RSC Applied Polymers

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DOI: 10.1039/D6LP00029K

The Rise of Polymeric Hydrogels, Sponges, and Electrospun Fibers as adsorbents for Microplastics Removal: Prospects for Sustainable Future

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Abstract

Microplastics (MPs) are ubiquitous and recognized as a significant environmental contaminant, owing to their persistent accumulation in terrestrial and aquatic environment. Microplastic pollution is primarily driven by rapid industrialization, improper disposal, and poor plastic waste management practices, and is extremely harmful to the environment and living species. To overcome this issue, sustainable approaches and mitigation strategies are necessary for removing MPs from environment. In this regard, various approaches have been developed so far including

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adsorption which is considered highly efficient. So far, various types of adsorbents, including metal-organic frameworks (MOFs), inorganic nanoparticles-based composites, 2D nanomaterials, and polymeric materials have been explored for the removal of MPs. Among adsorption-based materials, polymeric hydrogels, sponges, and electrospun fibers have recently gained significant attention due to their high porosity, tunable surface properties, and excellent adsorption capacity. Additionally, biodegradable polymer-based materials offer the possibility of removing MPs without causing any adverse impact on the environment as they do not generate any toxic byproducts upon degradation after their purpose is served. However, several factors including material reusability, long-term stability, and capability to degrade MPs must be resolved for their better performance. Hence, in this review, we have comprehensively and critically highlighted the recent advancements in polymer-based hydrogels, sponges, and electrospun fibers for MPs removal. We also summarize the facts associated with the contamination caused by MPs and explore recent MPs removal techniques, including physical methods, chemical methods, and biological methods. In addition, we describe the management strategies that can help mitigating issues of MPs-based environmental pollution. Finally, we discuss the current challenges associated with these materials for facilitating MPs remediation in a more efficient, scalable, and environment-friendly way for sustainable future.

Keywords: Microplastics, Hydrogels, Electrospun fibers, Adsorption, Water purification, Sustainability.

1. Introduction

Plastic-based products are more convenient and used worldwide on mass scale for diverse applications including healthcare, agriculture, electronics, construction, transportation, and most

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significantly packaging.[1] The journey of plastic began with the fabrication of first semisynthetic plastic *Xylonite*, which was produced by Alexander Parkes in 1861.[2] Following this, several plastics such as polyvinyl chloride (PVC), polyethylene terephthalate (PET), cellophane, polyamide (PA), and polyethylene (PE) were developed. They are extensively utilized because of their low cost, durability, versatility, ease of production, and ability to be moulded into various products that are integral to daily life.[3]

With exponentially growing population and rapid urbanization, the global plastic production has been reported to reach 359 million tons in 2018.[4] This demonstrates a substantial leap of 1.2-fold from 299 million tons in the short time frame of 5 years.[5] Despite several benefits, this rapid expansion of plastic utilization combined with inadequate handling and improper disposal of plastic waste has led to significant environmental accumulation causing detrimental effects.[6] It has been anticipated that ~31.9 million metric tons of plastic waste is released into the environment every year due to poor waste management.[7] Further, this plastic waste undergoes fragmentation via physical abrasion, chemical corrosion, and photooxidation, resulting in the formation of microplastics (MPs) ranging from 1 µm to 5 mm in size.[8] MPs have been identified as one of the emerging pollutants in aquatic environment. It has been found that ~89% of the Northeast Atlantic Ocean, ~70% of Jade Bay South–North Sea, and ~95% of Arctic Polar water contains MPs which is a serious matter of concern.[9]

Based on their fabrication, MPs can be of two distinct types i.e., primary and secondary. Primary MPs are generally added into personal care products as microbeads.[10] This also includes marine coatings, wear and tear of tyres, and microfibers coming from household laundry, and factories.[11] On the contrary, secondary MPs are generated by fragmentation of larger plastics via various processes, including physical degradation, biodegradation, photodegradation, oxidation,

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and hydrolysis.[12-13] They may also arise from the plastic waste of fishing, products abandoned during tourism, plastic products used in agriculture sector, and plastic garbage thrown away by residents during their daily life.[14] MPs are more susceptible to be carried from soil to aquatic environment via runoff where they accumulate in sediments, and are further ingested by the aquatic organisms posing a significant threat to human health through the consumption of seafoods.[15-16] It is to be noted that MPs are cytotoxic, and capable of inducing oxidative stress and inflammation that may disrupt vascular endothelial cells, and hamper immune and neurological functions in humans.[17] They can enter the brain through the blood-brain barrier and can affect the discharge of essential neuroinflammatory transporters such as chemokine and cytokine.[18] The MPs of size ≤20 μm can access the brain through blood vessels and lymphatics. The entry of MPs into the brain may destroy neurons and result in neural dysfunction. The adverse effects of MPs in the brain may be related to the pathogenesis of neurodegenerative disorders, including Parkinson's disease and Alzheimer's disease.[19] Moreover, MPs can undergo aging process which alters their size,[20] surface area,[21] hydrophilicity,[22] color,[23] and thermal stability and crystallinity.[24] These transformations further enhance the concern of deeper infiltration of MPs in ecosystem.[25] Aged MPs have higher affinity to adsorb heavy metals,[26-27] hydrophobic organic pollutants (e.g., pesticides, pharmaceuticals, polycyclic aromatic hydrocarbons) and polychlorinated biphenyls,[28] from water, which further increases their toxicity.

To combat the issue of MPs pollution, several methods have been explored so far including biological processes, membrane filtration, electrocoagulation, and chemical methods such as coagulation, flotation, and adsorption.[29-30] The biological processes exhibit limited removal efficiency for MPs. Although membrane filtration offers a viable solution for capturing large sized MPs, yet its performance is limited for smaller MPs. These smaller MPs can easily clog the

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membrane pores and hinder filtration process.[29] Chemical methods such as coagulation, flotation, and adsorption are more commonly employed owing to their simple operation, cost-effectiveness, and low energy consumption. However, the use of chemical reagents in water treatment makes coagulation and flotation less environment-friendly. On the contrary, adsorption is considered as most advantageous method for MPs removal owing to low cost, straightforward operation, and possible recyclability of adsorbents.[30] Previously, several adsorbent materials including layered double oxides, biochar, coffee grounds, and magnetic particles have been explored.[31-33] However, these materials tend to be ineffective against smaller MPs or nanoplastics (NPs), and often face limitations like poor efficacy, prolonged adsorption duration, and low reusability which hinders their large-scale implementation.[27, 30] Compared to above-mentioned traditional adsorbents, the polymer-based materials possess interconnected porous architectures, tunable surface functionalities and pore size, improved hydrophilic/hydrophobic balance, enhanced mechanical flexibility, and large specific surface area that altogether enhances the capturing efficiency and reduces the fouling behaviour.[34-35] The controlled pore size and tailored chemical functionalities also improve the interaction of such materials with MPs, and hence provide a multifunctional platform for designing next-generation separation techniques.

