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Fracture characteristics of SiC/graphene platelet composites

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ABSTRACT

Silicon carbide/graphene platelet (SiC/GPLs) composites were prepared using different weight percent of GPLs filler by hot pressing (HP) technology at 2100 °C in argon. The influence of the GPLs addition on bending strength, fracture toughness and related fracture characteristics was investigated. Both the bending strength and fracture toughness increased with increasing GPLs additives. The main fracture origins – strength degrading defects were pores at the low content of platelets and combination of pores and GPLs or clusters of GPLs particles in systems with a higher content of platelets. The fracture toughness increased due to the activated toughening mechanisms mainly in the form of crack bridging and crack branching, while the crack deflection was limited. The highest fracture toughness of 4.4 MPa $m^{1/2}$ was achieved at 6 wt.% of GPLs addition, which was ~30% higher than the K_{IC} value of the reference material.

1. Introduction

Silicon carbide (SiC) ceramics are attractive materials for many industrial applications such as cutting tools, bearings, mechanical seals, nozzles, turbine parts, heat exchangers, etc., due to their excellent combination of properties such as high hardness, high strength, high thermal conductivity, excellent wear and corrosion resistance, etc. [1–5]. Their wider applications however are still limited, because of the low fracture toughness and high brittleness. To improve the fracture toughness of SiC ceramics, first long and short carbon fibres were used and various processing techniques such as chemical vapour infiltration, liquid polymer infiltration, melt infiltration or spark plasma sintering (SPS) have been applied [6–8]. During the last decade, carbon nanotubes (CNTs) and carbon nanofibers (CNFs) have been used as toughening elements in micro-grained ceramics due to their excellent properties, such as low density, high aspect ratio, tensile strength, and an elastic modulus [9–14]. Recently, graphene-based nanostructures in the form of graphene oxide (GO) and graphene platelets (GPLs), also called graphene nanoplatelets (GNPs), multilayer graphene nanosheets (MGN) or graphene nanosheets (GNS), have attracted great interest as efficient reinforcement fillers for the toughening of advanced ceramics due to their capability for promoting toughening mechanisms [15–25].

The first Wang et al. [15] reported improved fracture toughness of GPL/Al_2O_3 composite, which was attributed to the nanosheets pulling out and bridging. According to their results, the fracture toughness of the GPL/alumina composite is about 55% higher than the unreinforced alumina material. Liu et al. [4] reported 30.75% increase in flexural strength and a 27.20% increase in fracture toughness for the Al_2O_3 ceramic composites by adding GPLs.

Walker et al. [16] applied aqueous colloidal processing methods to obtain uniform and homogeneous dispersions of GPL and Si_3N_4 ceramic particles which were densified using spark plasma sintering technology. They reported fracture toughness of 6.6 MPa $m^{1/2}$ for the composite with 1.5 vol.% of GPL which was significantly higher than the value measured for the monolithic silicon nitride with globular grains of alpha phase. The observed toughening mechanisms of the nanocomposites were in the form of graphene necking & crack bridging, crack deflection and graphene sheet

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pull-out. Graphene platelets added to silicon nitride (GPL/Si_3N_4) composites with various GPLs have been prepared by Dusza et al. [26] and the influence of the addition of GPLs on the microstructure development and fracture toughness was investigated. The fracture toughness of the composites is significantly higher compared to the monolithic silicon nitride with the highest value of 9.9 MPa $m^{1/2}$ in the case of composite reinforced by multilayer graphene nanosheets with the smallest dimension. The toughening mechanisms were similar in all composites in the form of crack deflection, crack branching and crack bridging.

In the case of SiC ceramics, Rahman et al. [27] applied polymer pyrolysis technique followed by spark plasma sintering technique to fabricate fine-grained bulk SiC and graphene-SiC systems by in-situ crystallisation of amorphous-SiC obtained from pyrolysis of the polymer precursor. The addition of graphene nanoplatelet affected the fracture toughness of the composites, which with 2 wt.% GPLs showed significant improvement (40% K_{IC}) over monolithic SiC samples thanks to different toughening mechanisms such as crack deflection and crack bridging. Li et al. [28,29] prepared SiC ceramics with different graphene contents using a solid-state pressureless-sintering method and investigated the effects of graphene on the fracture toughness and bending strength of the composites. The fracture toughness of samples was measured by a three-point bending and Single edge notched beam technique with a notch of 0.1 mm with and 1 mm depth. The fracture toughness increased remarkably with increasing graphene content up to 1 wt.%, thanks to the toughening mechanisms such as distortion and deflection of the graphene layers, the relative sliding between graphene layers, crack branching, etc. The highest fracture toughness of 5.65 MPa $m^{1/2}$ was obtained for the sample with 1 wt.% graphene. Very recently Belmonte et al. [18] developed reinforced SiC/graphene composites with graphene nanoplatelets and reduced graphene oxide as fillers. They measured fracture toughness by the surface crack in flexure (SCF) method by Knoop indenting at 100 N and then they performed three-point bending test. According to the results, the composite with 5 vol.% of rGOs filler shows outstanding toughness and strength improvement. Reduced graphene oxide led to an increase of fracture toughness to 8.3 MPa $m^{1/2}$, which is an increment of 162% comparing to monolithic SiC (3.2 MPa $m^{1/2}$). It seems that the lower dimensions and the better mechanical interlock to the matrix of rGOs, when compared to GNPs, support the excellent mechanical performance of SiC/rGOs composites. In the case of GNPs composites, filler contents up to 10 vol.% are required to promote a larger occurrence of crack shielding mechanisms and toughness improvement.

