Development and study of low-dimensional hybrid and nanocomposite materials based on layered nanostructures
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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2017

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Download date: 31-03-2020
Chapter 3
A bottom-up approach for the synthesis of highly ordered fullerene-intercalated graphene hybrids

Much of the research effort on graphene focuses on its use as a building block for the development of new hybrid nanostructures with well-defined dimensions and properties suitable for applications such as gas storage, heterogeneous catalysis, gas/liquid separations, nanosensing and biomedicine. Toward this aim, here we describe a new bottom-up approach, which combines self-assembly with the Langmuir-Schaefer deposition technique to synthesize graphene-based layered hybrid materials hosting pristine fullerene molecules within the interlayer space. Our film preparation consists in a bottom-up layer-by-layer process that proceeds via the formation of a hybrid organo-graphene oxide Langmuir film. The structure and composition of these hybrid fullerene-containing thin multilayers deposited on hydrophobic substrates were characterized by a combination of X-ray diffraction, Raman and X-ray photoelectron spectroscopies, atomic force and scanning electron microscopies and conductivity measurements. The latter revealed that the presence of C$_{60}$ within the interlayer spacing leads to an increase in electrical conductivity of the hybrid material as compared to the organo-graphene matrix alone.

This chapter is based on the article: "A bottom-up approach for the synthesis of highly ordered fullerene-intercalated graphene hybrids", by A. Kouloumpis, K, Dimos, K. Spyrou, V. Georgakilas, P. Rudolf and D. Gournis. Frontiers in Materials 2015, 2, 10
3.1 Introduction

The outstanding mechanical, thermal and electrical properties of graphene have attracted a lot of scientific effort aimed at exploiting them in the development of new hybrid nanostructures with well-defined dimensions and behaviour, which contain graphene as building block.\(^1\),\(^2\) In fact, graphene sheets can be used as templates for the synthesis of novel intercalated carbon-based materials suitable for various applications in gas storage, gas/liquid separation, heterogeneous catalysis, energy storage, Li-ion batteries, supercapacitors, nanosensing and biomedicine.\(^1\),\(^3\)–\(^7\) Combining graphene's properties with the extraordinary properties of fullerenes\(^8\),\(^9\) by incorporating the latter into well-defined graphene-based hybrid thin multilayers continues to be a challenging new field for developing novel hybrid nanocomposites.

Here we describe a new bottom-up layer-by-layer approach for the production of graphene hybrid materials where graphene acts as the structure directing interface and reaction media. This method, based on combining self-assembly with the Langmuir-Schaefer (LS) deposition technique, uses the graphene nanosheets as a template for incorporating \(\text{C}_{60}\) molecules in a bi-dimensional array and allows for perfect layer-by-layer growth with control at the molecular level.\(^10\) Similar synthetic protocols have been reported during the last decade for the development of hybrid multilayers and monolayers using the Langmuir Blodgett (LB) technique. Layered materials like clay minerals,\(^10\)–\(^20\) graphene and/or graphene oxide,\(^13\),\(^21\)–\(^23\) or other carbon-based nanostructures\(^24\)–\(^26\) have been used to produce hybrid thin multilayers with unique properties. Clay minerals with amphiphilic species and other complexes were combined to fabricate monolayers and multilayers used as photoprobes,\(^27\) sensors,\(^18\) catalysts\(^20\) and photomagnetic films.\(^17\),\(^28\) Moreover, the optoelectronic and mechanical properties of graphene can be modified, tuned, or enhanced by layer-by-layer assembly techniques such as the LB method, as has been reported in various studies over the last 3-4 years (for a review see\(^29\)). High performance dye-sensitized solar cells,\(^30\) and transparent conductive films\(^31\) are some examples of the uses of hybrid graphene-based thin multilayers. Finally, the advantages of the precise control and the homogeneous deposition over large areas makes the LB technique...
promising for preventing carbon-based nanostructures from agglomerating during the film synthesis, as demonstrated for fullerene derivatives or carbon dots.  

Our film preparation approach involves a bottom-up layer-by-layer process that starts with the formation of a hybrid organo-graphene Langmuir film and proceeds via two self-assembly steps to create a layered structure hosting fullerene molecules within its interlayer space (a schematic representation of the synthetic procedure is illustrated in Scheme 3.1). The composition, structure and transport properties of the fullerene-containing hybrid thin multilayers deposited on hydrophobic substrates were characterized by X-ray diffraction, Raman and X-ray photoelectron spectroscopies, atomic force and scanning electron microscopies and electrical conductivity measurements.