In the given scenario, polymeric hydrogels, sponges, and electrospun fibers-based adsorbents have emerged as a promising platform to remove MPs from ecosystem. In recent years, a few review articles have been published covering various types of adsorbents, including metal-organic frameworks (MOFs), metal nanoparticles-based composites, and 2D nanomaterials for the removal of MPs.[30, 36-37] However, very limited attention has been given specifically to polymer-based adsorbents, including hydrogels, sponges, and electrospun fibers. Moreover, biodegradable polymer-based materials offer the possibility of removing MPs without creating any adverse

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impact on the environment. Unlike engineered nanomaterials, covalent organic frameworks, and metal organic frameworks, these biocompatible and biodegradable polymer-based materials do not produce toxic by-products upon degradation. Usually, such polymeric materials offer the absorption of MPs through physical interactions such as electrostatic interactions, dipole-dipole interactions, hydrogen bonding, and π-π stacking interactions. Consequently, they are being widely explored as environmentally benign alternatives for microplastic remediation. Hence, a comprehensive review dedicated to these polymeric materials and their use for the removal of MPs can provide critical insights into their design, adsorption mechanisms, and practical applications, thereby guiding future research and development in this field.

Herein, we described the impact of MPs contamination and provided a comprehensive overview of current advances in polymeric hydrogels, sponges, and electrospun fibers-based membranes for eliminating MPs from water. Further, we discuss various management strategies that could help mitigating detrimental effect of MPs. Lastly, we highlight the key challenges associated with these materials and emphasize the need for continued research to improve their performance for MPs remediation on a large scale.

2. Background and motivation to control MPs pollution

The use of plastic-based products has significantly risen over the last 50 years. The annual production of plastic was 322 million tons in the year 2015 and is expected to reach 670 million tons in 2040.[38] It may be noted that instead of high production and rising demand of plastic-derived products, only 9-10% of the plastic waste has been recycled, and most of the waste is discharged into natural water resources that can be deposited in landfills.[39] The exact size of MPs is still not defined by the scientific community, and the MPs can show a maximum length of 5 mm in the

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form of fibres, spheres, films, fragments and foams.[40-41] The shape of MPs depends upon their source of origin. For example, most of the fiber-shaped MPs are produced by the apparel industry, while film-shaped MPs are produced by packaging industry. Similarly, sphere-shaped MPs are generated by crushing media, waste generated by abrasives and resin pellet leakage from the transportation industry.[41]

It may be noted that the studies have shown that an individual absorbs 39,000 to 52,000 MPs every year.[42] The microparticles can penetrate the gastrointestinal tract and can cause an inflammatory effect.[43] The chronic exposure to MPs can destroy the immune system and increase the risk of cancer. Moreover, smaller-sized MPs possess the potential for bioaccumulation and can exacerbate health issues **(Fig. 1)**.[44-45] Therefore, MPs removal from different sources is very crucial to safeguard our environment. Nowadays, different techniques, including physical, chemical, and biological methods, are being used to remove the MPs. This review mainly focuses on the advances of Polymeric Hydrogels, Sponges, and Electrospun Fibers for the removal of MPs.

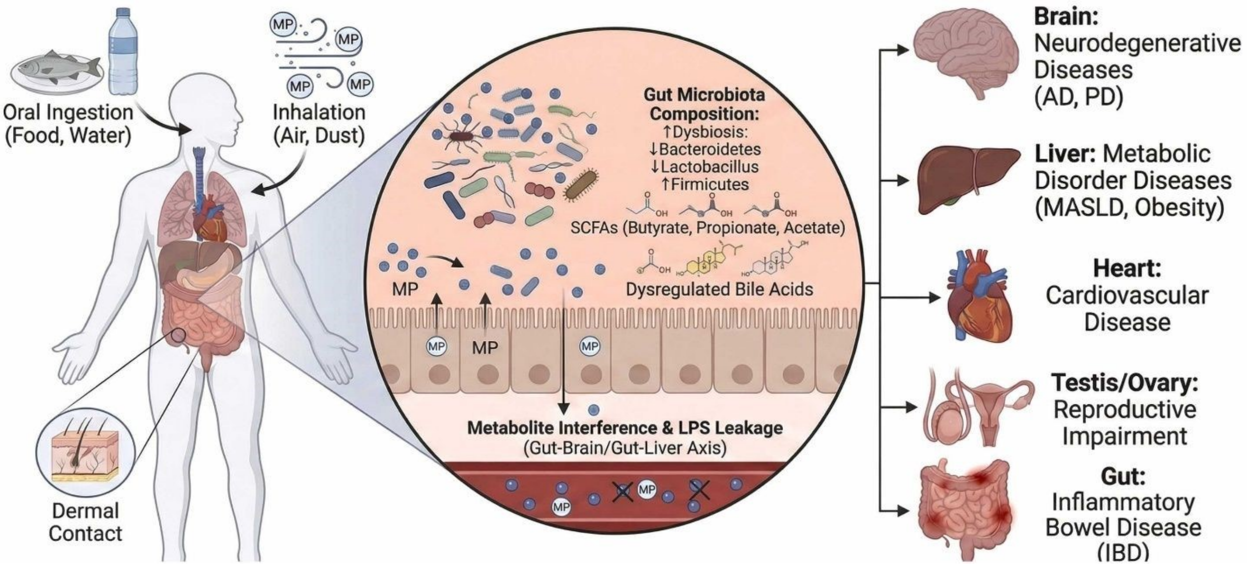

Fig. 1: Different pathways for human exposure to MPs and accumulation in distinct organs. Adapted with permission.[45] Copyright 2026, Publisher Elsevier.

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3. Adverse impact of contamination by MPs

Origin of MPs from different sources and their increasing amount in water has emerged as a critical problem, owing to their harmful effects on aquatic life and human health **(Fig. 2)**. It has been reported that over 250 aquatic species are affected by the ingestion and exposure to MPs.[46] It is evidenced by the presence of MPs in their digestive tracts, which leads to abnormal growth, structural and physical damage, disruption of normal feeding habits, and suppression of innate immunity.[47] Eventually, the consumption of such seafood by humans results in accumulation of MPs in their body. This has been recently indicated by the findings suggesting the occurrence of MPs in human stool, tissues, and body fluids.[48-52] Long-term exposure of MPs can also cause skin irritation, cytotoxicity, neurotoxicity, reproductive complications, Alzheimer disease, and chronic inflammation which increases the risk of cancer.[47-48] MPs can also affect cell metabolism in humans by altering the activity of gut microbiota, which reduces the absorption of nutrients in the gut.[53] Furthermore, MPs can also act as a vector for bacteria by providing substantial surface area for colony formation and proliferation.[54] This can result in tissue damage and increase the risk of infections.[54] Therefore, immediate intervention and development of novel technological alternatives is of prime importance for mitigating the issues related to MPs.

