Electrical and Optical Properties of Controlled Reduced Graphene Oxide Prepared by a Green and Facile Route

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Introduction

Since the emergence of graphene, numerous studies have been conducted on graphene and its derivatives, due to their remarkable properties. Graphene oxide (GO) and reduced-graphene oxide (rGO) are considered as the two main derivatives of graphene [1]. Additionally, when arranged in a stacked configuration, they would form many other derivatives including monolayer, few-layer, and multilayer graphene-based materials, each exhibiting distinct characteristics and properties [2].

Most graphene synthesis methods involve the utilization of complex and costly techniques, as well as the need for three-dimensional crystal, posing difficulties for its operation and investigation [3]. On the other hand, free-standing samples offer more flexibility and versatility. In this regard, we have developed a green and facile route towards the synthesis of free-standing rGO samples, based on the modified Hummer's method, which uses the least possible chemicals. In addition to structural analyses, the obtained samples were subjected to electrical and optical examinations to ascertain their distinct features. The interesting properties of these samples, along with their low chemical content, make them a great candidate for many applications, including medical uses such as biomedical imaging, contrast agents, and monitoring electrodes [4, 5].

Materials

Aqueous GO solution has been initially prepared by a modified Hummers method. Pre-treatment of graphite powder favored a lower amount of reactants during synthesis. Also, the reaction time was prolonged to 34 h, which provides a good GO exfoliation without compromising the quality of the dispersion [6-9]. The concentration of the solution was 12 mg/mL, which was then air dried overnight. This was followed by thermal reduction processes to obtain rGO free-standing layers [10]. We produced three distinct rGO samples, designated *rGO1*, *rGO2*, and *rGO3*, using the described method. Notably, this approach makes no use of any mechanical or shear force. Finally, it should be mentioned that these three rGO samples were fine-tuned by adjusting the initial reactant concentration and annealing temperature.

Structural Properties

The multi-stacked layer architectures of the rGO free-standing thin films are immediately apparent from their FESEM cross-sectional views. Each stacked layer is about between 2 to 3 µm thick and is made up of multiple rGO planes. Moreover, while looking at the surface morphology of the samples, it can be seen that they have a relatively smooth sheet-like surface with few wrinkles.

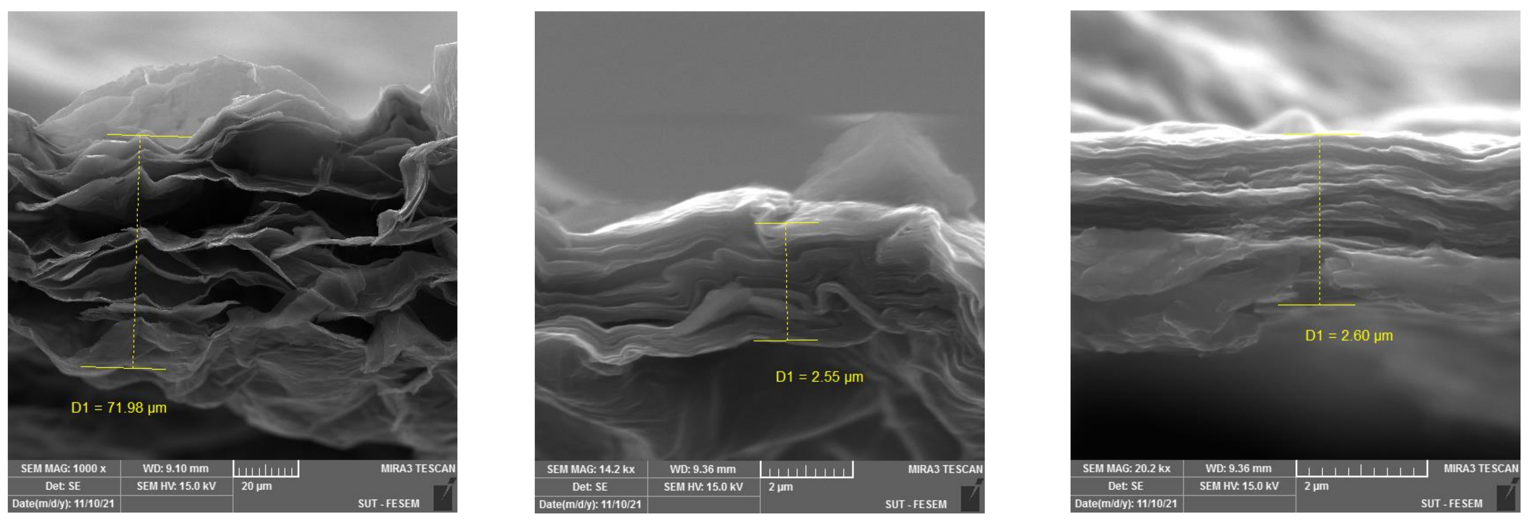

Figure 1. FESEM images of *rGO1*, *rGO2*, and *rGO3* (in order from left to right).

The Raman spectra of the samples disclose a shift of the G bands towards lower wavenumbers compared to the G bands of the GO, which is indicative of a lower oxygen content in the rGOs. In addition, the intensity ratio between the D band and the G band shows that the number of surface defects is small [11].

Optical Properties

rGO1 and *rGO2* were measured and analyzed by FTIR spectroscopy, within the wavelength range of 2.5 to 25 µm, in order to investigate their optical properties. Their resulting absorbance spectra are shown in Figure3. These results highlight the significant absorption values of the samples exceeding 90% across the entire spectrum, indicating their potential use as spectrally wide absorbers.