Based on the reported results, it seems that the dimensions and properties of the used carbon-based fillers are very important in the toughening process. The authors of the above mentioned reports used different GPLs with average thickness changing from approximately 2 nm reported by Walker et al. [16], through to a thickness of approximately 8–10 nm reported by Wang et al. and Fan et al. [15,17] up to approximately 20–30 nm reported by Ramirez et al. [20] and reported minimum information only as regards the other dimensions of the GPLs used. On the other hand, mentioned in many reports on ceramic + GPLs composites is the problem of the cluster formation/overlapping of GPLs during the processing. Also, the different ceramic matrix used in these systems can contribute to the toughness of the composite in the different level, so the toughening by GPLs is not easy to predict as it is visible for Si_3N_4/GPLs systems with α–Si_3N_4 and β–Si_3N_4 matrix, respectively [16,26]. This is possible in the case of composites with a very brittle matrix in which the matrix contribution to the toughening is negligible.

The aim of the present contribution is to investigate the influence of the GPLs addition on bending strength, fracture toughness and related fracture characteristics of SiC/graphene platelet composites with a very brittle matrix.

2. Experimental materials and methods

2.1. Experimental materials

Composites based on silicon carbide with varying amount of GPLs were prepared from the following commercially available powders: submicron silicon carbide powder H.C. Starck SiC UF-15, graphene Gn(12) of Graphene Laboratories, Inc., USA and amorphous boron Fluka Chemie15580. The average grain size of SiC powder was D50 = 0.6 μm, graphene flakes consisted of 30–50 monolayers (average thickness around 12 nm) and the average lateral particle size was around 4.5 μm (1.5–10 μm), the purity of graphene powder was 99.2%. Sintering of silicon carbide requires usage of some sintering additives. The most common and successful are boron and carbon. In these composites, 0.5 wt.% of boron and 3.5 wt.% of graphene were used as sintering additives. The amount of graphene added as dispersed phase was varying between 0 and 6 wt.% of GPLs (the total amount of graphene between 3.5 and 9.5 wt.%). Powders were homogenised in the rotary-vibratory mill for 2 h in isopropanol alcohol environment. Silicon carbide milling medium was used to avoid contamination. After drying and granulating, powders were densified using HP processing route (Thermal Technology LLC). Samples of diameter 50 mm were sintered at 2100 °C under 25 MPa pressure in an argon atmosphere. The holding time was 1 h and the heating rate was 10 °C/min.

2.2. Experimental methods

Apparent densities of the sintered samples were determined by the Archimedes method in distilled water. Specimens for microstructure examination were prepared by routine ceramographic procedure, they were cut, ground and polished to a 1 μm finish.

The distribution of GPLs in the SiC matrix was determined from fracture surfaces by field emission scanning electron microscopy (FIB-SEM ZEISS AURIGA Compact). The microstructures of the materials at higher magnifications were characterised using transmission electron microscopy by (JEOL JEM 2100F UHR) with an accelerating voltage of 200 kV in STEM-BF mode. Thin foils for purposes of TEM observations were prepared using standard preparation methods including cutting, grinding, polishing, dimpling and final step of thinning procedure was done by (PIPS Model 691 Gatan) operated at 4.5 kV with ion-beam angles of 4° and 3°.

Mechanical characterization was performed in terms of measurement of basic mechanical properties of bulk materials such as hardness and fracture toughness using indentation methods. Hardness was determined by Vickers indentation (Wolpert Wilson 432 SVD Vickers Hardness Tester) under a load of 9.81 N with a dwell time of 15 s. In order to determine the indentation toughness at least 10 Vickers imprints per specimen were introduced with the load 98.07 N. The indentation toughness was calculated from the lengths of radial cracks and indents diagonals using a formula (Eq. (1)) valid for semi-circular crack systems as proposed by Anstis et al. [30].

$$K_{IC} = 0.016 \cdot \left(\frac{E}{H}\right)^{1/2} \cdot \left(\frac{P}{c^{3/2}}\right) \quad (1)$$

Where K_{IC}–indentation toughness (MPa $m^{1/2}$), 0.016–material-independent constant for Vickers-produced radial cracks, E–Young's modulus (GPa), H–Vickers hardness (GPa), P–indentation load (N), c–half-length of the radial crack (m).

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