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# Enhancing Liquid Phase Exfoliation of Graphene in Organic Solvents with Additives

*Randhir Singh Bhoria*

## Abstract

Graphene is the wonder carbon nanomaterial with excellent electrical, mechanical, chemical and optical properties suitable for the fabrication of modern electronics devices such as supercapacitors, sensors, FET etc. Liquid phase exfoliation is the economical, safe, facile method of graphene synthesis without the requirement of harmful chemicals, toxic gases. However, the low concentration of graphene (<0.01 mg/ml) obtained by this method limits its application in various fields. Various techniques have been employed for enhancing the graphene concentration in certain organic solvents. Addition of additives and salts can enhance the graphene concentration in organic solvents to some extent. In this chapter, the earlier work done in enhancing graphene concentration is explained. Further, this technique is employed for graphene concentration enhancement in solvents by using new salts and additives. The results obtained with various additives are compared and it was found that by adding anthracene in NMP solvent graphene concentration increases upto 0.04 mg/ml. This process can be easily scaled up for better performance, and resulting high concentration graphene can be used for the fabrication of the efficient modern electronics devices.

**Keywords:** graphene synthesis, liquid phase exfoliation, additives, concentration enhancement, organic solvents

## 1. Introduction

Graphene is the most studied and explored nanomaterial with exceptional mechanical, electrical, optical and chemical properties. Graphene was discovered by Kostya Novoselov and Andre Geim via mechanical exfoliation method. Graphene has 2D hexagonal honeycomb structure made up of carbon atoms. Graphene is highly transparent as it reflects 2.3% and transmits 97.7% of light falling on it which makes it highly useful for making transparent conducting electrodes. Other exotic properties of graphene are high carrier mobility ( $200,000 \text{ cm}^2 \text{ v}^{-1} \text{ s}^{-1}$ ), Young's modulus of 1.0 TPa. Graphene is approximately 200 times more conductive than copper and 100 times stronger than steel. In addition to this it is very flexible in nature as it can be stretched to 20% of its original length.

Because of its very high electrical conductivity, high transparency and flexibility it is being used for the fabrication of wide variety of devices such as flexible transparent displays, energy storage devices etc. Graphene can be prepared by various methods such as mechanical exfoliation, electrochemical, liquid phase exfoliation

and CVD method. This chapter uses liquid phase exfoliation method for the synthesis and concentration enhancement of graphene, after discussing its relative advantages over other techniques.

## 2. Graphene synthesis methods

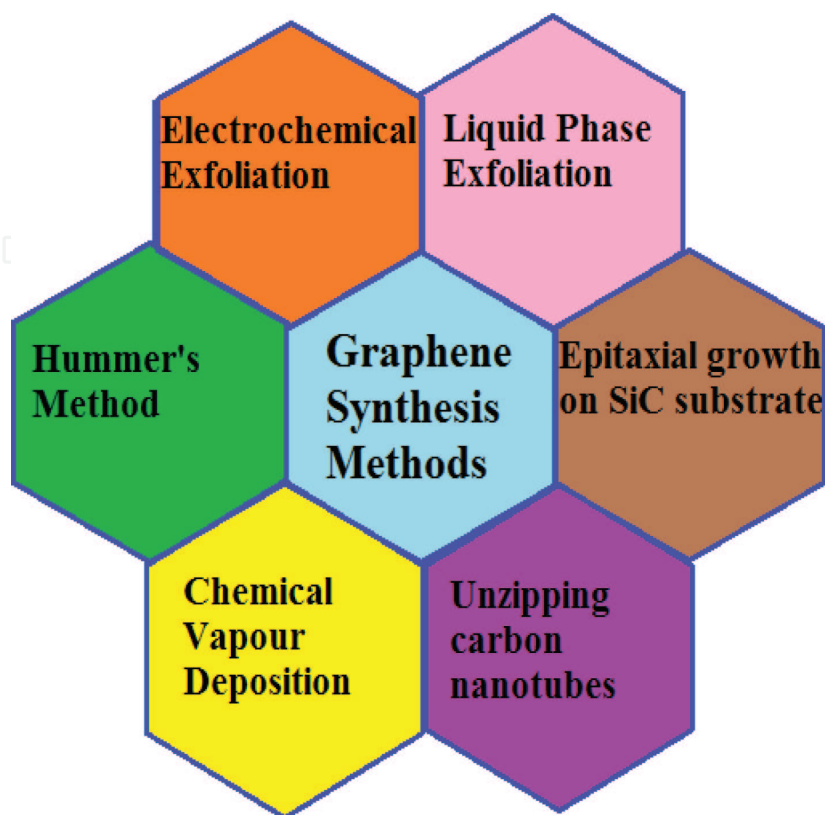
After the discovery of graphene a lot of research has been done for finding a suitable technique of graphene synthesis. Graphene synthesis method should be facile, economical and can be performed easily in the laboratory and should not require sophisticated equipment. Some of the most prominent graphene synthesis methods are described here (**Figure 1**).

### 2.1 Mechanical exfoliation method

Mechanical exfoliation method is the oldest and the simplest method, can be easily used in the college lab to prepare graphene of few micrometer length. It is called scotch tape method because here, a scotch tape repeatedly peels off various layers of graphene from the graphite source [1]. The disadvantage of this method is that it is time consuming and does not give graphene sheets of uniform thickness. Moreover, it is not a scalable method to produce high quality graphene sheets.

