

Research on Graphene/Polyacrylonitrile Composite Nanofiber Membranes

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Abstract: In recent years, electrospun nanofiber membranes have attracted extensive attention due to their advantages of high-efficiency particulate filtration and low pressure drop. However, traditional polymer nanofiber filter membranes have certain limitations in mechanical strength, thermal stability, and functionality. Graphene, with its outstanding two-dimensional structure, mechanical properties, and surface activity, is known as the "king of new materials". Incorporating it into the polyacrylonitrile (PAN) nanofiber matrix can form a synergistic composite membrane, effectively enhancing the comprehensive performance of the filter membrane. This paper comprehensively reviews the research progress of graphene/PAN composite nanofiber membranes: covering the background and significance of PM_{2.5} filtration, the preparation methods of graphene composite electrospun membranes, their microstructure and performance characterization, as well as their application effects in air purification and other fields. Literature results show that graphene/PAN composite membranes can achieve a pressure drop of less than 70 Pa while maintaining a fine particulate matter filtration efficiency of over 99%, and have mechanical strength several times that of the original membrane. Moreover, this composite membrane exhibits excellent stability and durability in harsh environments such as high temperatures and corrosive conditions. Based on the analysis of existing literature data, this paper summarizes the mechanism by which graphene improves the performance of PAN nanofiber membranes and looks forward to their application prospects in high-efficiency air filtration and related fields.

Keywords: Graphene; Polyacrylonitrile; Nanofiber Membrane; Electrospinning; Air Filtration

1. Introduction

PM_{2.5} particulate matter pollution in the air poses a serious threat to the environment and human health, and its control has become a global focus. Conventional air filters such as HEPA filters and melt-blown electret nonwoven fabrics can effectively intercept particles, but they often have high air resistance and high replacement costs. Some filter materials have poor moisture and heat resistance and cannot work for long periods under harsh conditions. The development of filter membrane materials with high filtration efficiency, low energy consumption, and good environmental tolerance is of critical value [1]. Electrospinning technology can produce polymer nanofiber membranes with submicron diameters. These membranes have a dense pore structure and are breathable, and have been widely used in high-efficiency air filtration [2]. However, single polymer nanofiber membranes, such as PAN membranes, generally have limited mechanical strength and insufficient heat and moisture resistance, which limits their practical application.

Graphene and its derivatives, such as graphene oxide, have opened up new paths for enhancing the performance of nanofiber membranes due to their unique two-dimensional sheet structure, extremely high specific surface area, and excellent mechanical and electrical properties [2]. Graphene sheets can be introduced into polymer matrices as functional fillers and closely combined with polymer chains through interfacial interactions, endowing composite materials with outstanding properties [3]. Research shows that there is a strong interfacial interaction between graphene and PAN matrices, which can form stable nanocomposite structures, enhancing the mechanical strength and structural stability of the membrane. Incorporating graphene into electrospun PAN nanofibers is expected to prepare composite nanofiber filtration membranes with high efficiency, low

resistance, and excellent durability [1]. This article will discuss the research status of graphene/PAN composite nanofiber membranes, covering preparation methods, structural and performance characterization, and their advantages in air filtration. Through a review and analysis of relevant literature, the research progress and development direction in this field will be clarified.

2. Literature Review

2.1 Research Overview of Graphene-Based Membrane Materials

Graphene and its derivative materials are widely used in the field of membrane separation, including applications in environmental treatment such as water treatment and gas purification [2, 4]. Compared with traditional membrane materials, graphene-based membranes have a unique layered stacking morphology, adjustable physical and chemical properties, and excellent molecular sieving characteristics [5]. Lopez-Fernandez et al. [2] reviewed that graphene-based membranes, with their extremely strong mechanical strength and ease of processing, can construct various forms of membrane structures (such as free-supported membranes, layered membranes, and nano-composite membranes), and have shown breakthrough progress in water purification and air purification. For example, Wei et al. [5] reported a layered reduced graphene oxide (rGO) membrane, by applying a low positive potential to cause water molecule intercalation between graphene layers and electrochemical expansion of the closed nanochannels, thereby significantly increasing water flux. This method overcomes the problem of the easy dense stacking of graphene sheet layers in pure graphene membrane sheets, which leads to low permeability, and is of great significance for constructing high-flux nanofiltration membranes [5].

