Quantifying the influence of graphene film nanostructure on the macroscopic electrical conductivity

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Abstract

Graphene films have emerged as a promising nanostructured material class to exploit graphene’s outstanding nanostructured properties on the macroscale. Their potential applications include solar cells (Eda et al 2008 Appl. Phys. Lett. 92, 233305; Müllen et al 2008 Nano Lett. 8, 323–7), antennas (Zhang et al 2018 Electronics 7, 285; Song et al 2018 Carbon 130, 164–9), or electromagnetic interference shielding (Zhou et al 2017 Nanoscale 9, 18613–8; Wan et al 2017 Carbon 122, 74–81; Wang et al 2018 Small 14, 1704332), all of which require a high electrical conductivity. While an outstanding electrical conductivity is a key feature of pristine graphene monolayers, the transfer to the macroscale is challenging. Here, we combined theory and experiment to quantify the impact of specific structural graphene film properties. We synthesized graphene films with systematically varied flake sizes, studied their electrical conductivities, and found excellent agreement to simulations with a three-dimensional random resistor network model. In a further percolation-type study, we computed the critical share of non-conductive elements in a graphene film \( \theta_c = 10\% \) where a substantial loss of electrical conductivity occurs. We prepared mixed films from graphene and graphene oxide to validate the threshold experimentally. In combination, experiments and simulations provide a coherent picture of how the graphene film microstructure is related to the macroscopic electrical conductivity (Rizzi et al 2018 ACS Appl. Mater. Interfaces 10 43088–94; Rizzi et al 2019 Comput. Mater. Sci. 161, 364–70). Our findings provide valuable insights for the production of highly conductive graphene-based macro-materials.

1. Introduction

In recent years, graphene has made its way from research laboratories into the first commercial products [1, 2]. Production methods have evolved to a point where graphene is obtained on a ton scale, although not yet of the highest quality [3]. Besides the seemingly natural application of graphene on the nanoscale, more and more bulk nanostructured materials are being developed as well. Examples include graphene fibers [4, 5], foams and aerogels [6, 7], or graphene-based composites [8]. Application-wise, one of the most promising macro-materials is the graphene film [1, 2], a flat layered assembly of graphene flakes. In contrast to graphite, the flakes are not orderly stacked, but twisted and shifted against each other. Graphene films are mechanically flexible, they can be bent or folded without breaking [9], they are electrically and thermally conductive [9, 10], and they are robust to many chemicals [11]. The first films were synthesized in 2007 by Dikin et al [12], at that time in the form of graphene oxide (GO) films. Since then, numerous possible applications have been studied.
electrodes [13, 14], antennas [15, 16], electromagnetic interference shielding [17–19], flexible radiofrequency filters [20], heat spreaders [9, 21], and many more. Graphene-based thermal management films are already being used in smartphones, which represents a major large-scale application [2].

For various graphene film applications, the electrical conductivity plays an important role. Recently, we presented the first theoretical model that describes how the microstructural material parameters of graphene-based conductors influence their macroscopic electrical conductivity [22]. Here, we report on systematic experimental studies on this topic, which are backed by extensive simulations. Two major nano- and microstructural aspects are investigated; the average flake size of the employed graphene flakes as well as the behavior of randomly distributed non-conductive flakes in functionalized graphene films. We establish quantitative relationships and present for the first time experimental data on the conductivity of graphene films in good agreement with microstructural simulations. In combination, experiment and simulation provide a coherent picture of the structure–property relationship regarding the electrical conductivity. Our findings are valuable for material optimization and can also be transferred to graphene-based fibers [23, 24], which are structurally similar to thin films.

2. Methods and materials

2.1. Materials

Aqueous graphene dispersion (concentration 1 mg ml⁻¹) was purchased from Sixonia Tech GmbH. Aqueous GO dispersion (concentration 25 mg ml⁻¹) was purchased from Graphenea S.A.

2.2. Preparation of graphene films with systematically varied flake sizes

Graphene dispersion was centrifuged in a liquid cascade with a Hettich Rotofix 32A centrifuge. In brief, multiple centrifugation steps were employed, beginning with the highest rotational speed. After each centrifugation step, the supernatant was collected, while the sediment was redispersed and centrifuged again at a lower rotational speed. Thus, successively increasing particle sizes were obtained. The exact centrifugation protocol is shown in figure S1 in the Supplementary Information available online at stacks.iop.org/NANOX/1/020035/mmmedia.

Each collected supernatant was vacuum-filtrated through a separate polyethersulfone (PES) membrane filter. After drying for 24 hours at room temperature, the films could be peeled from the membrane filters and were freestanding.