### 2.2 Chemical exfoliation method

Chemical exfoliation method uses harmful acids for the exfoliation of graphene sheets from graphite source. When graphite powder is mixed in solution of sulfuric acid and nitric acid, the inter-planar distance between individual graphene sheets



**Figure 1.**  
*Various synthesis methods of graphene.*

increases and hence exfoliation occurs [2]. The technique of increasing the inter-planar distance between graphene sheets is called as intercalation [3]. The advantage of the intercalation is that after intercalation, the intercalated graphite can be easily exfoliated via sonication. Other processes such as ultrasonic heating [4], acid treatment [5] are used for synthesis of graphene nanoribbons.

### 2.3 Chemical vapor deposition (CVD) method

CVD technique is used to produce good quality graphene on various substrates such as copper, nickel, cobalt etc. Here, the substrate is placed inside a furnace and hydrocarbon gas is passed at high temperatures. The carbon present in the hydrocarbon gas gets deposited on the substrate to form a graphene layer. Usually a mixture of hydrogen, argon and methane gas is passed through the furnace at temperature of 750–1200°C (**Figure 2**).

The CVD method is useful for scalable synthesis of high quality graphene. Using this method graphene has been successfully deposited upon various substrates such as Ni [6], Rh [7], Pt [8–11], Ir [12], Ru [13–16], Pd [17], Cu [18–21] using methane and ethylene. Graphene has also been prepared by using table sugar as a solid carbon source [22].

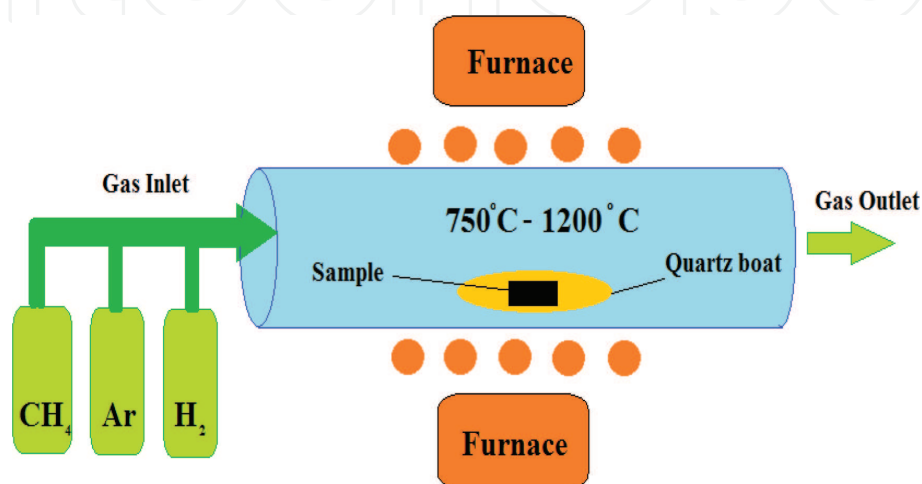
### 2.4 Epitaxial growth on SiC

Graphene layer can be grown on the silicon carbide substrate by heating at high temperature greater than 1100°C (**Figure 3**). The thickness of the graphene film which is prepared on SiC substrate depends upon size of the silicon carbide substrate because Si atoms are desorbed from surface during this process [23–25].

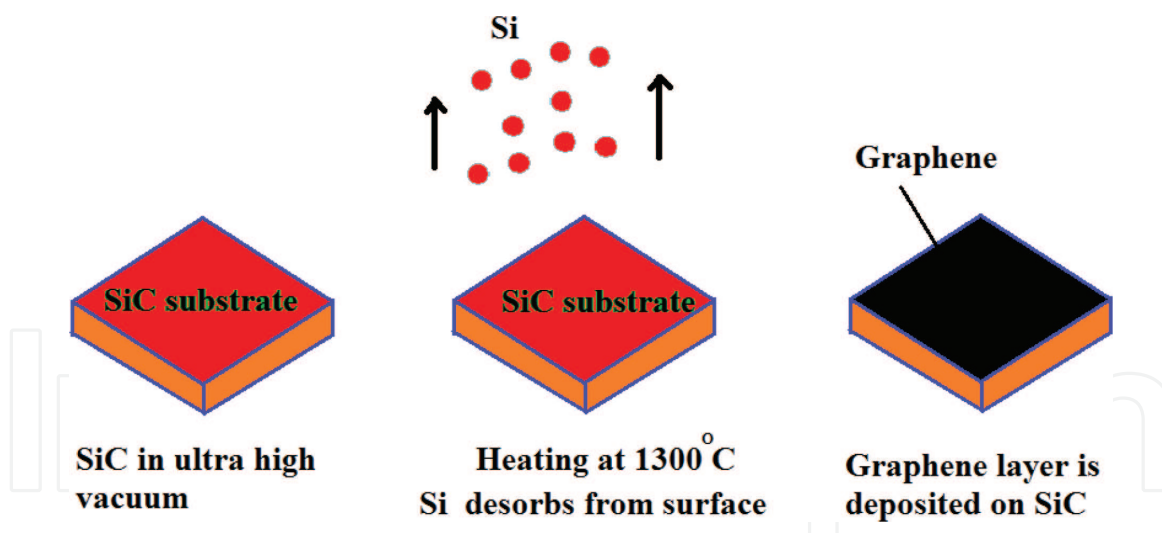
Whether SiC face is silicon or carbon terminated, graphene layer thickness and carrier density changes accordingly [26].