In addition to pure graphene membranes, more recent studies have focused on graphene/polymer composite membranes to combine the functional properties of two-dimensional graphene and the processing advantages of polymer matrices [6]. These composite membranes can retain the excellent performance of graphene while also possessing flexibility and ease of molding, and are regarded as strong candidates for environmental treatment

membrane materials. Especially in the field of air filtration, the combination of graphene and polymer nanofibers has aroused great interest. This article focuses on reviewing the research on graphene and polyacrylonitrile nanofiber composite membranes.

3. Preparation and Structure of Graphene/PAN Composite Nanofiber Membrane

3.1 Core Electrospinning Technology for Graphene/PAN Composite Nanofiber Membranes

Electrospinning is the Core Technology for Preparing Graphene/PAN Composite Nanofiber Membranes. Typical method involves dispersing a certain proportion of graphene or graphene oxide (GO) into the PAN spinning solution and forming nanofibers containing graphene through high-pressure electrostatic jetting. Fan et al. [1] used multi-needle electrospinning combined with water bath drawing to coat silver on nylon (SCN) fibers as the core material and uniformly coat a layer of PAN nanofibers, achieving the continuous production of multi-strand composite micro-nanofiber yarns. Simulation studies have shown that process parameters such as the receiving distance significantly affect the fiber morphology: increasing the receiving distance can reduce the fiber diameter and improve the uniformity of fiber dispersion. When the distance is approximately 100mm, smooth and separate fiber structures can be obtained on the surface. Under optimized conditions, the porosity of the obtained PAN nanofiber coated yarns is approximately 38.9%, and the mechanical properties such as tensile strength reach the best. This provides a process reference for the large-scale preparation of graphene/PAN composite membranes.

3.2 Microstructural Changes in Composite Fiber Membranes: Fiber Morphology and Interface Bonding with Graphene Incorporation

In terms of the microstructure of the composite fiber membrane, the morphology of the fibers and the interface bonding undergo significant changes after the introduction of graphene.

[3] Figure 1 shows the influence of GO doping on the morphology of PAN nanofibers before and after thermal treatment: (b) and (c) are scanning electron microscope images of pure

PAN nanofibers before and after thermal oxidation at 280°C, where it can be seen that the un-doped PAN fibers undergo large-scale melting and adhesion during thermal oxidation (the yellow circle marks the melting area); while (d) and (e) are the morphologies of PAN fibers doped with a small amount of GO before and after the same thermal treatment, the presence of GO effectively inhibits the melting and merging of fibers, and the fiber network structure remains clearly separated after thermal oxidation. The transmission electron microscope images (f) and (g) further confirm that GO doping promotes the formation of "node welding" connections at some fiber intersections instead of large-scale fusion, which enhances the integrity and stability of the fiber network at the microscopic level. This indicates that graphene forms a good interface bonding with the PAN matrix at the nanoscale, and can prevent structural collapse during subsequent processing or use.

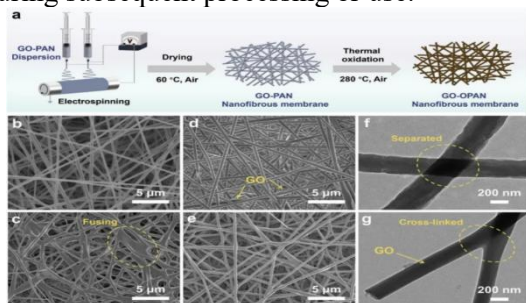


Figure 1. Preparation of GO-Doped PAN Nanofiber Membranes and Comparison of Their Microstructures Before and After Heat Treatment