Scheme 3.1. Schematic representation of the synthetic procedure followed for the synthesis of the hybrid GO/C\textsubscript{60} multilayer film consisting in a Langmuir Schaefer deposition (1\textsuperscript{st} step) combined with two self-assembly steps (2\textsuperscript{nd} and 3\textsuperscript{rd} step).
3.2 Experimental Section

3.2.1 Materials

Sulfuric acid (95-97%), nitric acid (65%), potassium chlorate (≥99%) and powder graphite (purum, ≤ 0.2 mm) were supplied from Fluka. Octadecylamine (ODA, ≥99%), acetone, methanol and ethanol were purchased from Sigma-Aldrich while fullerene (C_{60}, powder, 99% C) was obtained from Alfa Aesar. Ultrapure deionized water (18.2 MΩ) produced by a Millipore Simplicity® system was used throughout. The Si-wafer (P/Bor, single side polished, purchased from Si-Mat) substrates were cleaned prior to use by 15 min ultrasonication in water, acetone and ethanol. All reagents were of analytical grade and were used without further purification.

3.2.2 Synthesis of graphene oxide

Graphene oxide (GO) was synthesized using a modified Staudenmaier method:$^{13,32,33}$ 10 g of powdered graphite were added to a mixture of 400 mL of 95-97% H_{2}SO_{4} and 200 mL of 65% HNO_{3}, while cooling in an ice-water bath. 200 g of powdered KClO_{3} were added to the mixture in small portions under vigorous stirring while cooling in the ice-water bath. The reaction was quenched after 18 h by pouring the mixture into ultrapure water; the oxidation product was washed until the pH reached 6.0 and finally dried at room temperature.

3.2.3 Preparation of hybrid graphene/fullerene multilayers

A KSV 2000 Nima Technology Langmuir Blodgett trough was used for the preparation and deposition of multilayers at a temperature of 21 ± 0.5°C. The surface pressure in the LB trough was monitored with a Pt Wilhelmy plate. The deposition protocol is schematically illustrated in Scheme 1. A GO suspension in ultrapure water (0.02 mg mL^{-1}) was used as subphase. 200 μL of a 0.2 mg mL^{-1} ODA solution in chloroform/methanol 9/1 (v/v) were spread onto the water surface using a microsyringe to achieve the hybridization of GO sheets by covalent bonding via the amide functionality. After a waiting time of 20 min to allow for solvent evaporation and GO-surfactant functionalization to occur, the hybrid ODA-GO layer was
compressed at a rate of 5 mm min\(^{-1}\) until the chosen stabilization pressure of 20 mN m\(^{-1}\) was reached. As in any classical Langmuir Blodgett experiment, the applied pressure pushes the surfactant molecules along the water surface; the grafted graphene oxide sheets will simply follow that movement and therefore become packed, depending on the surface tension established in the trough.\(^{13}\) This pressure was maintained throughout the deposition process. Layers were transferred onto the hydrophobic Si-wafer substrates by horizontal dipping (this way of transferring is known as Langmuir-Schaefer technique), with downward and lifting speeds of 10 and 5 mm min\(^{-1}\), respectively\(^{23} - 1^{st}\) step in Scheme 3.1. After the horizontal lift of the substrate, the 2\(^{nd}\) step of the deposition protocol consisted in a surface modification of the GO nanosheets, induced by bringing the surface of the transferred Langmuir film (ODA-GO) in contact with a solution of ODA surfactant (self-assembly) dissolved in methanol (0.2 mg mL\(^{-1}\))\(^{13}\) as illustrated in Scheme 3.1. In the 3\(^{rd}\) and final step, the hybrid organo-GO film was lowered into a solution of C\(_{60}\) in toluene (0.2 mg mL\(^{-1}\)) for the formation of a hybrid graphene/fullerene monolayer by self-assembly. A hybrid multilayer film was constructed by repeating this 3-step-dipping cycle 40 times, as shown in Scheme 3.1 (sample denoted as ODA-GO-ODA-C\(_{60}\)). Each time when the substrate was lowered towards the liquid surface, it was allowed to touch the air-water interface or the solution surface in a very gentle dip of max 0.5 mm below the surface for 90 s. After each deposition step the sample was rinsed several times by dipping into ultrapure water to eliminate any weakly attached molecules that remained from the deposition step and dried with nitrogen flow to avoid contaminating the Langmuir film air-water interface or the solution employed in the following step.\(^{10, 21}\) For comparison an organo-GO hybrid multilayer (40 layers) was also fabricated under the same experimental conditions without the 3\(^{rd}\) step of C\(_{60}\) incorporation (sample denoted as ODA-GO-ODA). Moreover, a reduction and an annealing step (4\(^{th}\) step) were also performed in both multilayers in order to convert the deposited GO to graphene and thus to increase the conductivity. For this, the deposited multilayers were immersed into an aqueous solution of NaBH\(_{4}\) (1 mg mL\(^{-1}\)) for 10 min and annealed at 700 °C for 1h under argon (samples denoted as ODA-rGO-ODA-C\(_{60}\) and ODA-rGO-ODA). This treatment results in a partial reconstruction of the graphene mesh and consequently causes a drastic increase in conductivity.\(^{13}\)
3.3 Results and Discussion