### 2.5 Hummer's method

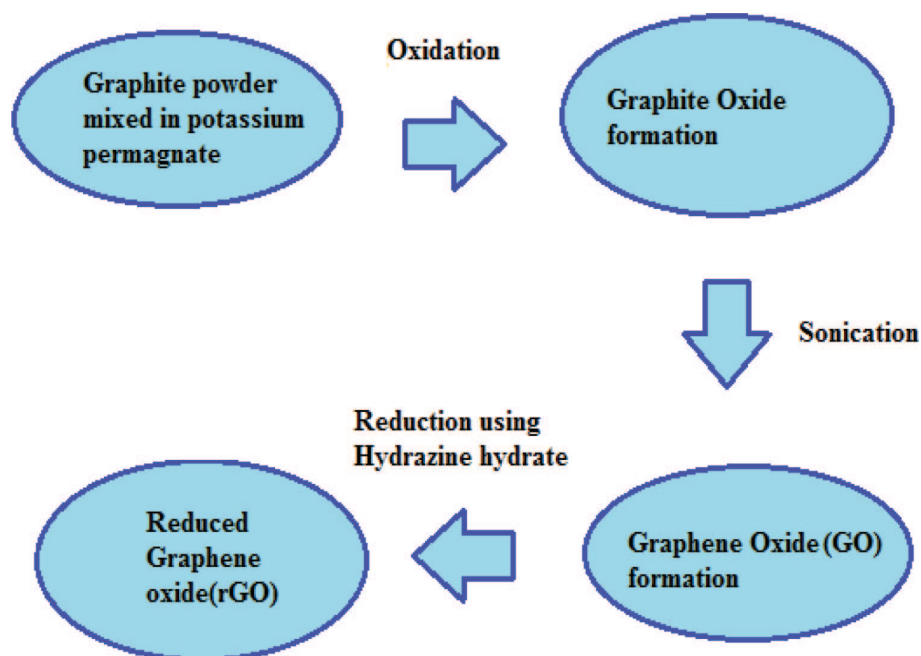
Hummer's method is a well-known method to prepare high yield graphene. Here, graphite powder is first converted to graphite oxide with potassium permanganate, which is further converted into graphene oxide (GO) via sonication process. Graphene oxide is later reduced to reduced graphene oxide (rGO) using various reducing agents such as hydrazine hydrate (**Figure 4**).



**Figure 2.**  
Schematic illustration of CVD method of graphene synthesis.



**Figure 3.**  
Process of epitaxial growth of graphene on SiC surface.



**Figure 4.**  
Synthesis of reduced graphene oxide using Hummer's method.

Most of rGO properties match with graphene, but due to structural defects in rGO, it does not produce high quality graphene. Moreover, because of the use of harmful and toxic chemicals, it is also not safe method.

## 2.6 Electrochemical exfoliation method

Electrochemical exfoliation method is another method for producing graphene from graphite rod in much shorter time as compared to CVD and Hummer's methods.

Typically, a platinum wire acts as cathode and graphite rod acts as anode (Figure 5).

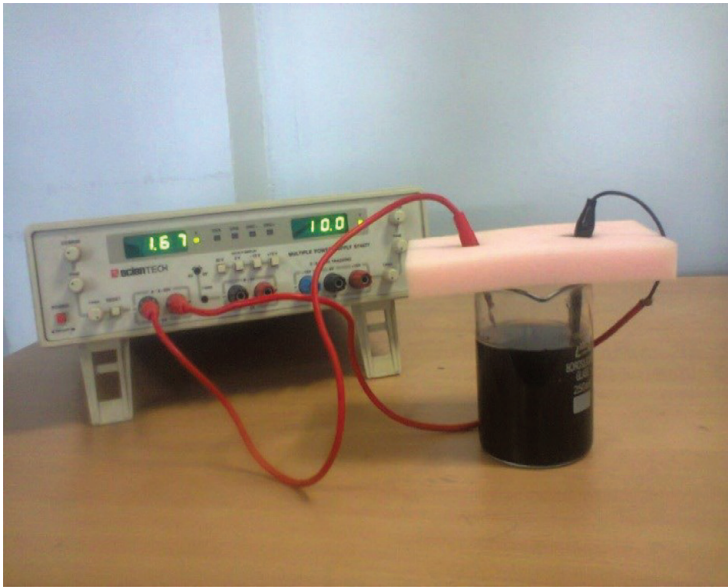
Both anode and cathode are dipped in an electrolyte solution which is usually an acid solution such as sulfuric acid, phosphoric acid etc. After applying 10 V DC between anode and cathode, graphene exfoliation starts [27]. After 2 h graphene nanosheets accumulated in electrolyte are filtered, washed and dried for characterization using SEM, TEM etc. In this method of graphene synthesis also uses various toxic and harmful acids and chemicals.