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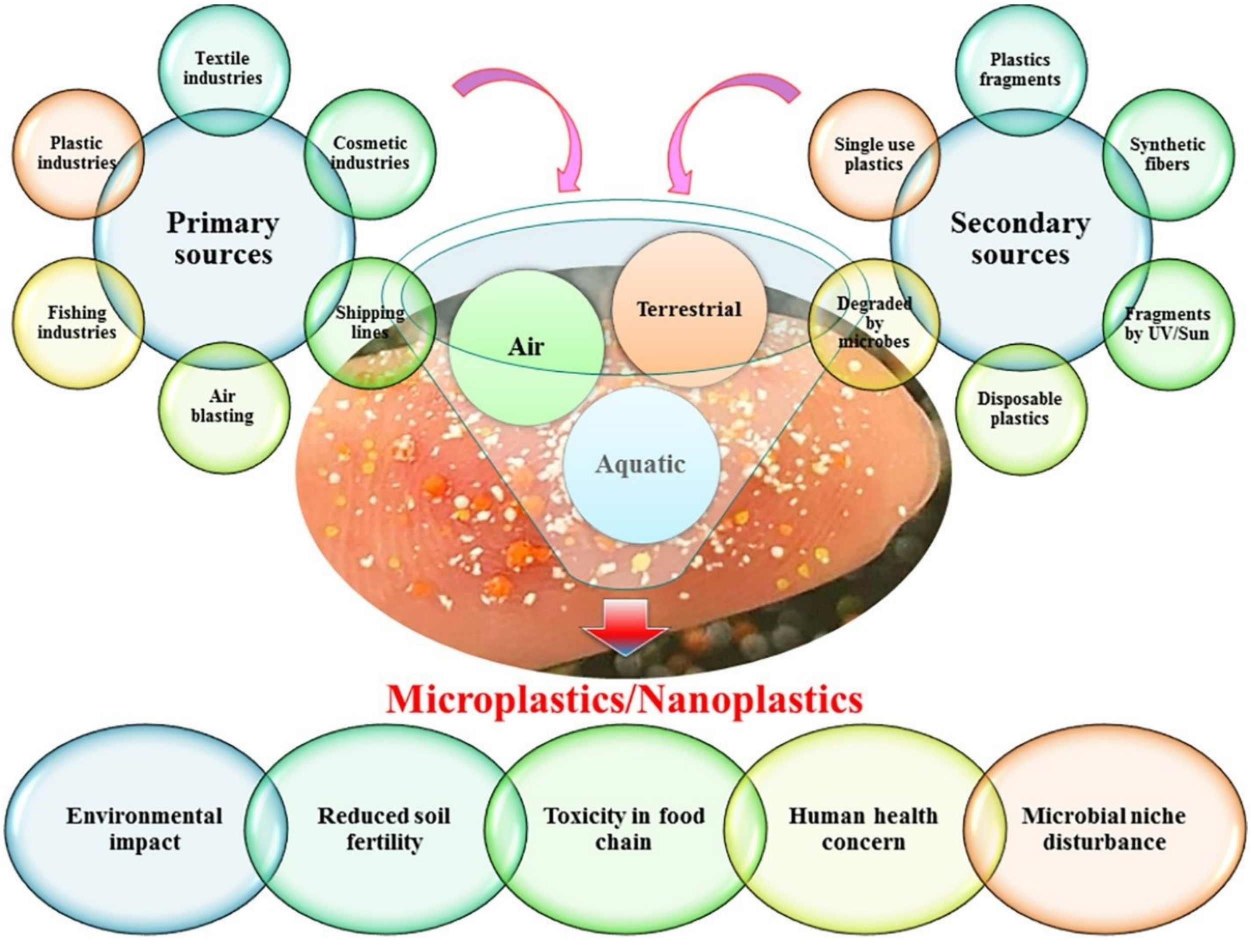

Fig. 2: Sources of MPs/NPs and their harmful impacts. Adapted with permission.[47] Copyright 2022, Publisher Elsevier.

4. Techniques for the removal of microplastics

In wastewater treatment plants, the most widely used methods may be physical, chemical, and biological methods. All these methods possess specific advantages as well as limitations for the recovery of microplastics from wastewater. A comparative analysis of these methods is summarized in **Table 1**.

4.1. Physical Methods

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The physical methods employed for the removal of MPs usually retain the chemical and biological properties of MPs. The commonly employed physical methods may include magnetic separation, froth flotation, filtration, adsorption, the oil film method, etc.

Magnetic separation is an effective method for the removal of MPs that uses a variety of materials, including carbon nanotubes, iron nanoparticles and magnetic seeds. The major controlling factors for magnetic separation may include hydrogen bonding, electric friction and complexation. The separation of MPs is possible due to a variety of removal mechanisms, including electrostatic interactions, hydrophobic interactions, π–π interactions, hydrogen bonding and complexation. Once magnetized, MPs can be effectively separated from water using an external magnetic field. Shi *et al.* reported the synthesis of modified maifanite through high temperature and acid treatment for the effective removal of MPs using the magnetic separation method. The modified maifanite demonstrated 98.46% removal efficiency of polystyrene under the influence of rerating magnetic field and recyclable upto 25 operational cycles.[55]

Similarly, the Froth flotation technique with ultrafine bubbles is an effective physical method for removing small MPs, including polylactic acid, polystyrene, polybutylene succinate, and polyethylene terephthalate from wastewater.[56] In this technique, there is selective adherence of bubbles to the required minerals, and the primary parameters required for froth flotation include surface wettability, hydrophilic interior, and hydrophobic outer surface tending to float in the form of froth aggregations.[57] A study carried out by Jiang *et al.* demonstrated that the efficacy of the froth flotation process for the removal of MPs can be enhanced by using cationic and anionic surfactants.[58]

Filtration of MPs can be carried out either by using a sand filter, disc filter or biochar filters. The suspended materials such as MPs can be removed with the help of rapid sand filters that can

