

Research on the Preparation and Property Control of Composite Materials Based on Single-Walled Carbon Nanotubes

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Abstract: Single-walled carbon nanotubes (SWCNTs) possess a unique electronic structure, high mechanical strength, and excellent thermal conductivity, making them a focus of research in the field of composite materials. Therefore, this paper first summarizes the preparation methods of SWCNT-based composite materials; then analyzes the property control strategies of SWCNT-based composite materials, and proposes methods to improve their mechanical, electrical, and thermal properties by controlling the defect degree and spatial arrangement, as well as conducting surface functional modification; finally, combined with the latest research results, focuses on the application breakthroughs of SWCNT-based composite materials in fields such as conductive agents for lithium-ion batteries, aerospace structures, and flexible electronic devices, so as to provide theoretical guidance for the industrial preparation of high-performance composite materials.

Keywords: Single-walled carbon nanotubes; Composite materials; Preparation; Property control

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1. Introduction

As an “important member” of the carbon nanomaterial family, single-walled carbon nanotubes (SWCNTs) have a tube diameter of only 0.4–3 nm but can reach several micrometers or even longer in length, exhibiting an extremely high aspect ratio. This unique geometric structure endows SWCNTs with outstanding physicochemical properties, making them a promising reinforcing phase in the field of composite materials. However, the industrial application of SWCNTs faces major obstacles, such as the high cost of preparation, difficult chirality control, and severe agglomeration. In recent years, composite materials have gradually evolved from single-component to multi-component and multi-functional development, bringing new possibilities for the industrial application of SWCNTs. Through composite material design, measures such as controlling the defect degree and spatial arrangement, and carrying out surface functional modification can realize the “optimizing the performance” of SWCNT performance, meeting the diversified and personalized

application demands in different fields ^[1].

2. Preparation methods of SWCNT-based composite materials

2.1. Preparation of SWCNTs

At present, the chemical vapor deposition (CVD) method is the mainstream technology for the industrial preparation of SWCNTs. Its core principle is the decomposition and growth of SWCNTs on the surface of catalysts (Fe, Co, Ni-based alloys) using carbon source gases (such as methane and ethylene) ^[2]. Moreover, according to different catalyst supply modes, the CVD method can be further divided into the substrate method, the supported catalyst method, and the floating catalyst method. A comparison of the characteristics of SWCNTs prepared by different CVD methods is shown in **Table 1**.

Table 1. Comparison of characteristics of SWCNTs prepared by different CVD methods

Preparation Method	Principle	Advantages
Substrate Method	Catalyst nanoparticles are deposited on the substrate, and growth conditions are controlled for directional growth	Precise control of growth direction and arrangement, well-aligned and aligned arrays
Floating Catalyst Method	Catalyst precursor and carbon source gas are injected into the high-temperature zone, and SWCNTs are generated and deposited via gas-phase reaction	Continuous and large-scale production, low cost
Plasma-Enhanced CVD Method	Plasma is introduced into traditional CVD to lower the reaction temperature and promote growth	Low-temperature preparation, improved crystal quality, reduced defects

Substrate Method: Catalyst nanoparticles are deposited on the surface of a substrate (such as a silicon wafer), and then directional growth of SWCNTs is achieved by controlling the growth temperature and carbon source flow rate. This method enables precise control over the growth direction and arrangement of SWCNTs, resulting in neatly structured and aligned arrays, which have potential applications in electronic devices, sensors, and other fields.

Floating Catalyst Method: A catalyst precursor (such as ferrocene) and carbon source gas are co-injected into a high-temperature reaction zone (900–1600 °C). SWCNTs are generated via gas-phase reaction and deposited on the surface of a collector along with the air flow. The advantage of the floating catalyst method is that it enables continuous and large-scale production with relatively low cost, making it suitable for industrial applications ^[3].

Plasma-Enhanced CVD Method: Plasma is introduced into the traditional CVD reaction. The high-energy active particles of plasma reduce the reaction temperature and promote the decomposition of carbon source gas and the growth of SWCNTs. This method not only allows the preparation of SWCNTs at a lower temperature but also effectively improves the crystal quality and reduces defects of SWCNTs, enhancing their performance. It is often used in the preparation of electronic devices and energy storage materials ^[4].

2.2. Preparation of SWCNT-based composite materials

As shown in **Table 2**, the two preparation methods of SWCNT-based composite materials each have their own advantages.

