Characterization of Thin Films of Graphene Nanosheets Prepared with Low-Cost Technique

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Abstract: Exploring and adjusting techniques to synthesis Graphene Nano-sheets (GNS) becomes strongly pursued due to the unique physical and mechanical properties in the desired applications. In this work, Multi-layer thin films of (GNS) were deposited on glass slides using a non-premixed flame technique. The aim was to prepare the homogeneous structure of films with fewer defects. Propane gas was used at monitored temperatures (400-500)°C, under atmospheric pressure. Morphological characteristics of (GNS) were analyzed by optical and Scanning Electron Microscopy (SEM). Furthermore, Ultraviolet-Visible (UV-Vis) spectrometer was used and showed high transmittance via the prepared films which close to nano-size thickness, it increased with increasing the temperature corresponding to about 13 and 11 layers at 400°C and 500°C respectively. The x-ray diffraction (XRD) test illustrated the polycrystalline pattern of (GNS) with sharp peaks at 2θ= 14.22° and 2θ=17.03° that corresponds to the interlayer distances of 6.22 Å and 5.2 Å respectively. However, the broad peak at 2θ= 25.9° for (002) direction matches up with an interlayer space of 3.43Å which refers to ordinary graphite layers presence. The concluded results provided an essential advantage of producing Multilayers of (GNS) with high characteristics.

Keywords: Graphene synthesis, Non-premixed flame, Graphene multi-layers characteristics

1. Introduction

Graphene represents a flat single layer that having sp2 hybridized carbon atoms firmly packed to dual dimensions honeycomb lattice structuring. Also, graphene is an example of the best exciting dual dimension material that is being examined nowadays [1]. It could likewise be regarded to be the fundamental foundation block of graphite material having entirely various dimensions. Graphene could be loaded to 3D graphite, twined to 1D carbon nanotubes (CNTs) as well as wrapped into 1D fullerene [2]. The sp2 hybridization makes strong bonds as well as defines a honeycomb lattice forming for graphene, and the Pz (π) orbitals create a delocalized π-system that controls the conducting features/charge movement of graphite [3]. Various methods are available for preparing a few multi-layer graphenes, for example, micromechanical exfoliation, atmospheric pressure graphitization of silicon carbide, epitaxial growth in ultrahigh vacuum, chemical oxidation of graphite as well as chemic vapour deposing by utilizing transitional kinds of metal as a catalyst [4]. However, synthesis by flame is a cheap, rapid, and high heat synthetic method. This method involves the merely one-step procedure, bringing about in a large-scale method to fabricate varied materials. It is including low-dimension carbon material similar to amorphous carbon thin films, CNTs, as well as fullerenes [5]. There are
many attempts for making a few layers graphene by flame, for instance, Z. Li et al. were made a study about the growth of graphene on copper foil via utilizing an ethanol burner, graphene hadn’t been shown because of the low-temperature as well as the existence of oxygen in the flame [6]. Regarding different experimentation, Z. Li et al. managed to get ready graphene successfully on a nickel foil using two different burners, alcohol burner encircled the substrate for all the period, therefore such flame prevented oxidation also worked for to be the carbon seed and butane burner provided the additional heating of the substrate besides worked for to be the carbon basis for graphene growing [7]. Also, N. K. Memon et al. formed few-layer graphene on nickel and copper foils via benefit from a flame synthesis method at atmospheric pressure [8]. N. G. Prikhodko et al. examined the production of few-layer graphene by propane flame on a nickel substrate at atmospheric pressure and low pressure. They illustrated the forming of graphene layers on a nickel substrate that includes 3 to 10 layers [9]. The aims of this work are preparing and characterizing of multi-layer graphene on glass slides efficiently with simple and low cost.

2. Experimental Work

Multi-layer graphene (MLG) films were settled on a glass slide having a thickness of ~1 mm through the flame synthesis method as shown in Figure 1. The experimental setup used in the current work depends on a non-premixed flame burner, as the blending of fuel (Propane gas) and oxidizer (air) happened after igniting, such procedure happened at atmosphere pressure. Glass slides were cut to parts that each one has dimensions of 2×2 cm$^2$ and the glass slides had been put in a clean beaker containing (HCl acid and distilled water) 5%) for 10 min to get rid of oil or dust which can exist on the surface of the substrate. After that, glass slides have been placed in a clean beaker which contains distilled water for 10 min. Then, the slides have dried by air blowers, followed by rubbing those using soft papers. The glass slides were located in the substrate holder, then the propane burner turned on and pointed towards the slide for 10 min. The distance between the burner and glass slide has been fixed at about 8cm to deposit graphene on the slide’s surfaces. At the temperature (400°C and 500°C) with utilizing a thermocouple type (Autonics, Model: TC4H-24R), the propane burner has switched off. Finally, cooling down the glass slides to the lab’s temperature.

Figure 1. The experimental setup of the flame synthesis technique.
Transmittance optical microscopy type (Olympus Bx 60) was used to imaging the samples at 500x and 1000x. Scanning electron microscopy (The VEGA Easy Probe) had been utilized for analyzing the topography and structure at a magnification of 5000x to 25000x. Further characterization has been done using the spectrophotometer of UV-Vis spectrometer type (Lambda 750, Perkin Elmer). This test had been employed to observe the optical spectrum of the graphene films at ranges between wavelengths of 350 nm to 1200 nm.

