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# Dispersing Carbon Nanotubes with Graphene Oxide in Water and Synergistic Effects between Graphene Derivatives

Ling Qiu,[a] Xiaowei Yang,[a, b] Xinglong Gou,[a] Wenrong Yang,[c] Zi-Feng Ma,[b] Gordon G. Wallace,[d] and Dan Li*[a]

Graphene sheets and their tubular variants, carbon nanotubes (CNTs), are renowned for their extraordinary electrical, mechanical and thermal properties.[1] Macroscopic assemblies of these nanostructures are promising for a wide range of applications, particularly as nanostructured electrodes for a variety of energy storage and conversion devices.[2] Large-scale production of both CNTs and graphene is being actively pursued in industry and both are now commercially available on the ton scale. However, as-produced graphene and CNTs are generally in an agglomerated powder form. To fabricate them into macroscopic films, fibres or composites for practical applications, the resulting agglomerates need to be well dispersed in solvents to allow the use of cost-effective solution processing techniques. To realise the dispersion, traditional powder dispersing strategies, which usually involve chemical modification and/or the use of surfactants or polymeric dispersants, are adopted.[3] The dispersants traditionally used, however, are electrically insulating and often need to be removed from the final products.[2a] The use and subsequent removal of dispersants adds an extra manufacturing cost and also deteriorates the mechanical properties of the resulting structures.[2a] New efficient dispersion strategies are highly desired to promote the large-scale industrial use of these carbon nanostructures.

Manufactured by different approaches, CNTs and graphene are usually supplied in a single-component form. Given similarities in structure and physical properties between CNTs and graphene, their hybridisation would presumably have little adverse effect on most of their applications, and in some cases, may lead to useful synergistic effects.[2c,4] Herein, we illustrate that the chemical and electrical synergies between two graphene derivatives, graphene oxide (GO) and CNTs, can indeed offer an amazingly simple strategy for processing graphene/CNT nanohybrids, in which no chemical functionalisation of CNTs is required and no organic solvents, toxic chemicals and surfactants are involved.

GO, an oxygenated graphene molecule, is an inexpensive precursor currently used for large-scale production of chemically converted graphene.[5] It can be readily prepared by exfoliation of chemically oxidized graphite.[6] According to recent studies,[6] a GO sheet consists of two types of randomly distributed regions: aromatic regions with unoxidised benzene rings and regions with aliphatic six-membered rings. Such a special molecular structure makes GO behave like an amphiphilic molecule and allows the formation of Langmuir–Blodgett films on a water surface in the absence of other amphiphilic molecules.[7] The oxygen-containing groups render GO sheets hydrophilic and highly dispersible in water, whereas the aromatic regions offer active sites to make it possible to interact with other aromatic molecules through π–π supramolecular interactions. Such a unique chemical structure prompted us to explore GO as a dispersant to suspend unfunctionalised CNTs in water and to develop a new solution processing strategy for making graphene/CNT hybrids. Recent studies show that CNTs functionalised through acid treatment are actually coated by partially oxidised CNT fragments through π–π interactions.[8] Strong non-covalent binding between GO and some conven-

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[a] L. Qiu,+ X. Yang,+ Dr. X. Gou,+ Dr. D. Li
Department of Materials Engineering
ARC Centre of Excellence for Electromaterials Science
Monash University, VIC 3800 (Australia)
Fax: (+61) 3-9905-4940
E-mail: dan.li2@monash.edu

[b] X. Yang,+ Prof. Z.-F. Ma
Department of Chemical Engineering
Shanghai Jiao Tong University
Shanghai 200240 (China)

[c] Dr. W. Yang
Australian Key Centre for Microscopy and Microanalysis
University of Sydney, NSW 2006 (Australia)

[d] Prof. G. G. Wallace
ARC Centre of Excellence for Electromaterials Science
Intelligent Polymer Research Institute
University of Wollongong, NSW 2522 (Australia)

[+] These authors contributed equally to this work.

