

Graphene/Silicon Heterojunctions: Preparation, Performance Optimization, and Applications

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ABSTRACT

The unique photoelectric properties of graphene/ Silicon (Gr/Si) heterojunction make this structure a very interesting candidate to promote optoelectronic technology. This paper reviews the latest progress in the synthesis, optimization and application of Gr/Si heterojunction in three main fields: photovoltaic conversion, optical sensing and electrochemical energy storage. For synthesis, chemical vapor deposition (CVD) is still the best method to obtain uniform and high-quality graphene on large surfaces, and its combination with wet or dry transfer protocols allows the construction of reliable heterojunctions. The performance of the equipment can be improved by many means, such as chemical hybridization to adjust the carrier density, modify the interface to reduce recombination, photon management structure to increase absorption, and accurate control of graphene thickness. In order to capture solar energy, the efficiency of Schottky Gr/Si battery has increased from the initial 1.5% to more than 15% now. The photoelectric sensors constructed on these isomers have broad spectral coverage, high responsiveness and fast time response. In addition, Gr/Si composites show strong electrochemical performance when configured as electrodes of supercapacitors or lithium ion batteries.

Keywords: Graphene/Silicon Heterojunction; Schottky Junction; Photovoltaic Devices; Photodetectors; Energy Storage; Interface Engineering; Chemical Doping.

1. INTRODUCTION

Rising worldwide energy consumption and increasingly urgent environmental constraints continue to motivate exploration into sustainable power generation and next-generation electronics. Among many studied material architectures, the heterojunctions formed between graphene and silicon (Gr/Si) are very interesting. This is due to their complementary physical qualities and their increasing practical uses. Graphene is a two-dimensional carbon isomorphism with atoms arranged in a hexagon. It has special characteristics: very high carrier mobility, almost complete transparency (single layer over 97%), flexibility and adjustable surface potential. All these characteristics make graphene an ideal partner for silicon. Silicon has been the main material in modern semiconductor manufacturing.

In 2010, Li and his co-authors demonstrated the first photovoltaic device based on Schottky Gr/Si configuration, and recorded the first power conversion efficiency (PCE) of only 1.5%. Later improvements, through doping, interface control and optical engineering, raised the benchmark to more than 15%.^[1] Under the contact of Gr/Si, Schottky barrier naturally grows, forming an internal electric field, which promotes the efficient dissociation of excitons and the extraction of carriers. Under illumination, the field guides photoexcited electrons into the silicon volume, while the upper layer of graphene collects corresponding holes. This load separation behavior is the core working principle shared by Gr/Si solar cells, photoelectric sensors and related energy equipment.

The research of Gr/Si heterogeneous solar cells has made great progress in recent years. Huang et al.^[2] comprehensively reviewed the progress of Gr/Si heterojunction photovoltaic devices; Iqbal et al.^[3] talked about the revolutionary potential of graphene silicon nanocomposites using solar energy; Li et al.^[4] studied van der Waals heterostructure based on graphene and dichalcogenide transition (TMD) for photovoltaic and photoelectrochemical energy conversion; Kumar et al.^[5] summarized the latest development of graphite-based solar cells.

The purpose of this review is to provide the latest development in Gr/Si heterogeneity research and logically organize a thorough review: from preparation methods to performance optimization, and then discuss photovoltaic, sensor and energy storage applications. By exploring these interrelated aspects, we try to provide useful insights for researchers working in this rapidly developing field.

2. PREPARATION METHODS FOR GRAPHENE/SILICON HETEROJUNCTIONS

The fabrication of high-quality Gr/Si heterojunctions constitutes a critical prerequisite for achieving optimal device performance. The preparation process typically comprises two principal stages: graphene synthesis and heterojunction assembly. Multiple approaches have been developed for each stage, with distinct advantages and limitations characterizing each method.