3. Results and Discussion

3.1. X-Ray Diffraction Pattern

In Figure 2, the XRD pattern for graphene film prepared by flame technique showed that the graphene’s structure is polycrystalline having dual strong sharp diffraction peaks at 2θ= 14.22° and 2θ=17.03° corresponds to an interlayer distance of 6.22Å and 5.2Å, correspondingly. It is attributed to a low degree of crystallinity, similarly more or fewer defects exist that may indicate the existence of carbon dioxide CO$_2$. The reason for such may be due to the broadened stacking of more wrinkled of graphene sheets otherwise having a disorder in graphene layers that exist at the areas of the edge [10]. Moreover, there is a weak and broad peak at 2θ= 25.9° for (002) direction which matches up to an interlayer space of 3.43Å that is in line with the interlayer space of regular graphite that having interlayer space equals to 3.35 Å.

![Figure 2. XRD pattern of graphene film prepared by the flame technique](image_url)
3.2. Optical Microscopy Characteristic:

Figures 3- (a) and (b) shows the optical microscope imaging of graphene films which have been deposited on glass slides at 400°C at 500x and 1000x magnification. Moreover, Figure 4-(a) and (b) illustrate the other optical microscope imaging of graphene films have deposited on glass at 500°C for 500x and 1000x magnification. The colour of the graphene film changes depending on its thickness. From these figures, it can be observed the formation of amorphous carbon impurities particles with the graphene films due to present carbon oxide and some impurities from the environment.

![Figure 3](image1.png)

**Figure 3.** Optical microscope imaging of graphene film deposited at 400°C for (a) 500x and (b) 1000x.

![Figure 4](image2.png)

**Figure 4.** Optical microscope images of graphene film deposited at 500°C for (a) 500x and (b) 1000x.

3.3. Scanning Electron Microscopy (SEM) Analysis

Graphene films were characterized by SEM for analyzing the topographic nature of the graphene as well as the micro/nanostructure of the layers. The images can be seen in Figure 4 and Figure 5 in two magnification scales. The observed structures for those films were dense and the graphene films were irregularly distributed on the glass slides with increasing
temperature from 400°C to 500°C. It is found that the film’s density of graphene layers of the specimen prepared at 500°C was greater than that prepared at 400°C. Flame synthesizing is determined by the generating of amorphous carbon particles which were dispersed arbitrarily to graphene films.

![Figure 5](image1.png)

**Figure 5.** SEM images of graphene films have been prepared at 400°C for different magnifications at (a) Scale =2µm, (b) Scale =5µm.

![Figure 6](image2.png)

**Figure 6.** SEM images of graphene films have been prepared at 500°C for different magnifications at (a) Scale =2µm, (b) Scale =5µm.

3.4. Spectroscopic Characteristics

Results of UV-Vis tests of multi-layer graphene (films have deposited on glass slides) showed different transmittance characteristics whereas temperatures changed. This behaviour introduced in the graphs against wavelength as shown in Figure 7 and Figure 8. The films have a high transmission at long wavelengths approximately (70 – 80 %), and decreasing transmission to (60%) at short wavelengths. It is worth mentioning that the shift of the curve near-maximum
transmission varies from one sample to another sample based on the GNS particle size distribution. Furthermore, the effect of the different temperatures 400 °C and 500 °C on transmission percentage, that graphene films reached to (75 %) in the visible region at 500 °C, however, at 400°C the transmittance is (70%). This is attributed to a slight increase in the transmittance in this spectral range, resulted in higher transmittance which can be ascribed to the improvement of structural homogeneity as well as crystalline of the particles.

Figure 7. UV-Vis spectrum of graphene film prepared at 400°C.

Figure 8. UV-Vis spectrum of graphene film prepared at 500°C.
Increase crystallinity of film’s structure leads to a decrease in the scatter of light waves which result to improve in films’ transparency. On the other side, the estimation of the number of layers could be made by the transmittance spectrum of multi-layer graphene film because all graphene layers absorb merely 2.3 % of visible lighting. At temperature 400°C, the transmission was noticeable which places in wavelengths (400-700) nm, which approximately represents 70 %. This relates to ~13 layers of graphene which can be grown consistently transverse the substrate of the glass. At 500°C the transmission in the noticeable area (400-700) nm approximately forms 75 %, which correlates to ~11 layers of graphene are grown, so the film was appeared more transparent and colourless.

Conclusions

The current investigation proves that Flame synthesis of graphene multilayers graphene using a non-premixed flame burner to be a suitable technique as it has low cost and low chemical impacts. The deposition of thin films of graphene (GNS) on glass slides (substrate) is difficult at a high temperature which leads to several heterogeneous features with cracks in glass slides. Therefore, the novelty of this work was using a gradual heating process on the glass substrate which allows thin films of graphene (GNS) to deposit gently. The homogeneity of graphene layers accumulation and distribution on the glass surface with monitoring affects significantly the crystalline structure. These results will improve the optical characteristics of graphene films that have prepared by this technique.

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