

# Investigation of Alternative Techniques for Graphene Synthesis

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## Abstract

In recent years, a great deal of concentration has addressed the electronic and morphological characteristics of carbonaceous substances. Nowadays, particularly, graphene is one of the most popular materials in condensed-matter physics and materials science. It is used in different fields such as desalination of seawater, smartphones, computers, satellites, planes, cars, building materials, obtaining protective coatings and rust-free cars, nuclear clean up, transistors, sensors, electron microscopy, Li ion batteries, super capacitors, and bionics. Mechanical cleaving (exfoliation), chemical exfoliation, chemical synthesis, and thermal chemical vapor deposition (CVD) synthesis are the most commonly used methods today. Some other techniques are also reported such as unzipping nanotube and microwave synthesis. In graphene synthesis, starting material is usually graphite. On the other hand, different starting materials such as rice husks, fenugreek seeds, hibiscus flower petals, camphor, alfalfa plants, petroleum asphalt are used as a carbon source for graphene synthesis. In this study, alternative methods for graphene synthesis specially microwave irradiation and ultrasound energy were studied, and the performances of the final products were compared with the help of different characterization techniques. Advantages and drawbacks of these methods were clearly discussed for enhancing the understanding of the graphene synthesis phenomena.

**Keywords:** graphene synthesis, graphene characterization, microwave irradiation, ultrasound energy

## 1. Introduction

Graphene is a 2D material, which was firstly discovered by Geim and Novoselov in 2004. They won Nobel Prize in Physics by synthesizing graphene including of  $sp^2$  carbon bonds via Scotch-tape method in 2010 [1, 2]. Graphene is a thin nanoplatelet, which can be produced by cleaving of graphite. Graphite can be downed into the single graphene sheet level [3]. Graphene is a one atomic layer having 0.34 nm thicknesses. Graphene is a single layer of carbon atoms organized in a honeycomb lattice [4]. It is the block of graphite that is used in pencil tips, but graphene is an extraordinary matter with a multitude of astounding specialties that named it as wonder material [5]. It is a hexagonal shaped plane consisting of  $sp^2$ -carbon atoms [6, 7]. Graphene can be seemed as either uncoiled single-walled carbon nanotubes or a wide atomic sheet of graphite. Graphene has superior mechanical strength, thermal

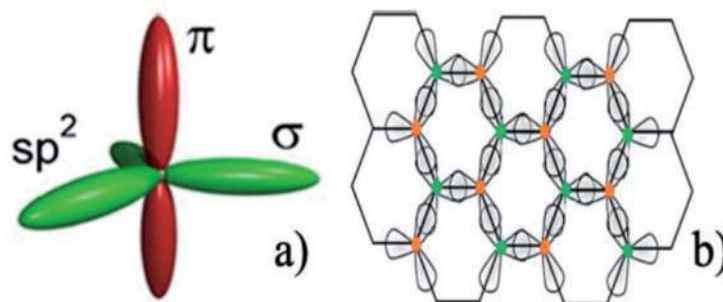
conductivity, optical transparency, high mobility, room temperature quantum Hall effect and great electronic properties like Dirac-particles having a linear dispersion, transport energy gap and simply absorption coefficient of lights, thus it will become the favorable prospect after the silicon time [8, 9]. It is the thinnest substance at one atom thick, and also fabulously strong around 200 times stronger than steel [5]. Apart from that, graphene is a superb conductor of heat and electricity and has exciting light absorption capabilities. It is truthfully a material with wide potential for integrating in nearly any industry.

Graphene is a highly varied material and can be merged with other materials (involving gases and metals) to synthesize various materials with different exceptional qualities. Researchers proceed to examine its unexplored properties and possible applications such as touchscreens (for LCD or OLED displays), computer chips, transistors, batteries, supercapacitors, energy production, DNA sequencing, water filters, antennas, solar cells, and spintronics. This new 2D material has a prominent importance in present day. It is a quickly developing subject that flourishing novel concepts at incredible speed [10]. Graphene is extensively used substance in electronic industry such as field-effect transistor, transparent electrode, etc. The recent developments in surface area, optical, magnetic, and mechanical properties of functionalized graphene and the unique electronics have arisen new attitude of green technology and creative discovery for present complications such as photonic and electronic usages for ultrahigh-frequency graphene-based apparatus, anode for Li-ion battery, material science, ceramics, light natural gas tanks, medical science, sensors to identify sickness, supercapacitor, solar cell, desalination of seawater, smartphones, computers, satellites, planes, cars, building materials, obtaining protective coatings and rust free cars, nuclear clean up, transistors, sensors, electron microscopy, and bionics.

Graphene molecular structure includes of  $sp^2$  hybrid carbon atoms that were presented in **Figure 1a**.  $Sp^2$  hybrids supply  $\sigma$  bonds with adjacent carbon atoms. Each of  $\sigma$  bonds has the length of 1.42 Å. Excellent mechanical characteristics of graphene are obtained under favor of  $\sigma$  bonds.