2.1 Graphene Synthesis Methods

2.1.1 Chemical Vapor Deposition (CVD)

Among the available manufacturing methods, chemical vapor deposition (CVD) is most commonly used to create large graphene sheets. They are of good quality for Gr/Si equipment. In a typical CVD process, a gas such as methane or ethylene is placed on a catalyst metal such as copper or nitrogen. This metal is very hot, between 800 and 1000 degrees Celsius. At this high temperature, carbon enters the metal. Then, when the system cools, the carbon aggregates to form graphene. The obtained films are very regular, their thickness can be selected, and they have almost no defects. The experimental study of Torres et al. ^[6] shows that when these thin layers made of CVD are transferred to the silicon platform, it will improve the filling factor and reduce the series resistance.

2.1.2 Mechanical Exfoliation

The earliest reported isolation of graphene, achieved by Geim and Novoselov in 2004, relied on micromechanical cleavage—often colloquially termed the adhesive-tape technique. In practice, a piece of highly oriented pyrolytic graphite (HOPG) is repeatedly peeled with tape until atomically thin carbon remnants remain. Crystallographically, this procedure produces graphene with exceptionally low defect densities and superior electronic properties; however, it is inherently limited by small sample dimensions—usually under 100 μm —and very low throughput. For these reasons, micromechanical cleavage remains largely confined to exploratory physics studies and is rarely adopted for industrial-scale production.

2.1.3 Liquid-Phase Exfoliation

Liquid-phase exfoliation (LPE) offers a solution-processable alternative in which bulk graphite is ultrasonically agitated within a selected liquid medium to weaken interlayer cohesive forces and release individual or few-layer flakes. Suitable dispersing media range from polar aprotic solvents such as N-methyl-2-pyrrolidone (NMP) and dimethylformamide (DMF) to surfactant-stabilized aqueous formulations. Attractive features of this route include straightforward scale-up, comparatively low cost, and compatibility with established coating technologies—including spin coating, spray deposition, and inkjet patterning. A trade-off exists, however: flakes obtained via LPE tend to carry higher defect concentrations and more restricted lateral sizes than their CVD counterparts.

2.2 Heterojunction Assembly Methods

2.2.1 Direct Transfer Method

The direct transfer method constitutes the most frequently employed approach for assembling Gr/Si heterojunctions. In this technology, CVD graphene is grown on a metal as a catalyst (usually copper or Ni) and first covered with a polymer to support it (usually polymethyl methacrylate, PMMA). Then, the following catalyst metal is dissolved with a special liquid (copper, FeCl_3 or Ni HCl). This allows graphene films to float on the liquid surface. Then, we moved graphene and PMMA to the silicon substrate. After that, PMMA was dissolved in acetone to obtain Gr/Si heterojunction. This method can well control the number and placement of graphene layers, but great care must be taken to avoid staining or folding.

2.2.2 Interface-Engineered Wet Transfer

The wet transfer of that interface engineering change order include changing the transfer proces to add a layer between graphene and silicon. Thin layers such as sodium hexagonal nitride (h-BN), alumina (Al_2O_3) or alumina (TiO_2) can be used to improve the quality of the joint. Meng and Al. ^[7] It shows that placing h-BN between graphene and silicon will increase PCE from 3.75% to 4.40%. This improvement is due to the reduction of interface recombination and the increase of Schottky barrier.

3. PERFORMANCE OPTIMIZATION OF GRAPHENE/SILICON HETEROJUNCTIONS

The performance of Gr/Si heterojunction equipment depends on several important parameters. These parameters are Schottky barrier height, series resistance, shunt resistance and interface quality. We have developed different optimization strategies to improve these parameters and improve the overall efficiency of the equipment.

3.1 Chemical Doping

Chemical addition is one of the best methods to modify the working function of graphene. This is helpful to optimize Schottky Barrier height on Gr/Si interface. Pure graphene has a work function of about 4.5 eV^[8]. This creates a fairly low Schottky barrier when it comes into contact with N-type silicon (its electron affinity is 4.05 eV). If we do P-type hybridization on graphene, we can increase its work function to 5.0 eV or higher. This provides greater built-in potential and better load separation.

