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Experimental investigation into the crack propagation in multiphase tantalum carbide ceramics

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ABSTRACT

Tantalum carbide ceramics with high volume fractions of the ζ-Ta_4C_3 phase have been shown to exhibit high fracture strength and toughness as compared to those in absence of this phase. In this work, we investigated how microcracks propagated in this these high toughness ceramics using Knoop and Vickers microindentation. The Knoop indentations demonstrated that cracking preferentially occurred parallel to the lath structure in ζ-Ta_4C_3; however shorter cracks did form between the laths when a sufficient driving force was present. The resulting crack path was tortuous providing direct evidence for toughening through crack deflection; however, the microscale nature of the work cannot rule out crack bridging as a toughening mechanism as well. Plasticity is also observed under the indents, but is likely a result of the high confining pressures that occurred during indentation allowing for plastic flow.

1. Introduction

Ultra high temperature ceramics (UHTC) from the transition metal group IVB and VB carbides, borides, and nitrides are characterized by chemical inertness, high hardness, moderate oxidation resistance, high temperature strength retention, and high melting points [1]. In addition, these materials can exhibit high fracture toughness, especially when multiphase microstructures are present [2–11]. Recently, high volume fractions of ζ-Ta_4C_3 in a tantalum carbide matrix has shown extraordinary high fracture toughness values [3,4] which maybe the highest reported outside those of the transformation toughened ceramic systems [12,13]. To date, the toughening mechanisms, particularly the crack pathways, of the tantalum carbides have yet to be fully elucidated and are the subject of this paper.

The phases of tantalum carbides are categorized by their carbon content relative to the stoichiometric TaC phase. The γ-TaC phase has a B1, or rock-salt structure, where tantalum atoms are arranged in a face-centered-cubic lattice with carbon atoms occupying the octahedral interstices. For sufficiently carbon deficient compositions, the rhombohedral ζ-Ta_4C_3 and/or the trigonal Ta_2C phases can form [14]. The Ta_4C_3 phase precipitates out as nanoscale laths in the TaC phase forming a crisscross pattern [15,16]. This results in this phase maintaining a close-packed plane orientation relationship with the TaC matrix. Since TaC has four variants of the {111} planes, an intersecting pattern of Ta_4C_3 laths results [5]. If Ta_4C_3 precipitates out of the Ta_2C grains, the laths are parallel to one another because Ta_2C has only one variant of close-packed planes, i.e. {0001} [5].

Prior research on tantalum carbides has shown that variations in microstructures, such as grain size, morphology, and phase content, significantly alter the fracture strength from 200 MPa to 800 MPa and the fracture toughness values ranging between 2 MPa√m to 15 MPa√m [3,4,17]. The unusually high fracture strength and toughness is associated with compositions near the Ta_4C_3 phase field, with its corresponding lath based microstructure, irrespective of the fabrication process – vacuum plasma spraying, reaction based hot isostatic pressing, reaction hot pressing and spark plasma sintering. Recently, Sygnatowicz et al. [17] reported the rising-crack-growth resistance or R-curve behavior for a series of C/Ta ratios that spanned 0.5–0.7. Their results revealed materials with a high fraction of Ta_4C_3 had shorter initial crack lengths, higher crack-extension resistance, and a greater R-curve slope. Using scanning electron microscopy (SEM), they noted the fracture behavior broke along the weak Ta_4C_3 lamellae interfaces and

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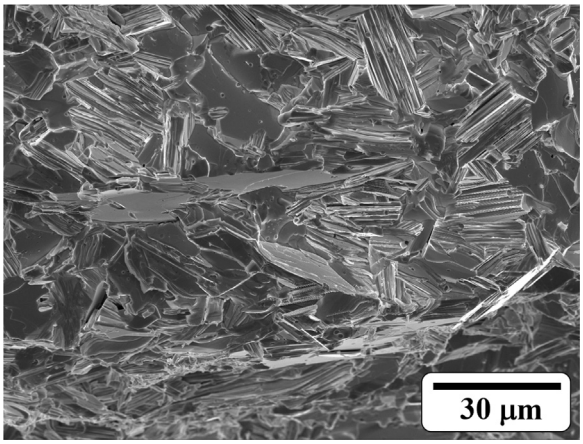

Fig. 1. Secondary electron SEM micrograph of a K_{IC} tested fractured surface. Note the complex cracking within the microstructure evident by cleaved grains and lamella within the grains.

suggested that lamellae bridging plays an important role in the crack growth. Fig. 1 is a SEM micrograph of a fracture surface of a similarly high volume fraction Ta_4C_3 carbide taken from the K_{IC} tested material for this study (with the details to be forthcoming). The micrograph reveals a multitude of cleaved surfaces with fractured lamellar features within each grain. The polycrystalline microstructure with clear secondary phase lamella in the grains has created a complex distribution of the load within the material from the bulk test. There is no clear singular transgranular or intergranular crack path that can be deciphered from the fractography surface. Given that crack propagation behavior has the potential to influence fracture toughness through processes like crack deflection, understanding the microscopic origins of crack propagation within these grains is critical in understanding the potential toughening mechanisms in these materials. By using indentation, where the load is more localized and can be limited to a small number of grains as well as orientated to a specific direction of the grain's morphology, these microstructural insights can then be ascertained, as done in prior studies where either the fracture mechanism [18–21] and/or fracture anisotropy [22–24] in the ceramic was being investigated. In this paper we further prior bulk testing investigations of Ta_4C_3-rich microstructures by studying how cracks propagate within these materials under indentation using electron microscopy.