3.3.1 Structure control of hybrid ODA-GO layer

The surface pressure versus molecular area (π-Α) isotherms of ODA monolayers in pure water and on GO dispersion are shown in Figure 3.1 (right). The curves show the change in the slope corresponding to the phase transitions of ODA-GO sheets from gas to condensed-liquid and then to solid state during the compression process.\textsuperscript{21} In the absence of GO, the π-Α isotherm is a smoothly increasing curve with a lift off area of 32.8 Å\(^2\). When adding a small amount of GO (0.02 mg mL\(^{-1}\)) to the aqueous subphase, the lift-off area increased to 52 Å\(^2\), verifying that GO flakes cause a stabilization effect on the ODA layer\textsuperscript{21} through chemical grafting (covalent bonding) of the terminal amine groups of ODA to the epoxide groups on the top side of the GO sheets. The amine end groups interact via a ring opening reaction (nucleophilic substitution reactions) of the epoxide groups of GO.\textsuperscript{13, 34, 35} At surface pressures above 40 mN m\(^{-1}\) the monolayer collapses due to the formation of bilayers at approximately 26 Å\(^2\).\textsuperscript{21}

![Figure 3.1](image_url)

**Figure 3.1.** (Left panel) π-Α isotherms of ODA monolayers in pure water and on an aqueous dispersion of 0.02 mg mL\(^{-1}\) GO. (Right panel) The black curve corresponds to the trough area covered by the hybrid ODA-GO Langmuir film at the air-water surface and the blue curve shows the surface pressure throughout (around 20 mN m\(^{-1}\)) the deposition of a 10-layer ODA-GO-ODA-C\(_{60}\) hybrid film.
The successful transfer of the hybrid Langmuir films onto the Si-wafer substrate can be deduced from the plot of the pressure measured at the surface of the subphase in the LB trough versus time.\textsuperscript{10} Figure 3.1 (right) shows the time dependence of the total trough area of the hybrid Langmuir film. As the substrate is dipped into the subphase, this area reduces due to the transfer of one hybrid layer from the air-water interface to the substrate during each dip. This transfer is visible as a sharp step in the curve.\textsuperscript{10, 11} If the step height (which gives an area value) is equal to the substrate surface area, the transfer ratio is 1 and the substrate surface is 100% covered by the hybrid layer each time it is lowered into the subphase. A different transfer ratio suggests a multilayer transfer or an incomplete coverage of the substrate.\textsuperscript{10} The curve shown in Figure 3.1 is a typical one recorded during the deposition of a 10-layer hybrid ODA-GO-ODA-C\textsubscript{60} film. The transfer ratio was very close to 1 throughout the deposition, testifying to the successful transfer of the hybrid Langmuir film at each dip of substrate into the LB trough.

