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Graphene Polymer Composites in Flexible Materials

Heng Jin[1, *]

and Boxuan Zhang[2]

[1]College of Chemistry and Materials Science, South Central Minzu University, Wuhan, China
[2]The Affiliated International School of Shenzhen University(YHV), Shenzhen, China

*Corresponding author: 202121132006@mail.scuec.edu.cn

Abstract:

Flexible materials have made remarkable development in recent years and are widely used in various fields such as flexible sensors and wearable devices. Existing flexible sensors, wearable devices, flexible electronics and other devices, however, still have problems such as low precision and poor mechanical properties. The emergence of graphene provides a new idea to solve these problems. The composite of graphene, which has excellent electrical conductivity, mechanical strength and flexibility, with polymer materials can improve its electrical and mechanical properties without affecting the flexibility of the materials. In this paper, we mainly introduce the relevant properties of graphene and polymer materials, list the common composite preparation methods of graphene and polymer materials, and the current status of graphene polymer composites. At this stage, graphene polymer composites are widely used in flexible electronics, wearable devices, sensors, folding screens, etc., but there are still some properties that need to be improved and enhanced

Keywords: Flexible materials; graphene; applications.

1. Introduction

Graphene, as an emerging two-dimensional material, receives close attention in the field of flexible materials due to its unique physical and chemical properties. The research direction of graphene in flexible materials mainly includes sensors and electronic devices, biosensors and drug delivery systems. Internationally, the research on graphene started earlier, and the main directions include: graphene electrode materials, energy storage devices, and only textiles and so on. However, despite the progress made in the research of graphene flexible materials, there are still challenges: the production technology of graphene has not yet matured, and the cost of large-scale production is high; the production technology of high-quality graphene has not yet matured; and the interfacial compatibility between graphene and other materials needs further research. Polymer materials, as a kind of materials widely used nowadays, are organic materials with polymer compounds as the main components. The relative molecular mass of polymer compounds is generally above 5000, and some are even as high as several million. Polymer chain due to internal rotation, can produce a very large number of conformations, so that the main chain bending and flexible, plastic deformation ability. Polymer is an ideal flexible material. When the polymer is compounded with graphene, the polymer material will produce a certain deformation when subjected to a certain external force, which will lead to the deformation of the surface graphene, which in turn affects the electrical properties of graphene, and provides a new way of thinking for the production of flexible electrodes, flexible sensors folding screen, and wearable flexible devices.

This paper introduces the structure and properties of graphene and some polymers, and describes the unique advantages of each of them. By integrating the relevant experimantal research and application progress on graphene/polymer composites in recent years, this article further reveals the prospects of graphene/polymer materials for a wide range of applications in the fields of flexible wearable sensors and foldable screens. And finally, it summarises the directions of future research, with a view to providing theoretical support and technological references to the researchers in the related fields.

2. Structure of Graphene and Polymer Materials

2.1 Graphene Structure and Properties

In 2000-2004, Andrey K. Heim and Kosta Novoselov, two scientists from the University of Manchester, used the 'tape tearing method' to peel off graphene flakes, and for the first time obtained single-layer graphene samples, and after a series of tests, it was found that graphene has excellent mechanics, electromagnetism, optics, thermodynamics and other advantages. After a series of tests, it was found that graphene has excellent mechanical, electromagnetic, optical, and thermodynamic properties, and the reason lies in the special structure of graphene [1].