Table 2. Comparison of two preparation methods of SWCNT-based composite materials

Preparation Method	Operation Process	Advantages
Physical Mixing Method	SWCNTs and matrix solution are mixed by ultrasonication and stirring, followed by casting and hot-pressing molding	Simple operation, low cost
In-Situ Polymerization Method	SWCNTs are dispersed in a monomer solution, and polymerization is initiated to form a coating layer	Improved dispersibility, enhanced interfacial bonding, excellent performance

2.2.1. Physical mixing method

This method involves uniformly mixing SWCNTs with matrix materials in a solution through ultrasonication, mechanical stirring, and other means, and then fabricating the composite material via casting, hot-pressing, and other processes. The method is simple to operate and low in cost, but the agglomeration of SWCNTs tends to degrade the performance of the composite material. To solve this problem, researchers can adopt measures such as surface functional modification and dispersant-assisted dispersion according to application requirements. For example, when preparing SWCNT/epoxy resin composites, SWCNTs can be surface-functionalized with a surfactant first to enhance their dispersibility in the solution, then fully mixed with the epoxy resin solution under stirring, and cast and cured to obtain a composite material with relatively excellent performance ^[5].

2.2.2. In-Situ polymerization method

The in-situ polymerization method disperses SWCNTs in a monomer solution and initiates a polymerization reaction to form a polymer coating layer on the surface of SWCNTs. This method can improve the dispersibility of SWCNTs and enhance the interfacial bonding force between SWCNTs and the polymer matrix, yielding composite materials with better comprehensive performance. It is frequently used in the preparation of high-performance fibers and biomedical materials. For instance, SWCNT/polyimide composites are novel materials prepared by the in-situ polymerization method, which can retain 85% of their initial strength at a high temperature of 300 °C and can be used in aerospace thermal protection structures.

3. Property control strategies of SWCNT-based composite materials

3.1. Defect engineering regulation

Defects in SWCNTs significantly affect the electronic structure and mechanical properties of the material. Therefore, in practical applications, defects are introduced in a controlled manner by means of plasma etching, electron beam irradiation, etc., to modify the performance of SWCNT-based composite materials. In the property control of SWCNT-based composite materials, the defect engineering regulation strategy is generally used to regulate the mechanical and electrical properties of materials.

In terms of mechanical property regulation, the defect engineering strategy can improve the toughness of SWCNT-based composite materials to a certain extent. For example, introducing appropriate defects during the preparation of SWCNT/epoxy resin composites allows the material to better disperse stress under external force, avoiding premature fracture caused by stress concentration.

In terms of electrical property regulation, the defect engineering strategy can adjust the electrical conductivity of SWCNT-based composite materials. This is because defects can act as carrier scattering

centers and affect the transport path and mobility of carriers. Proper introduction of defects can create more scattering and hopping paths for carriers in SWCNT-based composite materials, thereby improving the electrical conductivity of the material. For example, introducing an appropriate amount of defects via plasma etching during the preparation of SWCNT/polyaniline composites can significantly enhance the electrical conductivity of the material.

However, more defects are not better. Excessive defects will destroy the structural integrity of SWCNTs, leading to a sharp decline in the mechanical properties of the material, making it difficult to meet application requirements^[6-7].

3.2. Surface functional modification

The surface chemical state of SWCNTs directly affects their interaction with the matrix interface. Therefore, directional optimization of composite material performance can be achieved by introducing specific functional groups through covalent or non-covalent modification.

Covalent modification introduces active groups such as carboxyl and amino groups onto the surface of SWCNTs through acid oxidation and amination reactions, enhancing chemical bonding with polar matrices and thereby improving the performance of composite materials. For example, when preparing SWCNT/polyamide composites, acid oxidation can be used to introduce carboxyl groups onto the surface of SWCNTs, which react with amino groups in polyamide to form strong chemical bonds, significantly improving the interfacial bonding strength and mechanical properties of the composite material.

Non-covalent modification adsorbs aromatic molecules (such as pyrenebutyric acid, crown ether) or polyelectrolytes (such as polystyrene sulfonic acid) onto the surface of SWCNTs through π - π interactions or electrostatic interactions, aiming to improve the dispersibility of SWCNTs in solution and their compatibility with the matrix. This surface functional modification method can improve dispersibility while maintaining the integrity of the electronic structure of SWCNTs, and is commonly used in the preparation of composite materials with stringent requirements for electronic properties. For example, SWCNT/polythiophene composites used in some organic solar cells adopt this surface functional modification method.

3.3. Spatial arrangement regulation

The spatial arrangement of SWCNTs (e.g., random distribution, directional alignment) has a decisive influence on the load transfer efficiency in composite materials.

Randomly distributed SWCNTs provide a certain reinforcing effect in all directions in the composite material, resulting in relatively balanced performance of the material in different directions. However, the synergistic effect between SWCNTs is weak under this arrangement, and the load transfer efficiency is low. Therefore, although randomly distributed SWCNTs can improve the strength and toughness of the material to a certain extent, the improvement range is relatively small, and they are usually used in the preparation of ordinary plastic matrix composites.

