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A Review on the Preparation Technology of Graphene Thin Films

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Abstract: Graphene films have excellent properties and have great application potential in many fields. In this paper, the principle, operation process and characteristics of mechanical stripping method, chemical vapor deposition method, REDOX method and other mainstream preparation processes are described in detail from the traditional preparation process and the new preparation process. The film quality, advantages and limitations of each method are analyzed and compared. At the same time, the challenges faced by different preparation processes, such as the introduction of impurities, structural defects and their effects on the properties of graphene films, were discussed. In addition, the innovative development trend of preparation processes in recent years is also paid attention to. The aim is to provide theoretical reference and technical reference for the optimization and improvement of the subsequent graphene film preparation process, performance improvement and wider practical application through the systematic review of the existing research results.

Keywords: graphene film, Preparation technology, Mechanical stripping method, Chemical vapor deposition, REDOX method

1. Introduction

Since 2004, Andre Geim and Konstantin Novoselov of the University of Manchester in the United Kingdom have successfully stripped graphene from highly oriented pyrolytic graphite for the first time. Graphene films, as an important form of graphene materials, have many excellent properties, which make it show great application potential in many fields.

In terms of electrical properties, graphene films have extremely high carrier mobility, and their electron mobility can reach 100 times that of silicon at room temperature. In terms of mechanical properties, graphene films are among the strongest substances known, up to 100 times stronger than the best steel, and have excellent flexibility. In addition, graphene films also have high light transmittance, good chemical stability, excellent electrochemical properties, etc.

Based on the above excellent properties, graphene films have shown a wide range of application prospects in electronic devices, energy storage, composite materials and other fields. However, despite such attractive application prospects, there are still many challenges in the large-scale commercial application of graphene thin films, among which the preparation process is one of the key factors. At present, there are still some problems in the preparation process of graphene films, such as high preparation cost, low yield, difficult to large-scale production, and difficult to guarantee the consistency of film quality and performance. These problems have seriously restricted the industrial production and wide application of graphene films. Therefore, the research of

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graphene film preparation technology is of vital significance. In-depth research and optimization of the preparation process of graphene film can not only improve the quality and performance of graphene film, but also reduce the preparation cost and improve the production efficiency, so as to promote the wide application of graphene film in various fields. In this paper, by systematically combing and studying the current main methods of graphene film preparation, the principle, advantages and disadvantages of each preparation method and its impact on the performance of graphene film will be deeply discussed, which can provide theoretical guidance and technical reference for the further research and application of graphene materials, and provide technical support for the industrial production of graphene film.

2. Traditional preparation process

2.1. Mechanical stripping method

2.1.1. Process operation

Mechanical stripping is one of the earliest methods used to prepare graphene films, and the operation process is relatively simple, but extremely challenging. The method uses highly oriented pyrolytic graphite as raw material, and uses the adhesive force and mechanical force of the tape to achieve the stripping of the graphene film.

2.1.2. Application case

In their study, Farashiani et al. used an improved mechanical stripping method, namely shear stripping of flake graphite, to produce fewer layers of graphene, and investigated in depth its effects on the conductivity and properties of high-density polyethylene [1]. They first selected suitable flake graphite, which has a large lamellar size and a relatively regular crystal structure, as the raw material. Then, they treated the flake graphite with a specially designed shearing device. In the shearing process, by precisely controlling the size, direction and action time of the shear force, the graphite sheets are gradually separated under the action of the shear force, so as to obtain fewer layers of graphene. Compared with the traditional tape stripping method, this shear stripping method can control the number of layers and size distribution of graphene more effectively.

2.1.3. Analysis of advantages and disadvantages

The mechanical stripping method has some significant advantages in preparing graphene films. This method enables the preparation of high-quality graphene films with an intact crystal structure and extremely low defect density. In terms of electrical properties, its carrier mobility is extremely high, which can meet some applications that require demanding electron transport performance, such as high-speed electronic devices. However, the mechanical stripping method also has obvious disadvantages, which seriously limits its large-scale application. Due to its complete reliance on manual operation and random stripping of tape, the amount of graphene film that can be obtained at a time is very small, which is difficult to meet the demand for output in industrial production.

2.2. Chemical vapor deposition (CVD)

2.2.1. Process steps

Conventional chemical vapor deposition (CVD) produces graphene from a suitable carbon source, such as methane, by a chemical reaction on a substrate under high temperatures (usually over 1000°C) and a high vacuum. The metal copper is often used as the substrate, and the surface of the

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copper is catalyzed to form "active" carbon atoms that migrate on its surface and eventually splice together to form a graphene film [2].