3.1.1 Acid Doping

Combining with nitric acid (HNO₃), hydrochloric acid (HCl) or sulfuric acid (H₂SO₄) is a simple and effective method to hybridize P-type graphene. Acid treatment placed electron band drawing groups on the surface of graphene, which reduced Fermi level and increased working function. Feng et al.^[8] showed that the series resistance of Gr/Si solar cells was reduced from 6.11 to 4.07 Ω cm and the PCE was increased by 2.9% to 4.35% after being immersed in HNO₃ steam for several seconds. However, the stability of acid stimulants in the ambient air is a problem, because stimulant molecules will separate with time, which will lead to the decline of equipment performance.

3.1.2 Metal Chloride Doping

Compared with acid hybridization, hybridization with metal chloride, especially with gold chloride (AuCl₃), has superior stability. The doping mechanism involves the transfer of graphene charge to AuCl₃, the reduction of Au⁺ ions to Au nanoparticles, and the introduction of P-type stimulants to graphene at the same time. The output work of graphene can be accurately controlled by changing the concentration of AuCl₃. The reported efficiency of Gr/Si composites reaches 7.34%^[9], and the blending with Au and HNO₃ further improves the efficiency to 10.20%^[10].

3.2 Interface Engineering

Interface engineering plays a very important role in the performance of Gr/Si heterojunction devices. It controls how charges are transferred, recombined and moved to the interface.

3.2.1 h-BN Intercalation

Hexagonal carbon nitride (h-BN) has a layered structure, such as graphene, but it has a wide band gap (~6 eV) and excellent dielectric properties. When we put h-BN graphene between silicon, this increases the Schottky barrier height and reduces recombination to the interface. Meng et al.^[7] achieved a maximum efficiency of 10.93% in Gr/h-BN/Si solar cells. They combine the directly grown graphene/h-BN heterogeneous structure with the co-doping using gold nanoparticles and HNO₃. This record performance shows the synergistic effect of h-BN insertion and double chemical hybridization: the passive surface defect of h-BN layer is at the silicon interface, while the Au/HNO₃ co-hybridization strategy optimizes the working function of graphene and reduces the sheet resistance. In a word, this improves the voltage of the camera collection and open circuit.

3.2.2 TiO₂ Antireflection Layer

It is a broadband semiconductor. It has two functions: it enhances the interface properties, and it also acts as an anti-reflection coating. Shi et al.^[11] showed that the efficiency of Gr/Si solar cells was improved to 14.5% when a TiO₂ 2 anti-reflection layer was added through spin coating solution. The short-circuit current density is 32.7mA/cm, the filling factor is 72%, and the open-circuit voltage is 0.61V V.

3.2.3 MgF₂/ZnS Coatings

Ding et al.^[12] used a double-layer MgF₂/ZnS coating. These coatings not only have good anti-reflection performance, but also create color solar module for photovoltaic applications in buildings by changing the thickness of ZnS and MgF₂ layers. Multicolor Gr/Si solar devices reach a high PCE value, which is between 10.7% and 13.2%. PCE can be increased to 14.6% by optimizing MgF₂/ZnS antireflection coating.

4. PHOTOVOLTAIC APPLICATIONS OF GRAPHENE/SILICON HETEROJUNCTIONS

Gr/Si heterojunctions have been extensively investigated for photovoltaic applications, particularly as Schottky junction solar cells. The distinctive properties of graphene render it an attractive alternative to conventional metal electrodes and transparent conductive oxides.

4.1 Schottky Junction Solar Cells

Gr/Si Schottky junction solar cells represent the most widely studied photovoltaic devices based on this heterojunction system. The device structure consists of a graphene layer directly transferred onto moderately doped n-type or p-type silicon wafers, forming a Schottky barrier at the interface.