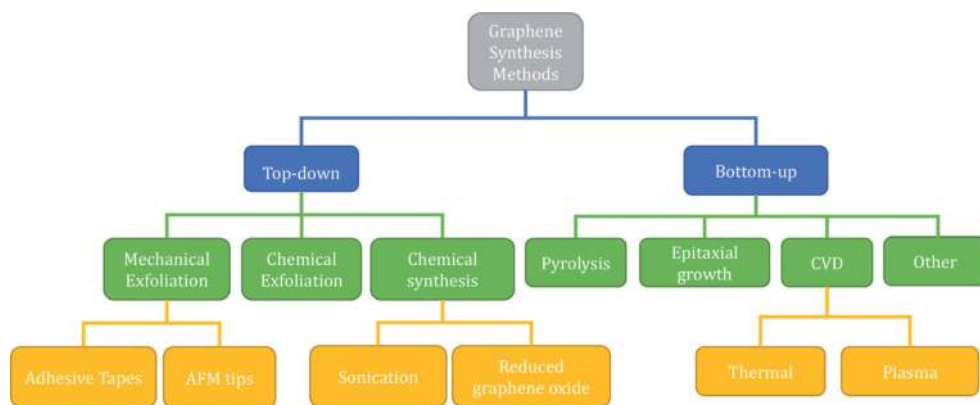
Graphene gathers much interest particularly after Geim and Novoselov win the 2010 Nobel Prize in physics by obtaining it in 2004. To produce high-quality graphene in high amount is not easy and affordable. Most companies are using chemical vapor deposition (CVD) based processes. Also, mechanical and chemical exfoliation and chemical synthesis are the most preferred ways today. Other methods are unzipping of a nanotube and microwave irradiation [11].

In graphene synthesis, starting material is usually graphite. But different starting materials are also used in literature such as; rice husks [12], fenugreek seeds [13], hibiscus flower petals [14], camphor [15], alfalfa plants [16], petroleum asphalt [17]. Graphene synthesis ways are primarily separated under two main groups entitling as bottom-up and top-down methods as seen as in **Figure 2** [11].



**Figure 1.**

(a)  $sp^2$  hybrids carbon atoms in graphene (b)  $sp^2$  hybrids of graphene carbon atoms connected to adjacent ones.



**Figure 2.**  
Flow chart for available methods for synthesis of graphene sheets.

In top-down approach, graphene is synthesized by using graphite or graphite-oxide with the help of different methods. In this method, carbon materials such as graphite, carbon nanotubes are starting substances, and they are peeled by using chemical, electrochemical or physical ways [18]. Main top-down techniques are micromechanical exfoliation, cleavage of graphite intercalated compounds (GICs), unzipping of carbon nanotubes (CNTs), arc discharge, cleavage of graphene oxide, and liquid phase exfoliation.

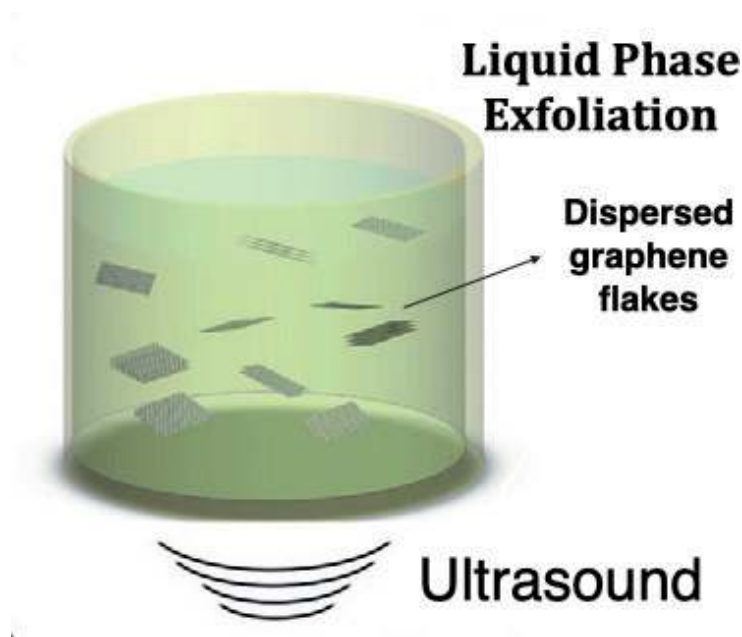
## 2. Liquid phase exfoliation

Liquid phase exfoliation is an efficient and productive way for synthesizing of single and few layered graphene. It has been considered as one of the most feasible approach for industrial production of graphene due to its scalability and low cost. Solvent – carbon source suspension was first sonicated for preparation of exfoliation. Due to not having defects and oxide groups in the graphene products synthesized by LPE, they are more suitable for use in the electronics industry than that are produced by other techniques.

The LPE can form a stable dispersion of monolayer or few-layer defect-free graphene, which only involves the exfoliation of natural graphite via high-shear mixing or sonication [19]. Prepared graphene dispersion was stabilized by used solvent. Solvent type has also importance in productivity of the graphene dispersion [20]. Solvent ensures both the stability of synthesized graphene mixture and its productiveness. Tetrahydrofuran (THF) and N,N-dimethyl-formamide (DMF) are advantageous solvents to get high quality of graphene merely they are poisonous and show low efficiency. Dibasic ester (DBE) is a nontoxic and environmental-friendly solvent and it was used for cleavage of graphite by Jiang et al. Its surface tension is  $35.6 \text{ mJ/m}^{-2}$  and solubility parameter is 9.7 [20].

Graphite can be exfoliated in liquid medium exploiting sound waves to form single layer, **Figure 3** [21]. Basically, exfoliation of carbon materials is a relatively economical and easy way to produce graphene [22].