In contrast, directionally aligned SWCNTs can significantly improve the performance of composite materials in specific directions. Directional alignment regulation arranges SWCNTs along a specific direction through magnetic field assistance, shear flow field, or template method, thereby enhancing the mechanical, electrical, and thermal properties of the composite material in that direction^[8-9].

For example, when preparing SWCNT/epoxy resin composites for aerospace applications, magnetic field-assisted directional alignment of SWCNTs can greatly improve the strength and stiffness of the material

in the flight direction to meet the high-performance requirements of structural materials for aerospace vehicles. When preparing SWCNT/polymer composites for flexible electronic devices, shear flow field directional alignment of SWCNTs can significantly improve the electrical and thermal conductivity of the material in specific directions, meeting the special performance requirements of flexible electronic devices^[10].

4. Application progress of SWCNT-based composite materials

4.1. Conductive agents for lithium-ion batteries

With the advantages of ultra-high electrical conductivity and low dosage, SWCNTs are gradually replacing traditional carbon black and multi-walled carbon nanotubes (MWCNTs) to become key conductive agents for high-energy-density lithium-ion batteries. For example, after adding SWCNTs as conductive agents to cathode materials of high-energy-density lithium-ion batteries such as LiCoO_2 and LiMn_2O_4 , the charge-discharge efficiency and cycle stability of the batteries are significantly improved. The reason is that SWCNTs have a unique one-dimensional structure, which can form an efficient conductive network between electrode material particles, thereby greatly reducing the internal resistance of the battery^[11].

4.2. Aerospace structural materials

The ultra-high strength and low density (1.3 g/cm^3) of SWCNTs make them ideal lightweight reinforcing materials for the aerospace field. For example, adding 0.1 wt% SWCNTs to carbon fiber/epoxy resin composites can significantly improve the interlaminar shear strength and fracture toughness^[12]. At present, SWCNT-reinforced carbon fiber prepreps have been adopted in the tail structure of some large passenger aircraft. The application of this prepreg reduces the weight of the tail while comprehensively improving its strength and fatigue resistance^[13].

4.3. Flexible electronic devices

The flexibility and high electrical conductivity of SWCNTs make them ideal materials for flexible electronic devices. For example, some flexible displays use SWCNT transparent conductive films instead of indium tin oxide (ITO) to achieve foldable and bendable functions, greatly expanding the application scenarios of display devices and improving design flexibility. Some wearable sensors use SWCNT-based composite materials as sensitive elements, which play an important role in human motion monitoring and health status detection. Their good flexibility ensures the fit between the sensor and human skin, improving the accuracy and comfort of detection^[14-15].

5. Summary and prospect

In summary, SWCNTs have a unique geometric structure, high mechanical strength, and excellent thermal conductivity, showing significant application advantages and great application potential in the field of composite materials. In industrial production, composite material design can solve the problems of high preparation costs, difficult chirality control, and severe agglomeration of SWCNTs, providing high-performance materials for various fields.

At present, the main preparation methods of SWCNT-based composite materials include physical mixing and in-situ polymerization; the main property control strategies include defect engineering regulation, surface

functional modification, and spatial arrangement regulation. Researchers need to select them according to specific production conditions and application requirements. In practical applications, the coordination of different preparation methods and property control strategies can make SWCNT-based composite materials exhibit more excellent performance, meeting the manufacturing needs of lithium-ion battery conductive agents, aerospace structures, and flexible electronic devices, and consolidating the development foundation of related fields.

In the future, with the deepening of research and continuous technological innovation, the application potential of SWCNT-based composite materials in more fields will be explored. Technicians can further improve the performance of SWCNT-based composite materials from the following aspects:

- (1) Precisely control the reaction conditions to achieve accurate control of the tube diameter, length, and chirality of SWCNTs, providing high-quality basic materials for the preparation of high-performance composite materials;
- (2) Gain a deeper understanding of the influence mechanism of defects on the performance of SWCNT-based composite materials, and improve the electrical, mechanical, and thermal properties of composite materials by introducing appropriate defects;
- (3) Develop more efficient and environmentally friendly surface functional modification technologies to improve the compatibility between SWCNTs and matrix materials and endow composite materials with new functions;
- (4) Adopt physical, chemical, or mechanical methods to align SWCNTs directionally and improve the anisotropic properties of composite materials;
- (5) Integrate the advantages of various disciplines through interdisciplinary cooperation to promote the research of SWCNT-based composite materials.

Disclosure statement

The authors declare no conflict of interest.

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