2.2.2. Problems exist

Although chemical vapor deposition (CVD) has significant advantages in the preparation of graphene films, there are also some problems that need to be solved. Side reactions are prone to occur during the preparation process, resulting in "intrinsic contamination" of the graphene surface [3]. The variety and content of gaseous species in the viscous layer will cause the formation of by-products, which will form amorphous carbon pollutants on the graphene surface.

2.3. Chemical REDOX method

2.3.1. Preparation process

Chemical REDOX method is an important method for the preparation of graphene film. Its preparation process mainly includes three key steps: the oxidation of graphite, the stripping of graphite oxide and the reduction of graphene oxide.

2.3.2. Effect on properties of the film

Although the chemical REDOX method has the advantages of low cost and suitable for large-scale production when preparing graphene films, it also has some adverse effects on the properties of graphene films. Because the graphene films produced by this method usually have structural defects, their carrier mobility is poor [4].

3. The new preparation process

3.1. Plasma enhancement method

3.1.1. Principle of plasma enhanced chemical vapor deposition (PECVD)

In the process of plasma enhanced chemical vapor deposition (PECVD), the reaction gas is first ionized by glow discharge to form plasma. The plasma contains a large number of high-energy electrons, which can provide the activation energy needed in the process of chemical vapor deposition, thus changing the energy supply mode of the reaction system. The collision of high-energy electrons with gas molecules can promote the chemical bonds of the reaction gas molecules to break and recombine, forming more active free radicals. These free radicals are more active than ground-state atoms or molecules and can form graphene and related products on catalyzed or uncatalyzed substrates at low temperatures. The various primary and secondary reaction products that reach the growth surface are adsorbed and react with the surface, accompanied by the re-release of gas-phase molecules. In this process, high-energy ions and free radicals in the plasma react chemically with atoms or molecules on the substrate surface to form a graphene film.

3.1.2. Application cases

Yuan Qianghua et al. used RF-PECVD equipment and found that there were many boundary defects in graphene films under the conditions of 3 Torr and 5 Torr [5]. When the methane/argon flow ratio was 10:30 and the growth time was 40 min, the graphene films with good quality were prepared. Xian Zhang et al. used plasma enhanced chemical vapor deposition equipment to prepare graphene film on the surface of Ti [6]. The results found that the prepared graphene film can

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effectively improve the corrosion resistance of medical titanium. In the biomedical environment, medical titanium may be eroded by corrosive media such as body fluids, and the existence of graphene film can significantly reduce the degree of such erosion, and improve the service life and safety of medical titanium.

3.1.3. Application effects and advantages

PECVD technology has the advantage of low temperature deposition. Compared to traditional CVD technology, PECVD is able to achieve high quality thin film deposition at lower temperatures. This has important implications for some materials or substrates that are sensitive to high temperatures. When preparing flexible electronic devices, the flexible substrates used (such as polyethylene terephthalate (PET), etc.) usually cannot withstand high temperature treatment, and the low temperature deposition characteristics of PECVD can avoid damage to the flexible substrate, making it widely used in the field of flexible electronic devices.

3.2. Electrochemical preparation method

3.2.1. Principles of electrochemical oxidation and reduction

Compared with the traditional chemical REDOX method, the electrochemical preparation method does not need to use strong oxidants and reducing agents, and its principle mainly involves electrochemical oxidation and reduction reactions. In the process of electrochemical preparation of graphene films, it is first necessary to carry out electrochemical oxidation. This step usually involves using the graphite as the anode and applying a voltage that moves the anions in the electrolyte towards the anode and inserts them into the layers of the graphite to form a graphite intercalation compound (GIC). At the same time, the water part of the anode breaks down to form oxygen, which also enters the graphite layers. Together, the graphite expands dramatically in volume and eventually falls off the surface to form graphene oxide (GO). After the electrochemical oxidation, an electrochemical reduction is required to reduce the GO to graphene. This step is usually done by applying a negative voltage to the electrode surface so that the graphene oxide accepts electrons, thereby reducing it to graphene.