The research by Si et al. also indicates that there is a strong interfacial interaction between aramid nanofibers and graphite layers, which can give the composite membrane excellent structural stability. Even when immersed in water for a long time or placed in acidic or alkaline solutions, its morphology and performance are almost unaffected [4]. Thus, the introduction of graphene significantly improves the structural strength and stability of PAN nanofiber membranes, laying the foundation for the preparation of high-performance filtration membranes. (a) Schematic diagram of the electrospinning preparation of GO-PAN composite nanofiber membranes and the formation of GO-OPAN membranes through thermal oxidation at 280°C in air; (b) and (c) SEM images of pure PAN nanofiber membranes before and after thermal oxidation, showing that thermal oxidation causes a large amount of fiber

melting and adhesion; (d) and (e) SEM images of GO-doped PAN nanofiber membranes before and after thermal oxidation, revealing that GO doping effectively inhibits fiber melting; (f) and (g) TEM images of GO-PAN composite fibers before and after thermal oxidation, where the separation at the fiber intersection nodes changes to local "welding" connection (indicated by the yellow circle), indicating that GO has induced the formation of cross-linking structures between the fibers.

The new structural design represented by multi-functional nanocomposite fiber membranes is also in the advancement stage. Just like Geng et al., they combined PAN nanofibers with SiO₂ nanoparticles and utilized the sol-gel reaction to generate a silica coating on the surface of the fibers, endowing the fibers with flame-retardant and heat-resistant properties [6]. Additionally, by introducing conductive nanomaterials such as carbon nanotubes (CNT) and MXene into the PAN matrix, a composite fiber membrane with both electromagnetic shielding and photothermal conversion functions can be fabricated. These functional modifications have expanded the application scope of graphene/PAN composite membranes [7].

4. Properties and Applications of Graphene/PAN Composite Nanofiber Membranes

4.1 In Terms of Filtration Efficiency and Pressure Drop Performance

The graphene/PAN composite nanofiber membranes exhibit excellent performance in the filtration of airborne particulates. Multiple research results indicate that this type of composite membrane can generally achieve a retention efficiency of over 99% for submicron particles [2]. For instance, the PMIA/PAN/SiO₂ multi-component nanofiber membrane prepared by Geng et al. achieved an efficiency of up to 99.69% in the 0.3 μm particle filtration test, and Wei et al. reported that the O-PASS nanofiber filter membrane achieved a removal rate of 99.98% under PM_{2.5} conditions [8]. Although the efficiency is very high, the air resistance of these nanofiber membranes remains very low. For example, the O-PASS filter membrane, when ensuring a nearly 100% retention rate, has a pressure drop of only about 68 Pa [8]. The corresponding quality factor is far superior to the performance indicators of traditional microfiber

filter paper. This is mainly because the nanofiber membranes have uniform pore size distribution and good connectivity, allowing the airflow to penetrate with low resistance, and the addition of graphene does not cause a large blockage of the pores, and can also assist in capturing particles through electrostatic adsorption and other effects, achieving the ideal filtering characteristics of high efficiency and low resistance.

4.2 Mechanical Strength and Durability: Most Thermoplastic Polymer Fiber Membranes

Such as pure PAN, often have insufficient mechanical strength. Under long-term airflow impact or high wind speed conditions, they may undergo deformation or damage. After the introduction of graphene, the mechanical properties of the composite membrane have been improved. On one hand, graphene sheets have high modulus and high strength, directly enhancing the fiber strength. On the other hand, the strong interface formed between graphene and the polymer matrix plays a role in nanoscale improvement and stress transfer, making the fiber network more stable. Taking the O-PASS filter membrane as an example [8], simple oxidation treatment did not significantly change the fiber diameter and morphology, but it increased the membrane's tensile strength from 1.51 MPa to 4.92 MPa, an increase of 226%, as shown in Figure 2, which presents the comparison of this mechanical improvement effect.