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cling to the surface of sand grains. In wastewater treatment plants, A rapid sand filter may consist of a series of sand filters having distinct materials of different grain sizes. For example, Chabi *et al.* fabricated a rapid sand filter that can effectively remove MPs of size < 10 µm with 98% removal efficiency and 97% desorption rate. The mechanism involved in this removal method was the synergic combination of physicochemical sorption.[59] Similarly, the disc filtration method is a frequently used method for the physical separation of MPs. In this method, the MPs removal is mainly dependent on the creation of cakes of sludge inside the filter panels as well as retention of particles in filters.[60] Disc filtration technique can achieve 98% efficiency for the removal of MPs as indicated by the reported study.[61] However, the removal efficiency can vary depending upon the water quality, filter design, pore size and effectiveness of the disc filters. Biochar-based filters further enhance the removal efficiency of the filtration process. They consist of different media, including anthracite sand and granular activated carbon. Duan *et al.* designed an iron-modified magnetic biochar based on sawdust that depicted 205 mg/g adsorption capacity for the removal of polystyrene following Elovich kinetics and Sips isotherm model. The fabricated filter maintained 72% removal efficiency even after six cycles of pyrolysis.[62] The oil film method is a hydrophobicity-based removal technique that is very effective for the removal of MPs. Wang *et al.* developed a superhydrophobic and multifunctional polyurethane sponge using polydimethylsiloxane (PDMS) and stearic acid-modified TiO_2 nanoparticles. The adsorption capacity of the material reaches 0.45 g/g within 10 s. In another study, magnetic cobalt ferrite particles were able to remove 100% of microplastics through the oil film method. The material also demonstrated 2.56 g/g capture capacity and 98% removal efficiency after 10 adsorption-desorption cycles.[63]

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Adsorption is the most favorable technique to remove MPs. A variety of adsorbent materials, such as iron oxide, graphene oxide, carbon nanotubes, etc., are being employed for the efficient removal of microplastics. For example, a study carried out by Heo *et al.* employed magnetic iron oxide nanoparticles demonstrating 98.2% adsorption efficiency for the removal of polystyrene MPs.[64] Similarly, reduced graphene oxide nanosheets were capable of removing 99.9 % of MPs through an adsorption process.[65] In short, the physical methods provide a reliable and non-destructive approach for the removal of MPs. The removal efficiencies can be enhanced by creating superhydrophobic–magnetic sponges, magnetic–biochar composites, and surface-engineered nanoparticles.[66] Out of these physical methods, adsorption is considered as the most versatile, cost-effective and highly efficient technique.

4.2. Chemical Methods

Chemical treatment methods are generally employed where the removal of MPs from wastewater is not possible with physical or biological treatment methods. The wastewater treatment using the chemical method may include a series of chemical reactions that help to selectively remove the MPs. Electrochemical oxidation, coagulation, flocculation, sol-gel techniques, photocatalytic degradation, and ozonization are frequently used chemical treatment methods for the removal of MPs.

Electrochemical oxidation is an affordable and sustainable method for the treatment of MPs present in wastewater. Instead of MP degradation, this method is suitable for the degradation of toxic dyes, organic pollutants, and medications. This method uses indirect cathode/anode oxidation and produces non-toxic degradation end products such as CO_2 and H_2O. Falco *et al.* demonstrated the fabrication of boron-doped diamond electrodes that can effectively remove 98.5% polystyrene

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MPs from wastewater at 25 mgL^{-1} initial MPs concentration, 90 min treatment time, and 8.07 $mAcm^{-2}$ current density at pH 4.[67] While the fabricated electrode showed high removal efficiency, the sludge generation during the removal process added operational cost related to the disposal of waste. In another study, the boron-doped diamond electrodes were also employed for the degradation of polyethylene terephthalate microplastics.[68] The removal efficiency was 81% in synthetic water and 95% in marine water with added salt due to the crucial role of chlorine species present in saline water. Moreover, the removal time was reduced from 12 h to 2h in saline water.

The coagulation/ flocculation method for the removal of MPs involves the neutralization of the charge present on the existing colloidal particles, which may form floccules and can be separated through sedimentation or filtration. For example, Zheng *et al.* employed polymeric ferric sulfate and Opuntia Milpa Alta particles for the removal of polystyrene MPs and achieved 93.6% removal efficiency at 20 mg/L Opuntia Milpa Alta concentration, 120 mg/L polymeric ferric sulfate concentration and 9.2 pH.[69] In comparison to chemical coagulation, the electrocoagulation method offers the use of metal electrodes, which is preferable over conventional coagulation techniques. For example, Sezer *et al.* suggested the optimization of the electrocoagulation method by Box Behnken Design and achieved 99% removal efficiency of MPs obtained from food packaging industry wastewaters under optimum conditions (i.e., current density: 3.16 mA cm-2, pH: 6.74, and time: 13.58 min).[70]

Ozonization is a tertiary treatment process used for the treatment of MPs present in the leftover residue of the coagulation process. The oxygen functionalities of the MPs can be degraded through ozonization. To improve the efficiency of ozonization, Hu *et al.* introduced 20 mM α-FeOOH into the ozonation system, and the mineralization efficiency in case of polyethylene MPs was found to be enhanced 3.27 folds due to the accelerated production of OH* free radicals.[71] In another study, the efficiency of ozonization was accelerated by implementing Co(II)-catalyzed ozonation. 1 mM Co^{2+} has reduced 20% turbidity of the wastewater and achieved 64% mineralization of MPs within 15 min of reaction time.[72] The major drawback of ozonization process is its high operation cost. In addition, the incomplete ozonization process can produce intermediate components that can be toxic to the environment and can lead to the generation of reactive oxygen species.

Sol-gel technique is a chemical process that involves the agglomeration of organosilanes into an inorganic-organic macromolecule before its removal from wastewater.[73] Using the sol-gel method, Pacaphol *et al.* demonstrated the removal of small MPs such as polyethylene, polyethylene terephthalate, polystyrene, polyvinyl chloride, polytetrafluoroethylene, polypropylene, and aged tyre rubber. An excellent MPs recovery rate (95-100%) was observed using tetraethyl orthosilicate as a floating media. However, the recovery rates were slightly reduced (82-98%) for finer particles of sizes < 40 µm.[74]

Photocatalysis is the most reliable and cost-effective chemical method for degrading toxic organic pollutants, including MPs. In this technique, the oxidation of MPs is carried out with the help of free radicals and reactive oxygen species produced by the semiconductor material while interacting with ultraviolet or visible light. Yang *et al.* prepared core–shell $BiO_{2-x}/CuBi_2O_4$ heterojunction for the effective degradation of polystyrene MPs. The results indicated that the complete surface of polystyrene was degraded in 30 days using the synthesized photocatalyst.[75] In another study, the photocatalytic degradation of micro polyethylene bags was studied using a ZnO-based photocatalysis-persulfate activation system. Hydroxyl and sulfate radicals were the major reactive species in the photocatalysis process, leading to a 50.91% mass loss ratio within 105 h.