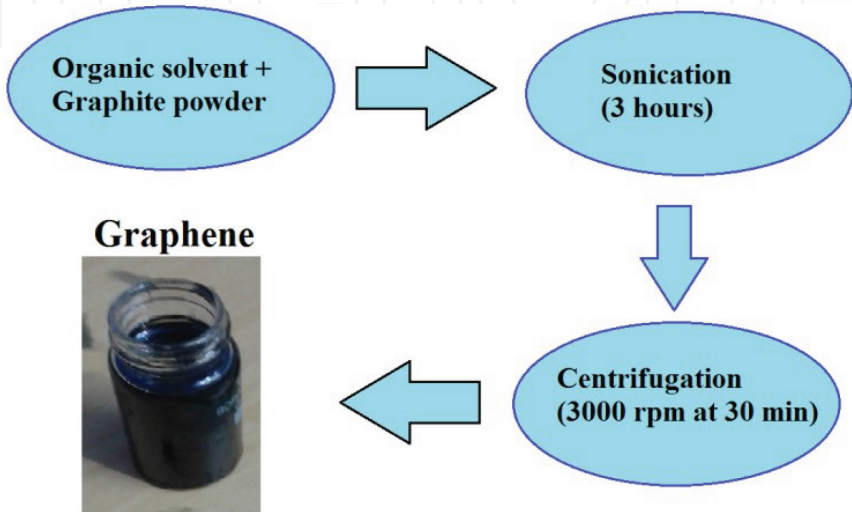
## 2.7 Liquid phase exfoliation

Liquid phase exfoliation of graphene uses sonication process to exfoliate graphene from graphite in solvents. Graphite has various layers of graphene attached by Van der Waals forces which is overcome if the solvent used has surface tension near 40–50 mJ/m<sup>2</sup> range. Some of the commonly used solvents are DMF (N,N-Dimethylformamide) and ODCB (ortho-dichlorobenzene). Typically, 2 g graphite powder is added to 300 ml of ODCB. This mixture is sonicated for 3 h. Then it is centrifuged for 30 min at 4000 rpm. (**Figure 6**) Finally supernatants are used for characterization using SEM and TEM [28].

**Table 1** shows that various synthesis methods have been explored by the researchers for graphene synthesis each having some disadvantages [29–34]. Hummer’s method usually gives high-yield graphene but suffers from defects and impurities in graphene structure. So, rGO prepared by Hummer’s method is not useful for the fabrication of electronics devices [35, 36]. Electronics devices fabrication requires good-quality defect-free graphene which can be easily prepared by



**Figure 5.**  
*Electrochemical exfoliation of graphene.*



**Figure 6.**  
*Various steps involved in liquid phase exfoliation of graphene.*

Synthesis method	Advantages	Disadvantages
Mechanical exfoliation method	Good quality, low yield	Not a scalable process, low yield
Chemical vapor deposition	High quality, large area graphene	High temperature and low vacuum conditions
Epitaxial growth on SiC	Large continuous film, good quality	High temperature and low vacuum conditions Not transferable
Hummer's method	High yield	High defects in graphene, harmful chemicals used
Electrochemical exfoliation	Lesser time, facile, economical	DC voltage and electrolytes requirements
<i>Liquid phase exfoliation</i>	<i>Easy, safe, high quality, economical</i>	<i>Long sonication time requirement, low graphene concentration</i>

*Note: The text is in italics to indicate that in this chapter liquid phase exfoliation method is used for the concentration enhancement of graphene.*

**Table 1.**  
*Various graphene synthesis methods with advantages and disadvantages.*

liquid-phase exfoliation, and cannot be produced by Hummer’s method [37–41]. The high quality graphene prepared by liquid-phase exfoliation is suitable for modern electronics device applications [42–47]. Graphene has high quantum capacitance [48], electrochemical properties [49] which can help to detect various explosives such as 2,4,6-trinitrotoluene [50], and generate chemiluminescence [51]. Although liquid phase exfoliation technique is the safe and environment friendly technique as compared to other graphene synthesis method, but its disadvantage is lower concentration of graphene obtained (usually less than 0.01 mg/ml). Therefore, there is a need of concentration enhancement of graphene for the fabrication of better and more efficient electronic devices.

3. Process of concentration enhancement

It has been experimentally found that graphene concentration can be enhanced by increasing sonication process time upto many weeks instead of hours which create defects in the graphene nanosheets [52]. Other approaches which have been utilized are mixed solvents [53–55], solvent exchange [56], solvothermal exfoliation [57, 58], intercalants [59–64]. It has been observed that Sodium hydroxide and naphthalene can enhance the graphene concentration in organic solvent [65, 66]. It has been experimentally found that solvents having surface tension near 40–50 mJ/m<sup>2</sup> are highly useful for graphene synthesis via sonication process. Some of the solvents satisfying this criterion such as 1-methyl-2-pyrrolidinone (NMP), benzyl benzoate (BB), 1,2-dichlorobenzene (ODCB), acetophenone (ACP), benzonitrile (BZN), dimethyl sulfoxide (DMSO) and 1,4-dioxane are used in concentration enhancement. It has been found that addition of organic salts can enhance the grapheme concentration in various organic solvents [67]. In addition to various salts, some additives such as phenolphthalein and anthracene have been explored for enhancing graphene concentration in solvents.

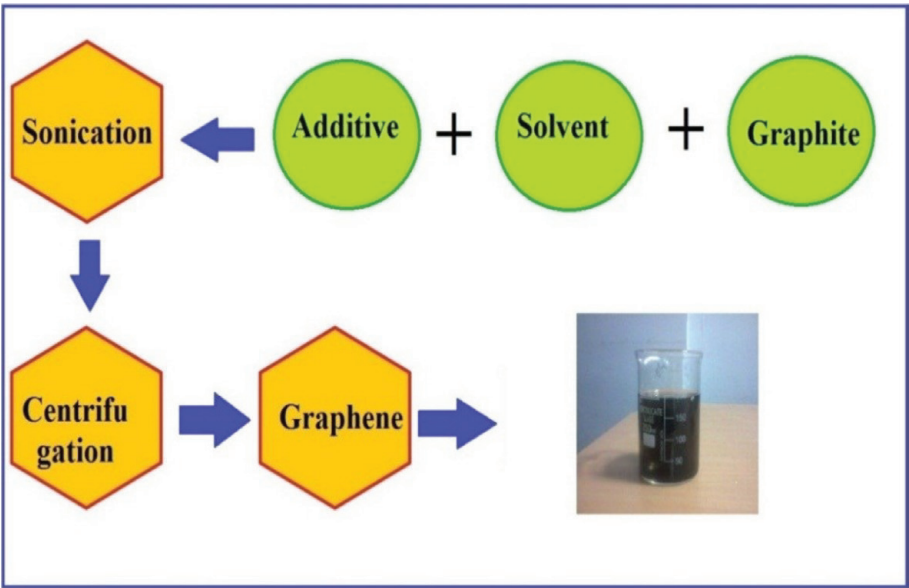
Initially, graphite powder is added to NMP, DMSO and CYN solvents (100 ml) with a concentration of 10 mg/ml. Then 100 mg of the additive or salt is added and sonicated for 3 h and centrifuged at 3000 rpm for 30 min (**Figure 7**). After centrifugation, supernatants are used for characterization using UV-Vis spectrum for finding graphene concentration by applying Lambert Beer’s law.