Graphene is a two-dimensional material consisting of carbon atoms through sp^2 hybridisation orbitals in a single atomic layer thickness. The carbon atoms are arranged in a honeycomb hexagonal lattice structure, and each carbon atom is linked to the other three carbon atoms through covalent bonds to form a strong and regular lattice. As a two-dimensional crystalline material, graphene has the advantage of a dense structure while being stable, and the suspended graphene can be measured by using nanoindentation on nanopores. The modulus of elasticity of graphene can be obtained as 1.0 TPa and the strength σint as 130 GPa, which gives an idea about the mechanical properties of graphene. The carbon-carbon bond length in graphene is about 0.142 nm, which gives graphene very high mechanical strength and elastic modulus. Graphene has one p-orbital electron for each carbon atom that is involved in the hybridisation due to sp^2 hybridisation these electrons form a π-electron cloud above and below the plane, this π-electron cloud gives graphene an extremely high electrical conductivity as the π-electrons are able to move around freely, resulting in the formation of an extremely conductive two-dimensional electron gas. In the Brillouin zone of graphene, the π-band intersects with the π*-band at the K and K' points to form a Dirac cone, which makes the graphene carriers behave as massless Dirac fermions in the low energy state, which gives them a unique electrical property. Graphene's crystal structure is perfect, which does not block the motion of the electron waves in the crystal, and thus has perfect electrical conductivity.

Graphene, as a single-layer, two-dimensional material, has a tremendous specific surface area of about 2630 square metres per gram. Even though graphene is only a single atomic layer thick, due to its ultra-thin structure, graphene transmits more than 97% of visible light. Graphene is considered to be one of the strongest materials known, with a Young's modulus of about 1 TPa and a tensile strength of up to 130 GPa, yet it also maintains good flexibility. There may be some defects in the structure of graphene, such as vacancies, five-membered ring or seven-membered ring defects, etc., which affect the electronic structure and physical properties of graphene.

2.2 Structure and Properties of Polymer Materials

In recent years, smart materials, self-healing materials, biodegradable plastics, conductive polymers and other functional polymer materials have been widely studied and applied. Polymer materials are widely and diversely used in flexible materials. Polyimide (PI) contains a large number of rigid aromatic rings in its molecular structure, which makes polyimide have high molecular rigidity and thermal stability, and the strong polarity of the imide ring increases intermolecular interactions. Due to its excellent thermal stability and mechanical properties, it is widely used in flexible circuit boards, flexible displays, and other high-temperature applications, where it can be used as a substrate material to support other conductive materials or functional layers.

Polyethylene Dioxythiophene (PEDOT) is a conductive polymer, widely used in light-emitting diodes, organic solar cells, and flexible sensors, suitable for the manufacture of transparent electrodes and conductive layers, which can be coated on a variety of flexible substrates.

Polyurethane (PU), and polydimethylsiloxane (PDMS), are used in the manufacture of flexible, breathable medical sensors and wearable devices. These materials offer excellent biocompatibility and comfort for long-term wear.

Poly-p-xylene (Parylene), polyimide (PI), and polycarbonate (PC) are widely used as substrate materials for flexible displays. These materials have excellent mechanical strength, heat resistance, and transparency, and can withstand repeated bending.

3. Graphene/polymer Composite Preparation Method

An environmentally friendly way of preparing highly

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conductive chemically reduced graphene oxide/polystyrene composites was reported by Long Gucheng, Junjie Shi et al. Firstly, a microsphere suspension of polymer was mixed with graphene oxide solution, which was graphene oxide adsorbed on the surface of microsphere, and the graphene oxide on the surface was reduced in situ by using hydriodic acid, and then hot pressed to produce polymer conductive composites adsorbed with reduced graphene. The method is simple to prepare, environmentally friendly, and can ensure that the reduced graphene is uniformly dispersed in the polymer microspheres, and composites with superior conductive properties can be obtained. Moreover, the method is widely applicable, not only to the preparation of reduced graphene/polystyrene composites but also to the preparation of other polymer composites [2]. However, the degree of graphene oxide reduction in the in situ reduction method is still the technical key of the method, and the selection of the reducing agent, the setting of the reduction temperature and the reduction time will have a large impact on the properties of graphene polymer composites

Zheng Yuan et al. prepared graphene/polyaniline composite film electrodes using the self-assembly method. Through the assembly of graphene and polyaniline on the electrodes, the electrode materials can be prepared easily and efficiently, and can be used directly for performance testing or detection. The electrostatic interaction between the positive and negative charges between the electrodes is the driving force, so that the positively charged lamellar polyaniline and negatively charged graphene oxide are attracted by electrostatic force. The study indicated that the properties of graphene/polyaniline composite films prepared on electrodes using an electrostatic layer-by-layer self-assembly technique were further improved. The self-assembly method can be used to prepare three-dimensional graphene/polyaniline hollow microspheres in addition to two-dimensional thin film materials [3].