The exfoliation step of the LPE can be conducted by the sonication of graphite in different solvents. There are two types of sonication: tip and bath sonication. In this study, tip sonication treatment was applied to the graphite-solvent dispersions. Epoxy/graphene composite shows better mechanical properties due to direct ultrasonication of tip sonication, that generates higher sound pressures and intensity compared to bath sonication which is indirect ultrasonication [23, 24]. The direct sonication of graphite in a solvent having similar surface energy to graphite enables a



**Figure 3.**  
*Liquid phase exfoliation.*

stable graphite dispersion [25]. Several studies have been performed in order to find the most appropriate solvent as well as the optimum operation conditions for the sonication process [26–29].

### 3. Experimental section

The experimental studies consist of two different methods; microwave (MW) energy-assisted method and ultrasound (US) energy-assisted method.

#### 3.1 Materials used

In microwave energy method; graphite (natural flake graphite, grade 3061; purchased from Asbury Graphite Mills, Inc., New Jersey) was used as starting carbon source. Different solvents were used such as 25% ammonia solution (Merck KGaA), N,N-Dimethyl formamide (Merck KGaA), ethylene glycol (ZAG Chemicals) and ethylene diamine (Merck KGaA). Chemicals used in the second cycle of experiments were of analytical grade; n-Hexadecane (Merck, 99.5%), dimethyl sulfoxide (Merck, 99.9%), sodium hydroxide (J.T. Baker, 99%), 1-octanol (Merck, 99%), perchloric acid (Merck, 70–72%), N,N-Dimethyl formamide (Merck, 99.8%), ethylene glycol (ZAG Chemicals, 99.3%), and ethylene diamine (Merck, 99%).

Chemicals used in the ultrasound method are as follows: Graphite fine powder (Extra pure, Asbury Inc., New Jersey), graphene nanoplatelets (XG Sciences, Michigan, US) Dimethyl sulfoxide - DMSO (Merck), N,N-Dimethylformamide - DMF (Merck), Perchloric acid 70–72% - PA (Merck).

#### 3.2 Preparation of graphene sheets via microwave method

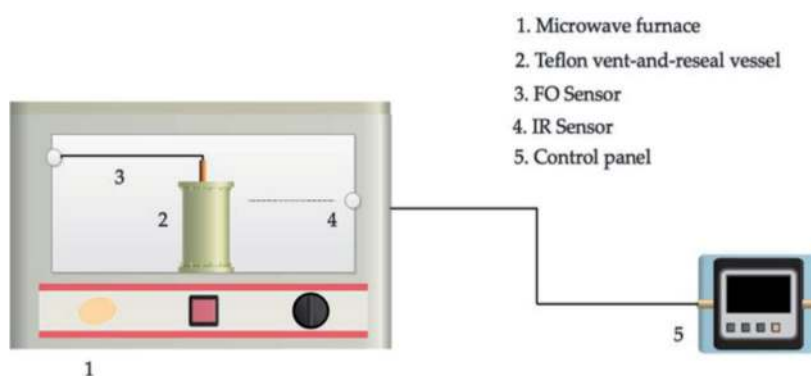
The procedure of MW treatment was summarized as following: First, natural graphite is added to ammonia, then obtained suspension was sonicated by ultrasound energy device (BANDELIN ® HD 2200 SONOPULS), under conditions 200 W,

35 kHz, mode 5 and 50% power for 10 min. Secondly, reaction was performed in Milestone Start-S model microwave oven for half an hour at 120°C temperature and 1 bar pressure by applying 50, 100 or 200 Watt energy. Pressure controller was active, and thermocouple was adjusted carefully as shown in **Figure 4**.

### 3.3 Preparation of graphene sheets via ultrasound method

0.3 g graphite was dispersed in 50 ml solvent such as DMSO, DMF and PA. Obtained dispersions were sonicated by the means of BANDELIN® HD 2200 SONOPULS (which is given in **Figure 5**) equipped with a VS 190 T sonotrode, 200 W, 50% amplitude for 3 hours.

Then, these dispersions were subjected to 60 minutes centrifugation (Elektromag, M 4812 P) at 3000 rpm to remove the unexfoliated part of graphite; after the heavier particles were settled down, supernatant parts were decanted and collected in separate vials.



**Figure 4.**  
*The experimental system with a multimode microwave furnace: Reaction was performed inside a Teflon vent-and-reseal vessel.*



**Figure 5.**  
*Ultrasound device.*

### 3.4 Characterization

Different characterization techniques were applied to the obtained final products via microwave energy method in order to determine their properties such as thickness, layer number, electrical conductivity. X-ray Diffraction (XRD) analysis was done via Rigaku D-Max 2200 Series equipped with Cu-K $\alpha$  radiation ( $\lambda = 1.54 \text{ \AA}$ ) at a scanning rate of  $3^\circ$  per minute. The tube voltage was 40 kV and the current were 40 mA. The intensity was determined over a  $2\theta^\circ$  angular range of  $2\text{--}90^\circ$ . Electrical conductivities of synthesized products were measured by Keithley 2400 Sourcemeter which is seen in **Figure 6**.