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The photocatalyst was recyclable up to many cycles, and the difference in the mass loss ratio of the first and fifth cycle was only 1%.[76]

4.3. Biological Methods

Biological methods of MPs degradation include the treatment of wastewater with different bacterial isolates. The extent of MPs degradation depends upon the type of microplastic and bacterial strain. In comparison to physical and chemical degradation methods, the biological methods of MPs degradation are rarely used. However, some studies have suggested the use of bioreactors and dynamic membrane techniques to remove low-density microplastic sources.[77] For example, the studies carried out by Corpuz *et al.* showed a 95% reduction of polyethylene MPs using an electrochemically enhanced living membrane bioreactor. Moreover, the live membrane bioreactor migrated the inhibition effect of MPs to PO_4-P and NH_4-N removals.[78] Lv *et al.* have shown the degradation of polystyrene MPs with Alcanivorax *xenomutans* and Halomonas *titanicae* marine bacteria. It was observed that only 4.5% and 1.9% of the plastic films were actually biomineralized with A. *xenomutans* and H. *titanicae* after 30 days of incubation.[79] In another study, P. *aeruginosa* and S. *haemolyticus* were isolated from landfill leachate that had shown the degradation of Polypropylene MPs. The maximum degradation efficiency achieved was 25.46% and 7.01% for S. *haemolyticus* and P. *aeruginosa,* respectively.[80] The degradation of polystyrene MPs using edible fungi was also studied by Yu *et al.* and the highest degradation rate (16.17 ± 8.87%) was found with Pleurotus *ostreatus* after 50 days of fungal degradation.[81]

It may be noted that several studies have reported different biological methods for the removal of MPs, still physical and chemical methods for the removal of MPs are more reliable, efficient and economical.

Table 1: Comparative analysis of different techniques for the removal of MPs

Method	**Key features of the method**	**Specific example**	**Type of target MP**	**Size range of targeted MPs**	**Surface charges on the adsorbent**	**Pore/ mesh size of the adsorbent**	**Degradation time**	**% MPs removal efficiency**	**Removal mechanism**	**Ref.**
Photocatalytic degradation	Durable and eco-friendly	Nb-doped SnO_2 QDs	Polyethylene	350 µm	−35 mV	-	7 h	28.9%	Interactive mechanism of dual defects and photocatalytic mechanism	[82]
Electrochemical oxidation	Highly efficient; does not require the use of toxic chemical agents	CeO_2–PbO_2-0.005 anode	Polyvinyl chloride	-	-	-	6h	38.67%	Ce(III)/Ce(IV) valence-modulated mechanism	[83]
Coagulation	Minimum energy uses for the removal of tiny MPs	Polyaluminum ferric chloride with Opuntia Milpa Alta particles	Polystyrene	2–10 µm	4.52 mV at pH 6 and −0.56 mV at pH 9	380 µm	60 min	94.8%	Charge neutralization and adsorption bridging	[84]
Electrocoagulation	Economical and energy-efficient; does not require chemical coagulants; efficient for removing small MPs	Al/Fe-graphene-Electrocoagulation (EC) system	Polyethylene Terephthalate, Polypropylene, and Polystyrene	45 µm to 250 µm	-	-	120 min	96 %	Adsorption and electrostatic interactions	[85]

Sand filtration	Cost-Effective and simple to operate	Natural quartz sand in a Rapid Sand Filtration (RSF) system	Polypropylene and Polystyrene	<10 µm	−30.1 mV and −32.8 mV	NA	20 min	84–98 %	Synergistic combination of physicochemical sorption	59
Disc filtration	Minimum energy consumption for the accelerative removal of MPs	Disc Filter	Polyethylene, polystyrene, polyester, polyvinyl chloride	> 300 µm	-	15 µm	-	89.7%	MPs removal by high-pressure back-flushing	86
Ozonation	Cost-effective and efficient	α-MnO2 and α-FeOOH	Polystyrene	< 1 µm	-	-	8h	16.5%	A free radical mechanism	71
Froth flotation	Low cost, and adaptable for quick operation	Flotation process combined 10 g/ton of sodium oleate	Polyvinyl chloride, acrylonitrile–butadiene styrene	0.074 µm to 5 µm	-33.1 t0 - 14.5 mV	-	24h	100%	Hydrophilization and electrostatic mechanism	87
Oil film separation	Affordable and simple to use	Castor Oil	Polypropylene, Polystyrene, polymethyl methacrylate, and glycol modified	0.3–1.0 mm	-	25 µm	4h	99%	lipophilicity-based protocol	88

			polyethylene terephthalate							
Biochar filters	Effective in achieving excellent adsorption capacities	Surface-engineered palm kernel shell biochar	Polyethylene, polyamide	159 nm–48 µm	+9.5 to +14.1 mV	0.6 to 1.18 mm	2h	96.12%	Electrostatic interaction, hydrophobic interaction, π-π interaction and hydrogen bonding	[89]
Adsorption removal	Cost-effective, easy to use and reusable upto many cycles	Polydopamine-modified sodium alginate hydrogel	Polyethylene, polystyrene	200 nm to 10 µm	−17.9 ± 2.21 mV	-	2h	99.6 %	Chemical adsorption, electrostatic interactions, hydrogen bonding and π-π interactions	[90]
Membrane bioreactor technology	Precise removal of contaminants at variable MPs concentration ensuring effective removal	Living membrane bioreactor containing biological layer as a membrane filter	Polyethylene	≤ 20 µm	-	0.04 µm	21 days	95%	Electrocoagulation and adsorption	[78]

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5. Hydrogels and their application in removal of MPs

In recent years, hydrogels have piqued the interest of scientists for addressing the critical issue of MPs pollution. Hydrogels leverage their high water-holding capacity and porous architecture to elevate the capture and removal of MPs. They can effectively eliminate MPs from water by simple adsorption, surpassing limitations of conventional removal techniques and unlocking new possibilities to improve water quality.

5.1. Salient features of hydrogels

Hydrogels are 3D crosslinked networks of polymeric chains that can absorb and retain significant amount of water without dissolving.[91-92] These materials exhibit exceptional properties including high porosity, structural flexibility, elasticity, and tunable functionality. Additionally, the possibility of incorporating the features of biocompatibility and biodegradability by a suitable choice of components and synthetic strategy, makes them an ideal candidate for green adsorbents.[93-94] **Fig. 3** summarizes a few examples of different building blocks (e.g., polymers, monomers, comonomers, and crosslinkers) that have been widely utilized for the fabrication of hydrogels.[95-102]

Hydrogels can be synthesized using various natural and synthetic polymers. Usage of synthetic polymers offers reproducibility and control over the molecular weight of polymer chains, while natural polymers are employed to utilize their inherent biocompatibility and biodegradability.[103-104] Additionally, a combination of natural and synthetic polymers is also frequently investigated to develop hybrid hydrogels that are benefitted from the advantages of both types of polymers.[103-104] Moreover, the incorporation of different functional groups (e.g., $–NH_2$, –