UV-Vis spectrum plots the variation of absorbance with wavelength. This technique is based on the principle that the absorption in a particular wavelength range is directly proportional to the color of the sample used for characterization. From the UV-Vis spectrum it is observed that at a particular wavelength the absorbance is maximum. For pure graphene sample the peak absorbance is obtained near 270 nm (**Figure 8**). As the impurities and functional groups are introduced in graphene the absorbance peak is shifted from 270 nm.

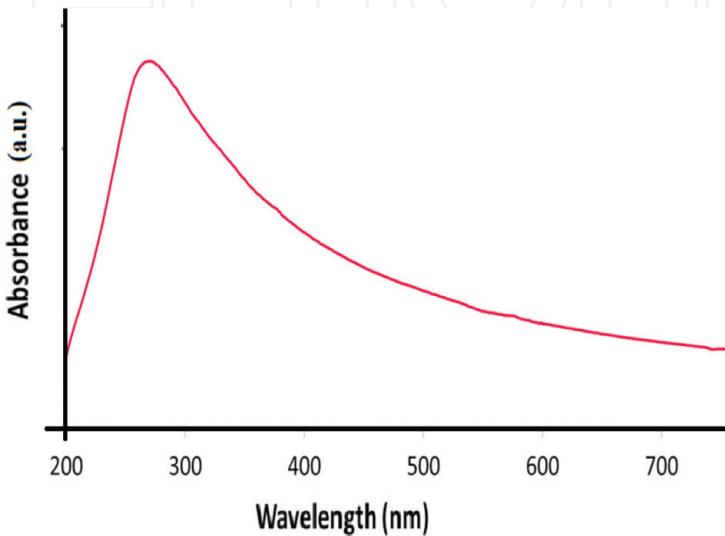
After plotting the UV-Vis spectrum curves we calculated the absorbance value at 660 nm wavelength. This absorbance  $A$  can be used to find the concentration of the graphene by applying Lambert Beer's law. So, according to the **Lambert Beer's law** the absorbance in terms of concentration is given by:

$$A = \alpha C l. \tag{1}$$

where  $A$  is the absorbance measured at 660 nm,  
 $l$  is the sample path length which is 1 cm,