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tional aromatic compounds has recently been used for drug delivery and biosensing.[9] All of these studies further suggest that using GO to interact with CNTs should be possible.

We used commercially available single-walled CNTs synthesised by high-pressure carbon monoxide conversion (HiPCO) to demonstrate this concept. It was found that sonicating a mixture of GO dispersion and CNTs powder (0.05 wt%, respectively) resulted in the formation of a black suspension. The suspension is very stable and no visible sediment was observed over two years. After being diluted by a factor of about 30, it looks like a "true solution" (Figure 1A). When the content of GO and CNT was increased to 0.5 wt%, a GO/CNT hydrogel was formed (Figure 1B). In contrast, no hydrogels were produced when only single components were used at the same concentrations. The formation of the GO/CNT hydrogel at such a low solid content indicates a strong interaction between GO and CNTs.

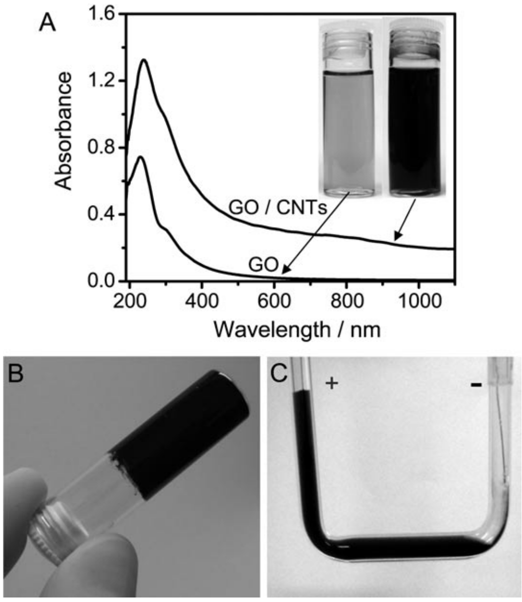

Figure 1. A) UV/Vis absorption spectra of diluted GO and GO/CNT dispersions. The inset shows photographs of the corresponding dispersions. B) GO/CNT hydrogel containing 0.5 wt% of GO and CNTs. C) Electrophoresis experiments on the GO/CNT dispersion containing 0.0025 wt% of GO and CNTs.

Like normal colloids, diluted GO/CNT dispersions displayed a Tyndall effect when a laser beam was passed through the sample (Figure S1 in the Supporting Information). Given that GO sheets are highly negatively charged when dispersed in water due to the presence of oxygen-containing groups,[6,10] we carried out electrophoresis experiments to further investigate whether GO sheets were attached to the CNTs. When an electric field was applied across the mixture dispersion, the black solution near the negative electrode gradually faded to colourless (Figure 1C), indicating that the negatively charged GO particles, together with CNTs, had migrated towards the positive electrode. After the electric field was withdrawn, the black colloidal particles gradually diffused back and eventually reformed as a uniform suspension. No separation between GO sheets and CNTs was found during these treatments. Given that the CNTs used had not been functionalised with oxygen-containing groups, there should be few charges on the CNTs; these observations suggest that CNTs are strongly attached to negatively charged GO nanosheets, which transport the CNTs under the influence of an electric field.

We envisage that during the dispersion process, large CNT agglomerates are first disassembled into smaller bundles or individual tubes by sonication. The dissolved CNTs then become associated with the GO sheets, such as other aromatic dispersants, through $\pi$–$\pi$ stacking interactions between the delocalised electrons in both the aromatic regions of GO sheets and the CNTs and remains dispersed due to the electrostatic repulsions between GO sheets. Distinct from conventional dispersants, the content of GO required to suspend CNTs can be very low. We have tested a range of GO/CNT ratios and found that stable suspensions can be obtained by adding up to 9 times (by weight) of CNTs in the 0.05 wt% GO dispersion. In contrast, when a commonly used surfactant, sodium dodecylbenzenesulfonate, is employed, a weight ratio of 10:1 (the surfactant to CNTs) is required.[2a] The exceptional dispersibility of GO is likely to be related to the high aspect ratio of GO sheets. The lateral sheet size of GO in our experiments is found to be a few hundred nanometers. Such large sheets result in strong steric effects and prevent aggregation of dispersed CNTs. The high aspect ratio of both GO and CNTs also facilitates the formation of a continuous network or gelation even at a relatively low concentration.

