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GRAPHENE SYNTHESIS AND ITS APPLICATIONS: A REVIEW

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Abstract:

Graphene is nothing but, a single layer of graphite crystal. Graphite crystal has stacks of carbon layers which are weekly bonded to each other. Due to this situation, these layers can easily slip over each other. Each layer has hexagonally arranged carbon atoms. These layers when appear as single, self standing material is graphene. It is the fundamental unit for other graphitic materials; hence we can say that, graphene is the basic building block for other graphitic materials. In this review paper, the emphasis was given on various methods employed for synthesis of graphene, its properties and applications. Here it is observed that, depending on the structure, various properties and applications of the graphene are changing, further due to more layers in graphene the entire structure of graphene becomes metallic in nature.

Key Words: Graphene, Exfoliation, Vapor Decomposition, etc.

1. Introduction:

In these days due to its exceptional properties like high current density, chemical inertness, high thermal conductivity, optical transmittance and many more, graphene has grabbed appreciable attention to be used as a next generation smart material. With the help of micromechanical cleavage technique, graphene was extracted from graphite in the beginning. This particular method was one of the easiest ways of production of high quality graphene crystallites. Some of the exceptional electrical properties of graphene have attracted application for future electronics such as ballistic transport, field emitter, transparent conducting electrodes and sensors. Graphene has a high electron mobility as well as low Johnson noise. But combination of excellent electrical property and low noise make graphene an excellent sensor, and is very efficient to detect absorbed molecules. Because of high electrical conductivity and high optical transparency graphene promotes as a suitable material for transparent conducting electrodes, which is required for the application in touch-screens, LCD's, organic photovoltaic cells and organic light emitting diodes.

These types of applications require growth of single-layer graphene on a suitable substrate and this is a very difficult work. Many researchers have done their work on graphene synthesis and most of these are based on mechanical exfoliation from graphite or thermal graphitization of Sic surface. But now days, chemical vapor deposition is preferred well. This article gives a brief overview of properties and various methods used in the synthesis of graphene and its applications.

2. Synthesis of Graphene:

Novoselov et. al. have been synthesized graphene in the year 2004. Before this work in the year 1999, as per literature survey there is a report of synthesis of grapheme [1]. But monolayer graphite synthesis was tried early in 1975, by B; Lamg et.al. They showed transformation of mono and multi layered graphite by thermal decomposition of carbon on single crystal pt. substrates [2].

2.1 Exfoliation and Cleavage:

Actually before going towards graphene, we must understand about graphite. Graphite is nothing but stacked layers of many graphene sheets which are bounded together by week Vander walls forces. Hence, normally it is possible to produce graphene from a high purity graphite sheet by breaking these bonds. In this technique mechanical or chemical energy is used to break weak bonds and as a result graphene sheets are separated from one another. In this case very first attempt was made by Viculies et. al., who have used potassium metal to intercolate a pure graphite sheet and then exfoliate it with ethanol to form dispersion of carbon sheets [3]. During sonication, the exfoliated nano-carbon sheets formed nanoscrolls. TEM analysis showed presence of 40 ± 15 layers in each sheet. Though these carbon nanoscrolls were much thicker than few layer graphene, this process showed that it is possible to separate layers of graphene from graphite with some modification.

As per Novoselov et. al; exfoliation is nothing but a repeated peeling process. Here, a commercially available high oriented pyrolytic graphite (HOPG) sheet of 1mm thickness was subjected to dry etching in oxygen plasma to make many 5 μm deep mesas [4]. This was then put on a photoresist and baked to stick the mesas to the photoresist. Then, a scotch tape was used to peel off layers from the graphite sheet. Thin flakes, attached to the photoresist, released in the acetone and transferred to a Si-substrate, were found to have single to few layer graphene sheets. This technique was later used to produce two dimensional atomic crystals of many

other materials, including BN, MoS_2, etc [4]. This process of producing graphene sheets was found to be very reliable and easy and thus, attracted the immediate attention of the scientific community [5, 6].