4.1.1 Device Operation Mechanism

Contact between graphene and n-type silicon establishes a thermodynamic equilibrium through electron transfer from the semiconductor into the carbon layer, which in turn creates a permanent internal field across the interface. The resulting Schottky barrier magnitude equals the offset between the graphene work function and the electron affinity of silicon. Upon light exposure, excitons generated within the silicon absorber are rapidly dissociated by this built-in field: minority holes migrate upward into the graphene sheet, while majority electrons are driven toward the rear silicon contact. In this dual capacity, graphene functions concurrently as an optically transparent current collector and as an active rectifying partner in the junction.

4.1.2 Performance Evolution

The first Gr/Si Schottky junction solar cell was reported by Li et al. in 2010 with a PCE of 1.5%^[1]. Through systematic optimization, efficiencies have improved substantially, with current values exceeding 15%.

4.2 Hybrid Electrode Solar Cells

Beyond serving as an active junction layer, graphene can be integrated with conventional transparent conductive oxides (TCOs) such as indium tin oxide (ITO) to form hybrid electrodes that combine the advantages of both materials. Torres et al.^[6] investigated hybrid transparent conductive electrodes composed of ITO combined with one to three graphene monolayers for silicon heterojunction solar cells. The hybrid electrode was inspected using a technique called THz-TDS. The results show that each time graphene sheets are added, they perform better electrically. Solar cells with these mixed electrodes have lower series resistance and better fill factor than reference cells using only ITO. Graphene monolayer devices on ITO are 1.6% more efficient than reference cells. This shows that the hybrid electrode based on graphene has the potential of the next generation of solar cells.

4.3 Flexible Solar Cells

A special advantage of Gr/Si heterojunction is that flexible solar cells can be manufactured by making the silicon substrate very thin. Graphene's high mechanical flexibility enables it to adapt to unbroken curved surfaces, and the silicon sheet (50-100µm) can also be bent into a small radius of curvature. Schottky Gr/Si junction flexible solar cells have reached more than 10% efficiency. These devices have very interesting uses, and normal rigid solar cells are not suitable, such as wearable electronic products, portable power supplies and building integrated solar panels. The main challenges faced by these Gr/Si flexible solar cells are to keep the silicon sheet firm, to ensure that the electrical contacts remain stable when folded, and to protect the equipment from environmental influences.

5. SENSING APPLICATIONS OF GRAPHENE/SILICON HETEROJUNCTIONS

The unique optoelectronic properties of Gr/Si heterojunctions render them excellent candidates for high-performance photodetectors and optical sensors. The broadband absorption of graphene, coupled with its high carrier mobility and tunable Fermi level, combined with the well-established photodetection capabilities of silicon, enables devices with high sensitivity, rapid response, and broad spectral response.

5.1 Photodetectors

Gr/Si heterogeneous photoelectric sensor works through the photoelectric effect of Schottky interface. When photons with energy greater than Schottky barrier are absorbed into silicon, electron hole pairs are created and separated by internal

electric field, resulting in photocurrent. The graphene layer is used as a transparent electrode to collect carriers, and it also helps photocurrent through the internal photoelectric emission of the heat carrier.

5.1.1 Broadband Photodetection

Gr/Si photoelectric sensor has a very wide range of responses, from ultraviolet (UV) to near infrared (NIR). The answers in ultraviolet and visible light mainly come from the absorption of photons in silicon. For the response in near infrared (wavelength over 1000 nm), this is because graphene has no band gap, which enables it to absorb photons, even if there is little energy. This broad response is an advantage in optical communication, imaging and spectroscopy applications.

5.1.2 High Sensitivity

The high fluidity of the carrier in graphene allows efficient collection of the photogenerated carrier, thus providing high responsiveness. Gr/Si photoelectric sensor shows a response value of more than 0.5 A/W in visible and near infrared regions. Responsiveness can further improve chemical hybridization or apply grid tension to change the level of Fermi graphene.