Representative AFM images of hybrid graphene oxide sheets (ODA-GO) deposited on Si-wafer with the LS method (at surface pressure 20 mN m\textsuperscript{-1}) during the first dip into the LB trough (1\textsuperscript{st} step in Scheme 3.1) are shown in Figure 3.2. In these images one recognizes high quality graphene oxide flakes with well-defined edges and a relatively low amount of cracks and wrinkles. The surface coverage is quite high; the GO platelets in the transferred layer are contacting each other, with hardly any overlap but only small voids between them, forming a nearly continuous, close-packed array. The average height of the flakes is 0.9-1.5 nm as derived from topographical height profile is larger than the value of 0.61 nm predicted for a single graphene oxide layer,\textsuperscript{36} as expected due to the ODA layer below the GO sheet. However, it is difficult to conclude anything on the orientation of the ODA molecules (straight or inclined) from these images because the height of single GO layers without ODA has also been found in the literature to be of the order of 1.1±0.2 nm (see for example\textsuperscript{37, 38}) probably due to presence of adsorbed water molecules.
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3.3.2 Characterization of hybrid graphene/fullerene multilayers

The X-ray diffraction pattern of a ODA-GO-ODA-C\textsubscript{60} hybrid multilayer (40 layers) is shown in Figure 3.3 in comparison with a ODA-GO-ODA hybrid multilayer (40 layers) that was synthesized under the same experimental conditions. The ODA-GO-ODA-C\textsubscript{60} hybrid multilayer shows a 001 diffraction peak at 2\(\theta\)=2.2° which corresponds to a \(d\)\textsubscript{001}-spacing of 40 Å. This value is higher compared with the corresponding value of the ODA-GO-ODA multilayer (\(d\)\textsubscript{001}=37.6 Å) testifying to the successful intercalation of the fullerene between the organo-graphene nanosheets. This increment does not correspond to the size of C\textsubscript{60} (~7 Å) implying that the fullerene molecules are accommodated between the double alkyl chains of the surfactant (see inset sketch) and not on top of it. Moreover, the absence of higher order (001) reflections in the XRD pattern of ODA-GO-ODA-C\textsubscript{60} suggests that the graphene oxide layers are not stacked in perfect registry but have slipped sideways and are turbostratically disordered.
A similar behaviour has been observed upon intercalation of fullerene derivatives in aluminosilicate clay minerals. This hypothesis is further supported by calculating the size of the coherently diffracting domains along the c axis (also called mean crystalline thickness, t) that give rise to the 001 diffraction peak, given by the Debye-Scherrer equation, \( t = \frac{K \lambda}{\beta \cos \theta} \), where \( K \) is a constant near unity (\( K = 0.91 \)), \( \lambda \) the X-ray wavelength (\( \lambda = 1.5418 \) Å), \( \theta \) the angular position of the first diffraction peak, and \( \beta \) the full width at half maximum of the 001 peak expressed in radians. The crystalline thickness (t) along c axis of the ODA-GO-ODA-C\(_{60}\) and ODA-GO-ODA multilayers is found to be 114 and 153 Å, respectively. Dividing these values with the corresponding \( d_{001} \)-spacings, the number of stacked layers can be calculated to be ~3 for ODA-GO-ODA-C\(_{60}\) and ~4 for ODA-GO-ODA, confirming the high degree of ordering of the produced multilayers.

Raman spectra of the hybrid ODA-GO-ODA-C\(_{60}\) and the ODA-GO-ODA multilayer samples are shown in Figure 3.4. Spectra are typical of GO materials, in that both exhibit the characteristic first-ordered G- and D-bands at around 1600 and 1350 cm\(^{-1}\), respectively. The G-band is associated with \( sp^2 \) hybridized carbon atoms and originates from the doubly degenerate zone center \( E_{2g} \) mode. The D-band is

