gardon gauge (TG1000-54, Vattel Corp., Christiansburg, VA). Oxidized samples were analyzed by X-ray diffraction (XRD: D8 Bruker AXS limited, Coventry, UK), and then mounted in epoxy and cut in half. The cross section of the oxidized face was then polished to 1 μm. The microstructures were characterized using scanning electron microscopy (SEM: Quanta, FEI, Hillsborough, OR) along with energy dispersive spectroscopy (EDS: Pegasus 4000, EDAX, Mahwah, NJ) for elemental analysis. Wavelength dispersive spectroscopy (WDS: SX100, Cameca, France) was used for the chemical analysis of W and Si. Transmission electron microscopy (TEM: Phillips CM200 FEI, Hillsborough, OR) samples were prepared by focused ion beam milling (FIB: DB235, FEI, Hillsborough, OR).

3. Results

3.1. Oxidation test parameters

The oxyacetylene torch test is one that is gaining commonality in use, but is not completely characterized. Sample

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