5.2 Gas Sensors

Gr/Si heterojunctions have also been investigated for gas sensing applications. The large surface area of graphene and its sensitivity to charge transfer from adsorbed gas molecules make it an outstanding sensing material. Li et al.^[14] studied the fabrication and performance of sensors based on graphene heterojunctions. When gas molecules adsorb onto the graphene surface, they can donate or accept electrons, shifting the Fermi level of graphene and altering the electrical properties of the heterojunction. The sensing mechanism involves modulation of the Schottky barrier height at the Gr/Si interface by adsorbed gas molecules. For oxidizing gases (such as NO₂ and O₂), electron withdrawal from graphene increases its work function, raising the Schottky barrier and decreasing current. For reducing gases (such as NH₃ and H₂S), electron donation to graphene reduces its work function, lowering the Schottky barrier and increasing current.

5.3 Biosensors

Gr/Si heterojunctions hold potential for biosensing applications due to the biocompatibility of graphene and the mature microfabrication technology for silicon. Li et al.^[15] investigated 3D graphene/low-dimensional material heterojunction optoelectronic sensing chips. Graphene's large surface provides many sites for biomolecule attachment. At the same time, its sensitive electrical response enables it to detect when biomolecules interact. If we add specific receptors (such as antibodies, aptamers or DNA detectors) to the surface of graphene, we can only detect the biomolecules we are looking for. When they are bound, this will change the charge distribution and the number of carriers in graphene. These changes can be seen in the current-voltage characteristics of heterojunction. Because of its high sensitivity and unmarked detection ability, Gr/Si heterogeneous biosensor is very attractive in medical diagnosis, environmental monitoring and food safety applications.

6. ENERGY STORAGE APPLICATIONS OF GRAPHENE/SILICON HETEROJUNCTIONS

Beyond photovoltaics and sensing, Gr/Si heterojunctions and related material systems have shown promise for energy storage applications, particularly in supercapacitors and lithium-ion batteries. Graphene's large surface area, excellent electrical conductivity and mechanical flexibility, coupled with the high theoretical ability of silicon, make these materials very interesting for the next generation of energy storage devices.

6.1 Supercapacitors

Supercapacitors, also known as supercapacitors or electrochemical capacitors, store energy. They are realized by separating charges at the position where the electrode contacts the electrolyte (i.e. double-layer capacitance) or by carrying out a very rapid chemical reaction on the surface (i.e. pseudo capacitance). Graphene-based materials are very good candidates to make supercapacitor electrodes. This is because they have a large surface area (theoretical value is 2630m²/g)^[16], and they conduct electricity well and are chemically stable.

Gr/Si structure can be used as electrodes of supercapacitors. Here, the silicon substrate provides mechanical support, and the graphene layer serves as the active electrode material.

Graphene's large surface area allows high capacitance. Its good conductivity also ensures low equivalent series resistance and high power density.

In addition, the Gr/Si interface can be modified to improve the load storage through the pseudo capacitance effect. Zhao and Zhu^[16] reviewed the cationic π interactions in graphene-containing systems and emphasized their importance for energy generation and storage.

The strong interaction between π electrons and cations leaving graphene leads to the redistribution of charges at graphene/electrolyte interface and the formation of pseudocapacitance. This pseudo capacity can greatly increase the energy density of graphene-based supercapacitan.

6.2 Lithium-Ion Batteries

Silicon is a promising material for anode of lithium ion battery. This is because it has a very large theoretical capacity (Li₄ is 4200mAh/g). 4Si), which is about ten times that of ordinary graphite (372mAh/g). However, silicon has a big problem: it will expand a lot (~300%) when charged^[17]. This creates mechanical stress, and particles may break and lose electrical contact. This is why the battery capacity has dropped so quickly.

Graphene can solve these problems. It forms a flexible and conductive matrix. It can withstand the volume increase of silicon. It keeps electrical contact when the battery is charged and discharged. Composite Gr/Si anode is made in many ways. For example, by mechanical mixing, by chemical vapor deposition, and by self-assembly. The graphene layer is coated with silicon particles. This will create a stable solid electrolyte interphase (SEI). This also prevents silicon from directly contacting the electrolyte. Zhang^[17] studied the energy storage properties of Graphdiyne/Graphene Heterojunction. He used first-principles calculations. The research shows that the heterojunction structure can improve the adsorption of metal ions. They can also improve the load transfer kinetics. This improves the performance of energy storage. These findings provide theoretical guidance. They are helpful to design better electrode materials. These materials are based on graphene heterojunctions.