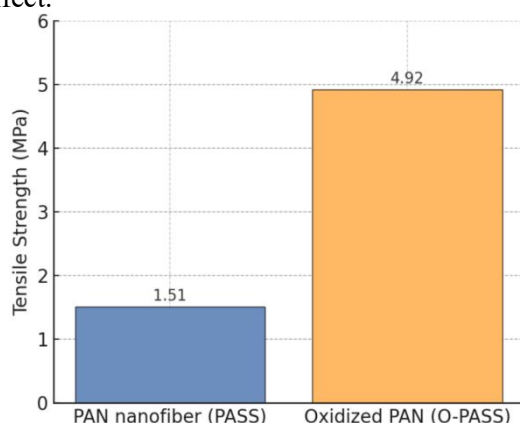


Figure 2. Comparison of Tensile Strength of PAN Nanofiber Membranes Before and After Oxidation Treatment

The oxidation polymerization reaction (such as introducing covalent bonds or cross-linking in PAN) significantly enhanced the mechanical strength of the membrane, which is beneficial for the graphene/PAN composite membrane to

maintain structural integrity under high wind speed, vibration and other working conditions.

The graphene/PAN composite membrane exhibits excellent durability stability under harsh environmental conditions. It possesses properties such as high temperature resistance, resistance to humidity and heat, and corrosion resistance. References [3,8] provide examples. For instance, the GO-OPAN composite membrane maintained its fiber structure and filtration performance even after undergoing 280°C high-temperature thermal oxidation [3]. When exposed to corrosive aerosols such as acid and alkali fumes for 30 days, the structure and ion conductivity of the composite membrane showed almost no change [4]. The antioxidant shielding effect of graphene and the "node welding" structure formed between graphene and PAN are regarded as the key mechanisms for improving the thermal resistance and flame retardancy of the membrane [3]. In contrast, pure polymer nanofiber membranes are prone to melting collapse when approaching their melting point or thermal oxidation temperature, and their filtration efficiency also drops sharply.

1. Multi-functional characteristics: By leveraging the functional properties of graphene, the composite membrane also exhibits additional functions that traditional filter materials cannot possess. For instance, graphene endows the PAN membrane with certain conductive properties, which can alleviate the attenuation of fiber electrostatic charges, thereby maintaining a long-term stable electrostatic dust collection capability; the functional groups (hydroxyl, carboxyl, etc.) on the graphene surface enhance the membrane's hydrophilicity, facilitating the realization of super-hydrophilic/water-repellent surfaces in oil mist filtration, effectively separating oil-water mixtures [6]. In practical research, Geng et al.'s composite membrane not only can efficiently remove particulate matter, but also can achieve a separation efficiency of >99.6% for emulsified oil/water systems [6], demonstrating the potential of "one membrane for multiple uses". Additionally, other nanomaterials can also be co-combined with graphene to confer special functions to the membrane: Liu et al. grew MOF crystal ZIF-8 and alginate-like ZnIn₂S₄ photocatalytic nanosheets on PAN nanofibers to construct a self-cleaning anti-fouling composite membrane, which has high-throughput separation ability for emulsified oil wastewater, and can also

photodegrade the attached organic pollutants, demonstrating excellent anti-pollution and self-cleaning properties. These achievements indicate that the graphene/PAN composite nanofiber membrane, as a platform material, can be optimized in structure and composition to meet the multiple performance requirements of filter materials in different application scenarios.

5. Argumentation Analysis

In conclusion, the graphene/polyacrylonitrile composite nanofiber membrane demonstrates significant advantages in the field of air particulate matter filtration. The results of the literature review clearly support the following argument:

The addition of graphene has effectively improved the insufficient performance of traditional electrospun nanofiber membranes. For polymer fiber membranes like pure PAN, although they have high filtration efficiency, their mechanical properties and heat resistance are lacking. As a nanoscale reinforcing phase, graphene sheets, relying on their extremely high strength and stability, form a stable interface with the polymer matrix, thereby enhancing the strength and environmental tolerance of the composite membrane [8]. Experimental data show that whether using a simple chemical modification method or directly doping graphene nanosheets, the mechanical properties of the fiber membranes can be significantly improved, and their stability in high-temperature environments or chemical media can also be enhanced [3,8]. This strongly indicates that graphene, as a functional filler, is effective in improving polymer filter membranes.

