Tri-layer enriched graphene sample by mechanochemical exfoliation of graphite: A one-step route for the production, processing and deposition as transparent films.

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Several efforts have been made in the last few years to produce graphene with yields high enough to be viable for scalable manufacturing. The majority of these efforts, however, resulted in graphene that was lacking in structural integrity or yield or purity. Besides the search for efficient bulk production methods for high-quality graphene samples, there are other important challenges in the field: one of them is related to the development of efficient processing routes to graphene. Graphene is a promising candidate for the next-generation opto-electronic devices, and these types of applications require the fabrication of large-scale, transparent thin films of graphene onto a number of different types of substrates, including plastics, flexible and stretchable substrates. Another challenge is the development of synthetic routes to produce size- and layer-monodispersed graphene.

We report here a novel route for the production of trilayer enriched graphene samples, obtained from graphite flakes, in which both the synthesis and processing as a transparent thin film (easily transferred to arbitrary substrates) are solved together in one single pot. The starting material should be any type of graphite sample, onto which a mechanical exfoliation processes are performed, followed by chemical/thermal exfoliation, yielding few-layer graphene (FLG) flakes. The chemical/thermal exfoliation is performed in a water/oil biphasic system in which the resulting FLG spontaneously self-assembles at the water/oil interface as a thin film that is easily transferred to an arbitrary substrate. The whole process consists in a mechanical peeling of graphite by rubbing it on the surface of a magnesium foil. The magnesium holding the peeled graphite is subsequently mixed to a heterogeneous liquid-liquid mixture formed from a toluene/aqueous hydrochloric acid solution under a ultrasonic bath. Immediately after the contact with the aqueous acid solution, the magnesium foil dissolves through a highly exothermic reaction. The carbonaceous material (characterized as FLG) remains dispersed in the water/toluene emulsion, and spontaneously agglomerates into a continuous and homogenous film located at the toluene/water interface, that can be easily transferred to an arbitrary substrate.

Figure 1a shows films deposited onto glass substrates (upper) and plastic substrates (bottom). The flexibility of the film on plastic substrate can be seen in Figure 1b. The film presents excellent optical transparencies (90% at 550 nm). The occurrence of FLG was confirmed by TEM, Electron Difraction and Raman spectroscopy. By analyzing 100 Raman spectra collected at different samples we generate occurrence statistics and estimate that our sample is composed of 59% of tri-layer AB-stacked graphene, which is one of the most efficient layer-controlled syntheses of tri-layer FLG described to date.

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Figures

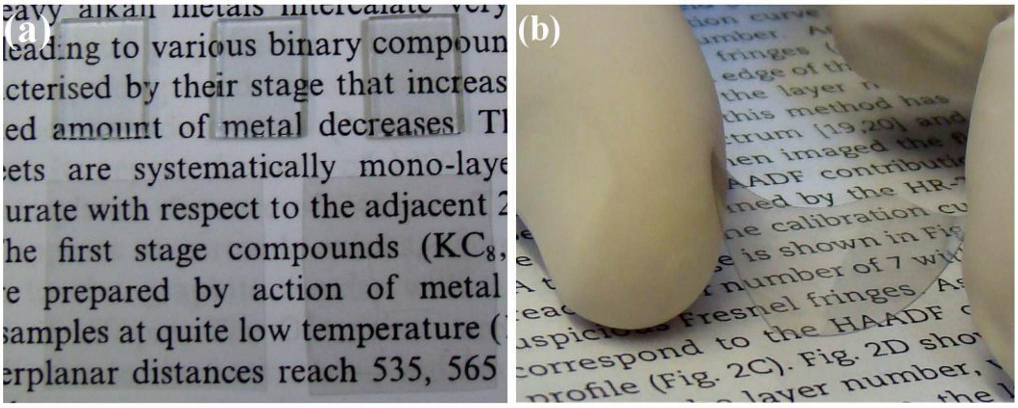

Figure 1 - A photograph of transparent graphene films deposited on quartz substrates (upper) and plastic (PET) substrates (bottom). For the quartz substrate from the left to the right it is shown the substrate itself (with no film deposited), and films prepared with 5 cm (middle) or 10 cm (right) of graphene. For the plastic substrate from the left to the right it is shown the substrate itself (with no film deposited) and a film deposited starting from 5 cm of magnesium foil. ***b,*** *A picture of the flexibility of the graphene film on the PET substrate.*