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Graphene nanoplatelet reinforced boron carbide composites with high electrical and thermal conductivity

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ABSTRACT

Monolithic B₄C ceramics suffer from poor machinability, high brittleness and low thermal conductivity. Here we intend to tackle these problems by incorporating graphene nanoplatelet (GNP) into B₄C matrix. Dense B₄C/GNPs composites containing 0–5 vol% GNPs were fabricated by hot-pressing using Ti₃AlC₂ as sintering aid. The electrical conductivity increased dramatically with the incorporation of GNPs, enhancing the machinability of B₄C composite remarkably by allowing electrical discharge machining. The establishment of a conducting network was revealed by conducting scanning force microscopy and a synergistic enhancement effect by conducting TiB₂ particles produced by reactions between B₄C and Ti₃AlC₂ and GNPs was responsible for the low percolation threshold. The GNPs also provided a good thermal transport channel and the thermal conductivity perpendicular to hot-pressing direction was significantly enhanced. Improvements on strength and fracture toughness by addition of GNPs were observed, and the mechanisms for the improvements include crack deflection and pull-out of GNPs.

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1. Introduction

The unique combination of light weight and extremely high hardness makes boron carbide (B₄C) an attractive material for multiple applications in wear resistant components, aerospace craft and light weight armors [1–3]. B₄C is also extensively used in nuclear reactors as control rods, shielding materials and as neutron detectors due to the neutron absorption ability of isotope ¹⁰B which owns a high neutron absorption cross-section over a wide range of neutron energies. Currently, they are the leading control rod materials in liquid-metal-cooled fast breeder reactors (LMFBR) and boiling water reactors (BWR) [3].

However, for practical applications, B₄C ceramics have several limitations such as poor machinability, low strength and toughness as well as inferior thermal conductivity and thermal shock resistance. It is challenging and costly to machine B₄C ceramics into intricate parts using traditional cutting techniques owing to their extremely high hardness and brittleness. Their relatively low strength and fracture toughness also limit the wider applications. In addition, B₄C ceramics suffer from the intrinsic low thermal conductivity and poor thermal shock resistance. As nuclear controlling

rods, B₄C ceramics easily crack due to thermal stress and helium gas during neutron irradiation, which will lead to the failure of the controlling rods and raise serious safety problems [4].

Many attempts have been made to overcome those obstacles by fabricating B₄C composites. In order to enhance the machinability, nano-sized hexagonal boron nitride (h-BN) has been incorporated into B₄C matrix [5]. However, the h-BN is detrimental to the hardness of B₄C ceramics and the B₄C/h-BN composites exhibit much lower Vickers hardness compared with monolithic B₄C ceramic [5]. Another way to enhance the machinability of hard ceramics without jeopardizing their unique hardness is to enable electrical discharge machining (EDM). EDM is an attractive alternative machining technique which realizes the low cost and highly flexible manufacture of hard ceramics, but a certain electrical conductivity is required [6,7]. Although B₄C is a semiconductor, its electrical conductivity does not suffice to enable the EDM. Consequently, it is intriguing to improve the electrical conductivity of B₄C ceramics for the utilization of EDM, through which the machinability of B₄C ceramics can be greatly enhanced with their hardness being well preserved. It has been reported that the electrical conductivity of B₄C ceramics could be increased significantly by adding over 25 vol% conductive TiB₂ particles [8]. While such a large volume fraction of TiB₂ phase is detrimental to both the light weight and mechanical properties of B₄C ceramics. For the enhancement of strength and toughness of B₄C ceramics, TiB₂, ZrB₂, SiC and Al₂O₃

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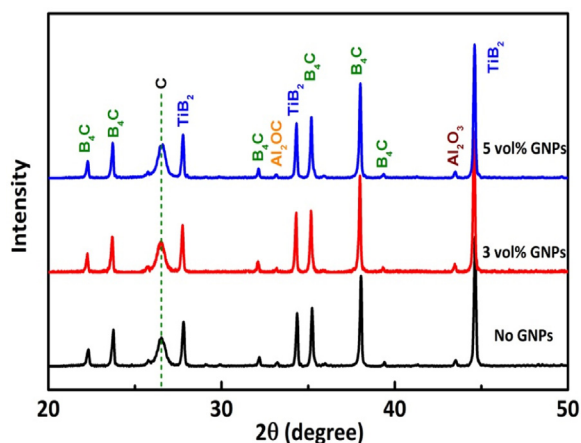


Fig. 1. XRD patterns of B_4C /GNPs composites with different amount of GNPs.

are commonly chosen as reinforcement phases and considerably high strength and toughness were obtained [1]. There are only few studies concerning the enhancement of thermal conductivity of B_4C by forming B_4C /copper cermet [9]. Although considerable improvement on thermal conductivity was achieved, the large volume fraction of copper and the low density of the cermet lead to serious degradation of the mechanical properties. In a word, the machinability, mechanical and thermal properties of B_4C ceramics can only be partially improved by fabricating traditional B_4C composites.

