## In situ growth of graphene within SiC ceramics by spark plasma sintering

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### Abstract

Over the last few years, there has been a growing interest in the development of new ceramics with enhanced mechanical properties and novel electrical and thermal functionalities. The outstanding electronic and physico-chemical properties of graphene make it ideal filler in the fabrication of conducting and robust ceramic composites. In this context, recent studies on graphene/ceramic nanocomposites have demonstrated outstanding improvements in the mechanical and electrical properties of alumina and silicon nitride ceramics [1-3] by the introduction of graphene nanoplatelets (GNPs) or graphene oxide (GO). At present, this type of graphene/ceramic composites have only been produced by mixing dispersions of GNPs or GO with ceramic powders in solvents, followed by a densification at high temperatures. Unfortunately, these experiments may lead to both graphene agglomeration (forming aggregated flakes) and its structural degradation. Although silicon carbide (SiC) is one of the most appropriate ceramics for structural applications due to its exceptional thermomechanical properties at high temperatures [4], no work has been reported on the fabrication of SiC matrix composites containing graphene. It is also important to point out that the fast generation of large-area and homogenous epitaxial grown graphene on SiC single crystals by thermal annealing have been reported [5,6].

In this work, a novel single step approach for manufacturing electrically conducting and well dispersed graphene/SiC nanocomposites with enhanced mechanical and electrical properties is explored. Epitaxial graphene (EG) was in-situ grown within either  $\alpha$ - or  $\beta$ -phase SiC ceramics during their densification by Spark Plasma Sintering (SPS). The in situ graphene growth mechanism is believed to be due to the simultaneous actions of the high temperature, the electric current passing through the graphite dies and specimen (Joule heating), and the partial vacuum, all of them involved in the SPS process.

The presence of few- layer epitaxial graphene (FLG) located between the grain boundaries of the bulk SiC has been confirmed using Raman spectroscopy combined with high resolution transmission electron microscopy (HRTEM) and conductive-scanning force microscopy (C-SFM). Raman spectroscopy mappings on the SiC nanocomposites (Fig. 1, left side) showed the presence of about 4 vol. % graphite-like structures dispersed within the SiC matrix. These structures were also observed on the fracture surface of the specimens using SEM. HRTEM examination showed that few-layer graphene domains were generated at grain boundaries, the number of layers being related with the crystal orientation (Fig. 1), whereas conducting and topographic SFM studies established a clear correlation between those FLG and conducting regions in the composites [7]. The developed conducting graphene network significantly enhanced the electrical and mechanical performance of SiC, reaching electrical conductivity values as high as  $1.02 \times 10^2 \, \text{S} \cdot \text{m}^{-1}$  and fracture toughness increases of 55%.

The approach presented here offers unprecedented opportunities for the fast manufacturing of graphene/SiC nanocomposites precluding the handling of potentially hazardous nanostructures and it widens possible applications, including micro- or nano-electromechanical systems, brakes, micro-turbines or micro-rotors.

### References

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**Figure 1.** FESEM micrograph of the fracture surface of a graphene/ $\alpha$ -SiC composite where multilayered graphene flakes are shown bending along grain boundaries, HRTEM examination showing fewlayer graphene domains at different grain boundaries and Raman spectroscopy mappings on scanned areas of 10  $\mu$ m x 10  $\mu$ m.