In another approach, mm-sized single to few layer graphene was produced by bonding bulk graphite to borosilicate glass followed by exfoliation, to leave single or few layers of graphene on the substrate [7]. In a somewhat different approach of exfoliation in a liquid phase, Stankovish et.al. Proposed to use hydrophobicity of graphite oxide (GO) and exfoliated GO nanosheets by ultra sonication in aqueous suspension and attempted reduction of the films in hydrazine hydrate at near about 100^0C for 24 hours [8, 9]. The process of exfoliation shows good promise to synthesize large scale graphene and some of the recent modifications in liquid phase are required. Due to oxidation-reduction process liquid phase exfoliation products are having defects, hence they are much poor in electrical properties.

2.2 Thermal Chemical Vapor Decomposition Method:

Graphene synthesis by this technique is very new. The very first report on few layer graphene synthesized by this method was found in the year 2006[(59)]. In this work camphor was used as a pre-cursor which is eco-friendly and low in cost. Treatment of camphor on Ni-foils was done. In this technique camphor was first evaporated at 180^0C and then pyrolyzed in another chamber of the CVD furnace, at 700 to 850^0C, using argon as the carrier gas. On natural cooling to room temperature few layer graphene sheets were observed on the Ni-foils. Graphene, produced by this particular technique found to have multiple folds. On the other hand in another approach, 1 to 2 mm thick graphene sheet was reported to be grown on Ni-subtrate by thermal CVD, while the same treatment failed to synthesis and graphene on Si. After that, Yu et.al. have reported three to four layer graphene formation on polycrystalline Ni foils through thermal CVD method [10]. Till this stage the researchers were depended on substrate for the synthesis of graphene. But Wang et.al. proposed a new method of growing substrate free few layered grapheme [11] . In their study they used MgO supported CO catalysts to grow graphene in a ceramic boat, at 1000^0C for 30 min, under a gas envelope of CH_4 and Ar. Obtained product was washed by concentrated HCL to remove MgO and CO, followed by distilled water and drying at 70^0C. It was noted that 500 mg of catalyst powder mixture can produce 50 mg of graphene, SEM, HRTEM and Raman spectroscopic analysis confirmed presence of rippled graphene sheets, having at least five layers.

Recent report of graphene synthesis has shown growth of single to few layer graphene on polycrystalline Ni film of 1-2 cm^2, by this CVD technique [12]. In this method, Ni film (500 nm thick) was evaporated on a SiO_2/Si substrate and was annealed in Ar+H_2 atmosphere, at 900 to 1000^0C, for 10 to 20 minutes. This annealing step produces Ni grains of 5 to 20 µm in size. After CVD at 900 to 1000^0C for 5 to 10 minutes, using 5 to 25 Seem CH_4 and 1500 Seem H_2 graphene was found to form on the Ni the size of each graphene being restricted by the Ni grain size. Graphene synthesized by this technique was suitable for various electronic applications. Also graphene synthesized and transferred into a glass substrate has shown as high as 90% optical transmittance and electrical properties also. By similar way Kim et.al. have shown growth of graphene on Ni-film (300 nm thick), which is deposited on SiO_2/Si substrate [13] was claimed by the author that the thickness of the Ni film was optimized for best quality of graphene. Graphene synthesis was performed in a CVD chamber using a gas mixture of CH_4, H_2 and Ar (50:65:200 seem), at a temperature of 1000^0C, followed by a rapid cooling (10^0C S^{-1}) in Ar envelope. It was found that the high cooling rate was important to minimize the number of layers onto other substrates. This graphene was later successfully transferred into flexible, transparent substrate, made up of polydimethyl siloxane (PDMS) without affecting its properties and showing 80% transparency in visible spectrum. Later on, it was observed by the authors that application of an aqueous $FeCl_3$ solution etches the nickel layers in a mild p^H value without causing formation of gaseous products which may damage the graphene sheets formed [13].

Another a very recent development in this area is a highly crystalline few-layer graphene was grown directly on 1 cm^2 area of polycrystalline Ni-substrates, by carefully controlling the gas ratio, growth time and temperatue [14]. Synthesis of Graphene by this way is confirmed by SEM images also. In further improvement in this synthesis process graphene was found to be synthesized on a 1 cm^2 area of Cu foil by thermal CVD process [15]. Graphene prepared by this route is of high quality. Due to limited solubility of C in Cu it was found that growth on Cu substrate was self-limiting. This process was claimed to be a surface-catalyzed process than a precipitation process. Hence, further development in graphene growth by thermal CVD has confirmed reproducibility of good quality graphene and successful transfer to many other substrates.