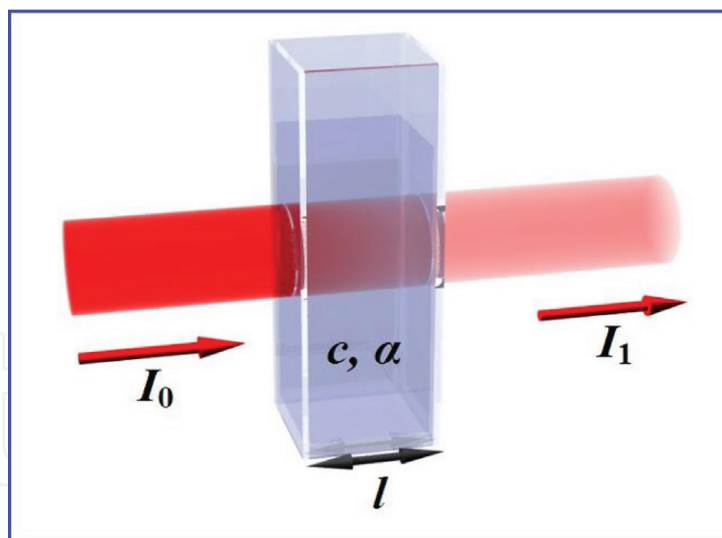


**Figure 7.**  
Schematic diagram of liquid phase exfoliation of graphene with addition of additive.

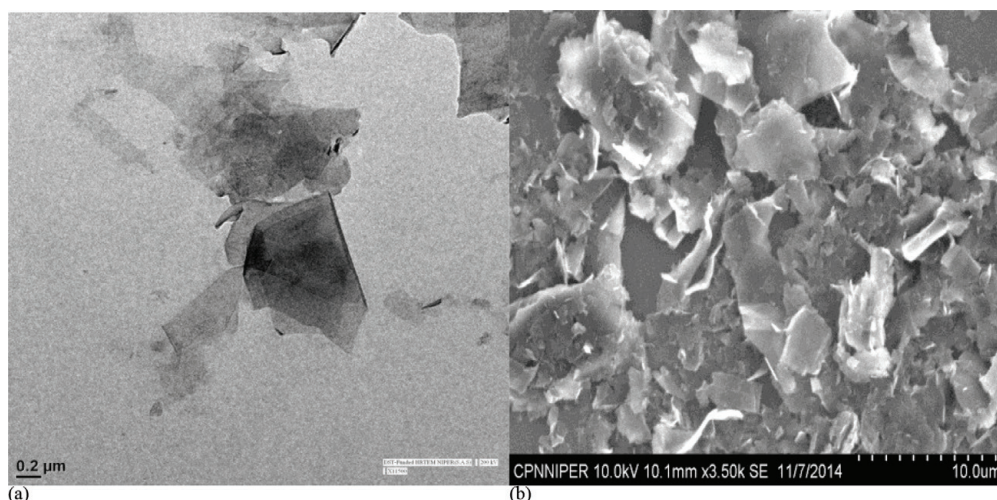


**Figure 8.**  
UV-Vis spectrum of graphene.





**Figure 9.**  
*Representation of Lambert-Beer's law.*



**Figure 10.**  
*(a) TEM images, (b) SEM images of graphene exfoliated in NMP solvent.*

**C** is the graphene concentration in the sample,

**$\alpha$**  is the extinction coefficient.

This process of UV-Vis spectroscopy is shown in **Figure 9** below.

The absorbance value at 660 nm in the UV spectrum of graphene is used to calculate the graphene concentration with absorption coefficient value  **$\alpha = 2460 \text{ ml/mg/m}$** .

It is observed that additives intercalate onto the graphitic layers which enhances graphene concentration by helping in the exfoliation process. The grapheme nanosheets produced by this process are used for the characterization using SEM and TEM. The TEM results of the grapheme nanosheets shows that single and few-layered, overlapped nanosheets have been produce (**Figure 10**).

**Figure 10** shows the SEM results of the graphene nanosheets produced in NMP organic solvent which indicates that graphene sheets size varies from 5  $\mu\text{m}$  to 20  $\mu\text{m}$ .

#### 4. Concentration enhancement with salts and additives

DMSO solvent is used for grapheme concentration enhancement by adding sodium tartrate (ST), sodium chloride (NaCl), potassium chloride (KCl), edetate

disodium (ED), sodium citrate (SC), naphthalene (N) and phenolphthalein (P) in it and analyzing their UV spectrum. The absorbance at 660 nm in UV spectrum is used for finding graphene concentration via Lambert Beer's law. By using this law graphene concentration is calculated and plotted in **Figure 11**. From **Figure 11** it is observed that SC and ST produces maximum concentration in DMSO.

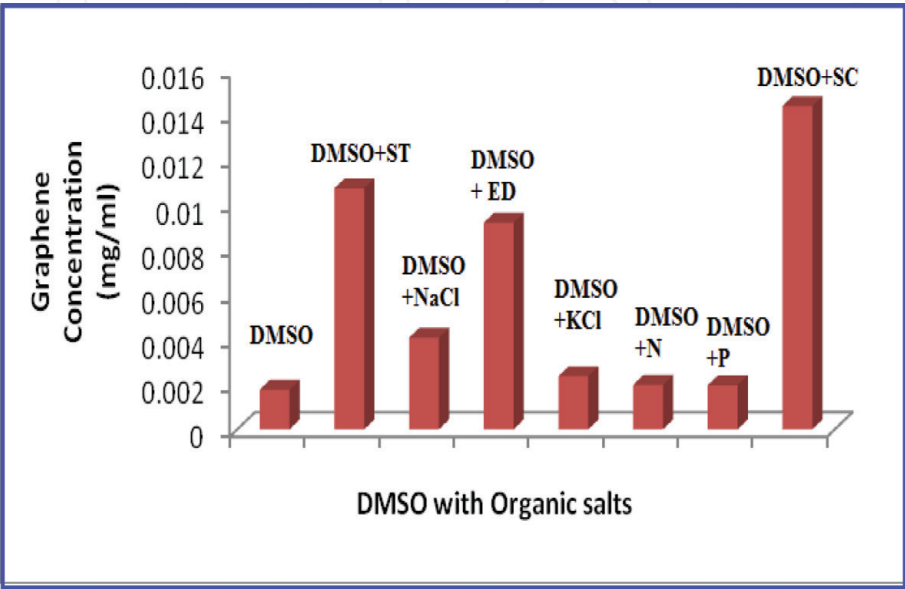
The main reason of the concentration enhancement is that additives intercalates onto graphene layers and helps in exfoliation process. It is also observed that lesser time is required for exfoliation after adding additives. Further, in contrast to organic salts, the inorganic salt KCl does not enhances the graphene concentration in DMSO. One reason for low concentration may be the aggregation of graphene nanosheets upon addition of KCl.

Firstly, the UV-Vis spectrum of the various graphene samples (with and without addition of additives in DMSO) is used to find the absorbance value at 660 nm. This absorbance value A, with  $\alpha$  the absorption coefficient is used to find graphene concentration as given by Beer's law,  $A = \alpha C l$ . Here, A is the absorbance at 660 nm, l is sample path length (1 cm), C is concentration and constant  $\alpha = 2460 \text{ ml/mg/m}$ . This procedure is repeated to calculate the graphene concentration in other solvents such as NMP, ODCB, BB, ACP, BZN etc. from their UV-Vis spectra.

