Insights on the synthesis of diamanes and diamanoid/graphene hybrids by hot-filament process

Fabrice Piazza[1], Marc Monthioux[2], Pascal Puech[2], Iann C. Gerber[3] and Kathleen Gough[4]

[1] Pontificia Universidad Católica Madre y Maestra, Autopista Duarte km 1 ½, Santiago de los Caballeros, Dominican Republic
[2] CEMES, CNRS, Université de Toulouse, 29, rue Jeanne Marvig, 31055, Toulouse, France
[3] LPCNO, CNRS, INSA, Université de Toulouse, 31400, Toulouse, France
[4] University of Manitoba, Winnipeg, MB R3T 2N2, Canada
* fpiazza@pucmm.edu.do

Genuine diamanes and diamanoids are promising versatile new wide band-gap semiconducting 2D materials for multiple applications such as band-gap engineering in microelectronics, room temperature single photon emission to be used in quantum information processing and sensing, nanoelectromechanical systems for implantable medical devices, and more. Genuine diamanes were recently made possible by the sp^2-C to sp^3-C conversion of bilayer graphene (2LG) using a pressureless and low-temperature hot-filament-assisted hydrogenation process, as demonstrated by Raman spectroscopy over area surfaces of up to 2000 μm^2. The actual full hydrogenation of 2LG into diamane is supported by first principle calculations. Because the 2LG used contains a mixture of AA and AB domains (for which the conversion is possible), together with randomly stacked layers (turbostratic domains, for which the conversion cannot take place), the hydrogenated material is under large stress, up to 10 GPa, assuming an average hydrostatic strain. By replacing 2LG with few-layer graphene (FLG), diamanoid/graphene hybrids were formed, because the most frequent stacking sequences are usually not suitable for the full conversion of the FLG into genuine diamanoids. Only the FLG partial conversion was then obtained, generating a configuration of twisted-2LG as evidenced by characteristic Raman features and electron diffraction patterns. Twisted-2LG are located at the interface between the upper diamanoid domain and the non-converted graphenic domain underneath. They were presumably formed following the relaxation of the stress resulting from the partial sp^2-C to sp^3-C conversion, estimated at around 33 GPa by Density Functional Theory calculations. C-H bonding in the surface of hydrogenated FLG, where half of the carbon atoms are bonded to a single hydrogen each, is directly evidenced by Fourier Transform Infrared microscopy. It is believed that dimensions are only limited by the domain dimensions in the starting material, not by the process.