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COOH, –OH, –CONH$_2$) inside the hydrogels during synthesis can enhance their interaction with the guest moieties including NPs and MPs, thus facilitating their effective removal.[105-109]

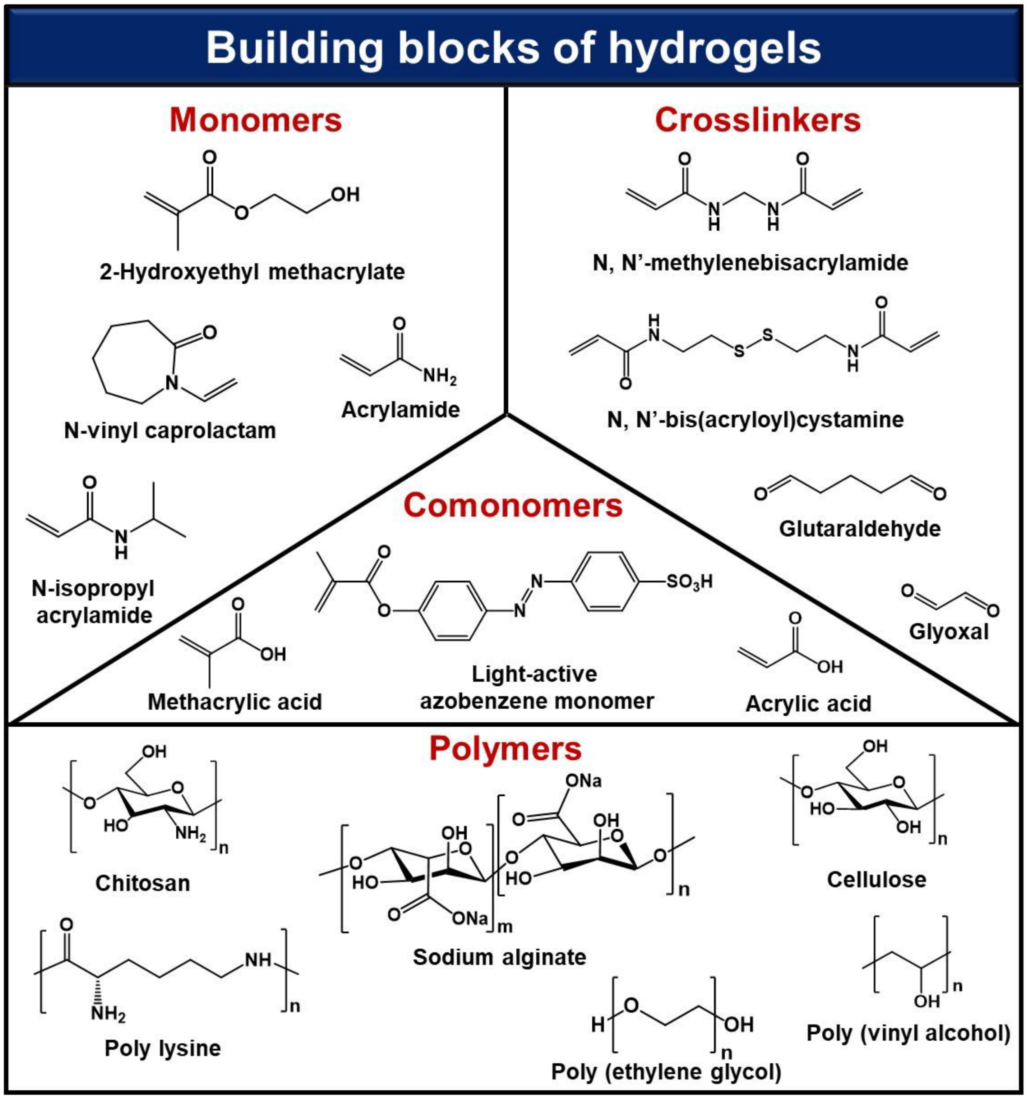

Fig. 3: Examples of key components utilized for the fabrication of hydrogel: Monomers, comonomers, crosslinkers and commonly used polymers.

Further, hydrogel networks are typically formed via physical or chemical crosslinking of polymeric chains.[110-111] Physical crosslinking involves hydrogen bonding, electrostatic interactions, hydrophobic/hydrophilic interactions, metal coordination,

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crystallization/stereocomplex formation, and π–π stacking interactions.[111] In the case of chemical crosslinking, formation of covalent or dynamic covalent bonds takes place providing a control over the mechanical strength and degradability of hydrogels.[110] Various dynamic covalent bonds including imine,[112] disulfide,[113] boronic ester,[114] and acylhydrazone[115] have been utilized in the literature for the fabrication of degradable hydrogels. These dynamic bonds get degraded under specific environmental conditions, reducing the risk of secondary pollution that is often associated with persistent synthetic materials.[116-118, 83-85]

5.2. Hydrogels for removal of MPs

In recent years, scientists have explored various natural, synthetic, and natural/synthetic hybrid polymers-based hydrogels to mitigate MPs pollution. However, the removal efficiency of hydrogels is strongly affected by the pore size of hydrogel, swelling behaviour, network density and overall accessible surface area.[119] In addition, the size, shape, and surface charge of the target MPs also affects the removal efficiency.[120]

Taking advantage of the attractive features of hydrogels mentioned earlier, Patel *et al.* reported aluminium and iron-incorporated ionotropic chitosan (CS) hydrogels for the removal of PET MPs.[121] Here, the integration of metal cations into CS hydrogels boosted their ionic charge, thus providing a straightforward and efficient method to enhance the adsorption capacity, while offering low cost and minimal environmental impact. Interestingly, a pH-based alteration of the adsorption capability of both Al-CS and Fe-CS hydrogels was observed here. At lower pH, the protonation of amino groups in CS combined with the formation of cationic metallic species (e.g., Al^{3+}, $Al(OH)_2^+$, $Al(OH)_2^+$, FeO^+) enhanced the overall positive charge of hydrogels, which in turn helped in achieving higher electrostatic interaction with anionic MPs, leading to a 70% adsorptive

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removal of PET MPs. While at higher pH, deprotonation of amino groups in combination with the generation of anionic hydrates (like $Al(OH)_4^-$, $Al(OH)_3$, FeO_2^-, FeO_2H) led to repulsion, thus reducing the adsorption efficiency. Moreover, upon increasing the doping concentration, adsorption efficiency was comparatively reduced which could be attributed to the cations occupying the pore regions of hydrogels. Finally, the successful removal of MPs from CS-based hydrogels using NaOH demonstrated the reversibility of adsorption–desorption, confirming that the process was governed by physical adsorption.[121]