2.2.1 Other Used Techniques:

- **By Using CNT's**

  Synthesis of graphene by using multiwall carbon nanotubes (MWCNT) as an initial material is recently in practice. This process is well known as unzipping of CNT's. In the initial study it was reported that MWNT's can be opened up longitudinally by using intercalation of Li and ammonia, followed by exfoliation in acid and supplying small amount of heat. In another experiment graphene nanoribbons were prepared by plasma etching of MWNT's partially embedded in polymer film [16]. In this method MWNT's opened up to form graphene. By another approach MWNT's were unzipped by a multistep chemical treatment, including exfoliation by concentrated H_2SO_4, $KMnO_4$ and H_2O_2, stepwise

oxidation using $KMnO_4$ and finally reduction in NH_4OH and hydrazine monohydrate ($N_2H_4.H_2O$) solution [17].

- **Thermal Decomposition on other Substrates**
  Monolayer graphene was synthesized on a single crystal Ru (0001) surface at ultra-high vacuum condition (4×10^{-11}Torr) [18]. Before the synthesis process, Ru crystal was cleaned by repeated cycles of Ar^+ sputtering and exposure to oxygen and heating to high temperature. Graphene was found to be formed on the crystal surface, either by thermal decomposition of ethylene at 1000k or by controlled segregation of carbon from bulk of the substrate. The single layered graphene was of high purity. Hence, graphene was synthesized on many other single crystal-transition metal surface, including Ir, Ni, Co, Pt, etc.
- **Plasma Enhanced Chemical Vapor Decomposition Technique**
  Synthesis of graphene through plasma enhanced chemical vapor decomposition (PECVD) is contemporary to that of exfoliation. The earlier report, by Obraztsov et. al., has proposed a dc discharge PECVD technique to prepare nano structured graphite like Carbon [19]. In this technique Si water and Ni, W, Mo, and some other materials were used as substrates and a gas mixture of CH_4 and H_2 with a total gas pressure of 10 to 150 Torr. The NG film prepared by this process looks thicker at most of the places, except at some twisted portions.

In the year 2004 the first report on single to few layer graphene by PEVCD was found [20, 21]. Fir this, a radio frequency PECVD system was used to prepare graphene on a variety of substrates, without any special surface preparation operation. The graphene sheets prepared in a gas mixture of 5-100% CH_4 in H_2, at 900W power and 680^0C substrate temperature was found to have subnanometer thickness and erected from the substrate surface. As per Zhu et. al., a growth mechanism for the graphene was processed in PECVD chamber. According to their proposal a thin graphene sheets were synthesized by a balance between deposition through surface diffusion of C-bearing growth species from precursor gas and etching caused by atomic hydrogen. The verticality of the graphene sheets obtained by this route is caused by the plasma electric field direction. In another experiment by doing some changes in the process, Shang et. al., have synthesized multilayer graphene nano-flake films (MGNF) on Si substrates, through microwave PECVsD [22]. Also, graphene synthesized by this method had highly graphitized knife-edge structure, having 2-3 nm thickness at the sharp edges. Here, graphene sheets obtained were roughly vertical to the substrate (Si) and reported to show very nice bio-sensing capability. Also, it was reported a very high growth rate of graphene by this process. On the similar manner, Tuan et.al., have prepared a high quality graphene sheets having 1 to 3 layer thick on stainless steel substrate at 500^0C by using microwave PECVD [23]. In this process a gas mixture of CH_4 and H_2 and a microwave of 1200W were used. Graphene prepared by this method were having better crystalinity than other methods. It was proved by SEM and HRTEM images by using PECVD method. Hence, PECVD technique has shown good results in synthesis of graphene on any substrate.