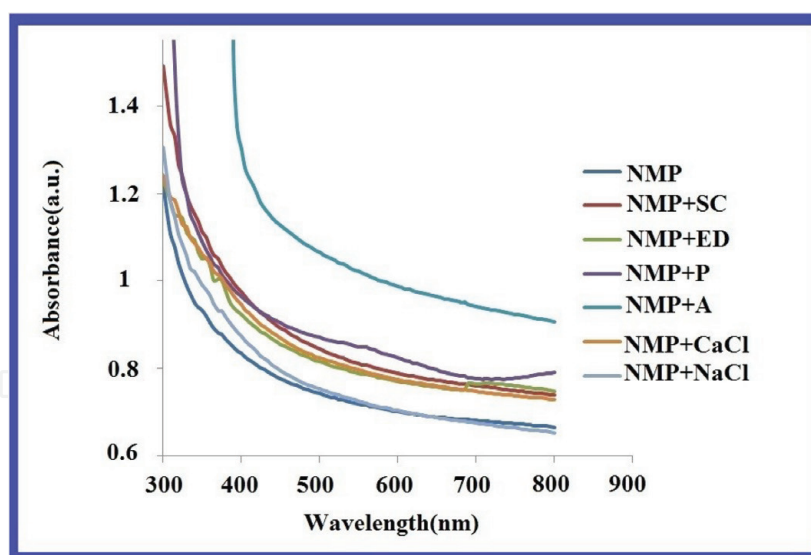
After repeating this procedure with the CYN solvents it is observed that in CYN maximum concentration is achieved by adding phenolphthalein additive. By adding phenolphthalein additive graphene concentration is increased upto nine times. But, as expected the inorganic salts such as CaCl, KCl does not enhance the graphene concentration. As compared to inorganic salts, organic salts are more useful for concentration enhancement with maximum concentration given by sodium citrate salt.

**Figure 12** shows the UV spectra of NMP with and without adding salts and additives. There is a great change in graphene concentration in NMP solvent after adding salts and additives in it with maximum graphene concentration of 0.08 mg/ml in NMP was observed by adding anthracene additive in it. In comparison to organic salts, inorganic salt KCl does not increase the graphene concentration in NMP. This low concentration may be again due to the aggregation effect.

**Figure 13** shows the effect of adding three additives anthracene, phenolphthalein, naphthalene in NMP, ODCB, BB, ACP, BZN, DMSO and 1,4-dioxane organic solvents. It has been observed that highest concentration is obtained by adding

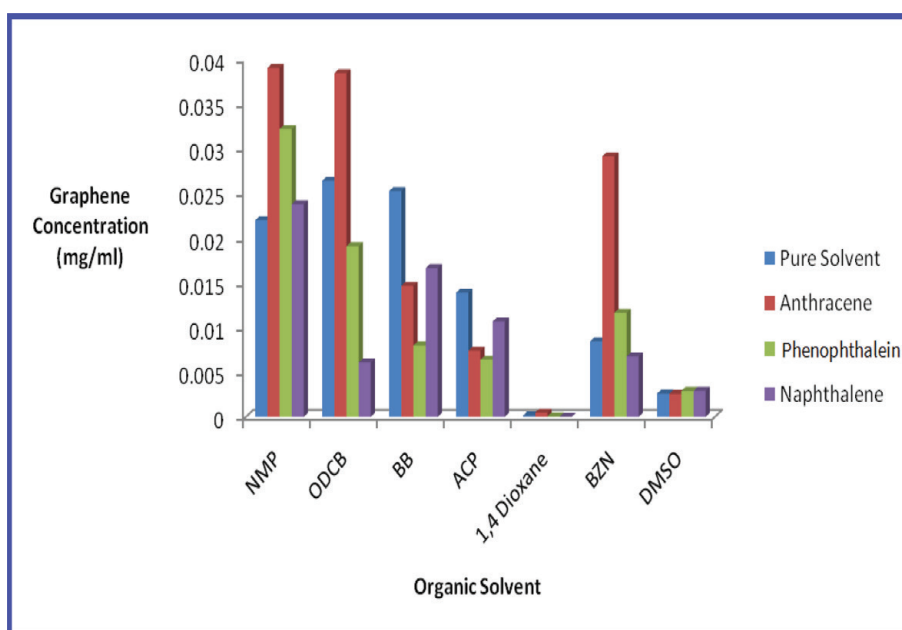


**Figure 11.**  
Concentration variation of graphene in DMSO solvent with addition of salts and additives.



**Figure 12.**

UV-Vis spectra of graphene in NMP solvent with and without addition of various salts and additives.



**Figure 13.**

Graphene concentration variation in seven organic solvents with and without addition of additives.

anthracene in NMP solvent, with 0.04 mg/ml concentration. Anthracene additive acts as molecular wedge between the individual edges of graphite and results in higher concentration. Anthracene when intercalates between the graphene layers, it increases the interplanar spacing between adjacent nanosheets and enhances the exfoliation process. It is also observed that by adding larger amount of anthracene additive (>100 mg) in the solvents, much lower concentration is obtained due to the aggregation effect. But as the additive amount decreases towards 100 mg, molecules are easily adsorbed onto graphene nanosheets which results in higher graphene concentration.

The structure of the additive and solvents also affects the concentration. Anthracene has structure of three benzene rings. Hence, those solvents which have benzene ring in its structure provides highest graphene concentration enhancement. It is found that solvents such as 1,4-dioxane and DMSO produces minimum graphene concentration as there is an absence of benzene ring structure in them [68]. NMP, BZN and ODCB exhibited maximum concentration of graphene

because of benzene ring structures. Two organic solvents BB and ACP have side chains in addition to benzene ring which restricts their intercalation between the individual graphitic layers and hence provides least graphene concentration as compared to other solvents [68].