Utilizing copper substituted polyoxometalate (Cu-POM) nanoclusters, Dutta *et al.* synthesized Cu-POM infused triple interpenetrating network hydrogel (pGel@IPN), which was composed of polyaniline (PANI), PVA, and chitosan (CS).[122] This hydrogel matrix exhibited excellent mechanical strength which can be assigned to strong electrostatic interactions between positively charged PANI and negatively charged Cu-POM. The hydrogel was further strengthened by crosslinking between hydroxyl groups of PVA and aldehyde groups of glutaraldehyde (GA) **(Fig. 4)**. pGel@IPN demonstrated high removal efficiency for both PVC and PP-based MPs which were labelled with Nile red for their easy detection and analysis via fluorescence microscopy. pGel@IPN showed ~95% and ~93% removal efficiency for PVC and PP MPs, respectively, at pH 6.5. Statistical analysis further revealed that the adsorption of MPs followed Langmuir model (R^2 >0.99), suggesting monolayer sorption of MPs on hydrogel surface. Notably, the adsorption capacity of hydrogel was high, reaching 321.87 mg g^{-1} for PVC and 144.29 mg g^{-1} for PP MPs. These hydrogels demonstrated good reusability to capture MPs up to five cycles. Further, UV-induced degradation of MPs was facilitated by taking advantage of the catalytic properties of Cu-POM, thus combining the concept of efficient capturing and degradation of MPs. Finally, after the

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hydrogel served its purpose, it was upcycled into carbon nanoparticles, which in turn were utilized as an adsorbent to remove Cr (VI) from contaminated water to move towards circular economy.[122]

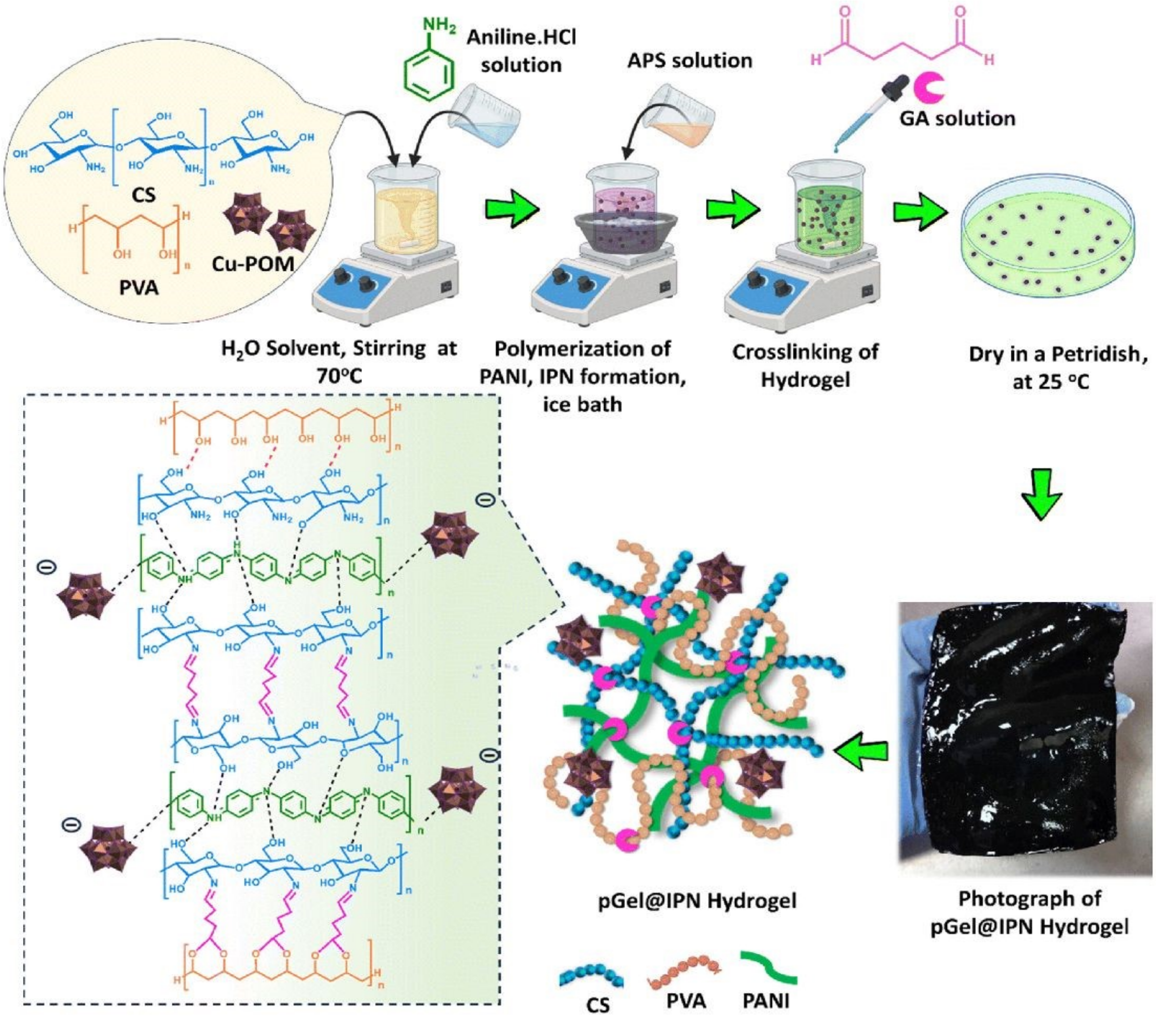

Fig. 4: Scheme showing the step wise synthesis of copper substituted polyoxometalate (Cu-POM) nanoclusters infused triple interpenetrating network hydrogel (pGel@IPN). Reproduced with permission.[122] Copyright 2024, Publisher Royal Society of Chemistry.

Developing biomass derived natural polymers-based hydrogels offers a sustainable approach to capture MPs for water treatment. In this regard, Li *et al.* utilized bamboo-derived lignin in combination with PVA to synthesize a hydrogel for the removal of PS MPs.[123] Here,

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lignin was subjected to phenolization and ammonization processes that markedly increased the phenolic hydroxyl and long-chain amino groups content in lignin, respectively. The incorporation of amino groups introduced dual functionality in lignin including pollutant adsorption over a broad pH range and improved encapsulation. Aminated lignin/epichlorohydrin/polyvinyl alcohol (LG-GH-PVA) hydrogel was synthesized via chemical crosslinking of aminated lignin with epichlorohydrin, as well as ionic crosslinking of PVA with Ca^{2+}. The adsorption process followed a pseudo-second-order kinetic model, while the equilibrium data exhibited the best fit with Langmuir isotherm confirming a monolayer adsorption with a maximum adsorption capacity of 288.6 mg/g. Notably, the hydrogel maintained 87.6% of its adsorption capacity even after five regeneration cycles which was attributed to π–π interactions between hydrogel and PS MPs in addition to hydrogen bonding between hydroxyl, carboxyl, and amino groups present on both the surfaces. The reported hydrogel demonstrated high adsorption of PS MPs in real-world systems, achieving efficiencies of 90.6–92.7% in lake water, 92.2–94.7% in pipeline water, 94.6–97.1% in river water, and 95.5–97.9% in sludge supernatant. In addition to PS MPs, the hydrogel effectively adsorbed four other MPs including PEI MPs, PP MPs, PVC MPs, and PA MPs even at a low concentration of 10 mg/L, presenting itself as a promising platform for MPs removal.[123]