Apart from HiPCO CNTs, we have tested single-walled CNTs prepared by laser ablation, multi-walled by chemical vapour deposition as well as carbon black. All of the carbon materials tested can be suspended in water by GO under sonication. We also found dried GO/CNT composites could be readily redispersed in water by mild sonication. These results clearly indicate that GO sheets can function as exceptional dispersants for carbon nanomaterials in aqueous solution without any additional organic solvents or pre-functionalisation involved.

Two groups have recently demonstrated that CNTs and GO can be co-dispersed in anhydrous hydrazine[4a] or DMF.[11] Graphene/CNT hybrid films can be fabricated from the hydrazine dispersion.[4a] However, in both cases, the CNTs employed were pre-functionalised with oxygen-containing groups, providing reasonable solubility in polar solvents, including DMF and hydrazine. The role of GO in the dispersion of CNTs therefore remains unclear. Additionally, both DMF and hydrazine are not environmentally friendly solvents.

Dispersing GO/CNT in water enables the use of cost-effective solution-phase processing methods to fabricate GO/CNT hybrid films. Like CNT buckypaper[2f,3a] and GO or graphene paper,[12] GO/CNT hybrid films can be readily

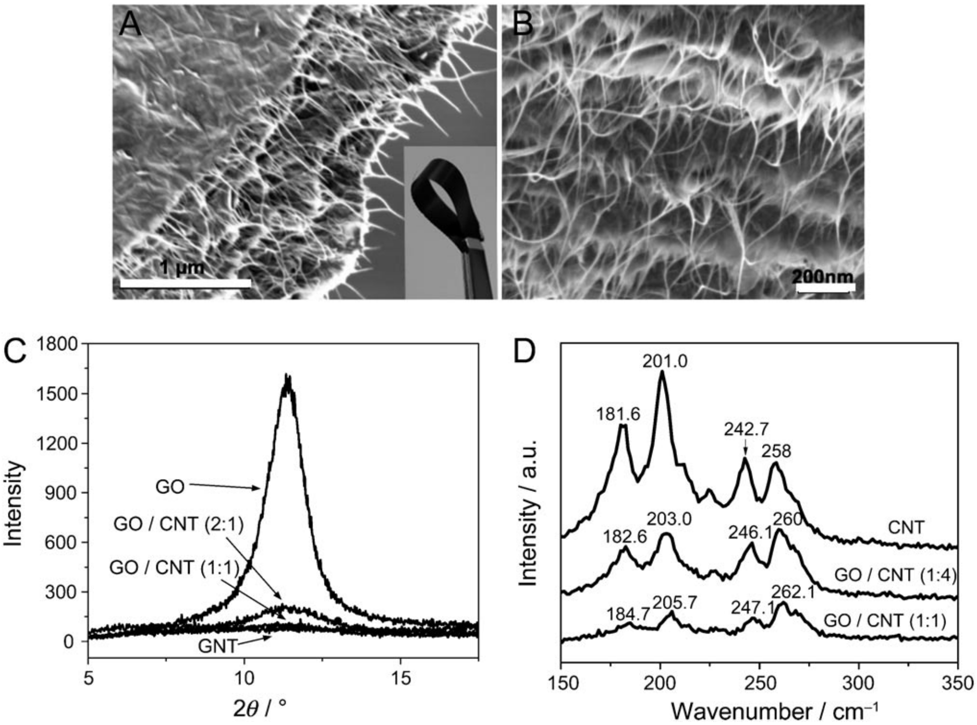

Figure 2. A, B) SEM images of the resulting GO/CNT hybrid film. The sample was torn apart to expose the cross section. As expected, the CNT protrusions on the edge were formed as a result of tearing. The inset in (A) shows a bent strip of the resulting film. XRD patterns (C) and Raman spectra (D) of the GO/CNT films with various weight ratios of GO and CNTs are also shown.

