Graphene/alumina (G/Al_2O_3) composites by Spark Plasma Sintering; a simple, fast and upscalable method

V.G. Rocha[a,b], A.Centeno[c], B.Alonso[c], A. Fernández[d], C.F. Gutierrez-Gonzalez[d], R. Torrecillas[d,e], A. Zurutuza[c]

[a] Department of Materials, Exhibition Road, London SW7 2AZ, United Kingdom

[b] ITMA Materials Technology, Parque Tecnológico de Asturias, 33428 Llanera, Spain

[c] Graphenea S.A. Tolosa Hiribidea 76, E-20018 Donostia-San Sebastián, Spain

[d] Centro de Investigación en Nanomateriales y Nanotecnología (CINN) (CSIC – Universidad de Oviedo – Principado de Asturias), Parque Tecnológico de Asturias, 33428 Llanera, Spain

[e]Moscow State University of Technology (STANKIN), Vadkovskij per. 1, Moscow, Moscow Oblast, Russian Federation

v.garcia-rocha@imperial.ac.uk

Abstract

Tough and electroconductive ceramics have a great potential to solve a wide number of material related challenges in high technology applications such as power generation, aerospace, transportation and military applications. The development of very complicated shapes and high accuracy components is especially challenging for ceramic materials due to their high hardness, low fracture toughness which has very often limited their applications. The reinforcement of ceramic materials with electroconductive second phases appears as an interesting alternative for manufacturing by Electro Discharge Machining (EDM) complex shape components from hard materials. However, low resistivity (<100 Ωcm) of the material is required to be shaped using this technology. In this work, graphene is incorporated in the alumina (Al_2O_3) matrix in order to make it electroconductive and improve it mechanical properties.

An outstanding dispersion of Graphene Oxide (GO) in the alumina matrix was achieved using a colloidal method and its consolidation by spark plasma sintering (SPS) allowed, in one-step, the in situ reduction of the GO during the sintering process. The sintered discs were cut along two directions: perpendicular and parallel to the pressure direction applied in SPS, as shown in Fig 1. Evaluation and optimisation of the graphene thermal reduction by SPS was performed by Raman spectroscopy [1,2]. Moreover, Raman spectroscopy is shown to be a powerful technique to study the orientation of the graphene in the G/Al_2O_3 composites. As it can be seen in Fig. 2 spectra there are important different peak intensities depending on the analysed orientation. The signal intensity in the surface perpendicular to the pressure direction applied in SPS (Fig. 2a) is much lower than in the parallel surface (Fig. 2b) indicating preferential orientation of the graphene in the composite. Raman parameters were determined in order to get further insight and to reveal the anisotropic structure of the composite.

As a result of the reduction process, non-conductive graphene oxide is transformed into a conductive material. Electrically conductive composites in both directions were achieved with an unexpected low amount of graphene. The percolation threshold of the as prepared composites was found to be around 0.22 wt%, indicated by the exponential decrease of the electrical resistivity up to 8 orders of magnitude in comparison to the monolithic alumina (15 vs 10^9Ωcm). The presence of graphene enhanced the mechanical properties of the raw alumina by nearly 50%. The resulting *R*-curves are shown in Fig. 3 and the clearly show the higher fracture resistance of the G/Al_2O_3 composite compared to the monolithic Al_2O_3. For the alumina, the initial fracture resistance is approximately 3.6 MPa $m^{1/2}$, however this value only rises up to 4.4 MPa $m^{1/2}$ showing a very soft R-curve behaviour that it may be caused by the crack bridging between the ceramic grains. The graphene nanoplatelets lying in the crack plane behind the tip act as ligaments bridging the two crack surfaces, which provide a stable crack growth until a steady-state toughness of 7 MPa $m^{1/2}$ approximately. The fracture toughness improvement was meant to be due to crack bridging reinforcement mechanism. This mechanism is confirmed using Scanning Electron Microscopy (Fig 3).

References

[1] Ferrari AC, Meyer JC, Scardaci V, Casiraghi C, Lazzeri M, Piscanec S, et al.. Phys Rev Lett **97** (2006) 187401.

[2] Malard LM, Pimenta MA, Dresselhaus G, Dresselhaus MS. . Phys Rep **473** (2009) 51–87.

