Research Progress of Microplastics Removal Technology in Water Environment

Yafei Wen^{*}, Weilin Guo^a

*2438107096@qq.com; ^achm_guowl@ujn.edu.cn

School of Water Conservancy and-Environment, University of Jinan, Jinan, Shandong province,250022, China

Abstract. Recently, the extensive origins, inadequate control, and challenging decomposition of microplastics (MPs) have posed a significant risk to both ecosystems and people health. Continuous advancement of various technologies is crucial for the efficient eradication of MPs pollution from the environment. Initially, a concise overview was provided in this review about the variety and origins of MPs found in aquatic environments. Afterwards, the effectiveness of diverse water treatment technologies in eliminating MPs was explored, including methods such as sorption, membrane separation, electrocoagulation, photocatalysis, peroxide oxidation, and the use of microorganisms or enzymes. This review establishes a scientific foundation for efficient elimination of MPs from water, offering valuable insights into effective removal techniques.

Keywords: MPs; Water environment; Harm; Removal technology

1 Introduction

Microplastics (MPs) are tiny pieces of plastic that can be found in the air, soil, and water. They are so small that their diameters range from 1 nm to 5 mm. In fact, research has shown that these particles have even been discovered in sediments from freshwater lakes in the Arctic region [1]. They may come from the degradation and weathering of larger plastic products (secondary MPs) or from materials of an original smaller size at the time of initial manufacture (primary MPs) [2]. There are different types of MPs that encompass polyethylene (PE), polypropylene (PP), polystyrene (PS), polyurethane (PU), polyvinyl alcohol (PVA), and more. According to research, the annual global production of plastic amounts to 300 million tons, with 13 million tons finding their way into rivers and oceans. If this trend persists, the amount of microplastics in water bodies will reach 250 million tons by 2050. Currently, the pollution caused by MPs is considered a highly alarming global environmental issue. Given a wide variety of sources available to MPs, they often traverse various avenues to negatively impact ecosystems and human well-being. Plastic waste in human daily life undergoes decomposition by microorganisms present in leachate, subsequently getting released into the groundwater within urban areas. Furthermore, microplastics generated by the plastic manufacturing sector and automobiles disperse in the air and subsequently settle into bodies of water via atmospheric deposition [3].

MPs in aquatic environments pose a grave danger to marine life due to their ability to migrate and transform. The level of threat is directly tied to the concentration of harmful chemicals

released from plastic additives and absorbed pollutants like metals, pesticides, and persistent organic pollutants [4]. MPs not only pose a threat to Marine life by acting as a potential reservoir for pathogen transmission in Marine habitats, but their impact on human health remains unclear. Marine organisms occupy a central position in the food chain and serve as a crucial source of nutrition in the daily human diet, thereby presenting a serious risk to human well-being. Commonly, MPs initially penetrate an organism gastrointestinal tract via ingestion and subsequently amass in various body organs via metabolic processes, leading to the generation of harmful consequences. MPs disrupt the balance between beneficial bacteria and harmful bacteria, resulting in decreased intestinal mucus secretion and an imbalance in the gut microbiota. This will also lead to a range of abnormal responses, including disrupted metabolism, inflammation, and impaired immune regulation. MPs can simultaneously induce toxicity in the reproductive system by impeding gamete binding and interfering with the fluidity of the gametes plasma membrane. Enveloping the embryo surface, causing a lack of oxygen for the embryo, thereby impacting the uptake of nutrients, leading to irregular growth and development of the offspring, as well as metabolic imbalances. Hence, it is crucial to research the effective elimination of MPs in water settings, while examining the pros and cons of various treatment methods and their workings. This will help lower the presence of MPs in water and prevent the negative impact they have on natural aquatic ecosystems [1].

In this study, the types and sources of MPs in water environment were briefly summarized, and then the physical, chemical and biological methods of MPs removal were reviewed and compared. Finally, the future development direction and trend of these methods were summarized and respected. This review aims to provide some technical reference and guidance for reducing the water environmental effects of MPs, in order to provide reference and reference for relevant researchers.