formed by vacuum filtration of the resulting dispersion through a filter membrane. SEM images of an as-fabricated film (Figure 2 A and B) reveal that CNTs and GO sheets are distributed evenly throughout the resultant structure. XRD analysis shows that the diffraction peak at $2\theta$ of about 10.9° (characteristic of stacked GO sheets[12a]) is much weaker than that observed for the CNT-free sample (Figure 2 C), indicating that the restacking of GO sheets into a layered structure has been inhibited by the attachment of CNTs. Raman spectroscopy (Figure 2 D) shows that the radial breathing modes of CNTs in the hybrid samples are shifted to some extent relative to the original CNT sample, also reflecting a significant interaction between CNTs and GO.[13]

The interaction between CNTs and GO makes the resulting hybrid paper very robust. The free-standing hybrid paper shown in Figure 2 A was found to have a Young's modulus of approximately 8.1 GPa, which is lower than that of CNT-free GO paper,[12a] but is much higher than that of GO-free buckypaper prepared by using a similar filtration method.[3a] It is worth noting that the preparation of CNT-only buckypaper requires chemical functionalisation of CNTs and the use of a significant amount of surfactants.[3a] The removal of the surfactants by washing is time consuming and is often difficult to take to completion. In contrast, in our case, surfactants and post-rinsing are not required, allowing substantial time and energy savings.

Interestingly, while GO offers solution processability and mechanical enhancement to CNTs, CNTs were also found to provide benefits to GO. GO is electrically insulating and needs to be converted into conductive graphene for most applications. GO can be deoxygenated to graphene by chemical reduction or thermal treatment.[5a] However, this process generally involves the use of toxic chemicals and/or high temperatures. An earlier report on ultrathin graphite oxide films[14] and a few recent studies[15] show that GO can also be converted to graphene by milder and greener electrochemical reduction processes. However, the insulating nature of GO limits the electrochemical reduction rate and is therefore problematic for large-scale production.[6] Our GO/CNT hybrid films fabricated by either spraying or filtration were found to be rather conductive (Table S1 in the Supporting Information). The incorporated CNTs can serve as an electrical conducting network, which was found to substantially facilitate the electrochemical reduction of GO.

Figure 3 A shows the cyclic voltammograms of a thin GO/CNT (1:1) film deposited on a glassy carbon electrode by drop-casting. During the first cycle, a remarkable cathodic peak appeared at around −1.1 V (vs. Ag/AgCl), which can be attributed to the irreversible electrochemical reduction of the functional groups of GO.[15b] This reduction peak almost disappeared in the following cycles. Note that the onset of reduction is about −0.3 V, which is lower than that of pure GO films (Figure S2 in the Supporting Information), indicating that the incorporation of CNTs can promote the electro-reduction process. This is in agreement with previous studies that CNTs can serve as conducting wires to promote charge transfer in electrochemical reactions.[16] Rapid reduc-