2. Experimental procedure

2.1. Specimen fabrication

The specimen studied was spark plasma sintered (SPS) using feedstock powder that was plasma melted and rapidly cooled from mechanically mixed Ta and TaC powders. The initial powder mix had an overall 0.67 carbon-to-tantalum ratio or sometimes denoted in the literature as $TaC_{0.67}$ [3,4]. This composition should yield a significant volume fraction of the Ta_4C_3 phase. The plasma-melted feedstock powders experienced an approximately 0.05–0.2 wt% loss because of carbon and powder carry-over. The melted button was crushed into powder which was subsequently SPS'ed at 2100 °C and 10 MPa for 1 h at an approximately 100 °C/min heating ramp rate. After sintering, the specimens were machined into mechanical test bars through electrical discharge machining (EDM) and electro-chemical mechanical polishing techniques.

2.2. Mechanical properties

The fracture toughness of each of the selected composition were determined using the (single edge notch beam method) SENB [25,26] and specifically, the SEVNB (single edge V-notched beam) methods [27]. The bar samples were prepared according to the ASTM C1161 standard with the bar sample being the standard ASTM C1161 size B flexural specimen with a ~0.20 mm wide machined notch that was 0.15–0.20 mm deep. The root of the machined notch is extended 0.10–0.20 mm deeper by polishing a V-notch with a razor blade using diamond paste as the abrasive. For the SENB, the bar samples were notched using a diamond blade with 0.18 mm blade thickness. The widths and depths of the notches ranged 0.20–0.25 mm and 0.20–0.22 mm, respectively. These tests were performed and averaged over three (3) specimens each to verify that the sample to be indented had a high K_{IC} similar to those reported in the introduction.

2.3. Deformation and crack propagation

Deformation and crack propagation studies were conducted at room temperature using two different hardness indenters. A Buehler Hardness Tester 1600–2007 with a Vickers tip at a load of 1000 g and a Wilson model MO hardness tester with a Knoop K-409 tip at a 5000 g load were applied. The Vicker's indent provide a load over a much smaller area where one would anticipate such an impression readily nucleates dislocations; in such a load condition, one can ascertain the material's plasticity capability to nucleate dislocations. In contrast, the Knoop indent, which is more commonly used in ceramic or brittle testing, places the load over a larger area to reduce the potential for catastrophic failure. The anisotropic nature of the Knoop indenter allowed us to align it relative to the microstructure feature from which we are able to ascertain the load direction and corresponding crack propagation to the grains and Ta_4C_3 lamella, a result that would not be attainable using Vickers, and a major emphasis of this paper. Both testers generated cracks in the specimens; however, several subtle differences in the microstructure response as a function of indenter type occurred and will be discussed later in this paper.

2.4. Microstructure characterization

Transverse and longitudinal specimens were cut from the SPS consolidated material and then mounted and mechanically polished using 3 µm diamond paste with a further polish for 24 h in aqueous 0.05 µm silica slurry using a Vibromet. Surface imaging was conducted with a FEI Quanta 3D dual focus ion beam (FIB) – scanning electron microscope (SEM). Both ion and electron imaging was used to capture the indented materials' surface structure. The dual beam FIB also allowed the material, with a crack, to be serial sectioned to reveal the 3D crack propagation within the volume [28]. Micrograph images were collected in an ion contrast imaging mode at 30 keV and 2.5 nA while serial section cutting at 100 nm slice thicknesses were done at 30 keV and 1 nA using an automated script program [29].

The phase and volume fraction analysis was done by X-ray diffraction (XRD) using a Bruker D8 discovery general area diffraction detector system (GADDS) with Cu-K_α radiation at settings of 45 keV and 40 mA. Prior to the XRD analysis, the specimen was crushed and ground into a powder using a mortar and pestle which ensured a random texture and sufficiently large sampling volume from the powder particulates. The collected peaks were compared with the lattice structure data in the international center for diffraction data (ICDD) [30,31]. The volume fraction of the phases were then estimated using Rietveld refinement [32] and found to be Ta_4C_3 70.6 ± 0.0 vol%; TaC 7.7 ± 1.1 vol%; and Ta_2C 21.7 ± 2.4 vol%.

Transmission electron microscopy (TEM) specimens were prepared using a FIB milling technique which allowed for site specific extraction and thinning. The FIB'ed TEM specimens were imaged in a FEI F20 Tecnai (scanning) transmission electron microscope ((*S*)TEM) operated at 200 keV and a CM200 TEM microscope at 200 keV.