The membrane composed of graphene and PAN achieves a balance between high efficiency and low resistance in terms of filtration performance. According to traditional beliefs, increasing filtration efficiency usually requires sacrificing resistance as a cost [8]. However, the composite nanofiber membrane has broken through this limitation. For instance, in a series of research reports, membranes based on graphene and PAN with filtration efficiency approaching 100% were mentioned, and their pressure drop remained at a relatively low level of several tens of pascals. This is due to the open network structure of the nanofibers and the possible electrostatic capture effect brought by graphene. The graphene layers can enhance the interception of sub-micron particles, and the

interception mechanism includes both physical obstruction and charge adsorption. This improves filtration efficiency without significantly sacrificing the porosity, and this efficient and low-resistance characteristic is of great significance for air filtration applications, confirming the feasibility and superiority of the graphene composite strategy.

Through the multi-functional composite design, the graphene/PAN membrane can achieve a performance combination that traditional filter materials cannot simultaneously achieve. For example, when the working conditions require both high-temperature resistance and flame retardancy, although using high-temperature-resistant engineering plastics is feasible, it is quite costly. However, the pre-oxidation treatment of PAN induced by graphene provides a low-cost path to achieve high-temperature filter materials [3]. For instance, for complex pollution conditions such as oil mist and organic aerosols, the graphene composite membrane can combine hydrophilic nanoparticles or photocatalysts to achieve the synergy of particle filtration and organic pollutant degradation [9]. These innovative research ideas show that by reasonably selecting the second-phase material and leveraging the functional diversity of graphene, the performance boundaries of PAN-based nanofiber membranes have been greatly expanded, making it feasible to meet special application requirements.

However, there are still some challenges and problems in this field that need to be studied and solved, such as how to uniformly disperse graphene in the polymer matrix and prevent agglomeration at high loads. This is one of the key difficulties in preparing large-area homogeneous composite membranes. Excessive graphene may increase the viscosity of the spinning solution and make it unstable, affecting fiber formation and the pore structure of the membrane. This needs to be paid special attention to in subsequent large-scale production. At the same time, the understanding of the mechanism by which graphene enhances performance also needs to be strengthened. Currently, most of the literature results are limited to performance characterization and lack systematic research on the interaction between graphene and PAN at the interface, as well as the influence of the charged properties on the filtration mechanism. By combining molecular dynamics simulation and experimental testing,

clarifying the quantitative relationship between the distribution morphology of graphene in fibers and filtration performance, mechanical properties, etc., will be able to guide the rational design of composite membranes. Long-term service reliability is also a factor to be considered in evaluating new filtration membranes.

Although graphene/PAN membranes perform well under laboratory conditions, in actual dusty air continuous impact, whether graphene sheets will fall off, whether the membrane resistance and efficiency change over time, etc., need to be verified by accelerated aging and field tests.

A large amount of literature evidence indicates that graphene/PAN composite nanofiber membranes have great potential in efficient air filtration. This composite nanofiber membrane successfully combines the performance advantages of graphene, a new type of nanomaterial, with the mature electrospinning technology, and produces a new type of filter membrane material. This material demonstrates outstanding performance in terms of filtration efficiency, pressure drop, mechanical strength, and environmental tolerance. In the future, as the understanding of the relationship between material structure and performance deepens and the process is continuously improved, graphene/PAN composite nanofiber membranes are expected to be widely applied in air pollution prevention and control, industrial dust treatment, and personal protection, contributing to the realization of clean air and sustainable development.

6. Conclusion

Through a review and analysis of multiple relevant domestic and foreign literature, the following main conclusions were obtained:

(1) Efficient filtration with low resistance and the following features are presented: The graphene/PAN composite nanofiber membrane, while maintaining a particle retention efficiency of over 99%, can also achieve a pressure drop of less than 100 pascals. Its quality factor far exceeds that of traditional filter materials [2]. This outstanding performance is attributed to the ideal balance achieved between the nanoporous structure of the nanofiber and the aerodynamic properties.