2 Types and Sources of MPs

MPs can be divided into primary MPs and secondary MPs (Table 1). Primary MPs are plastic particles that are released into the water environment in the form of small fragments via rivers, sewage treatment plants, and so on. These particles are discharged directly and contribute to pollution in water bodies. Numerous personal care products, including toiletries, cosmetics, detergents, and similar items, contain MPs. Plastic particles are manufactured and employed for specific purposes. Examples of such damage may arise from deterioration incurred during the production, utilization, or upkeep of sizable plastic goods, including the erosion of tires from driving or the wearing down of synthetic fabrics during laundering. Secondary MPs are formed from larger plastic particles through various natural processes, such as corrosion, aging, wear, and oxidative degradation, leading to the production of minuscule plastic fragments. Photodegradation and other weathering processes, like exposure to sunlight and natural decay, are responsible for degradation, which can occur in various scenarios such as agricultural insulation films or industrial composting [5]. Statistics suggest that a significant portion, approximately 80%, of the plastic waste found in oceans originates from terrestrial sources globally. These waste materials undergo substantial degradation to form secondary MPs.

MPs of various sizes are present in coastal regions and aquatic ecosystems worldwide, as they can disperse extensively through wind and oceanic flows. MPs originate primarily from the fragmentation or deterioration of plastic and the natural degradation and oxidation of plastic waste in the environment. Furthermore, as a result of improper disposal and haphazard discarding, plastic fragments are ultimately deposited within the soil and subsequently transform into MPs. It is necessary to implement measures to mitigate the discharge and disposal of plastic waste, including plastic particles found in cosmetics and cleaning products, raw materials used in plastic manufacturing, and plastic particles or powders utilized for air injection[6]. This is because MPs have the potential to cause harm to the water environment. The gradual fragmentation and UV degradation of larger plastic products under atmospheric conditions contribute to the secondary formation of MPs. The potential for different organisms to consume plastic waste is heightened, which can further negatively impact the ecosystem. Sewage treatment facilities are a crucial contributor to the discharge of MPs. After the treatment of wastewater, there is an effective removal of big plastic particles, allowing MPs to bypass the treatment system and accumulate in the aquatic environment. It should be emphasized that the ocean serves as a notable source of MPs due to the presence of multiple sewage treatment plants in close proximity.

Table 1. The classification, density, and primary sources of microplastics.

3 Different approaches and strategies for the removal of microplastics

3.1 Physical method

Due to its simple process, affordability, and high level of industrial utilization, the Physical method is extensively employed for removing MPs. The various methods comprise adsorption, membrane separation, electrocoagulation, flotation, screening, filtration, and other related techniques.

3.1.1. *Sorption.* **Sorption is a method widely used to remove MPs.**

The most popular adsorbents are mostly from natural environments, including activated carbon, zeolites, and biochar. These materials have strong affinity towards MPs, allowing them to effectively trap and remove the pollutants. The presence of hydrogen bonds and π - π interactions affects the overall structure and properties of the molecules involved. The specific mechanism by which biochar and activated carbon remove MPs in water depends on the surface properties of the particles and the adsorbent used. Steam-activated biochar (800 $^{\circ}$ C), which possesses increased porosity, has been identified as the optimal adsorbent for the efficient removal of MPs, owing to its high surface area and porosity [7]. Furthermore, when biochar and activated carbon are placed into the system to extract MPs When introducing biochar and activated carbon into the setup to eliminate microplastics, regarding their capabilities, they can also serve as filters.

3.1.2 Membrane separation.

Membrane separation technologies include membrane bioreactor (MBR), ultrafiltration (UF), reverse osmosis (RO), etc. During the treatment of MPs. The initial step in the process involves the bioreactor decomposing the MPs found within the sewage. In the sewage treatment process, the bioreactor effectively breaks down the MPs contained in the sewage and subsequently passes them through the porous barrier. Subsequently, the substance is driven through the membrane filter using a porous barrier typically allowing MPs to be captured and concentrated in the sludge within the membrane biological treatment system, due to the small pore size of approximately 0.1μm. The current treatment technologies have different removal efficiencies for MPs, but among them, the highest removal efficiency is observed. However, the expenses associated with its operation and membrane acquisition are excessively high, there has been an increasing recognition of its limitations in promoting its usage in wastewater treatment facilities, various techniques using biotechnology have been devised to address membrane pollution issues, which are becoming more commonly utilized due to their superior efficacy. It is required to make significant changes to the sentence without reducing its length and in line with the original sentence meaning: and more environmentally friendly than traditional physicochemical methods. Additionally, compared to conventional physicochemical approaches, it offers heightened environmental sustainability and preservation.