3.2.2. Preparation case

Wenyou Li et al. studied a method for rapid preparation of graphene oxide (GO) films. Using the N-type semiconductor transparent film glass of FTO as substrate, a certain thickness and partially reduced GO film were prepared on its surface by the combined method of anode electrophoresis and cathode electrochemical reduction [7]. And by adjusting the oxygen content of GO film to control its energy level structure. By improving the Hummers method, 10 mg of graphite oxide was dispersed in 20 mL of deionized water, and 10 mL of mixed liquid containing 0.02 mmol (Co to Ni ratio 2:1) was ultrasounded for 1 h [8]. The precursor electrode and pure graphite oxide electrode were prepared by drip coating ITO glass and drying. Using the former as the research electrode, the graphene/nickel-cobalt oxide electrode was obtained by calcination of -1.16V reduced graphite and 0.68V cobalt oxide ion in 0.5 mol/L KOH solution under 250 ℃ nitrogen for 2 h with ruthenium-titanium mesh and saturated calomel electrode.

3.2.3. Comparison with traditional REDOX method

Electrochemical preparation method has the characteristics of environmental friendliness. Because it does not use strong oxidants and reducing agents, it reduces the generation of waste and reduces the pollution to the environment. The strong oxidant and reducing agent used in the traditional

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REDOX method will produce waste containing heavy metal ions, acid ions and so on after the reaction, these wastes need special treatment, otherwise it will pollute the soil, water and so on. The waste generated by the electrochemical preparation method is mainly a small amount of electrolyte, which is relatively easy to deal with.

3.3. Metal-assisted non-transfer graphene film preparation technology

The method of metal film catalytic synthesis of non-transferable graphene film is derived from the traditional graphene film preparation process. The microscopic catalytic transformation mechanism of this method has many similarities with the traditional transition metal catalytic mechanism, but it also has its own particularity due to its variable configuration and composition [9]. This technology directly prepares high-quality and large-area graphene films on the dielectric substrate with metal catalyst, avoiding the complicated and time-consuming post-treatment transfer process in the chemical vapor deposition process of traditional transition metal catalytic synthesis of graphene films, and reducing the damage to the intrinsic properties of graphene films.

3.4. Plasma etching assisted chemical vapor deposition

3.4.1. Process principle and flow

The preparation process can prepare high-quality graphene films at a lower temperature. The decomposed active carbon atoms are partially condensed into graphene on the upper surface of the substrate, partially dissolved into the substrate, and diffused through the film into the interface between the substrate and the substrate. The diffusion of carbon atoms into the interface begins to nucleate and form graphene at the interface. This is followed by plasma cleaning, in which a hydrogen-argon plasma removes the graphene growing on top of the substrate to promote carbon diffusion to the basic-substrate interface. By repeating these steps, a super-flat single crystal graphene is produced at the interface. The resulting sample is removed from the reaction chamber and subjected to special treatment to deform the substrate, which can be easily removed with tweezers, leaving flat graphene on the substrate.

3.4.2. Preparation effect and application prospect

Xinyao Chen et al. designed a new insulating substrate growth system based on the traditional hot wall chemical vapor deposition system [10]. The microwave plasma etching and auxiliary CVD (MPEE-CVD) system is formed by adding the microwave plasma unit to make it have additional etching and auxiliary functions. With the help of this system, graphene films with controllable size can be directly prepared on silicon oxide and other insulating substrates. The crystal quality was characterized by Raman spectroscopy and scanning electron microscopy. This work opens up a new way to further improve the performance of graphene-based field effect transistor nanodevices.

The high quality graphene film prepared by plasma etching assisted chemical vapor deposition can effectively improve the interface contact between graphene and substrate, reduce the impurities between the interface, reduce the resistance and noise of the device, and improve the electronic mobility and switching performance of the device.

4. The effect of the preparation process on the performance of graphene film

Different preparation processes have significant effects on the electrical conductivity of graphene films. The conductivity of high-quality graphene films prepared by mechanical stripping method can reach more than 106 S/m. This is because the mechanical stripping method has little damage to the atomic structure of graphene during the preparation process, the arrangement of carbon atoms

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maintains a high degree of regularity, and electrons can be freely transmitted in it, thus making the conductivity very high.

The conductivity of high-quality graphene films prepared by CVD method is generally between 10-4105 S/m. In the preparation process, the purity of the reaction gas, the surface state of the substrate and other factors will affect the production of defects, which in turn affect the conductivity. If the reaction gas contains impurities, these may be incorporated during the graphene growth process, reducing the conductivity. However, by optimizing the preparation process parameters, such as precisely controlling the reaction temperature, gas flow, etc., the generation of defects can be reduced and the conductivity of graphene films can be improved.