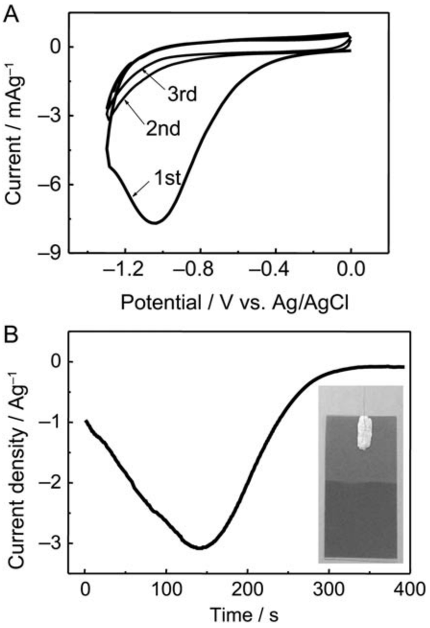

Figure 3. A) Cyclic voltammograms of a GO/CNT (1:1) film drop-cast on a glassy carbon electrode in PBS (0.1 M, pH 7.4) at a scan rate of 50 mV s$^{-1}$. B) Reduction current–time curve of a 12 μm thick GO/CNT (1:1) film with a potential of −1.1 V (vs. Ag/AgCl) being applied. The inset shows a photograph of a thin GO/CNT film deposited on a piece of plastic film by airbrush spraying. The bottom part has been immersed in PBS solution and electrochemically treated by applying a constant potential of −1.1 V through a platinum wire glued to the hybrid film with silver paste.

tion of GO in thick films can be achieved by applying a constant potential of −1.1 V to the film. As shown in Figure 3 B, when a potential of −1.1 V was applied, the reduction current increased with time initially as the reduced electrode become more conductive and the electrode surface area increased. The current then decreased with time as less GO was left unreduced. After a few minutes, the current dropped back to zero, indicative of the completion of the reduction. The inset presents a photograph of a thin GO/CNT film deposited on a plastic film by airbrush spraying. The bottom half was immersed in sodium phosphate buffered saline (PBS) solution with a potential of −1.1 V being applied. The immersed area became darker in appearance after just a few minutes, indicating that electrochemical deoxygenation of GO had occurred. In contrast, no clear colour change appeared after more than 3 h when the CNTs were absent in the GO film. The electrical conductivity of all of the hybrid films increased after the electrochemical treatment and the increment was dependent on the ratio of GO to CNTs (Table S1 in the Supporting Information).

After the electrochemical reduction, the GO/CNT films still remain free-standing and mechanically robust. Note that insulating binders that are traditionally used in many carbon-based devices, such as supercapacitors, are not required to make these films. These features make the resultant hybrid films very attractive for use as electrochemical electrodes in light-weight and flexible energy-storage and conversion devices. Of particular interest is that, as discussed below, synergistic effects were also observed in the devices based on the graphene/CNT hybrid films.

GO-derived graphene, due to its low production cost, has recently provided the basis of nanostructured electrodes for energy-storage/-conversion devices, such as supercapacitors[2b] and batteries.[2c] However, as we have demonstrated recently, graphene sheets, when assembled together, tend to restack as a graphite-like structure due to relatively strong intersheet van der Waals interactions.[17] Restacking makes the resulting films less porous and electrolyte infiltration more difficult when the structures are used as electrochemical electrodes. As described above, the restacking of graphene sheets in the hybrids can be inhibited by the attachment of CNTs. Thus, the intercalation of CNTs should facilitate the diffusion of electrolytes. This was confirmed by our electrochemical impedance analysis. As shown in the Nyquist plots (Figure 4 A), the projected length of the Warburg-type line on the real axis, which characterises the ion diffusion process from solution into the electrode structure, is related to the ratio of ER-GO and CNTs. The Warburg-type line became shorter as the ratio of ER-GO to CNTs decreased, reflecting improved ion diffusion across the film.

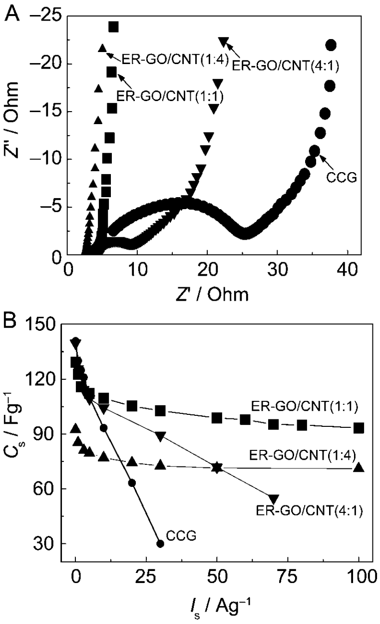

Figure 4. A) Nyquist plots of the electrochemically reduced GO (ER-GO)/CNT films. The high-frequency intercept at the $x$ axis corresponds to the internal or equivalent series resistance of the film electrodes. B) Gravimetric capacitance of the ER-GO/CNT films measured at various charge/discharge currents. The data for CNT-free chemically converted graphene (CCG) films are also presented for comparison.