(2) Mechanical properties have been enhanced: Graphene has a nanoscale enhancing effect, which significantly improves the mechanical

strength of the composite membrane compared to the pure polymer fiber membrane. The fiber network becomes more stable. Compared with the unmodified PAN membrane, the chemical oxidation modified composite membrane has increased its tensile strength by several times, and can maintain stability under high temperature, corrosion and other conditions. This stability has been mentioned in relevant studies [3, 6] and literature [8].

(3) Excellent environmental tolerance: All the shielding and protective effects of graphene enable the composite membrane to possess excellent high-temperature resistance, antioxidant properties, and chemical corrosion resistance. References [3, 6] indicate that the addition of graphene delays or avoids the structural degradation of PAN fibers in high-temperature environments or strong oxidizing conditions. Compared to pure PAN membranes, the composite membrane can maintain its filtration performance for a longer period of time and can be applied in special scenarios such as high-temperature waste gas purification.

(4) There is considerable potential for functional expansion: By introducing other functional nanomaterials or performing functional treatment on graphene, the composite membrane can achieve multiple functions beyond particle filtration, such as imparting antibacterial, anti-fouling, self-cleaning, and electromagnetic shielding properties. This meets the application requirements beyond air purification, providing a broad space for the research and development of new high-performance and multi-functional filter membrane materials.

In conclusion, the graphene and polyacrylonitrile composite nanofiber membranes have been experimentally proven to have significant advantages in the field of efficient filtration materials. In the future, it is necessary to strengthen the research on the process scaling-up and long-term reliability of their preparation, deeply analyze the mechanism of the synergistic improvement of graphene, continuously optimize the material formula and structural design. It can be foreseen that with the advancement of related research and the implementation of practical application demonstrations, graphene and PAN composite nanofiber membranes are expected to become the next generation of high-performance air filtration materials, playing a key role in environmental purification and human health

protection.

However, in the current practical applications of graphene/polyacrylonitrile (PAN) composite nanofiber membranes, there are still some technical challenges that need to be deeply studied and resolved. Firstly, as the amount of graphene added increases, the uniformity of its dispersion in the PAN matrix is often difficult to be effectively controlled. This leads to the tendency for graphene layers to aggregate or pile up during the preparation of the spinning solution. This aggregation phenomenon results in an uneven final nanofiber structure, manifested as an expanded range of fiber diameters within the membrane, uneven pore sizes, and possible dense block formation in certain local areas. This non-uniform structure directly reduces the overall capture efficiency of the membrane for particles and increases the air flow resistance during the filtration process, thereby significantly deteriorating the overall filtration performance and energy consumption of the membrane [6].

Secondly, the stability and durability of graphene/PAN composite nanofiber membranes in long-term operation or continuous exposure to real dusty air environments also have certain uncertainties and potential risks. As the usage time increases or the concentration of particles in the exposure environment rises, the microscopic structure of the composite membrane may gradually age, with graphene layers gradually detaching from the fiber surface or separating, and the connections between the fiber networks gradually weakening or loosening. This microstructural change not only leads to a decrease in the interception efficiency of particles by the membrane but also causes a significant attenuation in the mechanical strength of the membrane, thereby seriously affecting its reliability and safety in actual industrial application scenarios [8].

In conclusion, although graphene/PAN composite nanofiber membranes demonstrate significant performance advantages in the field of efficient air filtration, to truly achieve their large-scale practical application, in-depth and systematic research is still needed on the control of graphene dispersion and the reliability of long-term operation, as well as further optimizing the material preparation process and improving the understanding and control of the stability and performance degradation mechanism of the membrane structure, in order

to ensure that this type of composite membrane material can be applied in actual environmental governance and air purification fields in a long-term, stable, and reliable manner.

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