Graphene films prepared by REDOX method, due to the introduction of a large number of defects and oxygen-containing functional groups in the oxidation and reduction process, seriously destroy the conjugated structure of graphene, resulting in a significant reduction in electrical conductivity. In the oxidation process, the strong oxidizer introduces oxygen atoms between the graphite layers to form various oxygen-containing functional groups, which will change the electron cloud distribution of graphene and increase electron scattering, thereby reducing the conductivity. In the reduction process, although part of the oxygen-containing functional groups can be removed, some defects will remain, further affecting the conductivity

5. Conclusion and prospect

The research of graphene film preparation technology will be deeply explored in several key directions to overcome the current challenges. In terms of optimization and innovation of the preparation process, on the one hand, in-depth study of the microscopic mechanism of the existing process, such as the migration and growth process of carbon atoms on the substrate surface in chemical vapor deposition method, and the interaction mechanism between plasma, reaction gas and substrate in plasma enhancement method, will provide a more solid theoretical basis for the accurate regulation of process parameters. By accurately predicting the growth quality and performance of graphene films under different process parameters, the intelligent optimization of the process can be achieved. On the other hand, it is necessary to explore new preparation processes, such as the preparation technology based on molecular self-assembly, using the specific interaction between molecules to achieve the ordered growth of graphene under mild conditions, breaking through the limitations of existing preparation processes.

From the perspective of application field expansion, with the rapid development of communication technologies such as 5G, the application of graphene films in high-frequency communication devices will become a research hotspot. The preparation process of graphene film with low loss and high carrier mobility is studied, which is used to manufacture high-performance antennas and communication lines to meet the needs of communication technology for high-speed, high-frequency and low power consumption. In the field of energy storage and conversion, the development of graphene based electrode material preparation technology suitable for high-efficiency solar cells and fuel cells will provide new materials and technical support for solving energy problems. In the field of high-end equipment such as aerospace and automobile manufacturing, research will be carried out on the preparation process of graphene-reinforced composite materials with high strength, high modulus and lightweight.

References

[1] *2024-10-28, DOI: 10.1002/pen.26979Morteza Farashiani, Reza Shidpour, Mohammad Rajabi*

[2] *Hou Yuting. Structural regulation of graphene thin films prepared by copper substrate chemical vapor deposition [D]. University of electronic science and technology, 2024. DOI: 10.27005 /, dc nki. Gdzku. 2024.000336.*

DOI: 10.54254/2755-2721/2025.23755

[3] *Liu Xiao-Ting, Zhang Jin-Can, Chen Heng, et al. Preparation of ultra-clean graphene films [J]. Acta Physico-Chimica Sinica,2022,38(01):58-72.*

[4] *Chen X N. Study on the preparation of highly conductive graphene based on the destruction of janus-GO composite structure by chemical reduction [D]. Wuhan university of technology, 2021. DOI: 10.27381 /, dc nki. Gwlgu. 2021.001328.*

[5] *Yuan Qianghua, Ren Jiangfeng, Yin Guiqin. Effect of growth parameters of RF-PECVD on the quality of graphene films [J/OL]. Journal of vacuum science and technology, 1-7 [2024-12-06]. HTTP: / / https://doi.org/10.13922/j.cnki.cjvst.202403020.*

[6] *Zhang Xian-Ming, CAI Dingsen, Qian Shi. Medical titanium surface graphene films of PECVD was prepared and its performance technique [J]. Journal of surface technology, does 2024 (8) : 156-162 + 190. DOI: 10.16490 / j.carol carroll nki. Issn 1001-3660.2024.08.014.*

[7] *Li Wenyou, He Yunqiu, Li Yiming. Acta Phys. - Chim. Sin., 2015, 31 (3), 457-466. The doi: 10.3866 / PKU WHXB201501093.*

[8] *Wang Hong-Zhi, Jiang She-Sheng, Shi Yu-Lei, ZHANG Wei-Guo, Yao Su-Wei. Preparation of graphene/Nickel-cobalt oxide composite thin film electrode by electrochemical method and its properties. Chemical Industry & Engineering, 2018, 35(1): 63-68.*

[9] *Huang Yong, Guo Chongxiao, Ni Jiameao, et al. Research progress in preparation of metal-catalyzed non-transferable graphene films [J]. Materials Review,2024,38(15):188-202.*

[10] *Chen Xinyao, Tian Bo, Peng Dongqing, et al. Graphene growth on insulating substrate assisted by plasma etching [J]. New Chemical Materials, 2020, 48(5): 100-103.*