2.3. Preparation of mixed graphene/graphene oxide films

GO dispersion was diluted to a concentration of 1 mg ml⁻¹ by adding de-ionized water to the purchased dispersion. Subsequently, it was sonicated for 20 minutes at 20% power with a Bandelin Sonopuls HD4200 ultrasonic homogenizer. Graphene dispersion was centrifuged at 4000 revolutions per minute for 60 minutes. The supernatant was discarded and the sediment was redispersed.

The graphene and the GO dispersions were mixed with different ratios and vacuum-filtrated through aluminum oxide membrane filters. The GO share varied between 5% and 50%. After drying for 24 hours at room temperature, the films could be peeled from the membrane filters and were freestanding.

The procedure outlined above lead to homogeneously mixed graphene/GO films. Without the sonication step, the films were inhomogeneous and separated graphene and GO phases became visible.

2.4. Characterization

The sheet resistance $R_s$ of the thin films was measured with a non-contact eddy current device (SURAGUS EddyCus TF map 2525SR) that enables spatially resolved measurements. The film thickness $t$ was determined with a Mitutoyo 293-240-30 micrometer gauge. The electrical conductivity $\sigma$ of a graphene film was computed according to

$$\sigma = \frac{1}{t \cdot R_s}. \quad (1)$$

The weight of the films was measured with a Sartorius LA310S lab scale. From the weight and the geometric dimensions of a sample, the density $\rho$ could be computed. We defined the packing density $p$ of a graphene film as the ratio of its density to the density of a perfect graphite single crystal ($\rho_{\text{graphite}} = 2.26 \text{ g cm}^{-3}$), which can be regarded as an upper bound for the film density:

$$p = \frac{\rho}{\rho_{\text{graphite}}} \leq 1. \quad (2)$$

Raman spectra were recorded with a Horiba LabRAM Aramis confocal Raman microscope at a laser wavelength of 633 nm. We took measurements at five random positions on a given film and averaged the spectra. In all films
measured here, the Raman spectra recorded on a single film were similar and thus compatible for averaging. SEM micrographs were obtained with a ZEISS MERLIN field emission SEM.

2.5. Visualization of graphene flake sizes
Graphene dispersion was diluted to a concentration of 0.1 mg mL\(^{-1}\) and dripped onto a clean silicon piece. A PES membrane filter was placed on top of the sample. The filter paper prevented graphene flakes from agglomerating while the liquid evaporated. The water was sucked into the filter, leaving the flakes evenly spread over the silicon piece. After drying, the filter paper was removed and the silicon piece was imaged in a scanning electron microscope (SEM). The outlines of the flakes were marked with a commercial software which then computed the flake area. For each sample, 100-200 flakes were considered. Figure 1 shows an SEM micrograph of visualized flakes.

2.6. Random resistor network simulation
The simulations are based on a random resistor network model, which has been presented in detail elsewhere \[22, 25\]. Graphene flakes are modeled as randomly shaped polygons and are assembled to form a layered structure. From the geometric objects, a resistor network is created: The overlaps between flakes of adjacent layers correspond to nodes of the network, which are connected through the individual graphene flakes. The resistances of these connections depend on geometric properties: The distances between connected overlaps, the overlap areas, and their orientation towards each other. From the resistor network, the electrical conductivity of a given structure is computed. For the simulations presented here, the structures comprised a minimum of 50,000 graphene flakes. The major input parameters for the simulation are the overall packing density, the sizes of the graphene flakes, the in-plane conductivities and the out-of-plane conductivities of the individual flakes. The in-plane and out-of-plane conductivities can either be modeled according to statistical distributions or as uniform effective conductivities \[22\].

3. Results and discussion

3.1. General film characteristics
The synthesized graphene films are between 5 μm and 20 μm thick, which could be adjusted by selecting the appropriate volume and concentration of the dispersion that is filtrated. The films are freestanding, mechanically flexible and can be folded without breaking or degradation of the electrical conductivity. Optical images are shown in figure 2(a)–(c). Scanning electron micrographs revealed that some surface roughness is present, but on the inside, the flakes are arranged in a flat layered way (figure 2(d) and (e)). The films exhibit a

![Figure 1. SEM micrograph of graphene flakes evenly spread over a silicon surface. The inset shows the evaluation of \(N = 177\) flake sizes.](image)
spatially homogeneous sheet resistance and the highest electrical conductivities of the as-deposited films range around 70 000 Sm$^{-1}$ without any post-treatment, which is on par with state of the art literature results [26–28]. For higher conductivities, additional treatment such as mechanical densification, chemical doping, or thermal processing is required [11].

Raman spectra of the graphene films feature the typical D, G, and 2D-peaks of graphene-based materials around 1340, 1580, and 2660 cm$^{-1}$ respectively [29, 30]. An exemplary Raman spectrum of an as-deposited graphene film is shown in figure 2(f). The presence of a prominent D-peak is a sign of defects, whereas the distinct G and 2D-peaks are characteristic of material systems with an intact honeycomb lattice of carbon atoms [29, 30]. Overall, the structural film properties are in line with published data of high performance graphene films [9, 21, 31].

