Graphene and carbon nanotubes film organization with a new solution-based method: a substrate independent transfer for transparent electrode applications

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Abstract

Graphene and carbon nanotubes (CNT) have exceptional properties that make them fascinating objects for both academic and application-oriented studies. In particular, with the combination of their electronic, mechanical and optical properties, they are considered as potential candidates for new generations of transparent electrodes in o-PV cells, touch screens and flexible displays. However, such technologies rely on the capacity to form high-quality thin-films with a controlled morphology.

In order to address the related issues a low-cost and original method based on the transfer of surfactant-stabilized water films has been developed in our group. This *bubble deposition method* (BDM) proved very efficient to organize and transfer, under ambient conditions, dense and homogeneous monolayers of nano-objects such as nanowires and nanoparticles, over large areas. The BDM does not require sophisticated transfer processes and is compatible with a large panel of substrates (silicon, glass, polymers…), both hydrophilic or hydrophobic.

Recently we proved the usefulness of this approach to self-assemble carbon materials such as single-wall carbon nanotubes (SWNTs)[1] and graphene oxide sheets (GO)[2] into close-packed monolayers. Of particular interest is the compatibility of this technique with: *(i)* a pre-structuration of the substrate in micro-channels, such structuration leading to the specific increase of the deposition density within the channels (see figure 1)[3]; *(ii)* homogeneous transfers at the wafer scale using vertical water films in place of bubbles; *(iii)* a simple layer-by-layer approach, enabling the formation of thickness-adjusted films through multiple depositions. This layer-by-layer approach was extended notably to realize hybrid materials and, as a proof of concept, a stacked structure was formed by alternating SWNTs and GO layers[2].

Our results provide insight into important problems that impeded the development of SWNT and Graphene based devices. Indeed, in contrast with most methods (such a spin coating), BDM leads to the transfer of the full amount of engaged material. It is thus compatible with high added-value materials, such as SWNTs selected by chirality. We also present how this method can be used to aligned carbon nanotubes at various scales using the drainage of the water confined in the double surfactant wall of the bubble[1].

Despite the variety of existing methods there is still a lack of a simple, efficient and substrate-independent technique enabling the deposition of graphene sheets free of wrinkles. The Langmuir Blodgett approach is highly efficient to self-assemble a monolayer but the roughness of the films deteriorates rapidly when several layers are tentatively stacked. In contrast, we show that this drawback can be almost completely suppressed using our approach with both small (1-10 μm^2) and large (10-500 μm^2) GO sheets (see figure 2). As well as the precise control of the nano-objects assembly, the efficient chemical reduction of GO into graphene is still a pressing issue that limits the development of GO-based electrodes. We are currently investigating this point and will report our last results combining the BDM with a post-deposition reduction step.

The BDM is both versatile and scalable, and is adapted to a wide variety of applications. Of particular interest are conductive films that are optically transparent and yield adequate and uniform electronic properties. They could be used as replacement for ITO in both light emitting devices and photovoltaic ones. Concerning PV, one particularly interesting system is the carbon/silicon heterostructure that was shown to display very high efficiency of light-to-current conversion despite its simplicity. Using the BDM, ultra-thin and uniform films of both SWNTs and GO were deposited on silicon substrates and the mechanism of charge separation at the carbon/silicon interfaces is studied by the non-invasive Time Resolved Microwave Conductivity (TRMC) method.[3] This technique is based on the analysis of the

evolution of the microwave absorption of the studied samples containing mobile charges generated by a nanosecond laser excitation. The measured signal is proportional to the conductance change and consequently to the number of charge carrier and to their mobility. It allows studying the evolution of the lifetime of the photo-generated carriers as a function of the heterostructure properties. As an example, the charge carrier lifetime in the case of a modified silicon-nanotube junction (see figure 3) is 100 times longer than for the bare silicon. Such signature of an efficient charge separation at the carbon/silicon interface measured by TRMC is very helpful to understand and optimize nanotube-silicon solar cells.

References

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Figures

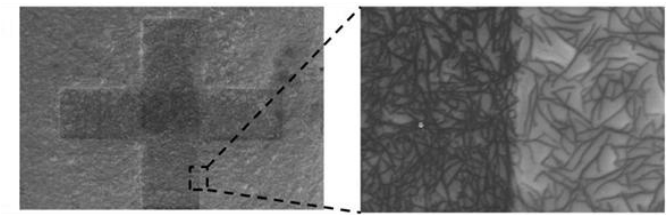

Figure 1. SEM images of a carbon nanotube film transferred on a lithographically patterned glass substrate.

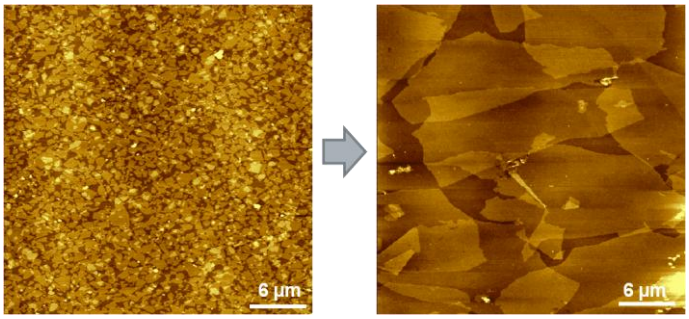

Figure 2. AFM images of a close-packed arrangement of small (left) and large (right) graphene oxide sheets in a dense and homogeneous monolayer film.

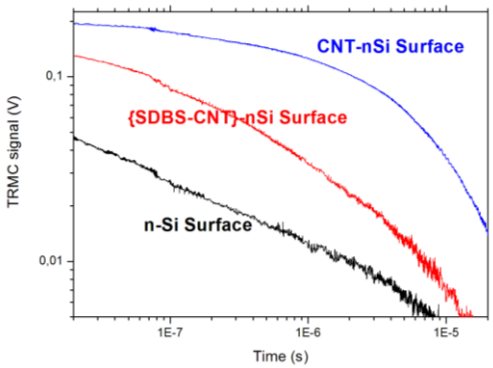

Figure 3. Amplitude of the TRMC signal of Si, {SDBS-CNT}-Si and CNT-Si surfaces