Materials and Design 145 (2018) 97–107

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Understanding the mechanical properties of novel UHTCMCs through random forest and regression tree analysis

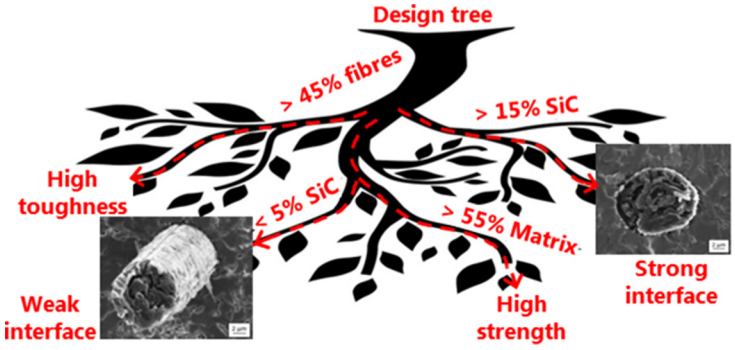
Antonio Vinci [a,*], Luca Zoli [a], Diletta Sciti [a], Cesare Melandri [a], Stefano Guicciardi [b]

[a] CNR-ISTEC, Institute of Science and Technology for Ceramics, Via Granarolo 64, I-48018 Faenza, Italy
[b] CNR-ISMAR, Istituto di Scienze Marine, Largo Fiera della Pesca 2, 60125 Ancona, Italy

HIGHLIGHTS

- The mechanical properties of ZrB_2/SiC-Cf composites were investigated.
- The most important parameter is the ratio between SiC and fibre content.
- High strength and toughness are achieved for high SiC/fibre ratio.
- Higher SiC contents promote a stronger fibre/matrix interface but limit pull-out.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 January 2018
Received in revised form 20 February 2018
Accepted 21 February 2018
Available online 23 February 2018

Keywords:
Ceramic-Matrix Composites (CMCs)
Mechanical properties
Statistical methods
Ultra-High-Temperature-Ceramics (UHTCs)
Fibre-matrix interface
Zirconium diboride

ABSTRACT

The microstructure and the mechanical properties of ZrB_2–SiC–Cf ceramic composites were investigated. The SiC phase, whose amount was varied from 5 to 20 vol%, was introduced in order to improve the densification, oxidation resistance and mechanical properties of the composite. The microstructure was analysed by SEM-EDS and image analysis. Increasing the amount of SiC from 5 to 20 vol% resulted in an improvement of the materials density, from 90% to 94%. The non-brittle 4-pt flexural strength ranged from 164 to 247 MPa, with no clear dependence on the amount of SiC added. The same holds true for the fracture toughness, ranging from 4.8 to 8.4 $MPa \cdot m^{0.5}$. In order to track the most important microstructural parameters affecting the properties, experimental data were analysed with the Random Forest and Regression Tree statistical models. The statistical analysis demonstrated that among the possible explanatory variables such as porosity, amount of SiC, fibre content, matrix content, SiC/fibre amount ratio, the one having a major influence on both the flexural strength and fracture toughness is the ratio between SiC and carbon fibre content.

1. Introduction

Ultra-High-Temperature-Ceramics (UHTCs), such as transition metal diborides, are a novel class of materials characterized by melting points exceeding 3000 °C, high thermal and electrical conductivity and good ablation resistance [1]. Among UHTCs, ZrB_2-based ceramics have been investigated as potential candidates for the fabrication of reusable Thermal Protection Systems (TPS) for aerospace applications owing to

* Corresponding author.
E-mail addresses: antonio.vinci@istec.cnr.it (A. Vinci), luca.zoli@istec.cnr.it (L. Zoli), diletta.sciti@istec.cnr.it (D. Sciti), cesare.melandri@istec.cnr.it (C. Melandri), stefano.guicciardi@an.ismar.cnr.it (S. Guicciardi).

https://doi.org/10.1016/j.matdes.2018.02.061
0264-1275/

their high thermal conductivity and relatively low density [1,2]. However, their low fracture toughness and poor thermal shock resistance pose major obstacles to their implementation [3]. Moreover, the oxidation resistance of ZrB_2 is low at temperatures above 1200 °C due to the formation of volatile oxides (B_2O_3) that leave behind a non-protective porous scale of ZrO_2 [4]. The addition of silicon carbide up to 30 vol% has been found to aid the sintering of ZrB_2 and to increase its oxidation resistance due to the formation of a viscous borosilicate glass with low vapour pressure [5–8]. Common routes to the sintering of UHTCs are hot pressing (HP), pressure-less sintering (PS), spark plasma sintering (SPS) and reactive hot pressing (RHP) [9,10]. Even though pressure-less sintering is an attractive route for the fabrication of more complex shapes, hot pressing remains the preferred route because of the high relative density of the final composite. The materials currently used in aerospace environments, based on C/C, SiC/SiC or C/SiC ceramic composites, display excellent thermomechanical properties but carbon fibres start oxidizing at temperatures as low as 500 °C, while SiC becomes chemically active above 1650 °C, resulting in high ablation rates and the sublimation of the ceramic matrix. By combining the good ablation resistance of a UHTC matrix and the high damage tolerance provided by carbon fibres, the above mentioned limits could be overcome.

Previous works done by Corral et al. [11] on UHTC coatings demonstrated the increase of ablation and oxidation resistance of C/C composites, while Tang et al. [11,12] achieved increased oxidation resistance by embedding 4–18 vol% of ZrB_2 particles in a C/SiC composite due to the synergy between B_2O_3 and SiO_2 that form a protective borosilicate glass.

