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A Review of the Current Research Status of Graphene for the Removal of Microplastics and Antibiotics from Water

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ABSTRACT

As highly representative pollutants among emerging pollutants, antibiotics and microplastics are characterized by strong bioaccumulation, difficult degradation, and high toxicity, and along with the increasing use, the crisis of synergy between the two is increasing. Graphene, a material with high specific surface area and high reactivity, has a wide range of application prospects in the field of water treatment, especially graphene oxide and graphene composites make the application of this material has a certain diversity. The research on graphene and its composites in water membrane treatment and catalytic degradation for the removal of microplastics and antibiotics in recent years is reviewed to explore the research trends and hotspots, and to explore the research prospects.

KEYWORDS

Graphene; antibiotics; microplastics; membrane; catalytic.

1. INTRODUCTION

In the context of the global technological era, human beings have not only invented new technologies, but also brought new pollutants and new challenges to the environment. With the modernization and large-scale development of animal husbandry, antibiotics, as a class of drugs that can prevent and treat animal diseases, have been widely used globally due to their effective, inexpensive, and spectral antimicrobial properties, especially in China, where antibiotic misuse has become the norm and the problem of exceeding the detection limits in the aquatic environment is serious[1]. Microplastics (MPs), on the other hand, have become a focal point of environmental issues because of their diverse sources, their popularity in life, their prevalence in the environment, and their strong adsorptive properties that can easily adsorb other pollutants and produce synergistic effects[2]. At the same time, both pollutants are highly bioaccumulative, difficult to degrade, and capable of affecting bioenergetic metabolism and gene expression, which have attracted much international attention[3].

There have been many studies on the removal of antibiotics and microplastics in the aqueous environment[4-6]. Nanotechnology is considered to be the most promising technology for water purification, in which carbon nanomaterials are one of the most concerned research areas[7]. Carbon nanomaterials represented by graphene also show excellent application prospects. Graphene is a honeycomb monolayer two-dimensional planar structure formed by carbon atoms in the form of sp2 hybridization, especially graphene oxide (GO), a monolithic dibasic nanostructure supported by the oxidation of natural graphene, which is rich in the π-π electronic system and rich in oxygen-containing functional groups[8]. Due to its huge specific surface area, strong adsorption capacity, and low biotoxicity, it is able to be combined with a variety of traditional methods (e.g., advanced oxidation, membrane separation technology, adsorption technology, etc.) for the removal of two kinds of pollutants, and is highly recyclable, so it has a wide range of prospects for application in the field of water treatment, and has become a hotspot of attention in the industry[9]. Graphene-based materials technology has a better application prospect, but nowadays only fewer reviews and researches focus on the unified removal of two types of pollutants, graphene-based materials technology has a greater hope for the unified removal of composite pollutants of microplastics and antibiotics, and most of the removal technologies for such pollutants are mostly focused on adsorption treatment, with less attention paid to the prospects of the application of membrane treatment and catalytic degradation, therefore The purpose of this paper is to summarize and review the membrane treatment and catalytic degradation of graphene and graphene composites for the removal of microplastics and antibiotics in the aqueous environment over the past few years, which is of great significance to provide certain ideas and theoretical support for this field.

2. MEMBRANE TREATMENT TECHNOLOGY FOR POLLUTANTS

Graphene itself has certain flexibility and is one of the materials with the highest mechanical strength known today, which enables it to be better integrated with membrane technology. The adsorption effect of adsorption treatment is easily affected by factors such as environmental pH, and the conditions are more stringent. In contrast, membrane technology has demonstrated its advantages in this respect. Its physical and chemical properties are relatively stable, less affected by the environment, and it has higher recycling potential and reusability. At the same time, due to the advantages of low cost and extremely high removal efficiency, there are many studies on the combination of graphene and membrane technology. However, if the GO membrane without any surface modification material is not introduced, it will easily cause secondary pollution. Therefore, many scholars have modified and modified the super-hydrophilic GO and achieved good removal effect.

Table 1 summarizes the removal efficiency of various membrane technologies for different types of pollutants. The data show that the removal efficiency of most membrane technologies for microplastics can be more than 95 %, which is an extremely excellent removal effect. It shows the superiority of membrane technology in the removal of microplastics. At the same time, many studies have found that the membrane has a good removal effect on microplastics in industrial wastewater[10 - 11], and its environmental protection and recyclability are also excellent. This largely ensures the application of membrane technology in the field of water treatment, indicating its excellent application prospects.

Zou et al. found that GO has a certain inhibitory effect on microbial uptake of antibiotics. GO and sulfamethoxazole (SMZ) can form a complex, which also reduces the uptake of SMZ by bacteria, thereby controlling the spread and pollution of antibiotic resistance genes[12].Therefore, GO has a wide application prospect in the direction of antibiotic removal. Nowadays, the field of membrane technology treatment of antibiotics is mostly aimed at the modification and preparation of GO, and shows its extremely effective removal effect in Table 2, with a retention rate of up to 100 %. In other studies, the removal efficiency of antibiotics is also above 70 %. Especially in the study of Gao et al., the membrane allows a large number of required nutrients to pass through[13], an effect that has not been paid attention to or cannot be achieved in the study of other graphene membranes, which provides a great possibility for its practical application.