2.3 Chemical Method:

There are some many methods are available for the synthesis of graphene rather than exfoliation and CVD process out of these techniques, chemical based technique is one of them. Here a part which is already covered in thermal chemical vapour decomposition as liquid phase exfoliation. In chemical methods graphene films are extracted chemically from graphite without exfoliation stage. Horiuchi et. al. synthesized graphene very first time by this technique, when they produced carbon nanofilms (CNF) from natural graphite [29]. Natural graphite was subjected to a series of oxidation and purification process, followed by dilution in methanol and several centrifugation stages to extract the thinnest sheets from the dispersion. Tn another chemical method, H_2SO_4 and HNO_3 were intercalated between the layers of graphite, followed by rapid heating to 1000^0C, so that explosive evaporation of the acid molecule could produce thin graphite sheets [23]. In the next step, intercalation the chemicals like Oleum and tetrabutylammonium hydroxide (TBA) were used to produce single and few layer graphene, after sonication in a surfactant solution. A best quality graphene was obtained by this technique. On the other hand, Choucairet. al., have reacted sodium and ethanol (1:1) to form a graphene precursor, it was then rapidly pyrolyzed [31]. Here, the production yield of 0.1 gm graphene per 1 mol of ethanol was obtained. The obtained product was in the form of suspended solid form. By different way chemical exfoliation technique was used to synthesis graphene. Here, graphite oxide was thermally expanded by rapidly heating it near about at 1050^0C, followed by a two stage reduction using hydrogen gas and N-methyl pyriolidone. In this study it was claimed that the number of layers in the graphene is dictated by the lateral size and the crystalinity of the input graphite.

3. Properties of Graphene:

Detailed discussion of properties of graphene is out of the scope of this work, which may be available in other review article. By this article the emphasis was given to introduce the basic properties of graphene.

3.1 Single and Bi-layer Graphene:

To understand the properties of graphene, it is necessary to know and understand the structure of graphene. In a single-layer graphene, a single two dimensional hexagonal sheet of carbon atoms is observed

[24]. On the other hand Bi-layer graphene has two sheets and few layer graphene has 3-10 layers of two dimensional sheets are observed. If, in a graphene structure more than 10 layers are present then these layers are considered as a thick graphene sheet and are scintifically less interested. Here, in case of Bi-layer and few-layer graphene, carbon atoms can be stacked in different ways, generating hexagonal or AA or AB and rhombohedral or ABC stacking. Basically graphene has Sp^2 hybridization. It shows a three in plane σ bonds and π orbitals perpendicular to the plane. While the strong sigma bonds work as the rigid backbone of the hexagonal structure, the out of plane π bonds control interaction between different graphene layers. To identify the thickness of graphene, one of the used tools is optical microscopy; it can be done through it on Si substrate with a 285 nm SiO_2capping layer, by using contrast spectra. However, the best way to understand the structure of graphene is by TEM. But, due to some drawbacks this tool may not be suitable always. Another suitable tool is Ramon spectra. Here, images obtained from different portions of graphene can give an idea about the thickness of the graphene sheet. One can find out the number of layers in the graphene film from the intensity, shape and position of the G and 2D bonds. Also, with increasing number of layers, 2D bond changes its shape and width.

In electronic structure single layer graphene shows bond overlap in two conical points in the Brillouin zone temperature- ambipolar characteristics, i.e, charge carriers can be alternated between holes and electrons depending upon the nature of the voltage. Anomalous Quantum Hall Effect (QHE) at low temperature and room temperature has also reported for single layer graphene structure.

Due to these unusual properties of single layer graphene it has made suitable for applications in electronics and quantum physics Bi-layer graphene shows a gapless state with parabolic bonds in contrast to conical bands of single layer graphene. Hence, it is considered as gapless semiconductor. However, Zhou et. al., has proved that an energy band gap of ~ 0.26eV is produced in grapheme, when it is epitaxically grown on SiC substrate [26]. Graphene synthesized by this type is more applicable in electronic industry. Hence, synthesis methods play important role in determining the structure and properties of graphene.

3.2 Few-Layer Graphene:

As compare to single layer graphene few –layer graphene shows very high surface area, the structure becomes increasingly metallic with more number of layers in it [27] also this few-layer graphene shows no gap as well as it shows good gas adsorption property. Further, it shows good capability to be functionalized by different covalent and non-covalent modifications, in order to solubilize them in various solvents. By reacting with concentrated sulphuric acid and Nitric acid this graphene becomes water soluble. Hence, all such kind of chemical behavior of the material through covalent modification was found to affect the electronic structure and also the properties of graphene. Hence, for further reactions, somewhat modification in the final product is essential, and it can be done effectively by wrapping it with polyethylene glycol, by this process material becomes water soluble and also without modifying its electronic structure. Rather than functionalization, decoration with metallic nanoparticles can become necessary to make the structure more applicable in optics, electronics and biotechnology area. Also, magnetic properties of graphene can be changed by chemical modifications.