We have fabricated prototype supercapacitors using the ER-GO/CNT hybrid films as electrodes to further demonstrate the benefits of hybrid structure. As revealed by the cyclic voltammograms and galvanostatic charge/discharge curves (Figures S3 and S4 in the Supporting Information), the incorporation of CNTs in graphene sheets had a significant effect on their electrochemical properties. Supercapacitors fabricated from ER-GO/CNT hybrids with a lower content of CNTs exhibited a higher specific capacitance at a low charge/discharge current (Figure 4B), which was likely to be due to the higher specific surface area of graphene than CNTs. The gravimetric capacitance for the graphene-only sample is approximately 140 $\mathrm{F g^{-1}}$ at a charge/discharge current of 0.1 $\mathrm{A g^{-1}}$, which is comparable to the value reported in the literature.[2b] However, the capacitance measured at high charge/discharge rates decreased rapidly (Figure 4B). The capacitance for the graphene-only sample dropped to 30 $\mathrm{F g^{-1}}$ at a charge/discharge current of 30 $\mathrm{A g^{-1}}$. The inhibition of ion diffusion due to restacking of graphene sheets was believed to be responsible for this deterioration in device performance. As presented in Figure 4, this problem can be simply solved by distributing CNTs throughout the graphene sheets. As the CNT content was increased, the capacitance became less affected by the value of charge/discharge currents. ER-GO/CNT (1:1) based supercapacitors displayed a specific capacitance of over 90 $\mathrm{F g^{-1}}$ at a high charge/discharge rate of 100 $\mathrm{A g^{-1}}$. Given supercapacitors are normally operated at high currents, such improvement is of significance.

Both our four-probe measurement (Table S1 in the Supporting Information) and impedance analysis (Figure 4A) reveal that hybrid films containing more CNTs are more electronically conductive. Recent studies[18] have shown that chemically reduced GO contains a considerable amount of defects, which limit the conductivity of chemically prepared graphene. The CNTs used in our hybrids are not chemically modified and therefore are highly conductive. The increased electrical conductivity may have also contributed to the improvement in the performance of supercapacitors.

In conclusion, we have demonstrated that GO can serve as a superior dispersant to disperse pristine CNTs into water to form stable suspensions through supramolecular interactions. The hybridisation of GO with CNTs can promote the electrochemical conversion of GO to graphene. These discoveries offer a very simple, cost effective and environmentally friendly strategy to address the long-standing processablity issue of CNTs. It also allows convenient integration of the 1D CNTs with the 2D graphene to form hierarchically structured carbon nanohybrids with enhanced performances. Given that CNTs are essentially rolled graphene sheets and GO is partially oxidised graphene, this work presents an excellent example that fascinating synergetic effects could exist between different graphene derivatives. We expect that chemical and/or electrical synergies between graphene derivatives will play an important role in the future development of carbon-based materials. Additionally, this work suggests that GO should be, more accurately, viewed as an amphiphilic molecule. This means that GO, like other conventional amphiphilic surfactants, may be further used to disperse other nanostructures, which will enable solution-phase processing of many new graphene-based nanohybrids.