Each sample was measured by applying following procedure; first, it was placed in a copper cylindrical container which has a copper cap and it was compressed by a hydraulic press under 50 bar for 30 min. The electrical resistivities of obtained products were determined by 4-point probe method. Synthesized powder sample were compressed in copper mold with the help of a joiner's clamp during the electrical conductivity measurement. The conductivity  $\sigma$  was then estimated according to  $\sigma = 1/AR$ . The Fourier Transform Infrared (FTIR) spectra of synthesized products were measured by Perkin Elmer Spectrum Two equipped with a germanium (Ge) crystal (Pike Gladi ATR Ge-ATR) in the range of  $650\text{--}4000 \text{ cm}^{-1}$ . The obtained powder was characterized via ultraviolet-visible (UV-Vis) spectroscopy. For UV-vis analysis, the dried filtrate



**Figure 6.** Electrical resistivity measurement system: (a) copper cylindrical container and a copper cap. (b) Electrical resistivity measurement set-up (joiner's clamp and copper container). (c) Keithley 2400 Sourcemeter.

which is dried on drying oven at overnight was dispersed in distilled water by agitating via a magnetic stirrer. After that an amount of dispersion was taken into the 10x10 mm vial then it was analyzed by comparing with the water which is reference sample. The spectrum has an operation range (UV Perkin Elmer, Lambda 35) of 200 to 700 nm.

Also, the synthesized products via ultrasound energy method were analyzed via different characterization techniques such as UV-vis spectroscopy, Atomic Force Microscopy, X-ray Diffraction and dynamic light scattering analysis. UV-vis spectral measurements were acquired using a Perkin Elmer Precisely Lambda 35 UV/vis Spectrometer. UV-Visible spectra (Perkin Elmer, Lambda 35) were measured from 200 to 800 nm. Samples for AFM were prepared by dropping the graphene dispersions onto glass pieces ( $0.7 \times 0.7 \text{ mm}^2$ ) and measurements were made in contact (tapping) mode, with  $10.00 \mu\text{m}$  scan size, and 20.35 Hz scan rate by using Digital Instruments Nanoscope. Samples for XRD were prepared by depositing onto glass pieces ( $0.7 \times 0.7 \text{ mm}^2$ ) and X-ray diffraction (XRD) patterns were obtained with a Rigaku D-Max 2200 Series equipped with Cu-K $\alpha$  radiation ( $\lambda = 1.54 \text{ \AA}$ ) at a scanning rate of  $3^\circ$  per minute. The tube voltage was 40 kV, and the current was 40 mA. Also, an extensive study of the particle size distribution was carried out by an analytical technique such as dynamic light scattering (DLS) method by using Malvern Zetasizer Nano ZS Laser Particle Size Distribution Meter.

## 4. Results & discussion

Microwave energy-assisted method and ultrasound energy-assisted method were studied, and the final products were obtained. Synthesized carbon products were analyzed by applying different characterization techniques such as XRD, AFM, TEM.

### 4.1 Microwave (MW) assisted method results

All the results of ammonia tests were summarized in **Table 1**. According to the results; sonication did not create a positive effect on electrical conductivity of final product. Lower temperature conditions give better yield and electrical conductivity results.

According to these results which were given in **Table 1**, low temperature showed better electrical conductivity results. Sonication step built a negative effect on electrical conductivity results. Also, after annealing step, electrical conductivity results slightly increased.

Another set of experiment were done in order to compare the effect of different solvents on graphene synthesis via microwave energy. The results of microwave tests that were conducted by using N,N-Dimethyl formamide (DMF), ethylene glycol (EG) and ethylene diamine (ED) were given in **Table 2**.

According to the results which were given in **Table 2**, the reaction yields of DMF, EG, and ED are 60, 88, and 75%, respectively. The electrical conductivity values of DMF, EG, and ED are 22.716, 6.0002, 7.0967 S/m, respectively. It can be concluded that; G-DMF shows better conductivity performance.

XRD spectra of natural graphite, MW assisted expanded graphite products which were obtained in different solvents such as ethylene glycol, ammonia, and DMF were given in **Figure 7**, respectively.

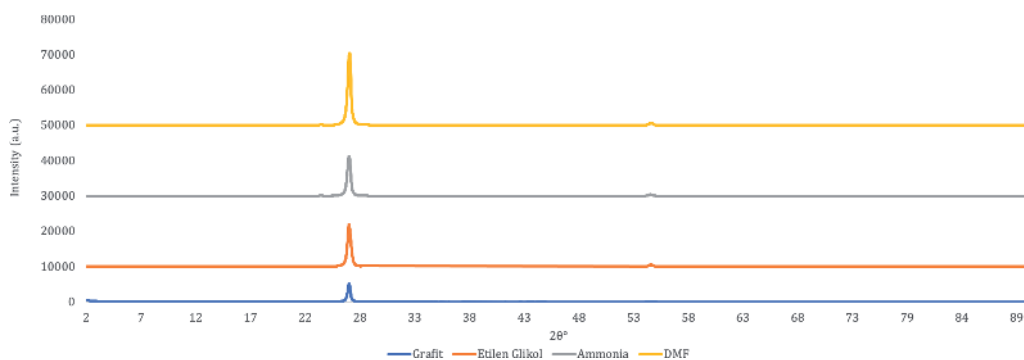
According to XRD results; all the spectrums show the 002 peak of graphite was predominant in all the four types of graphite, at  $2\theta^\circ = 26.44^\circ$  peak, which is characteristic for graphite. Natural graphite shows highest intensity peak at  $2\theta^\circ = 26.44^\circ$ . The intensity of other two peaks 101, 004 was low at all the spectrums. Layer