3.2. Flake-size dependence of the film electrical conductivity

In three batches, 20 graphene films with systematically varied flake sizes were prepared according to section 2.2. There was a time interval of two weeks between each batch. Figure 3 shows the electrical conductivities of the films as a function of the average flake size of the employed base material. Since the films vary in packing density $p$, the conductivities $\sigma$ are rescaled according to $\sigma \rightarrow \sigma \cdot p^{-1}$.

Across all batches, we observed a systematic increase of the film conductivity with the employed graphene flake size. Qualitatively, this behavior is known from the literature [32, 33]. Within our theoretical model, it can be explained as follows: In graphene films, the flakes themselves are expected to be much more conductive than the individual connections from one flake to another. Larger graphene flakes lead to fewer overlaps per unit length and larger overlap areas, which increases the overall conductivity [22].

In addition to the flake-size dependence, we also observed a decreasing conductivity from batch to batch, which is most likely linked to an aging process of the dispersion. Since the batches were processed each with two weeks in between, the graphene flakes are expected to have agglomerated more and more from one batch to the next. With an increasing layer number, the in-plane conductivity of the individual flakes decreases [34, 35]. As the in-plane flake conductivity directly translates to the macroscopic film conductivity [22], the behavior seen in figure 3 is the logical consequence.

We performed simulations with the random resistor network model described in section 2.6 and compared the results to the experiment as shown in figure 3. The measured packing densities, average flake sizes, and electrical conductivities were used as input parameters for the simulation. The remaining simulation parameters
are the effective in-plane \( \sigma_{\text{in}} \) and out-of-plane \( \sigma_{\text{out}} \) graphene flake conductivities. As explained above, the in-plane graphene flake conductivities are expected to differ from batch to batch, but can be assumed constant within a given batch. Under these conditions, a least-squares fitting routine was applied to each batch, and the solid curves in figure 3 were obtained. They are based on a uniform effective out-of-plane conductivity \( \sigma_{\text{out}} = 0.073 \text{ S m}^{-1} \) and the in-plane conductivities that are listed in table 1.

Note that these effective quantities are valid simulation parameters due to the self-averaging nature of the system at hand. The modeled graphene flakes are randomly shaped and oriented, and the individual flake conductivities vary according to statistical distributions. Thus, the system is disordered. However, there are always universal effective in-plane and out-of-plane conductivities that can be assigned to all the flakes, which reproduce the same overall film conductivity. Comprehensive studies on this topic have been carried out elsewhere [22, 36].

The theoretical model of the electrical conductivity of graphene-based conductors [22] strongly agrees with the experimental results. Moreover, the fitted effective in-plane and out-of-plane graphene flake conductivities are in line with literature data. The out-of-plane value is well within the range of twisted bilayer graphene (0.03 Sm\(^{-1}\)–3 Sm\(^{-1}\) depending on the twisting angle [37, 38]), which is expected for an assembly of randomly orientated graphene flakes. The in-plane values match literature results of electrical conductivities measured in undoped graphene films [18, 26, 39].

### 3.3. Non-conductive elements in conductive graphene films

One of the numerous advantages of graphene-based macro-materials is the potential for functionalization [5]. Foreign species can be intercalated between graphene layers and create new functionalities for applications. If those species are non-conductive however, only a certain amount can be incorporated before the material loses a substantial part of its electrical conductivity.
In a combined simulative and experimental study, we investigated graphene-based conductors, where a well-defined share of non-conductive elements is among the conductive graphene flakes. With the random resistor network model, we simulated 180 different geometries with a share of non-conductive elements between 0.5% and 50%. The packing densities varied between 0.7 and 0.9. The non-conductive elements were randomly distributed across the whole structure and not specifically clustered (see figure S2). The resulting overall conductivities were normalized to the potential maximum and subsequently averaged for each share of non-conductive flakes. Simultaneously, we synthesized three batches of graphene films with incorporated GO flakes as explained in section 2.3. Structurally, GO flakes are similar to graphene and can be homogeneously integrated into graphene films. However, the electrical conductivity of GO flakes is orders of magnitude lower than that of graphene flakes [40]. We varied the share of GO flakes from 5% to 50% and measured the electrical conductivity of the resulting films. To account for variations from batch to batch, the conductivities were normalized to 1. For this purpose, every batch contained a pure graphene film without GO flakes. These films always exhibited the highest conductivities within the respective batches and correspond to the maximum of the normalized film conductivity. Figure 4(a) compares the experimental results to the simulation. No fitting parameters are required for this comparison.

