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(Ta,Nb)C composites formed with graphene nanoplatelets by spark plasma sintering

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ABSTRACT

TaC-NbC with the addition of sintering additive (5 vol.% B_4C or 5 vol.% Si) and 3 vol.% Graphene Nanoplatelets (GNP) are consolidated by spark plasma sintering (SPS). GNP are aligned in a uniformly oriented direction perpendicular to the processing axis of the SPS equipment during consolidation. High load instrumented indentation is performed and the projected area of residual damage is compared to estimate relative fracture toughness. The projected residual damaged area after indentation in the surface direction (out-of-plane GNP orientation) was 89% greater in the TaC-NbC-5B_4C-3GNP sample, and 96% greater in the TaC-NbC-5Si-3GNP sample when compared to indentation in the orthogonal (in-plane GNP orientation) direction. The toughening mechanisms prevalent in the orthogonal indentation direction (crack bridging and crack deflection) result in greater energy dissipation than the prevalent mechanisms in the surface indentation orientation (sheet sliding, crack bridging, and crack arrest).
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1. Introduction

Ultra high temperature ceramics (UHTC) have long been considered candidate materials for high temperature aerospace applications. Among the UHTC materials, group IV and V carbides and borides are characterized as having high melting points (>3000 °C), exceptional refractory properties, and high strength. These qualities make them candidate materials for leading edges of hypersonic aircrafts which experience high temperatures due to enormous drag forces as well as nozzle inserts for rocket propulsion systems. In this study, a pre-alloyed Tantalum-Niobium carbide (TaC-NbC) made up of 80 wt.% TaC and 20 wt.% NbC was used. Both TaC and NbC have high resistivity to chemical erosion and show promise for use in non-oxidizing environments such as throat inserts for aluminum burning rockets. One of the biggest advantages of NbC addition is that the density is almost half that of TaC (7.78 g/cm^3 vs 14.50 g/cm^3). Because this system is being considered for high temperature aerospace applications, the reduction in weight due to the decrease in the density will make this system more viable and cost effective. Additionally, the 80-20 solid solution was commercially available for purchase through the manufacturer.

The strengths of UHTC materials are dependent on the density and final grain size. According to the Hall-Petch relationship, the yield strength increases as the inverse square of the grain size [1]. Due to the strong covalent bonds, low self-diffusion coefficients, and high melting temperatures of UHTC materials, achieving full densification is difficult. Consolidation methods previously used include conventional hot pressing [2,3], pressureless sintering [4,5], and high frequency induction heating [6–8]. While these methods have reached a final relative densification of up to 97.5%, they use higher temperatures (2100–2300 °C) and longer dwell times (30 min up to several hours) which can result in large grain growth. Consolidation by spark plasma sintering (SPS) has achieved full relative densification at a temperature of 1800–1900 °C and a dwell time of just 10 min [9–11].

Another route to increase densification during sintering is to use sintering additives. The use of additives such as TaB_2, $TaSi_2$, $MoSi_2$, SiC, B_4C, and Si has been shown to improve not only the densification of the sintered compact but also the mechanical properties [2,3,8]. The use of sintering additives (B_4C and Si) to enhance densification of TaC-NbC by SPS was studied and previously reported [12]. Nearly full relative densification (>99%) of TaC-NbC was achieved with a hold time as short as 3 min at 1850 °C.

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Even with full densification, the use of UHTC materials for aerospace applications is hampered by the material's low fracture toughness. During use, the materials would be subjected to immense vibrations as well as shock waves and possible impacts. All of these occurrences can cause cracking which could lead to failure.

Previous studies have demonstrated that the addition of GNP can increase the fracture toughness of a ceramic composite [13–17]. Nieto et al. [18] prepared a 5 vol.% graphene platelet reinforced tantalum carbide composite and reported a fracture toughness increase of 99% over the monolith. In our previous study [19], we stated that the increased toughness in composites reinforced with GNP can be attributed to three different regimes; (i) property changes during processing, (ii) increased load capacity prior to initial crack propagation, and (iii) crack propagation suppression mechanisms. Examples of regime (i) include increased densification and grain wrapping, which can inhibit grain growth during sintering [20,21]. Examples of regime (ii) include stress-shielding, sheet pull-out, bending, kinking, and sliding which all dissipate energy that would otherwise lead to crack propagation [22–24]. Regime (iii) toughening mechanisms would include crack bridging and crack deflection after initial propagation leading to mixed mode fracture [25,26]. While these toughening benefits have been observed previously, they have not been reported with respect to the orientation of the GNP in the composite which affects the overall toughening due to variation in loading on GNP. In addition, anisotropy of the GNP mechanical properties can also increase anisotropy in the composite. Our previous work studied the prevalent energy dissipation mechanisms of regime (ii) with respect to orientation by performing in-situ indentation on bulk graphene nanoplatelets in both the in-plane and out-of-plane directions [19]. When indenting in the orthogonal direction, the prevalent mechanisms included compressive reinforcement, bending, push-out, and pop-out while the prevalent mechanisms observed in the surface indentation direction included sliding, bending, kinking, and pull-out.