4. Applications:

All varieties of graphene, it may be of single, Bi- and few-layer, have fond several applications in field of electronics, energy storage devices, sensors, biotechnology, memory etc. Basically, all the applications are based on synthesis methods as well as structure and properties of the product formed. Applications of graphene in energy storage devices, such as batteries and super capacitors, are also observed. Few layer graphene have been used effectively as part of composite electrodes in Li-ion batteries [25, 28]. Further, graphene is used in field-emission, field effect transistors, transparent electrodes, Structural composites, Catalyst Supports, Polymer master batches, Functional links etc.

5. Conclusion:

If we observe the single layer graphene, at room temperature it shows ambipolar-characteristics, while in case of few-layer it shows no energy gap. The structure becomes increasingly metallic due to more layers in it. It shows a high surface area and good gas adsorption property. Synthesis of graphene by thermal chemical vapor deposition onto Ni thin film proved reproducibility of good quality graphene on a centimeter scale substrate also successful transfer to many other substrates including Si, glass and PDMS. Although graphene has showed exceptional electoral, optoelectric and chemical properties, due to these properties it is useful in many areas. Hence, it will work as future electric device with high speed, chemical stability and better functionality along with environmental friendliness.

6. References:

1. Novoselov, K. S., Geim, A. K., Morozov, S. V., Jiang, D., Zhang, Y., Dubonos, S. V., Grigorieva, I. V. and Firsov, A. A. (2004). Electric field effect in atomically thin carbon films. Science, 306: 666
2. Lamg, B. A LEED study of the deposition of carbon on platinum crystal surfaces. Surface Sci., 53: 317(1975).
3. Viculis, L. M., Mack, J. J. and Kaner, R. B. A chemical route to carbon nanoscrolls. Science, 299: 1361 (2003).