The most efficient and cost-effective process is ultrafiltration. This low-pressure membrane process effectively separates macromolecules weighing several thousand Daltons and small suspended particles of colloidal size from the solution. Polyethylene, the most prevalent polymer type among MPs, can be completely eliminated by an ultrafiltration membrane because of its significant size variations [8]. Ultrafiltration membranes are effective in removing polyethylene due to its particle size. Ultrafiltration and coagulation, when used together, have proven to be highly effective in eliminating 100% of MPs particles through ultrafiltration membrane treatment.

Reverse osmosis is an extensively utilized and highly prospective technique. The increased pressure in reverse osmosis membrane treatment can enhance its effectiveness; however, it has the potential to induce the formation of nano-plastic particles due to the crushing phenomenon. The removal efficiency of MPs through reverse osmosis (RO) is not high; however,

implementing a combined ultrafiltration (UF) and RO process can potentially enhance the removal efficiency of MPs.

3.1.3. Electrocoagulation.

Electrocoagulation is commonly employed in wastewater treatment facilities for the removal of MPs. By primarily discharging positively charged ions via metal electrodes, it functions as a coagulant, promoting the formation of clusters with suspended particles. The findings indicated that the elimination rate of MPs reached its peak at 98%. The pH 4 and a current density of 2 resulted in a 6% yield.88 mA/m [9]. Furthermore, the continual removal of residual mulching sludge, which rises to the surface after undergoing electrocoagulation, can be achieved through either overflow or skimming methods in an uninterrupted procedure.

3.2 Chemical method

The removal of MPs often involves chemical approaches such as photocatalysis and peroxide oxidation, which play a vital role in the process.

3.2.1. Photocatalysis.

Photocatalytic degradation of MPs involves a redox reaction, whereby light excites electrons within the low-energy zone of the photocatalyst (Fig.1). After the electron transition, valence electrons and holes generated by light in the catalyst give rise to the formation of superoxide radicals through their reaction with oxygen, while hydroxyl radicals are formed by the reaction of the light-generated holes with water molecules. The aforementioned active oxides have a notable effect on MPs, causing the breakdown of polymer chains and, in some cases, complete conversion into water and carbon dioxide through mineralization. Photocatalysis, being an eco-friendly technology utilized for the elimination of MPs, offers the benefits of affordability and sustainability. However, the challenges associated with regenerating the catalyst, limited selectivity, and low efficiency in degrading MPs hinder its widespread industrial implementation. The response of titanium dioxide catalyst to ultraviolet light is evident, whereas the activity of zinc oxide catalyst is higher in the presence of visible light [10].

The degradation of MPs through photocatalysis can be influenced by various factors such as the size and shape of the MPs, the pH of the solution, temperature, and other variables. As the size of MPs decreases, their contact area with the photocatalyst increases, leading to higher degradation efficiency. However, film-like MPs exhibit lower degradation efficiency. In an acid environment, the presence of H^+ encourages the incorporation of H^+ into the reaction system, thereby facilitating the interaction between the photocatalyst and MPs. Conversely, in an alkaline solution, the Coulomb repulsion effect hinders the degradation of MPs. Furthermore, exposure to frigid temperatures leads to the disruption of the outer layer of certain MPs, resulting in an augmented surface area and a subsequent enhancement in their interaction with the photocatalyst.