3. CATALYTIC DEGRADATION OF POLLUTANTS

In addition to the adsorption treatment and membrane treatment technologies that have been studied more, emerging pollutants can also be effectively decomposed or destroyed into non-toxic small molecule substances under sunlight, visible light or ultraviolet(UV)irradiation. As a promising catalyst, graphene has excellent conductivity in terms of storage and rapid electron transport due to its uniform dispersion of large specific surface area, narrow band gap energy and excellent conductivity. Now, some studies have explored its effect in the removal direction of microplastics and antibiotics.

Table 1 summarizes different photocatalytic technologies based on graphene materials. The results show that this technology can achieve a certain removal effect of microplastics in water, and individual technologies can also be efficiently removed, but the overall removal effect is not ideal. In addition, some scholars have studied the environmental toxicity of graphene-based nanocomposites as photocatalysts. In the study of El-Wakeil et al., the new material has less microbial toxicity to humans and the environment. It is a hybrid nanocomposite that is safe for humans and the environment.

In Table 2, we summarize the catalytic degradation efficiency of various graphene-based and graphene-based nanocomposites. The results show that different treatment technologies have different removal effects on different types of antibiotics. In the study of Kang et al. and Jiang et al., the catalytic degradation efficiency of nickel nanoparticles doped porous graphene (Ni @ NPG) and graphene-modified electro-Fenton catalytic membrane is excellent[14 - 15], but the degradation effect of other materials is not ideal. Because this kind of technology is extremely friendly to the environment, it is not easy to produce secondary pollution, and most of the photocatalytic degradation under ultraviolet irradiation is simple. It shows its good application prospect, so this kind of technology still has great potential.

Table 1 Removal of microplastics

Materials and methods	Processing principle	Pollutants	Removal effect	References
GO-PVA-based composite membrane	membrane	HDPE-MP	95%	Dey et al. [16]
Holey rGO-based membranes	membrane	MPs	99.9%	Yang et al. [11]
Graphene oxide tuned by laser bombardment	membrane	10μm polyvinyl chloride	99%	Sun et al. [17]
A covalent adaptable network based graphene in water-remediating membranes	membrane	MPs	97%	Sen et al. [18]
composite membrane rGO/PAN	membrane	MPs	82%	Fryczkowska et al. [10]
GO-CuO2、GO-MnO and GO-TiO2	Photocatalytic degradation	Polyethylene granules	48.06%、39.54% and 50.46%	I. Uogintė et al.[19]
Ag/TiO2/rGO photocatalyst	Photocatalytic degradation	MPs	76%	Fadli et al. [20]
a novel HNCP based on rGO@SiO2.Fe3O4	Photocatalytic degradation	terephthalate microplastic	64%	El-Wakeil et al. [21]

Table 2 Removal of antibiotics

Materials and methods	Processing principle	Pollutants	Removal effect	References
GO-MPD10.5 and GO/1GO-MPD0.5 membrane	membrane	ciprofloxacin	93.35 ± 3.62% and 95.48 ± 2.97%	Zhou et al. [22]
		ofloxacin	85.89 ± 6.52 and 88.21 ± 3.67%	
GO/CNC membrane	membrane	sulfamethoxazole	74.8％	Gao et al. [13]
		levofloxacin	90.9％	
		norfloxacin	97.2％	
GO-CMC film	membrane	oxytetracycline	102.05mg/g(the maximum adsorption capacity）	Juengcharoenpoon et al. [23]
		oxolinicacid	256.68mg/g(the maximum adsorption capacity)	
		trimethoprim	370.93mg/g(the maximum adsorption capacity)	
PS-DVB membrane	membrane	tetracycline	22.79mg/g(the maximum adsorption capacity) retention rate reached 100%.	Hu et al. [24]
		ciprofloxacin	46.69mg/g(the maximum adsorption capacity) retention rate reached 100%.	
Graphene sponge electrodes doped with atomic boron and nitrogen	Electrochemical degradation	sulfamethoxazole, trimethoprim, ofloxacin, and erythromycin	79%-98% in 10 mM phosphate buffer and 46%-85% in tap water.	Ormeno-Cano et al. [25]
Ni@NPG catalysts	catalytic removals	sulfachloropyridazine	100% SCP removal from water in 30 min	Kang et al. [15]
N-TiO2/graphene layered materials	Photocatalytic degradation	norfloxacin, sulfamethazine, oxytetracycline and chlortetracycline	Removal efficiency greater than 45%	Zhao et al. [26]
Graphene modified electro-Fenton catalytic membrane	Photocatalytic degradation	in-situ	Removal efficiency of 90%	Jiang et al.[14]

4. CONCLUSION

Graphene has shown unprecedented potential and feasibility in removing pollutants in water environment due to its huge specific surface area, strong adsorption capacity and easy modification. The research of modified graphene and three-dimensional graphene also makes the research results in this direction more diversified and practical. However, most studies now focus on the single removal effect and adsorption treatment of microplastics and antibiotics by graphene materials. There is a lack of relevant research on the composite pollutants and co-removal of the two, and there is a lack of diversity in treatment technology. In addition, in terms of evaluating the removal effect of graphene materials, the laboratory mostly uses simulated microplastics and simulated water containing antibiotics to conduct experiments, but does not evaluate the effect of graphene on the removal of two types of pollutants in the actual water environment according to the actual water body, resulting in the actual application. The treatment effect is not ideal. In addition, there are relatively few studies on the synergistic effect and mechanism of microplastics and antibiotics. In summary, further research is needed in the future, and the development of this work also has important theoretical guidance and practical significance.

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