Exp. No	Carbon source	Solvent	Sonication step	React. Cond.	Yield (%)	Elec. cond. (S/m)	E. cond. (After annealing) (S/m)
1	Natural graphite (0.5 g)	25% Ammonia	—	120°C, 1 bar, 50 watt	94	52.44	58.114
2	Natural graphite (0.5 g)	25% Ammonia	—	120°C, 1 bar, 50 watt	89	12.8	30.647
3	Natural graphite (0.5 g)	25% Ammonia	30 min mode 5 power 50%	200 °C, 1 bar, 50 watt	53.5	9.06	12.047

**Table 1.**  
Results of experiments that were done by using ammonia.

Exp. No	Carbon source	Solvent	Sonication step	React. Cond.	Yield (%)	Elec. cond. (S/m)
4	Natural graphite (0.1 g)	DMF (50 ml)	10', 200 W, 20kHz, mode 5, power 50%	30 min 180°C	60	22.7
5	Natural graphite (0.1 g)	EG (50 ml)	10', 200 W, 20kHz, mode 5, power 50%	30 min 180°C	88	6
6	Natural graphite (0.1 g)	ED (50 ml)	10', 200 W, 20 kHz, mode 5, power 50%	30 min 180°C	75	7.1

**Table 2.**  
Microwave tests that were conducted by using DMF, EG and ED.



**Figure 7.**  
XRD spectra of commercial graphite and the MW-assisted graphene products which were obtained in ethylene glycol, ammonia, and DMF.

numbers of final products calculating by using XRD data were presented at **Table 3**. Layer numbers of expanded graphite products, which were obtained in EG, ammonia, and DMF by using MW energy, were calculated as 1.5 for all solvents. Layer number of natural graphite was calculated as 1.75 by the help of XRD results.

The results of another experiment plan which covering the usage of wide scale of solvents including n-Hexadecane (n-Hexa), Dimethylsulfoxide (DMSO), Sodium



Code	Layer number
Ethylene glycol (EG)	1.5
Ammonia	1.5
N,N-Dimethyl formamide (DMF)	1.5
Natural graphite	1.75

**Table 3.**  
 Layer numbers of final products calculating from XRD results.

Hydroxide (50% aq.) (NaOH), 1-octanol (OCTA), Perchloric acid (PA), N,N-Dimethyl formamide (DMF), Ethylene glycol (EG), and Ethylene diamine (ED) were presented in **Table 4**.

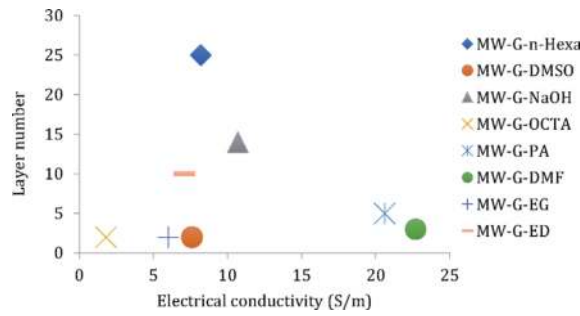
According to the results, MW-G-DMF showed the highest electrical conductivity. Electrical conductivities of MW assisted graphene products were higher when the used chemicals have 2–4 Debye (D) dipole moments. These results are compatible with the dielectric constants and surface tensions of the used chemicals. Layer numbers were calculated by Scherrer equation and the half-width of the diffraction line  $\beta(2\theta)$  (in rad) was taken as the experimental half-width ( $\beta_{exp}$ ) and was corrected for experimental broadening ( $\beta_{instr}$ ) as described in Saberi et al.'s study [30]. Layer numbers show distribution between 10 and 16. MW-G-EG showed the thinnest layer number with the value of 5.5, which is seen at **Table 4**. Solvents that have surface tension bigger than 40 mN/m show better layer number results. Briefly, as the surface tensions increased, layer numbers decreased. These results are supported with Hernandez et al.'s study [29]. Electrical conductivities of MW assisted graphene products were higher when the used chemicals have 2–4 Debye (D) dipole moments as seen as in **Table 4**. When the dielectric constants ( $\epsilon$ ) get larger, electrical conductivity values of synthesized products increased.

MW-G-PA showed the optimum electrical conductivity and layer number values for the MW assisted graphene synthesis as seen in **Figure 8**.

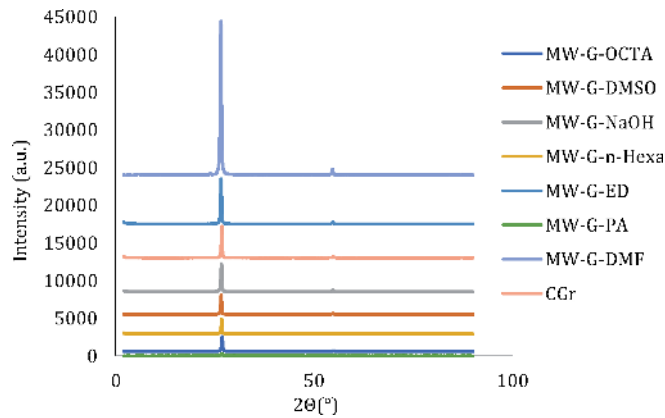
All XRD spectrums showed peak at 26.5° which can be seen in **Figure 9**. XRD spectra of MW- G-PA also proved that graphite peak at 26.5° shows minimum intensity.

