Manufacture and electromechanical characterization of highly conductive multilayer-graphene/polydimethylsiloxane flexible paper

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Multilayer graphene (MLG) micro- and nanosheets have been investigated for use as nanofiller in polymer composite in order to obtain multifunctional materials with enhanced electrical conductivity and mechanical properties [1-4]. In order to take advantage of the conductivity properties of MLG sheets, a large amount of nanofiller should be used [5]. Although, increasing filler loading alters the mechanical properties of the composite because of serious filler agglomeration [6, 7]. It has been shown that a promising approach to realize electrically conductive light-weight composite is to incorporate an electrically conductive graphene paper (GP), obtained by vacuum filtration of a nanofillers suspension, into the polymer matrix [8]. One advantage of infiltrating the GP with polymer is that the tensile modulus of the composite can be greatly improved as compared with either GP or neat polymer, without weakening the electrical properties of the highly continuous nanofillers network formed in the paper making process [8]. In this work we present experimental results related to the fabrication process and the electromechanical behaviour of a free standing, highly-conductive MLG paper impregnated with polydimethylsiloxane (PDMS).

We produce a graphene-based porous paper made of MLG microsheets through the thermal expansion of a graphite intercalated compound, the successive liquid-phase exfoliation of the resulting expanded graphite in a proper solvent, and finally the vacuum filtration of the MLG-suspension using a nanoporous alumina membrane [9]. Due to its porosity, the MLG paper can be impregnated by PDMS prepolymer in order to obtain a MLG/PDMS composites. The pre-polymer base and the crosslinking curing agent were mixed together at 10:1 volume ratio [10]. All samples were mixed using a mechanical stirrer and the mixed uncured PDMS was degassed in order to fabricate bubble free test samples. The degassed PDMS mixture was then spin coated in order to obtain a layer of ~376 μm. Finally the MLG paper was soaked in the uncured solution and then cured in a oven. Figure 1 shows photographs of a MLG/PDMS composite as prepared (Fig, 1a) and under bending (Fig. 1b) showing its good flexibility. The combined Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray (EDX) analysis of the produced flexible MLG/PDMS composite shows that the PDMS is well infiltrated into the porous structure of the paper, thus providing to the resulting composite flexibility and mechanical strength.

References

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The sheet resistance of the produced free-standing MLG paper and of MLG/PDMS composite paper was measured at room temperature through a four-point probe resistivity measurement system. The obtained results show that the free-standing MLG paper is characterized by a sheet resistance (Rs) of ~ 0,69 Ω/□, which does not increase significantly after polymer infiltration into the porous paper (Rs of ~ 0,76 Ω/□). Electromechanical behavior of MLG/PDMS composite paper has been investigated experimentally by measuring the variation of the DC electrical resistance during a tensile strength test. Electromechanical test was performed using an INSTRON 3366, universal testing machine equipped with a 500 N load cell and with the crosshead speed fixed to 10 mm/min (Fig. 1d). The specimens to be tested were realized following a modified version of the standard ASTM D 882 2002. The mechanical characterization shows that the break of the composite paper occurs after an elongation of ~ 11 mm that corresponds at a deformation of ~ 80%, like the neat polymer, but for an applied load which is twice the load at break of the PDMS (Fig. 1e). The obtained data demonstrate that the produced composite is characterized by an enhanced stiffness with respect to PDMS. Furthermore, the DC electrical resistance of the MLG/PDMS composite paper is nearly constant for small elongation. On the contrary, for larger values of elongation, an increase of the DC electrical resistance is observed (Fig. 1f).

In conclusion we have proposed an alternative viable approach to one consisting in the nano-filler dispersion in the polymer matrix, which allows to produce highly conductive and highly flexible carbon-based polymer composites. We finally demonstrate the remarkable electromechanical properties of the newly developed carbon-based nanomaterial by measuring the variation of the electrical properties during a tensile strength test.

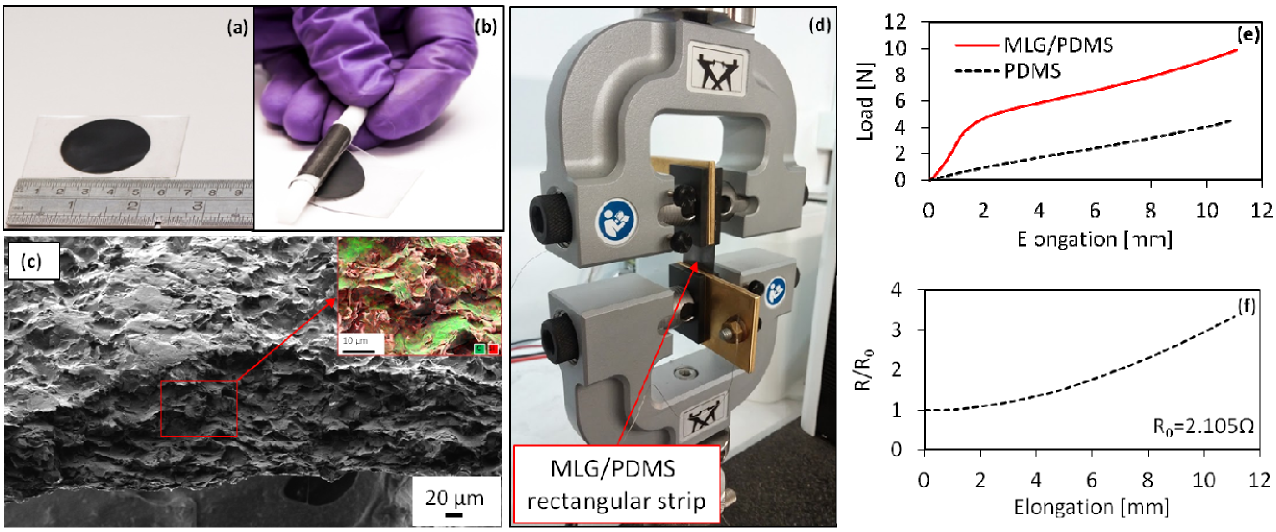

Figure 1: Photographs of a MLG/PDMS composite as prepared (a) and under bending (b); SEM micrographs of the fracture section of a MLG/PDMS composite paper (c) and corresponding EDX elemental analysis (insert); photographs of the INSTRON 3366, universal testing machine and MLG/PDMS rectangular strips of 24.3 mm×25 mm×0.5 mm for mechanical testing (d); load-elongation characteristics of MLG/PDMS composite paper and PDMS (e); DC normalized electrical resistance (R/R_0) - elongation characteristics of MLG/PDMS composite paper (f).