## Experimental Section

**Dispersion of CNTs by GO**: GO dispersions were prepared by using the procedure we reported previously.[12b] Unfunctionalised single-walled CNTs synthesised by HiPCO were purchased from Carbon Nanotechnologies. In a typical dispersing procedure, 0.05 wt % GO colloid (20 mL) was added to a vial pre-loaded with CNTs (10 mg). The mixture was then subjected to pulsed sonication for 1 h (2 s on and 1 s off) in an ice bath using a Vibra-Cell sonicator with a horn of 13 mm in diameter (VCX750, 750W, 30 % amplitude). A uniform and opaque black colloidal dispersion was obtained for characterisation and further processing. The concentration of GO and the weight ratio of GO and CNTs were varied to study their influence on the dispersibility. Electrophoresis experiments were carried out by applying a voltage of 30 V (DC power supply, GPR-6–010) for 30 min to a diluted dispersion of GO-CNTs (0.0025 wt % of GO and CNTs) and GO dispersion (0.0025 wt %) in a U-type tube with two platinum wires as electrodes. The CNT-free GO dispersion gave a similar result to GO/CNT dispersion shown in Figure 1C.

**Fabrication of GO/CNT films**: Free-standing GO/CNT papers were prepared by vacuum filtration of GO/CNT dispersion through an Anodisc membrane filter (47 mm in diameter, 0.2 μm pore size, Whatman) using a similar method that was previously used to make GO and graphene paper.[12,17] Unless specifically stated, the samples used for electrical, mechanical and electrochemical measurements were prepared by vacuum filtration.

**Electrochemical reduction of GO in GO/CNT films**: GO/CNT films prepared by drop-casting, vacuum filtration and air-brush spraying were all tested for electrochemical reduction. For the drop-cast film, 5 μL of as-obtained GO/CNT (1:1) dispersion was spread on a glassy carbon electrode (GCE) and the electrode was dried at ambient temperature. As-obtained GO/CNT-modified GCE was used as working electrode, a platinum wire as the counter electrode and the Ag/AgCl (KCl-saturated) electrode as the reference electrode. The potential was scanned from 0 to −1.3 V (vs. Ag/AgCl) at a scan rate of 50 $\mathrm{mV s^{-1}}$. For the electro-reduction of the films prepared by vacuum filtration, a platinum wire was first clipped onto the film and then a potential of −1.1 V (vs. Ag/AgCl) was applied to the film for about 7 min. PBS solution (0.1 M, pH 7.4) was used as electrolyte for all the electrochemical reduction experiments.

**Fabrication and test of supercapacitors**: Supercapacitors based on ER hybrid films were assembled with the two-electrode configuration by using a similar procedure that was recently reported by Hu et al.[2a] with aqueous $H_2SO_4$ (1.0 M) as the electrolyte. For comparison, supercapacitors based on chemically converted graphene paper were also assembled. The graphene paper was prepared by using the method we recently reported.[17] The electrochemical properties of the supercapacitors were studied by cyclic voltammetry, electrochemical impedance spectroscopy and galvanostatic charge-discharge cycling by using a VersaSTAT 3 potentiostat.

**Characterisation**: UV/Vis absorption spectra were measured with a Carry 300 UV/Vis spectrophotometer (Varian). XRD patterns were recorded on a Philips 1130 X-ray diffractometer (40 kV, 25 mA, $\mathrm{Cu_{K\alpha}}$ radiation, $\lambda = 1.5418$ Å) at room temperature. SEM images were obtained by using a Zeiss ULTRA plus scanning electron microscope at an accelerating voltage of 1 kV and a working distance of 2.9 mm. Raman spectra were obtained with an excitation line at 514.5 nm from an Ar laser (Renishaw Invia). Specially adapted research grade 10X Leica microscope focused the excitation beam onto the samples. The data acquisition time used in the measurement was 20 s. The electrical conductivity of the hybrid films was carried out on a Jandel 4-point Conductivity Probe by using a linear arrayed four-point head. The thicknesses of the films were determined on a JEOL 840A scanning electron microscope.

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**Keywords:** graphene · hybrid materials · nanotubes · supercapacitors · synergistic effects

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[19] After the acceptance of our manuscript, we noticed that Huang and co-workers have just reported that GO could serve as a surfactant to disperse CNTs (J. Kim, L. J. Cote, F. Kim, W. Yuan, K. R. Shull, J. Huang, *J. Am. Chem. Soc.* **2010**, *132*, 8180–8186).

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