Solvent	Dipole moment (Debye)	Dielectric constant ( $\epsilon$ )	Layer number	Surface Tension @ 20 °C (mN/m)	Elect. conductivity (S/m)
n-Hexadecane	0.06	2	15.81	27.47	8.174
Dimethylsulfoxide	3.96	46.7	12.36	43.54	7.581
Sodium Hydroxide (50% aq.)	6.832	57.5	10.33	74.35	10.664
1-octanol	1.76	3.4	14.02	27.6	1.784
Perchloric acid	2.146	115	10	69.69	20.619
N,N-Dimethyl formamide	3.86	36.7	15	37.1	22.716
Ethylene glycol	2.746	37	5.5	47.7	6.002
Ethylene diamine	1.83	16	10.61	42	7.097

**Table 4.**  
 Electrical conductivities, dipole moments, layer numbers and dielectric constants of MW supported graphene products.



**Figure 8.**  
Relation between layer numbers and electrical conductivity.

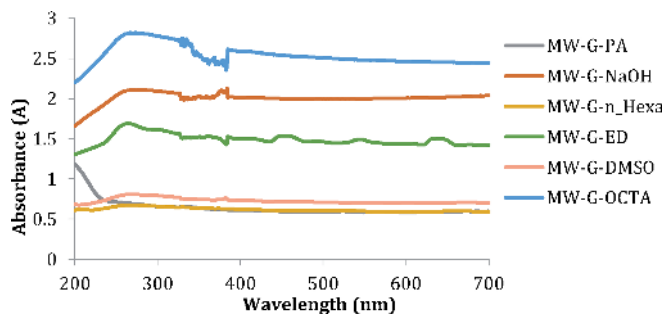


**Figure 9.**  
XRD spectra of MW-assisted graphene products.

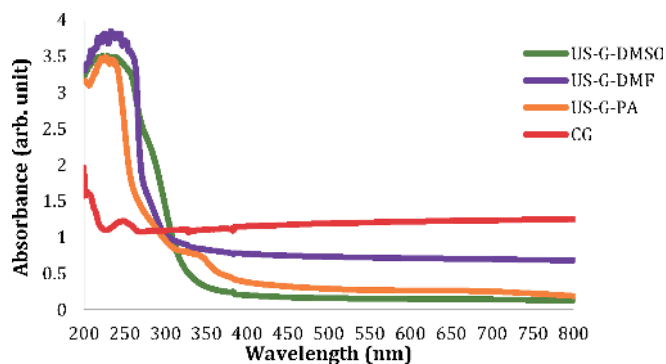
According to the UV–vis spectrums of MW-assisted graphene samples, which are presented in **Figure 10**, synthesized graphene samples, which were labeled as MW- G-PA, MW-G-NaOH, MW-G-n-Hexa, MW-G-ED, MW-G-DMSO, and MW-G-OCTA showed peak at 265 nm wavelength that referring  $sp^2$  C=C bonds. This result is in line with the previous literature [31].

#### 4.2 Ultrasound (US) assisted method results

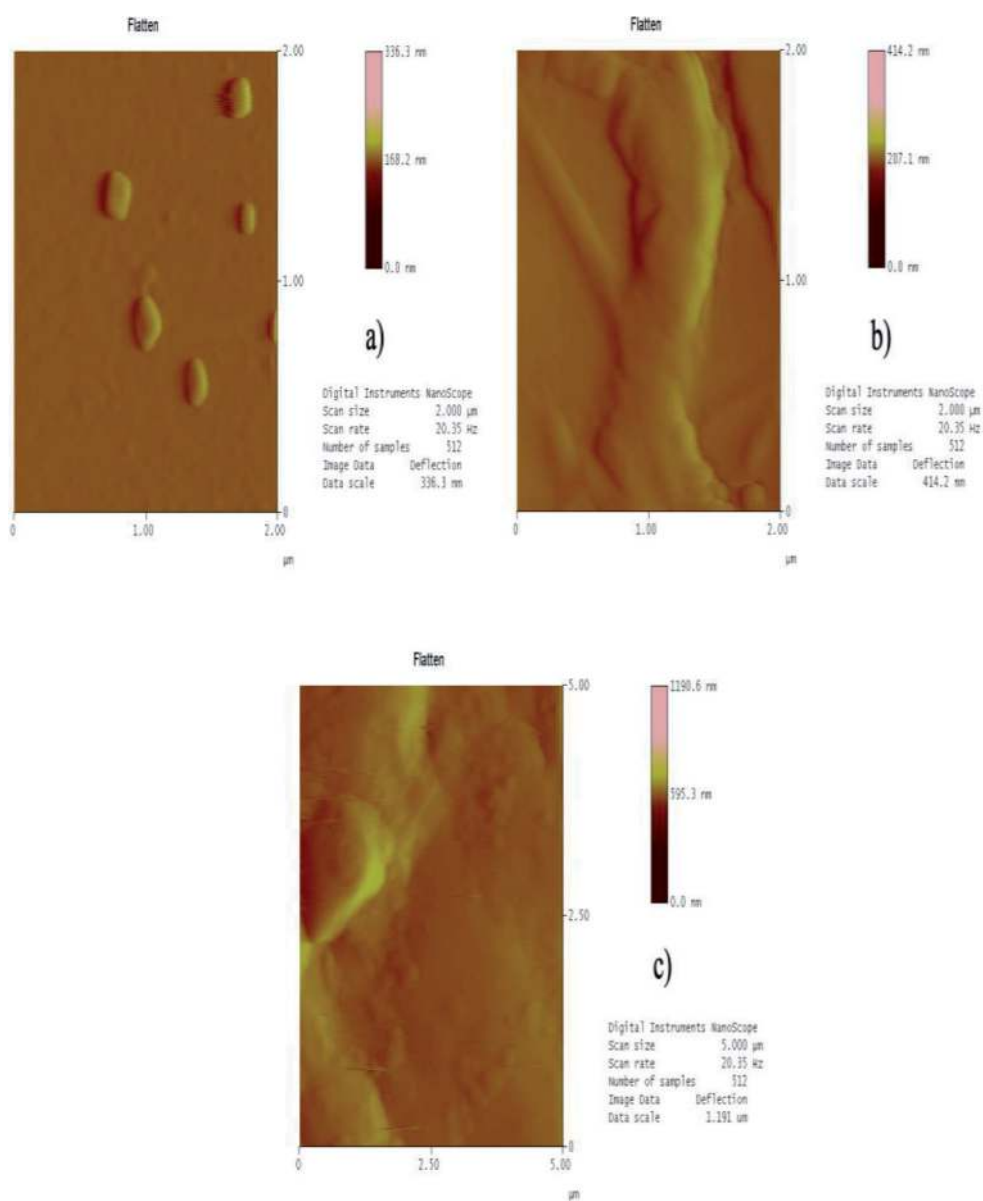
The US-assisted synthesized graphene products were characterized by using UV–vis spectroscopy, AFM Spectroscopy, and DLS analysis. UV–vis spectrums of US-assisted graphene products are presented in **Figure 11**. Coleman’s team calculated the absorption coefficient of graphene dispersion via UV/vis spectroscopy.



