Large crystalline graphene oxide sheets from helical ribbon carbon nanofibres

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Among the different methods that are currently known to obtain graphene, the chemical methods through the intercalation compounds of acids or/and oxysalts yielding graphite oxide, seems to be, at the moment, the most promising technique to produce large quantities and thus supply the increasing demand on graphene products. Commonly, natural graphite is used as a raw material[1], The subsequent exfoliation of the intermediate graphite oxide produces, more precisely, graphene oxide, by definition single layer or up 10 stacking layers. As graphite is composed by hundreds or thousands of graphene layers, complete separation of all the layers is difficult and therefore the yield in graphene oxide sheets becomes low. Other approaches have been use carbon nanotubes (CNT), which are also a graphitic material, with less number of graphene layers [2]. Considering these examples, we use helical ribbon carbon nanofibers (HR-CNF) to produce graphene oxide sheets.

The HR-CNFs are CVD-produced by Grupo Antolin (Burgos, Spain) by the floating catalyst method at industrial scale, in a continuous process. Structurally, these filaments have a wide hollow core and a highly graphitic structure, which is formed by a ribbon rolled along the fiber axis developing a continuous spiral. This ribbon is composed of around 5 stacked graphene layers and this is the key point of our production method, the structure of the CNFs is rolled graphene that has to be unzipped.

The production method consist of two steps, in the first step, the CNFs are unzipped by chemical oxidation with a modified Hummers reaction [3, 4], in the second step the layers of the obtained graphene oxide are even more separated and exfoliated by ultrasonication.

Characterization of this material seems to disagree when analyzing the bulk and just one graphene oxide layer. X Ray Photoelectron Spectroscopy (XPS) shows a high content of oxygen, while electron energy loss spectroscopy (EELS) analysis performed in one layer shows a much less oxidized material. Interestingly, both analysis detect N in the structure, thus the graphene oxide sheets are presumably N-doped. The single layer of graphene oxide was also analyzed by Raman spectroscopy, so the contributions of the sp2 and sp3 C domains are certainly these corresponding to just one layer.

This graphene oxide was introduced in an epoxy matrix [5]. The combination of O and N present groups in the structure of the graphene oxide resulted in impressive improvements in the fracture toughness and the fatigue life of the G-O/epoxy nanocomposites.

References

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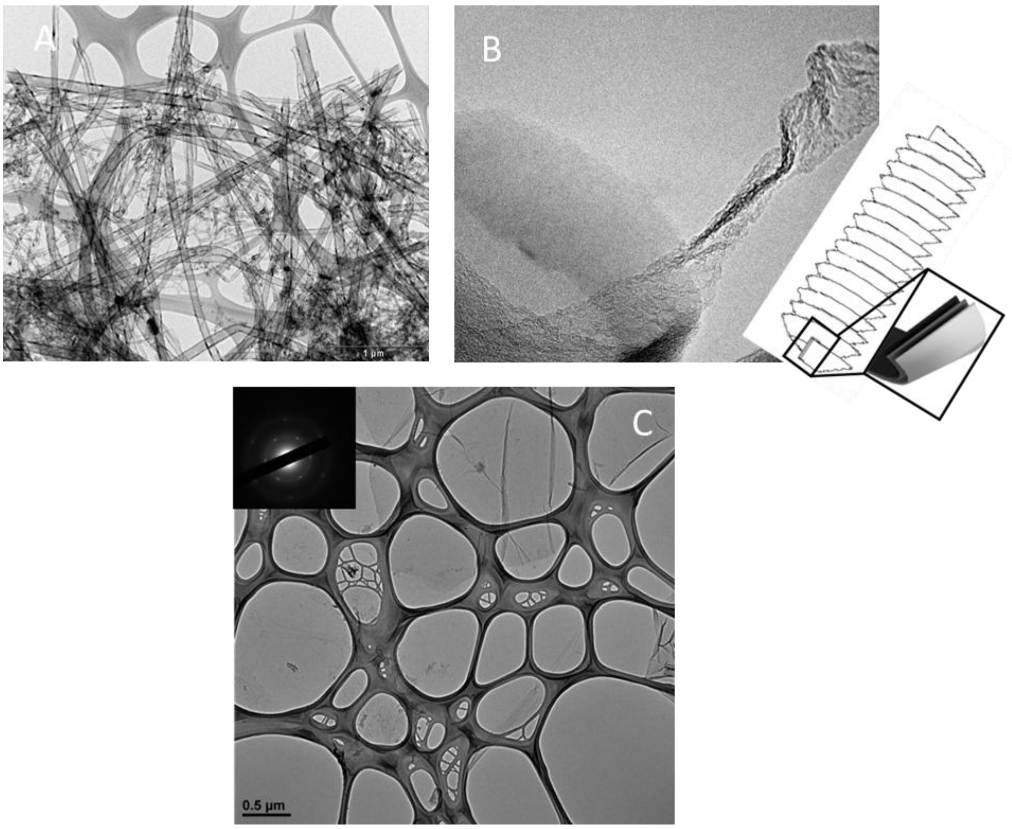
Figures



TEM images of: A) HR-CNF; B) evidence of the spiral structure of the HR-CNFs with a scheme of the structure; C) single layer graphene oxide obtained from the HR-CNFs