Figures

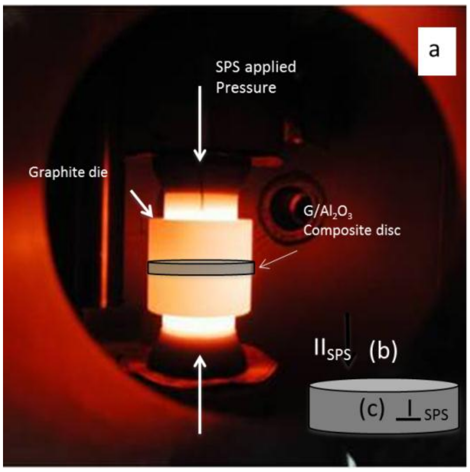

Figure 1. Image of SPS furnace and sketch of the sintered disc and the applied pressure

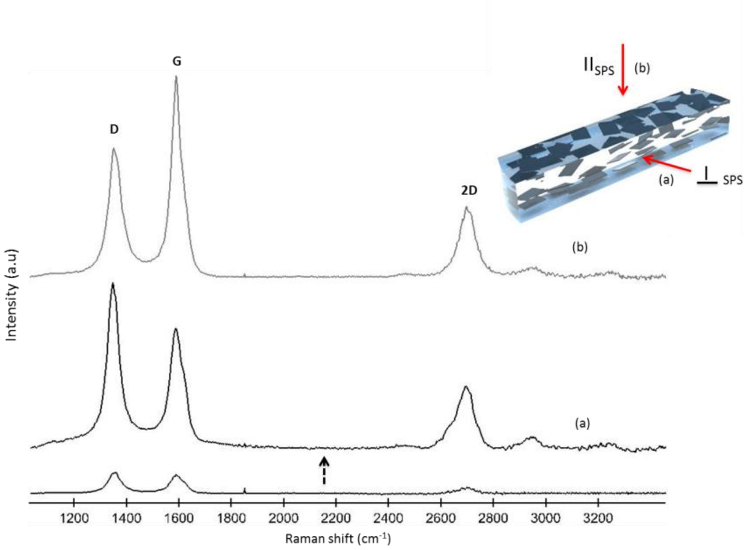

Figure 2. Raman spectrum of the 2-G/Al_2O_3 composite collected at two different orientations: (a) perpendicular to the pressure direction applied in SPS and (b) parallel to the pressure direction applied in SPS.

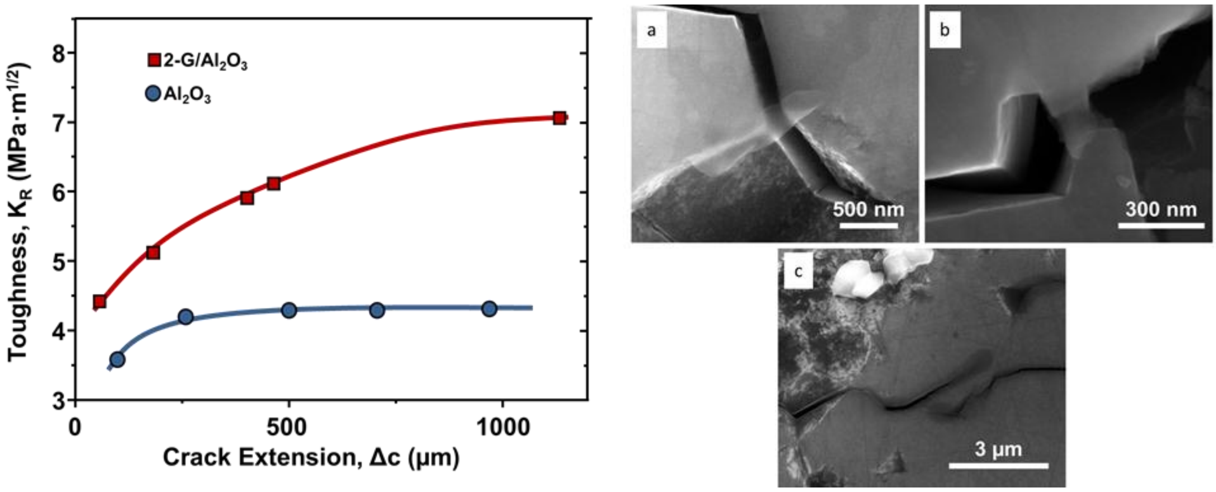

Figure 3. Comparison of the R-curves measured in the Al_2O_3 and G/Al_2O_3 materials (left). SEM observations of the reinforcement mechanisms of the G/Al_2O_3 composite (right).