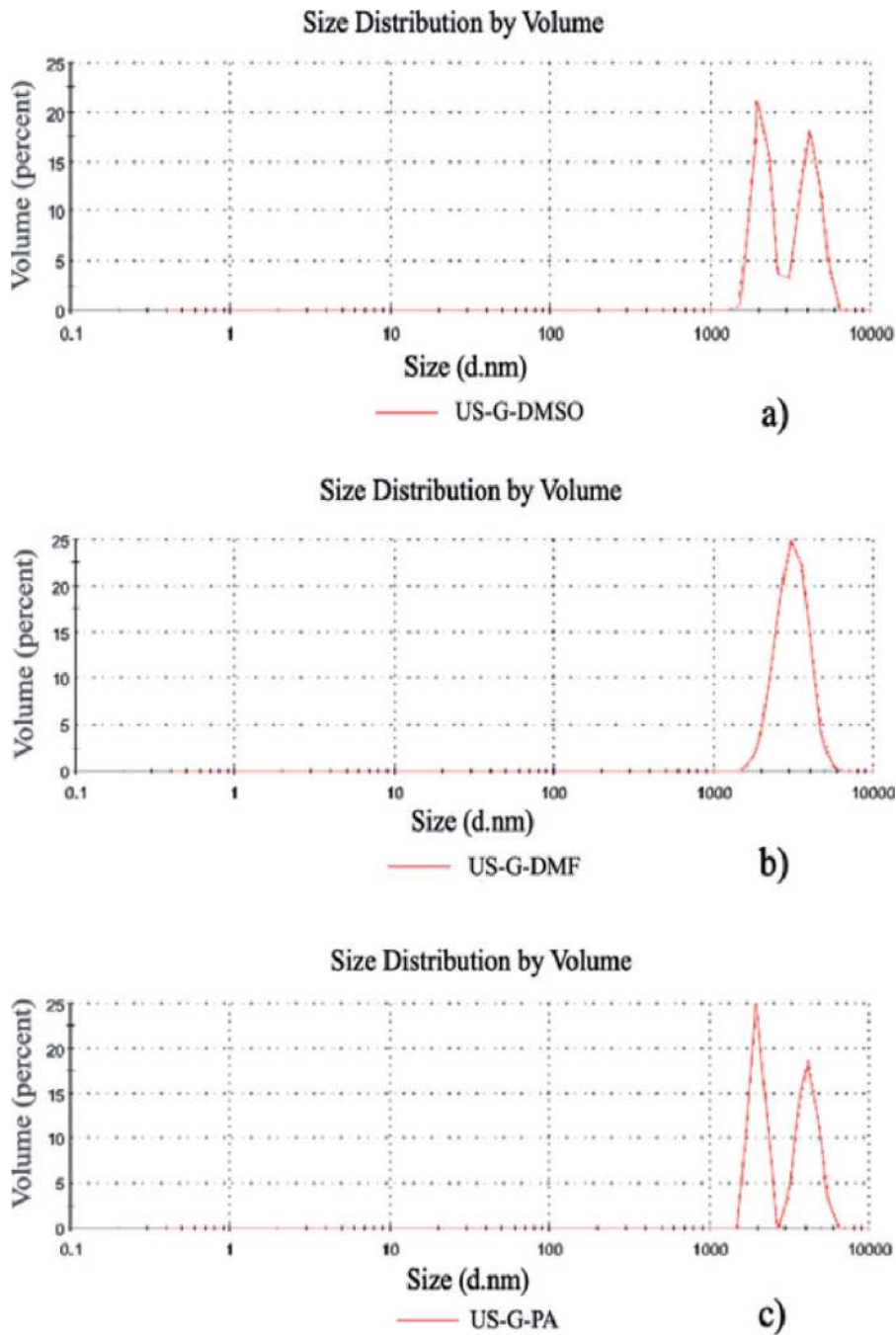
**Figure 10.**  
UV spectrums of MW based synthesized graphene products.