The melt blending method was collated and summarised by Md Abdullah Al Faruque et al. Melt blending graphene with thermoplastic polymer materials to make composites is an environmentally friendly, cost-effective and scalable approach. Melt blending does not require solvent, graphene treatment and modification. During melt blending, the degree of dispersion of graphene depends on the degree of melting of the polymer at that temperature, and melt polymerisation is suitable for composites of graphene with polyurethane [4], PET [5], polylactic acid [6], styrene, acrylonitrile [7] and isotactic polypropylene. However, excessive temperature of melting, defects and breakage of graphene can affect the properties of graphene/polymer composites.

4. Graphene/polymer Applications in Flexible Materials

4.1 Flexible Wearable Sensors

Bao et al. used electrospinning to prepare fibre mats with polylactic acid (PLA) and a spraying technique to fabricate flexible strain sensors based on poly(lactic acid) (PLA) fibre membranes by spraying graphene solution onto the surface of a fibre-like mould. Due to the advantages of electrospun fibres, the graphene/PLA composite exhibits excellent flexibility, can be bent, folded and twisted for long term durability and repeatability over 1200 cycles. Meanwhile, the contact angle of the composite product was measured, and it can be found that the contact angle of the composite material was slightly increased compared to PLA, indicating that the material has enhanced water resistance and is suitable for the preparation of wearable electronics. The prepared flexible strain sensors were fixed on different joints such as fingers and knees using double-sided adhesive, and it was found that the sensors could monitor the movement of such joints in real time and accurately, and the detected values quickly returned to the original values in the case of muscle relaxation, which indicated that the composite material had structural stability. In addition, the strain sensors can be operated at relatively low voltages, so they do not generate excessive heat and operate at temperatures that are within the comfort range for human health, making them safe and reliable. However, the dense matrix of the flexible device may cause discomfort to the human skin. Thus, future development can focus more on the solution of the breathability of graphene/polymer material flexible strain sensors [8].

Yimeng Li et al. prepared a flexible piezoresistive pressure sensor with simple structure, low cost and stable performance by replicating the surface microstructure of bionic ginkgo biloba onto a polydimethylsiloxane (PDMS) substrate and spin-coating graphene ink as a functional layer. The sensor has excellent contact ability, very high sensitivity due to the Ginkgo biloba microstructure and the two-dimensional material graphene functional layer, the response time of the sensor is only 80ms, and it can sensitively identify the objects of about 0.2 g. At the same time, the sensor can maintain good performance under 10,000 cyclic pressure tests. Moreover, due to the good biophilicity of polydimethylsiloxane (PDMS), the prepared sensor has flexibility and dexterity, and is suitable for wearing on all parts of the human body (fingers and wrists, etc.) [9].

4.2 Folding Screen

Vlasov, A. I., et al. have investigated a flexible touchscreen using graphene material with an integrated analogue-to-digital converter. Graphene is used as the main material for touch screens due to its excellent conductivity and flexibility. The integrated converter is able to convert touch signals into digital signals, which improves the response speed and accuracy of the touch screen. Graphene's high conductivity makes the touch screen more responsive. Graphene's flexibility allows the touchscreen to be bent and folded, making it suitable for wearable devices and flexible electronics. The integrated analogue-to-digital converter improves the accuracy of the touch signal and enhances the user experience. Users will have a smoother experience when using the touchscreen due to the fast response time and high accuracy. This touchscreen can be applied to various flexible electronic products, such as smartwatches and foldable mobile phones. However, the high production cost of graphene material may affect the marketing of the product. The technical complexity of the integrated analog-to-digital converter is high and needs to be further optimised and simplified. The future development direction may be how to reduce the production cost of graphene materials to make them more competitive in the market. Further optimise the integration technology of analogue-to-digital converters to improve production efficiency and product performance. Explore the application of graphene touch screens in more fields, such as medical devices, smart home [10].