7. CONCLUSIONS AND OUTLOOK

Gr/Si heterojunction is a very useful material platform, which has great potential in photovoltaics, sensing and energy storage. This review examines the latest developments in this field, preparation methods, strategies for optimizing performance, and different applications.

7.1 Summary of Key Achievements

Synthesis and assembly: Chemical vapor deposition continues to lead the production of graphene on Gra/Si platform, because it provides uniform coverage and layer thickness control in large wafer area. Complementary transmission protocols-wet, dry, and solution-based-have adapted to reliable heterogeneous structures. The inclusion of an intermediate buffer layer, such as hexagonal nitride, alumina, molybdenum disulfide, or sulfur dioxide, between graphene and silicon shows that it significantly improves the integrity of the interface.

Improvement of equipment: modifying carrier density and graphene surface potential leads to a significant increase in output characteristics through acidity, metal chloride or organic additives, mainly by increasing comprehensive potential and reducing sheet resistance. Passive interface and intake surface state reduce non-radiative recombination and accelerate carrier transportation. At the same time, the textured surface and anti-reflective dielectric coating enhance photon coupling in the active region, thus improving the conversion efficiency.

Solar energy, how to convert it: The efficiency of Schottky Gr/Si battery has increased from a record 1.5% in 2010 to more than 15% today. Improved by more than 10 times. Electrodes that mix graphene with common transparent conductive oxides produce less resistance and better fill factor. In addition, the design with a thin substrate allows the manufacture of foldable devices, which are very suitable for wearable devices and facilities on buildings.

Optical and chemical detection: photoelectric sensors made of Gr/Si architecture look very wide, ranging from ultraviolet to near infrared, and they are very sensitive and fast. They can be used for data transmission, image shooting or spectral analysis. But that's not all! The same heterogeneous binding platform also shows that it is very sensitive to detect gas or biomolecule targets.

Electrochemical storage: Gr/Si heterogeneous structure is competitive in the test of supercapacitors and lithium-ion batteries. Graphene skeleton provides three-dimensional conductivity, large surface for ion adsorption, and mechanical flexibility to absorb large volume changes related to lithium silicon.

7.2 Current Challenges

Although great progress has been made, there are still several challenges to be solved:

stability: chemical impurities of graphene are often unstable under normal conditions, resulting in the decline of performance over time. For practical application, it is very important to develop a stable doping method or packaging strategy.

Interface quality: Gr/Si interface is still the key factor limiting the performance of equipment. We need to further improve the interface engineering to reduce reassembly and optimize tape alignment.

Scalability: Although CVD graphene can be produced on a large surface, the transmission process and equipment manufacturing steps must be suitable for commercial applications, while maintaining high quality and low cost.

Theoretical understanding: A deeper theoretical understanding of transmission load, recombination and transmission process on Gr/Si interface is necessary to guide further optimization.

7.3 Future Perspectives

Looking into the future, several directions show the hope of promoting Gr/Si heterojunction technology:

Integration of new materials: Integration with Gr/Si heterojunction of other two-dimensional materials (such as TMDs, h-BN and MXenes) may allow new functions and improve performance through synergistic effect.

Advanced device architecture: New device architecture, such as series unit combining Gr/Si connector with other photovoltaic technologies, can achieve higher efficiency and wider application.

Machine learning and computational design: The combination of machine learning method and first-principle calculation can accelerate the discovery of the best material combination and equipment configuration.

Emerging applications: In addition to those we have seen, Gr/Si heterojunction may be useful in new fields, such as neuromorphological computing, quantum devices and flexible electronics.

In a word, Gr/Si heterojunctions has shown extraordinary potential in many application fields. It is very important to continue to study and solve current problems and explore new directions. This will realize the full potential of this exciting material platform and turn the findings in the laboratory into practical technology. These technologies will contribute to sustainable energy and advanced electronic products.

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