4. Novoselov, K. S., Jiang, D., Schedin, F., Booth, T. J., Khotkevich, V. V., Morozov, S. V. and Geim, A. K. Two-dimensional atomic crystals. PNAS, 102: 10451 (2005).
5. Datta, S. S., Strachan, D. R., Khamis, S. M. and Johnson, A. T. C. Crystallographic etching of few-layer grapheme. Nano Lett., 8: 1912 (2008).
6. Bangert, U., Bleloch, A. L., Wang, P., Nair, R. R., Geim, A. K. and Gass, M. H. Free-standing grapheme at atomic resolution. Nature Nanotechnol., 3: 676 (2008).
7. Shukla, A., Kumar, R., Mazher, J. and Balan, A. Graphene made easy: high quality large-area samples. Solid State Commun., 149: 718 (2009).
8. Stankovich, S., Dikin, D. A., Piner, R. D., Kohlhaas, K. A., Lleinhammes, A., Jia, Y.Wu, Y. Synthesis of grapheme-based nanosheets via chemical reduction of exfoliated graphite oxide. Carbon, 45: 1558 (2007).
9. Somani, P. R., Somani, S. P. and Umeno, M. Planer nano-graphenes from camphor by CVD. Chemical Physics Letters, 430: 56 (2006).
10. Yu, Q., Lian, J., Siriponglert, S., Li, H., Chen, Y. P. and Pei, S. S. Graphene segregated on Ni surfaces and transferred to insulators. Appl. Phys. Lett., 93: 113103 (2008).
11. Wang, X., You, H., Liu, F., Li, M., Wan, L., Li, S.Li, Q. Large-scale synthesis of few-layered graphene using CVD. Chem. Vapor Deposition, 15: 53 (2009).
12. Reina, A., Jia, X., Ho, J., Nezich, D., Son, H., Bulovic, V., Dresselhaus, M. S. and Kong, J. 2009. Large area few-layer graphene films on arbitrary substrates by chemical vapor deposition. Nano Lett., 9: 30 (2009).
13. Kim, K. S., Zhao, Y., Jang, H., Lee, S. Y., Kim, J. M., Kim, K. S.Ahn, J. H. Large-scale pattern growth of graphene films for stretchable transparent electrodes. Nature, 457: 706 (2009).
14. Chae, S. J., Günes, F., Kim, K. K., Kim, E. S., Han, G. H., Kim, S. M.Shin, H.-J. Synthesis of large-area graphene layers on poly-nickel substrate by chemical vapor dposition:wrinkle formation. Adv. Mater., 21: 2328 (2009).
15. Li, X., Cai, W., An, J., Kim, S., Nah, J., Yang, D.Piner, R. Large-area synthesis of high-quality and uniform graphene films on copper foils. Science, 324: 1312 (2009).
16. Jiao, L., Zhang, L., Wang, X., Diankov, G. and Dai, H. Narrow graphene nanoribbons from carbon nanotubes. Nature, 458: 877 (2009).
17. Kosynkin, D. V., Higginbotham, A. L., Sinitskii, A., Lomeda, J. R., Dimiev, A., Price, B. K. and Tour, J. M. Longitudinal unzipping of carbon nanotubes to form graphene nanoribbons. Nature, 458: 872 (2009).
18. Vázquez de parga, A. L., Calleja, F., Borca, B., Passeggi, M. C. G. Jr., Hinarejos, J. J., Guinea, F. and Miranda, R. Periodically rippled graphene: growth and spatially resolved electronic structure. Phys. Rev. Lett., 100: 056807(2008).
19. Obraztsov, A. N., Zolotukhin, A. A., Ustinov, A. O., Volkov, A. P., Svirko, Y. and Jefimovs, K. DC discharge plasma studies for nanostructured carbon CVD. Diamond Related Mater., 12: 917 (2003).
20. Wang, J. J., Zhu, M. Y., Outlaw, R. A., Zhao, X., Manos, D. M. and Holoway, B. C. Synthesis of carbon nanosheets by inductively coupled radio-frequency plasma enhanced chemical vapor deposition. Carbon, 42: 2867 (2004).
21. Wang, J. J., Zhu, M. Y., Outlaw, R. A., Zhao, X., Manos, D. M. and Holoway, B. C. Free-standing subnanometer graphite sheets. Appl. Phys. Lett., 85: 1265 (2004).
22. Malesevic, A., Kemps, R., Zhang, L., Erni, R., van Tendeloo, G., Vanhulsel, A. and van Haesendonck, C. A versatile plasma tool for the synthesis of carbon nanotubes and few-layer graphene sheets. J. Optoelect. Adv. Mater., 10: 2052 (2008).
23. Yuan, G. D., Zhang, W. J., Yang, Y., Tang, Y. B., Li, Y. Q., Wang, J. X., Meng, X. M.He, Z. B. Graphene sheets via microwave chemical vapor deposition. Chem. Phys. Lett., 467: 361 (2009).
24. Hass, J., de Heer, W. A. and Conrad, E. H. The growth and morphology of epitaxial multilayer graphene. J. Phys. Cond. Matter, 20: 323202 (2008).
25. Ni, Z. H., Wang, H. M., Kasim, J., Fan, H. M., Yu, T., Wu, Y. H., Feng, Y. P. and Chen, Z. X. Graphene thickness determination using reflection and contrast spectroscopy. Nano Lett., 7: 2758 (2007).
26. Zhou, S. Y., Gweon, G.-H., Fedorov, A. V., First, P. N., de Heer, W. A., Lee, D. -H.Guinea, F. Substrate-induced bandgap opening in epitaxial graphene. Nature Materials, 6: 770 (2007).
27. Morozov, S. V., Novoselov, K. S., Schedin, F., Jiang, D., Firsov, A. A. and Geim, A. K. Two-dimensional electron and hole gases at the surface of graphite. Phys. Rev. B, 72: 201401 (2005).
28. Subrahmanyam, K. S., Vivekchand, S. R. C., Govindaraj, A. and Rao, C. N. R. A study of graphenes prepared by different methods: characterization, properties and solubilization. J. Materials Chem., 18: 1517 (2008).

29. Horiuchi, S., Gotou, T., Fujiwara, M., Asaka, T., Yokosawa, T. and Matsui, Y. Single graphene sheet detected in a carbon nanofilm. Appl. Phys. Lett., 84: 2403 (2004).
30. Yuan, G. D., Zhang, W. J., Yang, Y., Tang, Y. B., Li, Y. Q., Wang, J. X., Meng, X. M.He, Z. B. Graphene sheets via microwave chemical vapor deposition. Chem. Phys. Lett., 467: 361 (2009).
31. Choucair, M., Thordarson, P. and Stride, J. A. Gram-scale production of graphene based on solvothermal synthesis and sonication. Nature Nanotechnol., 4: 30 (2009).