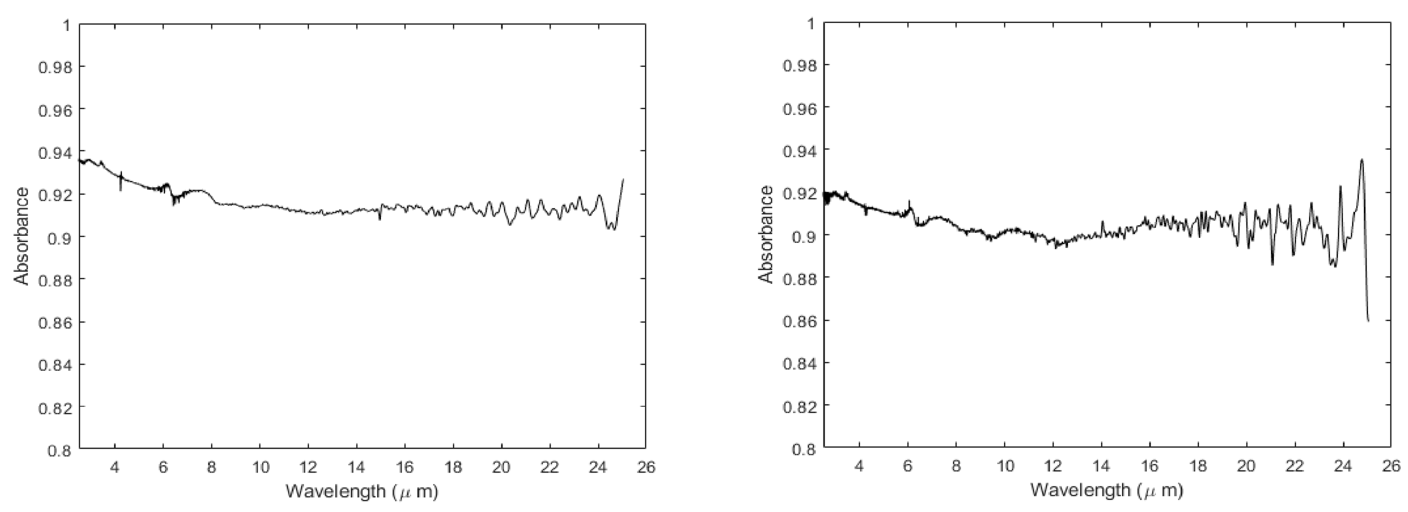

Figure 3. FTIR spectra of *rGO1* (left) and *rGO2* (right).

Electrical Properties

We first measured the samples' resistance values in various directions at room temperature using the four-point probe technique. These tests were also repeated on several different days. All values measured for each sample differ just slightly, which indicates that the samples are homogenous and thermally stable. The recorded measurements exhibit an average value of approximately 19.38, 294.25, and 5849.7 Ω for *rGO1*, *rGO2*, and *rGO3*, respectively. These results indicate that rGO samples can be produced with any desired conductivity over a broad spectrum, which brings us the versatility and tunability. The samples were also analyzed by AC measurements and no phase change was observed between voltage and current passing through them. As a result, these thin-films are laterally pure-resistive.

It can be seen from the electrical resistance versus temperature (RT) curves of the samples that with an increase in temperature, the resistance of *rGO1*, *rGO2*, and *rGO3* decreases at a rate of 0.35, 0.55, and 0.65 %/K, in turn, implying semiconducting behavior in the rGOs. It is worth noting that the sample with higher conductivity and therefore higher reduction degree, suggesting a lower carbon to oxygen ratio, has a lower temperature coefficient of resistance, implying a decrease in its semiconducting energy gap [12].

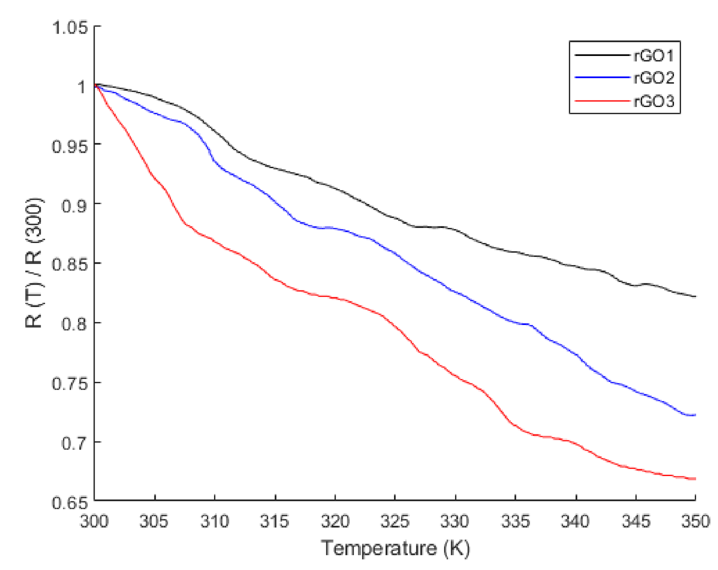

Figure 2. RT curves of *rGO1*, *rGO2*, and *rGO3*.

Conclusions

By using a modified version of Hummer's method, which is a facile, versatile, and eco-friendly approach, three self-standing rGO samples were successfully synthesized. Despite the absence of shear stress in the production process, their cross-sectional FESEM images validate the attainment of a well-organized stacking arrangement. Also all samples displayed minor surface defects and a smooth sheet-like structure with few wrinkles. According to our electrical and optical investigations, the samples showcased semiconducting behavior and substantial absorption values exceeding 90% in a wide infrared range. Different features exhibited by each sample indicate that it is possible to fine-tune their properties, thereby enhancing their potential for a variety of applications, including electronics, thermoelectric, and optoelectronics devices. For instance, the mentioned green synthesis route, coupled with high optical absorption of samples even in electrically conductive ranges, makes them a clean and favorable option for many biomedical applications.

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†Presented at the 10th International Electronic Conference on Sensors and Applications

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