The experiments are in line with the simulations. In both cases, the total conductivity generally decreases with an increasing number of non-conductive elements. On the one hand, this is due to the reduced amount of current-carrying material inside the conductor. On the other hand, the more non-conductive elements are incorporated, the longer the current path through a structure becomes. The non-conductive parts act like dead material, which the current has to circumvent. Beyond this qualitative agreement, a considerable drop of the conductivity occurs around a critical share $\theta_c$ of 10% non-conductive flakes. Despite some statistical fluctuation, this holds true for both the experiment and the simulation. In the experiment, above a 20% share of GO flakes, the measuring device could not detect a conductivity $>0$ anymore.

Upon closer inspection of the simulated data points, the conductivity drop follows a power law as it is known from percolation theory [41]. It has the mathematical form

$$\sigma \propto \left(1 - \frac{1 - \theta}{1 - \theta_c}\right)^{-t_c},$$

with the critical exponent $t_c$. Figure 4(b) shows the simulated data points on a logarithmic scale, together with equation (3), where $\theta_c = 0.1$ and $t_c = 4.1$. Commonly, percolation studies investigate conductive elements in a non-conductive host. In that case, a steep increase in the conductivity is observed as soon as the conductive elements form a closed path through the whole structure. The scenario investigated here is different: The conductivity drops drastically as soon as the overall current path is severely constricted and not as soon as the non-conductive elements have formed a closed network from side to side. Thus, $\theta_c$ and $t_c$ can be very specific to the system under consideration. For the type of structure modeled here however, both parameters are valid for all the simulated packing densities, flake size distributions, and conductivities. The strong agreement with the experimental results suggests that the mathematical model accurately describes the behavior of such conductors.
experiment as well as the universality of $\theta_c$ in the simulations clearly indicate that for a high electrical conductivity, the amount of non-conductive elements in a graphene film has to remain well below 10%.

To study a related microstructural aspect, we varied the spatial homogeneity of the GO content inside the graphene films. By skipping the sonication step as explained in section 2.3, we obtained inhomogeneous thin films where separated graphene and GO phases occurred. Figure 5(a) shows the frontside and the backside of such an inhomogeneous film. The phases can be distinguished with the naked eye, but the pattern that is visible on the frontside does not continue throughout the complete thickness of the film. From the back, the inhomogeneity is still visible but the distribution of graphene and GO is clearly different. A spatially resolved eddy current measurement revealed that the sheet resistance of the mixed films does not fluctuate by more than 5 percent over the entire film surface. This indicates that the graphene and GO phases are randomly distributed inside the films and an overall average conductivity can be reasonably determined.

For 10 films with varying GO content $\theta$ between 10% and 50%, the conductivity results normalized to a pure graphene film are shown in figure 5(b). They are compared to the previously synthesized homogeneous films and the theoretical maximum conductivity $\sigma_{\text{max}}$, which is given as

$$\sigma_{\text{max}} = \sigma_{gr}(1 - \theta)$$

where $\sigma_{gr}$ denotes the conductivity of a pure graphene film without GO. For the inhomogeneous films, there is also a conductivity drop around $\theta = 10\%$, although not as severe as for the homogeneous films. When the GO content reaches 50%, the inhomogeneous films retain more than 20% of their electrical conductivity.

The results are explained as follows: In the inhomogeneous films, there is always a well-connected network of conductive graphene flakes that is not interrupted by the non-conductive GO flakes. Consequently, the conductivity of these films remains finite, even with higher shares of GO flakes $\theta > 20\%$. Nevertheless, the flake distribution is not optimal for current flow, which can be seen from the comparison with the theoretical maximum conductivity in figure 5 (b). These findings illustrate that not only the properties of the nanoscopic components themselves, but also their mere spatial arrangement has a considerable influence on the macroscopic film properties.

4. Conclusion

In two separate studies, we have investigated specific nano- and microstructural aspects of graphene films that directly translate to their electrical conductivity. We studied the influence of the average graphene flake size and the incorporation of non-conductive GO flakes in a graphene film. For quantitative results, we have combined conductivity simulations with experiments for the first time in this field. We performed the simulations with a random resistor network model [25] and we synthesized graphene and mixed graphene/GO films via vacuum filtration. Overall, a coherent picture of the relationship between the graphene film microstructure and the electrical conductivity emerged.
We found that large graphene flakes are essential to obtain highly conductive macroscopic films. If graphene films are functionalized with non-conductive or non-doping chemical species, the total share of those elements needs to remain well below 10% to preserve a high electrical conductivity. The greatest conductivity loss occurs when homogeneously distributed non-conductive elements are incorporated inside a graphene film. The experimental results have been quantitatively explained with a previously presented model of electrical conduction in graphene-based materials [22]. Furthermore, they are transferable to any material with a structure similar to graphene films. A prominent example are graphene fibers, which can be synthesized with a flat, belt-like shape where the individual graphene flakes are arranged layerwise like in a thin film [23, 24].

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