**Figure 11.**  
UV-vis spectra of CG, US-G-DMSO, US-G-DMF, and US-G-PA products.



**Figure 12.**  
The AFM images of (a) US-G-DMSO, (b) US-G-DMF, and (c) US-G-PA drop casted onto glass piece showing the homogeneous structure of the pristine graphene nanosheets.



**Figure 13.** Lateral size results of synthesized samples, (a) US-G-DMSO, (b) US-G-DMF, (c) US-G-PA.

Concisely, with the help of the Beer–Lambert law, absorption coefficient ( $A = \alpha cl$ ) of graphene could be found by using dispersion at specific concentrations [29, 32–35]. UV–Vis absorbance spectroscopy was conducted at fixed wavenumbers of 253 nm for graphene. A piercing peak at 210 nm can be noticed and one more peak around 226 nm with a little bit less intensity of absorption peak is also observed due to  $\Pi$ - $\Pi^*$  bondings of the C-C aromatic rings.

The obtained graphene samples, which are labeled as US-G-DMSO, US-G-DMF and US-G-PA, show peak at 265 nm wavelength that referring  $sp^2$  C=C bonds [31].

AFM characterization of final graphene products (US-G-DMF, US-G-DMSO, US-G-PA) were conducted to determine the optimal growth condition by measuring surface roughness and thickness. The AFM images of US-G-DMSO, US-G-DMF, and US-G-PA were presented in **Figure 12**. The Ra values of US-G-DMSO, US-G-DMF, and US-G-PA are 2.937, 6.343, and 10.103 nm, respectively. The Rq values of US-G-DMSO, US-G-DMF, and US-G-PA are 3.471, 8.046, and 11.748 nm, respectively. The RMS values of US-G-DMSO, US-G-DMF, and US-G-PA are 5.675, 8.842, and 11.910 nm, respectively. Vertical distance denotes the thickness of graphene and it is determined for US-G-DMSO, US-G-DMF, and US-G-PA as 1.638, 2.151, and 10.754 nm, respectively. The layer numbers were calculated via following equation:  $N = (t_{\text{measured}} - 0.4)/0.335$ .

The layer numbers of US-G-DMSO, US-G-DMF, and US-G-PA are calculated as 4, 5, and 31, respectively. According to AFM results, best result was obtained with DMSO. All these results confirmed that the US-G-DMSO materials had fewer layers and defects.

Although these techniques can determine the size of graphene products, dynamic light scattering (DLS) is also helpful to measure the lateral size. It is an easy and quick method for evaluating the size of graphene samples [36]. The size distribution of the synthesized graphene samples using DLS are shown in **Figure 13**. Z-average hydrodynamic radius (Rh) of US-G-DMF is 3846 nm, Rh of US-G-DMSO is 6930 nm, and Rh of US-G-PA is 7137 nm. According to these results, DMF provides graphene products with smallest lateral size.

## 5. Conclusion

Microwave (MW)-assisted method was developed. Although many solvents have been studied, carbon product, which was synthesized in DMF, showed the highest electrical conductivity. Electrical conductivities of MW-assisted graphene products were higher when the used solvents have 2–4 Debye (D) dipole moments. These results are compatible with the dielectric constants and surface tensions of the used chemicals. Layer numbers show distribution between 10 and 16. EG has minimum layer number with the value of 5.5. Solvents that have surface tension bigger than 40 mN/m show better layer number results. When the dielectric constants ( $\epsilon$ ) get larger, electrical conductivity values of synthesized products increased. As the surface tensions increased, layer numbers decreased. PA showed the optimum electrical conductivity and layer number values for the MW-assisted graphene synthesis. According to the UV-vis spectrums of MW assisted graphene samples. The obtained graphene samples, which were labeled as MW-G-PA, MW-G-NaOH, MW-G-n-Hexa, MW-G-ED, MW-G-DMSO, and MW-G-OCTA showed peak at 265 nm wavelength that referring  $sp^2$  C=C bonds.

Ultrasound (US)-assisted method was studied. Graphene samples were easily synthesized via solution-based process. According to the UV-vis spectrums, all graphene products gave peak at 265 nm wavelengths, which may be caused by the ultrasonication required for proper suspension using the solution-based process. Also, as a result of AFM analyses, US-G-DMSO has four layers, US-G-DMF has five layers and US-G-PA has thirty-one layers. It can be understood that DMSO shows better solvent effect on graphite exfoliation by sonication process. Z-average hydrodynamic radius (Rh) of US-G-DMF is 3846 nm, Rh of US-G-DMSO is 6930 nm, and Rh of US-G-PA is 7137 nm. It can be concluded that, DMF provides graphene products with smallest lateral size.

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## **Conflict of interest**

The authors declare no conflict of interest.

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