In the past few years, a new class of materials labelled UHTCMCs (Ultra High Temperature Ceramic Matrix Composites) has been developed that consists of a UHTC rich matrix reinforced with carbon fibres. The main limitation lies in the difficult infiltration of long fibres with the ceramic powders suspensions and the densification of the final composite. Sciti et al. [13–15] successfully sintered UHTCMCs without the aid of expensive techniques such as CVI or PIP, by employing slurry infiltration coupled with vacuum bagging and hot pressing. Preliminary studies on the oxidation behaviour of these composites have shown their promising oxidation resistance up to 1500 °C [16], but no mechanical characterization has been carried out.

In this work, carbon fibre reinforced ZrB_2/SiC composites were fabricated by slurry infiltration and hot pressing. SiC was added in amounts ranging from 5 to 20 vol%, while carbon fibre content was varied between 35 and 50 vol%. Due to process limitations, such as powder suspension rheology, it was difficult to adjust the fibre content for some compositions. Pitch fibres were chosen on the basis of previous studies by Silvestroni et al. [17] that showed the different reactivity of fibres with the surrounding ceramic matrix. The aim of this work is to investigate the mechanical behaviour of ZrB_2/SiC composites and identify the key parameters affecting mechanical properties. Two machine learning techniques, Random Forest and Regression Tree, were employed in order to rank the key explanatory variables and quantify their effects on the mechanical properties [18]. These techniques are widely applied in several fields like ecology [19–22], medicine [23–26] or sociology [27–30]. Few examples can be also found in material science [31,32].

2. Experimental

2.1. Materials

Commercially available powders were used for the fabrication of ceramic composite materials: ZrB_2 (H.C. Starck, grade B, Germany, specific surface area 1.0 m^2/g, particle size range 0.5–6 μm, impurities (wt%): 0.25C, 2 O, 0.25 N, 0.1 Fe, 0.2 Hf), α-SiC (H.C. Starck, Grade UF-25, Germany, specific surface area 23–26 m^2/g, D50 0.45 μm Italian retailer: Metalchimica). Unidirectional high modulus carbon fibres (Granoch Yarn XN80-6K fibres; tensile modulus of 780 GPa and tensile strength 3.4 GPa, 10 μm diameter. Supplier: Angeloni) were used as carbon preforms.

2.2. Process

Powder mixtures containing ZrB_2 and SiC ranging from 5 to 20 vol% were prepared by wet ball milling of the commercial powders and then dried with a rotary evaporator. The composites were prepared through slurry infiltration of unidirectional carbon fibre preforms and hand lay-up in a 0–90° configuration. Hot pressing cycles were carried out at 1900 °C, using a pressure of 40 MPa and a holding time of 10 min, on the basis of previous studies [13]. For each composition, three variants, a, b and c, were fabricated by adjusting the powder suspension rheology according to three fixed values labelled as a,b,c (not explicitly indicated in this work). In order to obtain different contents of carbon fibres. Due to process limitations, it was not possible to obtain fibre contents higher than 40% for some compositions as shown in Table 1.

2.3. Microstructure analysis

The microstructure was analysed on polished and fracture surfaces by field emission scanning electron microscopy (FE-SEM, Carl Zeiss Sigma NTS Gmbh Öberkochen, Germany) and energy dispersive X-ray spectroscopy (EDS, INCA Energy 300, Oxford instruments, UK). For each composition, three pellets (variants) were prepared by varying the fibre content from 35 to 50% and maintaining the hot pressing parameters unchanged. Samples were prepared for microscopy by cutting cross sections, mounting them in epoxy resin, and then polishing down to a 0.25 μm finish with diamond abrasives, using semi-automatic polishing machine (Tegramin-25, Struers, Italy). The polished samples were then washed with ethanol in an ultrasonic bath, dried under IR light and cleaned with a plasma cleaner (Colibrì Plasma RF 50 kHz, Gambetti, Italy) at 40 W for 5 min. The starting compositions (e.g. the

Table 1
Flexural strength (mean ± 1 standard deviation) and physical characteristics of samples ZS5, ZS10, ZS15, ZS20.

	σ (MPa)	Density (g/cm^3)	Porosity (vol%)	Fibre (vol%)	Matrix (vol%)	SiC/fibre Ratio	SiC vol% in matrix
ZS5 a	198 ± 12	3.649	12.9	40.7	46.4	0.123	5
ZS5 b	207 ± 24	4.194	7.2	35.2	57.6	0.142	5
ZS5 c	247 ± 14	4.055	9.5	35.4	55.2	0.141	5
ZS10 a	218 ± 30	3.616	9.5	45.2	45.3	0.221	10
ZS10 b	171 ± 1	4.205	4.1	37.7	58.2	0.265	10
ZS10 c	196 ± 28	3.864	9.8	37.8	52.3	0.264	10
ZS15 a	190 ± 33	3.315	10.2	50.8	39.0	0.295	15
ZS15 b	167 ± 4	3.617	6.4	48.2	45.3	0.311	15
ZS15 c	186 ± 49	3.952	7.5	36.9	55.6	0.406	15
ZS20 a	221 ± 19	3.391	8.7	49.3	42.0	0.405	20
ZS20 b	247 ± 12	3.762	6.0	42.7	51.3	0.468	20
ZS20 c	164 ± 2	4.003	5.4	36.6	58.1	0.547	20