![Figure 3.3. XRD patterns of ODA-GO-ODA-C\(_{60}\) and ODA-GO-ODA hybrid multilayers (40 layers).](image_url)
correlated with sp³ hybridized carbon atoms, as it requires a defect for its activation by double resonance, thus indicating the presence of lattice defects and distortions.⁴⁰, ⁴¹ The ratio of the D- to G-band intensities (I_D/I_G) is indicative of the quality of the graphitic lattice. This ratio is equal to 1.06 for the ODA-GO-ODA-C₆₀ multilayer and 1.04 for the ODA-GO-ODA multilayer, implying that fullerene intercalation into the interlayer space of the organo-graphene nanosheets does not influence the GO structure. In addition, the shape of the two spectra in the 2D region is alike; both exhibit two broad peaks at ~2700 and ~2930 cm⁻¹. The low intensity 2700 cm⁻¹ peak is attributed to the 2D (or else G') vibrational mode, which is an overtone (second order) of the D peak. The second peak at ~2930 cm⁻¹ is assigned to the mode D+D', originating from the combination of phonons with different momenta, thus requiring defects for its activation; its intensity agrees with the defective nature of the GO lattice revealed by the high I_D/I_G ratios of both samples.⁴⁰-⁴³ The typical pentagonal pinch mode (A_g(2)) of C₆₀ that is expected between 1440 and 1470 cm⁻¹⁴⁴ is not visible since it is superimposed on the broad D band of graphene oxide. Moreover, upon reduction and annealing (see Figure 3.4 inset), the GO sheets show a noticeable decrease in the D/G ratio from 1.06 to 0.76.

**Figure 3.4.** Raman spectra of ODA-GO-ODA-C₆₀ and ODA-GO-ODA hybrid multilayers (40 layers), Inset: Raman spectrum of the ODA-rGO-ODA-C₆₀ hybrid multilayer after reduction and annealing.
This observation suggests that while most of the oxygenated groups are removed (in the form of CO or CO$_2$), the relative amount of disordered sp$^2$-hybridized atoms is still high. The later could be explained either by the fact that amine molecules covalently bound to graphene survive the reduction and annealing procedures (contributing to an enhanced sp$^3$ hybridization) or that vacancies created during production of GO remain unchanged by the reduction process and ultimately define the intact graphene regions.$^{13, 37, 45}$

The C1s core level X-ray photoelectron spectra of the graphene/fullerene hybrid multilayers before (ODA-GO-ODA-C$_{60}$) and after reduction and annealing (ODA-rGO-ODA-C$_{60}$) in comparison with pristine GO and the organo-GO multilayers (ODA-GO-ODA) are shown in Figure 3.5. The spectrum of ODA-GO-ODA displays four main contributions at 285.0 eV, 286.1 eV, 287.5 eV and 289.3 eV corresponding to different carbon environments while for ODA-GO-ODA-C$_{60}$ an additional contribution appear at 290.5 eV. The peak at 285.0 eV is attributed to the C-C bonds of the aromatic ring of GO as well the organic carbon-carbon groups of ODA attached to graphene and represents 53.7% of the total carbon 1s intensity of ODA-GO-ODA (Figure 3.5 top-right). This contribution is more intense in the case of ODA-GO-ODA-C$_{60}$ (Figure 3.5 bottom-left) because in addition to photoemission signal due to the C-C bonds of ODA and GO as also that from the C$_{60}$ cage appears at this binding energy. The increased intensity of this peak to 57.9% of the total C1s signal testifies therefore to the successful incorporation of C$_{60}$ into the layered structure. C-O and C-N both contribute to the photoelectron peak centred at 286.1 eV for ODA-GO-ODA and 286.5 eV for ODA-GO-ODA-C$_{60}$; in both spectra its intensity is higher than in the corresponding spectrum of the GO starting material due to the chemical grafting of the amino groups of ODA with the epoxy groups of GO. Similarly, the contribution at 287.5 eV in the ODA-GO-ODA spectrum (at 288.0 eV for ODA-GO-ODA-C$_{60}$), assigned to the epoxy groups of GO, is smaller than for the GO starting material, attesting that the epoxy groups have reacted with the amines via chemical grafting. The peak at 289.3 eV in the ODA-GO-ODA spectrum (at 289.1 eV for ODA-GO-ODA-C$_{60}$) is attributed to carboxyl groups created during the oxidation of GO. The additional peak at 290.5 eV in the ODA-GO-ODA-C$_{60}$ spectrum is due to the C1s shake up features of C$_{60},^{46}$ which result from π-π* transitions excited in the
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photoemission process; this peak therefore gives additional support to the presence of fullerenes in the hybrid multilayer.\textsuperscript{10, 47} Finally, upon reduction and subsequent annealing at 700°C the C1s spectral intensity due to oxygen-containing functional groups of graphene oxide (Figure 3.5 bottom-right) is significantly lower as compared to before the treatment, indicating a partial reconstruction of the graphene network expected to result in a better electrical conductivity (see below). It is noteworthy to mention that a new photoemission peak appears in the same spectrum at 282.9 eV; we attribute this peak to C-Si bonds formed as a result of the annealing procedure. Finally, no contribution in N1s photoelectron peak was observed in both ODA-rGO-ODA and ODA-rGO-ODA-C\textsubscript{60} samples indicating that annealing didn’t cause any nitrogen doping in the graphene layers.