Usman Khan et al. investigated the potential of graphene in e-skin and touchscreen applications. Graphene has high electrical conductivity and mechanical strength. The unique properties of graphene are utilised to develop new electronic devices. The electrical conductivity of graphene is several times higher than conventional materials. Graphene's strength gives it an advantage in flexible electronic devices. Improved sensitivity and durability of electronic devices. The use of graphene materials reduces energy consumption. However, the current high production cost of graphene limits its large-scale application. And some application technologies are not yet fully mature and need further research. The future development direction is in reducing the production cost of graphene through technological innovation. Explore the application of graphene in more fields, such as energy storage and biomedicine [11].

5. Conclusion

Based on the application of flexible materials at home and abroad, and the advantages of graphene/polymer composites in flexible devices, this study reviewed the structure and properties of graphene two-dimensional materials, and the common methods of composite modification of polymer materials and graphene. The advantages and applications of graphene composite with polymer materials in flexible wearable sensor devices and folding screens are described, while the shortcomings of the composite materials are pointed out. Future research needs to further optimise the preparation process of composites, as well as the surface modification of graphene. And the synergistic mechanism with polymers are explored in depth, to develop composites with more stability and functionality, and comfort, in order to respond to the growing market demand.

Authors Contribution
All the authors contributed equally and their names were listed in alphabetical order.

References

[1] Wang Bailu Starting from the Nobel Prize winning exploration of pencil lead - the discovery, physical properties, and application prospects of graphene. Exploration of Physics Teaching: Middle School Teaching and Research Album, 2012,30 (11): 1-3,11

[2] Long Gucheng, Tang Changyu, Man Changzhen, et al. Preparation and Performance Study of High Conductivity Electrochemical Reduction Graphene Oxide/Polystyrene Composite Materials. Plastics Industry, 2013 (3): 4.

[3] Zheng Yuan, Luo Jing, Liu Xiaoya Preparation and Application of Graphene/Polyaniline Composite Materials. Polymer Bulletin, 2017 (6): 19-26

[4] Kim, H.; Miura, Y.; Macosko, C.W. Graphene/Polyurethane Nanocomposites for Improved Gas Barrier and Electrical Conductivity.Chem. Mater. 2010, 22, 3441–3450.

[5] Shabafrooz, V.; Bandla, S.; Allahkarami, M.; Hanan, J.C. Graphene/polyethylene terephthalate nanocomposites with enhanced mechanical and thermal properties. J. Polym. Res. 2018, 25, 256.

[6] Chieng, B.W.; Ibrahim, N.A.; Wan Yunus, W.M.Z.; Hussein, M.Z.; Silverajah, V. Graphene nanoplatelets as novel reinforcement filler in poly (lactic acid)/epoxidized palm oil green nanocomposites: Mechanical properties. Int. J. Mol. Sci. 2012, 13: 10920–10934.

[7] Steurer, P.; Wissert, R.; Thomann, R.; Mülhaupt, R. Functionalized graphenes and thermoplastic nanocomposites based upon expanded graphite oxide. Macromol. Rapid Commun. 2009, 30: 316–327.

[8] Bao, Z., Liu, Z.&Sun, B. manufacture flexible, biocompatible, and permeable graphene/PLA strain sensors for wearable devices. Fiber polymer, 2024

[9] Li Yimeng, Hou Xiaojuan, Zhang Liaoyuan, etc Graphene/PDMS biomimetic Ginkgo biloba leaf microstructure flexible piezoresistive pressure sensor. Micro Nano Electronic Technology, 2020,57 (3): 198-203

[10] Vlasov, A. I., Terent'ev, D. S., & Shakhnov, V. A. Graphene flexible touchscreen with integrated analog-digital converter. Russian Microelectronics, 2017: 46(3), 192–199.

[11] Khan, U., Kim, T.-H., Ryu, H., Seung, W., & Kim, S.-W. (2016). Graphene Tribotronics for Electronic Skin and Touch Screen Applications. Advanced Materials, 29(1), 1603544.