The motive of this study is to gain an understanding of the anisotropy of the GNP reinforced ceramic composite resulting from the anisotropy of the toughening mechanisms of GNP. This will be achieved using a combination of instrumented indentations and in situ imaging using an electron microscope.

2. Experimental procedure

2.1. Powder preparation

Tantalum-Niobium Carbide powder (80 wt.% TaC, 20 wt.% NbC; $(Ta_{0.67}Nb_{0.33})C$) was purchased pre-alloyed from the manufacturer listed in Table 1. 5 vol.% Boron carbide (B_4C) nanopowder and 5 vol.% Silicon (Si) nanoparticles were the chosen sintering additives based on their high temperature properties when considering possible secondary phases formed during the sintering process [12]. 3 vol.% graphene nanoplatelets (xGNP-M-5) with a thickness between 6 and 8 nm, an average diameter of 5 μm, and a typical surface area of 120–150 m^2/g were used to increase the toughening of the composite.

Each composite powder was prepared using tip-sonication of the mixture in acetone for the sample powders prepared with GNP and B_4C with GNP and in Methanol for the sample powder prepared with Si with GNP. The powders were split into three 600 mL beakers and filled to 450 mL with their respective solutions and tip-sonicated (Model VCX750, Sonics & Materials, Inc., Newtown CT) using a ¾ inch high-gain probe at 100% frequency for 45 min each. The mixtures were then dried overnight at 80 °C. The powders were crushed using an agate mortar and pestle and dried for an additional 4 h to ensure no liquid remained.

2.2. Consolidation by spark plasma sintering

Spark plasma sintering (SPS) (Model 10-4, GT Advanced Technologies, Santa Rosa, CA) was used to consolidate the prepared powders. The powders were loaded into a graphite die that was covered in 4 layers of graphite felt in order to provide thermal insulation. SPS was performed in a vacuum atmosphere (pressure $<2 \times 10^{-2}$ Torr) at 1850 °C and 60 MPa with a heating rate of 100 °C/min and a hold time of 10 min for the TaC-NbC-3GNP and TaC-NbC-5B_4C-3GNP samples and a hold time of 3 min for the TaC-NbC-5Si-3GNP sample. The difference in hold time for the TaC-NbC-5Si-3GNP sample was outlined in our previous study as to reduce the occurrence of grain growth [12]. The temperature of the sample during sintering was measured by pyrometer focused on a hole drilled halfway through the graphite die where the powder was being sintered for improved accuracy. At the end of the hold, the power is shut off, allowing for the die and sample to cool. The sintered samples were cylindrical in shape with a thickness of approximately 5 mm and a diameter of 20 mm.

2.3. Microstructure characterization

Sintered samples were ground using SiC paper to remove graphite that surrounded the surfaces as a result of SPS processing. Apparent bulk density measurements were carried out using the water immersion technique which employs Archimedes principle as well as using a Helium gas pycnometer (Accupyc 1340, Micromeretics Instrument Corporation, Norcross, GA). The relative density was calculated as a percentage of true density, which was measured for each starting powder mixture using the Helium gas pycnometer. Phase components of the sintered samples were analyzed using X-ray diffraction (XRD) (Bruker D5000) using Cu-Kα X-rays at a scan rate of 0.5°/min. and an SEM equipped with a field emission gun and energy dispersive X-ray spectroscopy (EDS) (JSM6330F, JEOL USA, Inc., Peabody, MA). Fracture surfaces were prepared by cutting off a 5 mm section of each sample using a low speed diamond saw and then fracturing the cut sections using a hammer strike. The cross-sections and surfaces of the remaining samples were then ground using diamond paper to the 15 μm level followed by polishing using a diamond suspension to a 0.25 μm finish. The morphologies of the powders and fracture surfaces were analyzed using a SEM after being gold-coated for greater electrical conductivity and image quality.

2.4. Mechanical property characterization

Vickers hardness was measured by indenting on the polished cross-sections and top surfaces of the sintered samples. Indentations were carried out using a microhardness tester (Model LM810AT, LECO Corp., St. Joseph, MI) at a load of 1000 g with a dwell time of 10 s. A total of 8 indents were performed on each sample and the average and standard deviation are reported. Instrumented indentation was performed using a linear, screw driven micro-load frame (SEMtester 1000, MTI Instruments Inc., Albany, NY). The micro-load frame has a load capacity of 4500 N with an accuracy of 0.2% and a linear movement resolution of 20 nm. The load frame is controlled using an interface and MTESTQuattro software (ADMET, Norwood, MA) which also outputs the load-displacement data. Indentation was performed with an approximately 1 μm, 120° conospherical diamond tip loaded across from the prepared sample. The indentations were carried out using load control with a loading function of 100 N/min. up to 500 N followed by a dwell of 5 min at 500 N load before being unloaded at 100 N/min. The

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