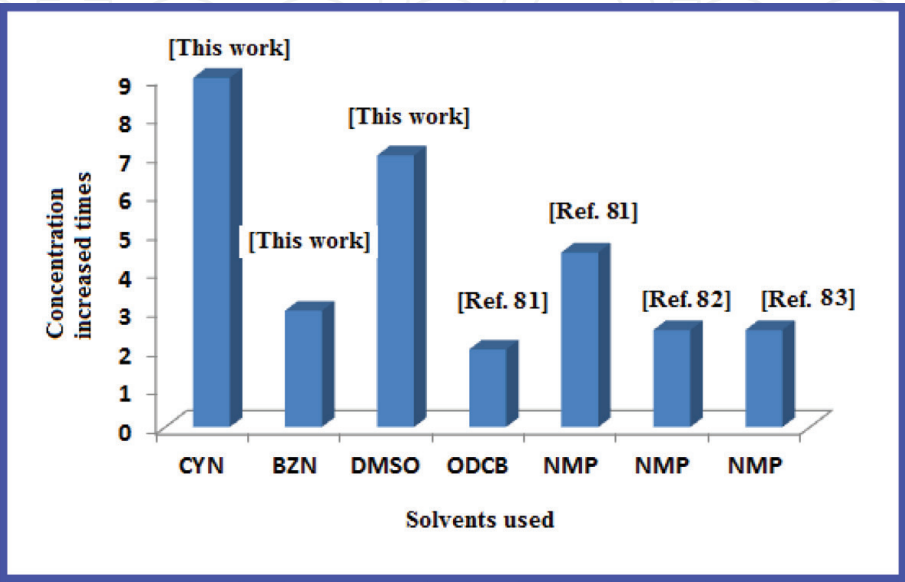
## 5. Results and discussions

Various additives and salts have been explored for graphene concentration enhancement in NMP, ODCB, DMF, CYN, DMSO solvents. It has been observed that addition of phenolphthalein in CYN increases graphene concentration from 0.005 to 0.045 mg/ml. With addition of SC salt in DMSO solvent, graphene concentration increased from 0.002 to 0.015 mg/ml which is comparable to earlier published works [27].

The effect of adding Anthracene additive in solvents NMP, DMSO, ODCB, BZN, ACP, BB and 1,4-dioxane have been explored and it has been observed that the concentration depends upon additive-solvent structures and interactions. Because the molecular structure of anthracene has three benzene rings, hence solvents with benzene ring structure produced maximum concentration such as NMP, BZN, ODCB solvents. The results obtained with various additives are compared and it was found that by adding anthracene in NMP solvent graphene concentration increases upto 0.04 mg/ml.

This result is in agreement with earlier published work on naphthalene additive [26]. Here, it is found that anthracene is more useful in BZN solvent (three times concentration) than naphthalene in NMP solvent (two times concentration) [26]. The effect of anthracene in concentration enhancement is most prominent in 1-methyl-2-pyrrolidone (NMP), orthodichlorobenzene (ODCB) and BZN solvents as compared to others. With addition of anthracene, the graphene concentration in NMP and ODCB solvents is increased to 0.04 mg/ml.

From **Figure 13** it is observed that benzyl benzoate (BB) solvent is not very effective in concentration enhancement with anthracene. The least concentration is exhibited by the organic solvents acetophenone, benzyl benzoate, 1,4-dioxane and DMSO. With addition of anthracene, the graphene concentration in NMP and ODCB solvents is increased to 0.04 mg/ml [28]. From **Figure 14** a comparison of



**Figure 14.**  
*Comparison of the enhanced concentration in this work with earlier published works.*



the graphene concentration enhancement in this work with the earlier published results has been made. The criterion of comparison is the how many times the concentration is increased with additives in various solvents such as CYN, BZN, DMSO, ODCB, NMP etc. From **Figure 14** it is observed that by adding additives graphene concentration is increased to approx. nine times in CYN solvent, approx. three times in BZN solvent, approx. seven times in DMSO solvents after experimental verification. On the other hand by observing earlier published results, graphene concentration was increased from 2.5 to 4.5 only in NMP solvent [69–71].

## 6. Conclusion


Generally liquid phase exfoliation produces graphene in low concentration ( $<0.01$  mg/ml) which is not suitable for electronics device fabrication. Various additives and salts have been explored for enhancing graphene concentration in organic solvents such as DMSO, CYN, NMP, ODCB, DMF etc. In this study new additives such as phenolphthalein and anthracene have been found to enhance the graphene concentration in solvents such as NMP, ODCB, DMSO and BZN etc. By adding phenolphthalein in cyclohexanone (CYN) solvent the graphene concentration was increased from 0.005 to 0.045 mg/ml and by adding sodium citrate (SC) organic salt, the graphene concentration in dimethyl sulfoxide (DMSO) was increased from 0.002 to 0.015 mg/ml. Further, the effect of addition of anthracene additive in seven organic solvents NMP, DMSO, ODCB, BZN, ACP, BB and 1,4-dioxane have been studied. It was observed that the additive-solvent interactions affect the graphene production yield. Since, the molecular structure of additive anthracene having three benzene rings matches with structure of the solvents such as NMP, BZN, hence causes enhancement in the graphene concentration. The results obtained with various additives are compared and it was found that by adding anthracene in NMP solvent graphene concentration increases upto 0.04 mg/ml. A significant concentration enhancement (three times) was observed with addition of anthracene additive in BZN solvent as compared to two times enhancement with addition of naphthalene in NMP solvent.

## Author details

Randhir Singh Bhoria  
University Institute of Engineering and Technology, Kurukshetra University,  
Kurukshetra, India

\*Address all correspondence to: [mr\\_randhir\\_singh@yahoo.co.in](mailto:mr_randhir_singh@yahoo.co.in)

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