Fig.1. The photocatalytic degradation mechanism of microplastics on Zno-PT (a) and BiOCl-X (c) under visible light irradiation is depicted in the schematic diagram. The infrared spectrum of low-density polyethylene was analyzed after being exposed to ZnO and Zno-PT catalysts for a period of 175 hours (b). Through the utilization of photocatalysis, the pathway for the degradation of polyethylene sulfide microplastics (MPs PE-S (d)) has been determined [11][12].

3.2.2. Peroxide oxidation

Peroxide oxidation generally employs a catalyst to activate either persulfate or hydrogen peroxide (known as Fenton oxidation), leading to the formation of highly reactive oxidizing free radicals like sulfate or hydroxyl groups. These radicals are responsible for the degradation and mineralization of MPs. The peroxide oxidation process can rapidly degrade MPs, resulting in a higher degradation rate, and the resulting byproducts can serve as a source of carbon for promoting the growth of algae. However, the current focus of studies on the degradation of MPs mainly lies in laboratory research, and the efficacy of treating real-life polluted water remains restricted.

The degradation of MPs by peroxide oxidation is greatly influenced by variations in temperature and pH levels. High temperatures play a key role in breaking down MPs by increasing their molecular size and promoting the formation of free radicals, ultimately boosting the decomposition process. The efficiency of the advanced oxidation reaction is affected by the pH level, with an optimal pH range required for MPs degradation. In the case of the Fenton reaction, a higher pH level hampers the generation of hydroxyl radicals and leads to the creation of iron hydroxide precipitates, ultimately decreasing the system ability to undergo oxidation [13].

3.3 Biological method

The process of removing MPs using biological techniques is a comparatively new method. The application of biological methodologies for the removal of MPs is a relatively recent technique. Biodegradation is a fundamental aspect of utilizing biological techniques to facilitate the breakdown of MPs in aquatic environments through the introduction of microorganisms capable of degrading them. During the microbial degradation process, MPs serve as nourishment for the development of biofilms. As biofilm develops, it causes pitting and cracking in MPs, resulting in weakened structural integrity. Bacterial enzymes can target both specific and non-specific fragments of the degraded MPs during this process. Furthermore, certain types of plankton, including water fleas and mollusks, as well as aquatic plants, possess the capability to ingest and absorb MPs. These MPs can then be further eliminated through biological consumption [14].

Although previous research has concentrated on biodegradable MPs, it is crucial to overcome the obstacles at each stage of the process to enhance their biodegradability and establish a viable and effective treatment approach. The degradation of MPs by one type of bacteria often results in harmful substances that hinder the growth of microorganisms, but the collaboration between various microorganisms and enzymes can also impact the breakdown and utilization of MPs by microbial communities. Despite the excellent removal effect and environmental friendliness of biological methods, further research and exploration are required since the current studies are still at an early stage.

4 Conclusion and prospect

MPs have led to severe water pollution due to their widespread dispersion and challenging removal methods. MPS adverse effects on the environment and human health are a result of its high stability, capacity to break apart easily, and ability to adsorb various pollutants. Hence, selecting an appropriate technique to eliminate microplastics from water is imperative. There have been multiple research endeavors in the past few years that have put forth a range of techniques to eliminate MPs from water, such as adsorption, membrane separation, electrocoagulation, foam flotation, filtration, photocatalysis, peroxide oxidation, as well as employing microorganisms or enzymes. Despite the efficacy of several techniques in reducing the presence of MPs in aquatic ecosystems, there remain unresolved challenges. To effectively address the worldwide issue of pollution caused by MPs, the forthcoming research should focus on investigating the following key areas:

(1) Currently, the primary emphasis of research on the elimination of MPs lies in larger particle sizes, with limited investigation conducted on smaller particle sizes. However, further research is required to understand the additional study needed for small particle size MPs, as they possess a larger specific surface area and higher biological toxicity.

(2) There are still certain drawbacks in the application of current techniques for eliminating MPs, like the generation of harmful by-products through advanced oxidation and challenges related to membrane contamination.

(3) Currently, the main focus of the research on the removal of MPs lies within laboratory settings, indicating certain drawbacks when it comes to implementing these techniques in realworld scenarios. Moving forward, it is imperative to enhance pilot testing or conduct more extensive research to validate and reinforce the findings.

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