CVD synthesis of a MWCNT-graphene composite

I. Martin-Fernandez¹, N. Mestres², P. Godignon¹

¹Instituto de Microelectrónica de Barcelona (IMB-CNM, CSIC), Campus UAB, E-08193 Cerdanyola del Vallès, Spain ²Institut de Ciència de Materials de Barcelona (ICMAB, CSIC), Campus UAB, E-08193 Cerdanyola del Vallès, Spain inigo.martin@imb-cnm.csic.es

Carbon nanotubes (CNTs) and graphene have generated great interest for device applications due to their particular physicochemical properties and, thus, big efforts are being dedicated to the optimization of their synthesis. These efforts, however, are not only leading to solve their growth mechanism, but also to the finding of new material architectures aimed to possible new functionalities [1]. This way, for example, a new composite material based on multi-walled (MW) CNTs and multi-layer graphene was presented recently [2].

This contribution presents a CNT-graphene composite similar to that reported in [2] but where a single graphene layer is formed on top of the CNT array.

The CNT-graphene composite is formed by a vertically aligned array of nanotubes and by a top film that is formed during the same synthesis process. The synthesis of the composite is performed by thermal CVD and, first, a few nanometer thick layer of platinum (that is a non-conventional catalyst material for CNTs and graphene) needs to be deposited on the substrate to catalyze its growth [3]. The synthesis of the composite is performed at 800°C and CH_4 is used as the carbon feeding gas.

The composite is dark colored. The vertically aligned CNTs are MW, analogous to those synthesized in previous works [3]. Their diameter is ~10 nm, and their walls are parallel and present low density of defects. The top film is smooth and flexible, and its roughness is related to the MWCNT array underneath. Cracks on the top film are only observed close to positions where the composite had been modified because of scratches or because of squashing. The SEM images in Figure 1 show (a) a shifted flake of the top film on top of the MWCNT array as a result of a scratch on the composite, and (b) a tilted view of the composite at a section that had been fabricated by FIB.

The composition of the composite was analyzed by EDX and micro-Raman spectroscopy. These analyses were performed at different positions of the sample to discriminate the different components of the composite The EDX analyses determined that the top layer is made of carbon. The Raman analysis was performed on the squashed composite area shown in Figure 2(a-b). The Raman spectra, A-F, in Figure 2(c) are related to the positions A-F in Figure 2(a). The low frequency region in these spectra (1200-1800 cm⁻¹) is dominated by the typical spectral features of MWCNTS. The large relative intensity of the G' peak in the spectra for positions A, E and F may only match that of graphene [4]. In particular, the Raman spectra could be attributed to a single layer graphene [5,6] or to a few layers graphene film where the layers are folded or arranged according to a disordered stacking [7].

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Figures



Figure 1: SEM images on the composite material. (a) Shifted top film flake on top of the CNT array. (b) Tilted view of the FIB made section of the composite.



Figure 2: Raman analysis of the composite material at a squashed area. A-F in (a) denote the positions from which the Raman spectra A-F in (c) were obtained.(b)Detail of the folding of the top film.