Carbon-based nanomaterials such as carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) have attracted tremendous attentions since their discovery. They exhibit superior properties such as high aspect ratios, outstanding mechanical properties as well as exceptional carrier mobility and thermal conductivity [10–15]. Consequently, they are becoming the most attractive

nanofillers for tougher polymer/ceramic composites with higher electrical and thermal conductivity [16–34]. Reasonably low CNTs content was reported to be sufficient for the transition from an insulating polymer/ceramic matrix to a conducting composite [16,17]. However, the poor dispersability of CNTs within the polymer/ceramic matrices due to their tendency to aggregate into bundles makes the homogeneous dispersion of CNTs a big challenge [35]. Moreover, the toughening effect by CNTs addition in ceramic composites is still under debate and the thermal conduction enhancements associated to the presence of CNTs seems more subtle [36–38]. Compared with CNTs, GNPs which refer to stacks of graphene layers with thickness in the range of few nanometres to tens of nanometres seem more appealing [39]. GNPs can be achieved by mechanical or chemical exfoliations from pristine graphite for large scale production, thus GNPs are more cost-effective compared to CNTs [15,39,40]. More importantly, GNPs exhibit a better dispersability in ceramic matrix and a higher resistance to structural damage at elevated temperatures and consequently they are more favourable for high-temperature ceramic matrix [24]. GNPs provide reinforcement in two directions and are expected to be more efficient in improving mechanical properties than unidirectional CNTs. In addition, it has been demonstrated GNPs provide a higher thermal transport enhancement than CNTs in polymer composites owing to the reduced thermal interface resistances [22,41]. There are a number of reports on GNPs/ceramic composites and most of them focused on the mechanical reinforcement by GNPs. Extraordinary toughening enhancement is demonstrated in Si_3N_4 , ZrO_2 , SiO_2 , and Al_2O_3 ceramic matrices by adding a small amount of GNPs [24,28,42,43]. Crack deflection, crack bridging by GNPs and pull-out of GNPs were identified as the main toughening mechanisms. Considerable papers focusing on the electrical properties of GNPs/ceramic composites have also been published in recent years [26–32]. The incorporation of certain amount of GNPs into insulating Al_2O_3 and Si_3N_4 matrices gives rise to increments of up to 14 orders of magnitude in

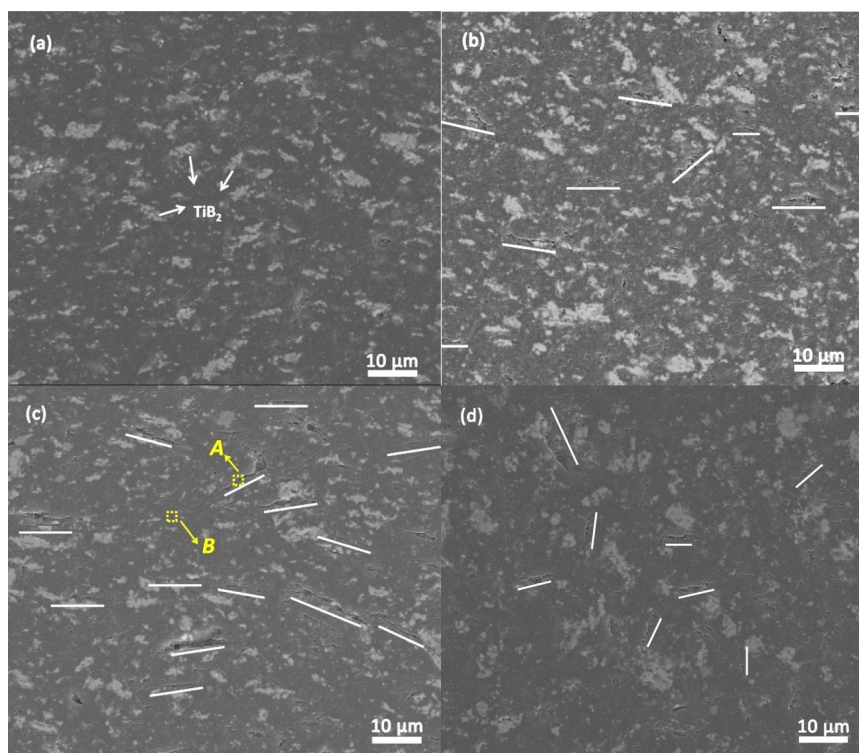


Fig. 2. Polished surfaces of B_4C /GNPs composites with different amount of GNPs. (a) no GNPs, parallel to hot-pressing direction; (b) 3 vol% GNPs, parallel to hot-pressing direction; (c) 5 vol% GNPs, parallel to hot-pressing direction; (d) 5 vol% GNPs, perpendicular to hot-pressing direction.

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