![X-ray photoelectron spectra of the C1s core level region of GO, ODA-GO-ODA and graphene/fullerene hybrid multilayers (40 layers) before (ODA-GO-ODA-C\textsubscript{60}) and after reduction and annealing (ODA-rGO-ODA-C\textsubscript{60}).](image)

Figure 3.5. X-ray photoelectron spectra of the C1s core level region of GO, ODA-GO-ODA and graphene/fullerene hybrid multilayers (40 layers) before (ODA-GO-ODA-C\textsubscript{60}) and after reduction and annealing (ODA-rGO-ODA-C\textsubscript{60}).
The sheet resistance measurements of the ODA-GO-ODA and ODA-GO-ODA-C\textsubscript{60} multilayers gave values of about 15-20 MΩ and 10 MΩ, respectively. These values were remarkably reduced to 20 kΩ and 4 kΩ, respectively, after the reduction and annealing treatment was applied in order to convert the deposited GO to graphene.

These values are mean values of all measurements performed in different spots on both samples. By taking into account the thickness (as estimated from AFM section analysis) of the produced films (70 nm for ODA-rGO-ODA and 90 nm for ODA-rGO-ODA-C\textsubscript{60}) the electrical conductivity of the produced films was estimated 714 S·m\textsuperscript{-1} and 2800 S·m\textsuperscript{-1} for the reduced and annealed samples without and with the C\textsubscript{60} respectively. It is worth to notice that the produced multilayer films have a quite uniform and smooth surface as revealed by SEM measurements (Figure 3.6). According to the roughness analysis of the surface shown in the inset of Figure 3.6, the mean roughness (RMS) is estimated around 3 nm. The poor electrical conductivity of both the ODA-GO-ODA and the ODA-GO-ODA-C\textsubscript{60} multilayer is due to the presence of ODA and of the oxygen-containing groups decorating the graphene oxide sheets. However, the reduction and annealing steps substantially improved the electrical conductivity by removing effectively these moieties. The difference in sheet resistance between the two multilayers indicates a strong influence of the presence of C\textsubscript{60}. It is clear that the presence of C\textsubscript{60} inserted between graphene layers increases the electrical conductivity by an order of magnitude.

Figure 3.6. Representative SEM image of ODA-rGO-ODA-C\textsubscript{60}. Inset: surface roughness analysis of the same sample as shown by AFM micrograph.
3.4 Conclusions

In conclusion, a highly controllable layer-by-layer synthetic approach for the production of a new class of hybrid carbonaceous structures was discussed. A 40-layer thick film consisting of organo-modified graphene oxide layers accommodating pure fullerene molecules (C\textsubscript{60}) in the interlayer space was successfully fabricated by using a combination of the Langmuir-Schaefer deposition method with two self-assembly steps from solution. Initially, the effectiveness of this method in terms of coverage, uniformity and single-layer-level control of the assembly of the first organo-modified graphene oxide layer was confirmed by \pi-A isotherm measurements and AFM microscopy. For the hybrid multilayer sample X-ray diffraction measurements revealed the presence of the fullerene molecules within the interlayer space between the turbostratically layered organo-graphene oxide nanosheets and confirmed the high degree of ordering of the produced structure. The existence of fullerenes in the hybrid multilayer system was confirmed by X-ray photoelectron spectroscopy while Raman spectroscopy showed that the intercalation of the fullerene within the interlayer space of the organo-graphene oxide nanosheets did not affect the structure of GO itself. Finally, a considerable improvement (an order of magnitude) of the electrical conductivity of the hybrid multilayer with respect to the organo-graphene analogue was observed due to the presence of C\textsubscript{60} inserted between graphene oxide layers. This graphene/fullerene hybrid material constitutes a novel hybrid system that could ideally be used in diverse applications such as transparent electrodes, thin film transistors, supercapacitors or lubricants.
3.5 References


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