Exploring other natural polymers, Leppanen *et al.* developed cellulose-based hydrogels by using two different grades of cellulose nanofibrils including native cellulose nanofibrils (CNF) and TEMPO-oxidized CNF.[124] These hydrogels were utilized to capture both cationic and anionic PS particles (1 μm and 100 nm) using a microfluidic setup coupled with fluorescence microscopy **(Fig. 5a-b)**. It was reported that the adsorption and removal of PS particles were influenced by the hygroscopic cellulose network and large surface area of hydrogels.[124] The removal mechanism was based on the attractive surface interactions when capillary forces were not assisting the

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capturing process. The larger surface area and enhanced surface interactions were the driving forces for the cohesion between material surface and NPs here. Next, Yi *et al.* developed dual crosslinked chitin (Ch) nanofibrils-based hydrogels by using formaldehyde for chemical crosslinking and ammonia fumigation for physical crosslinking using freeze thaw method **(Fig. 5c).**[125] The dual crosslinked hydrogels exhibited higher mechanical strength and filtration efficiency as compared to individual physically or chemically crosslinked hydrogel. Moreover, a syringe-based filtration device using the dual crosslinked nanoCh hydrogels was prepared which could achieve almost complete removal of MPs (size ~3 μm) with highest flux of 8.22 mL cm^{-2} h^{-1} owing to the larger pore size and robust channel structure, offering scalability for large-scale water purification. Finally, after filtration, the SEM images of nanoCh hydrogel (top side and down side) were captured which clearly depicted the presence of adsorbed MPs on the top surface indicating their effective removal **(Fig. 5c).**[125] The porosity of the dual-crosslinked nanochitin hydrogels was 257.5 $m^2.g^{-1}$ and carries rich pores in the microscale range explaining the effective particle interception both on the surface and within the hydrogel structure with a flux of 8.22 mL $cm^{-2}h^{-1}$.[125] Overall, the use of these systems for the elimination of MPs could pave a new pathway for next-generation portable wastewater treatment methods.

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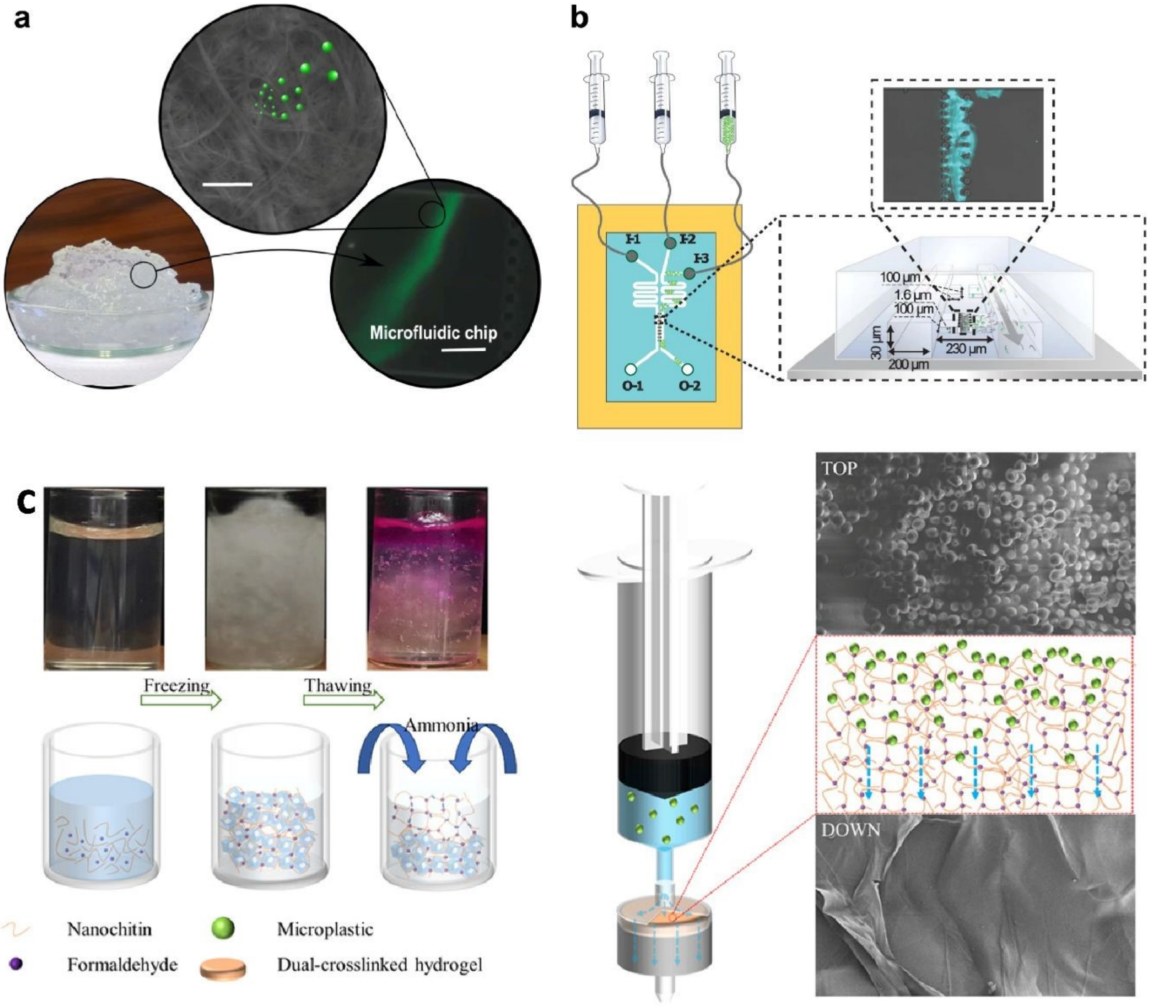

Fig. 5: (a) Schematic representation of a proof of concept for capturing fluorescently labelled MPs by CNF hydrogel using a microfluidic set-up coupled with fluorescent imaging (Scale bar in SEM image is 1 µm and 25 µm in microfluidic chip image). (b) Microfluidic setup for CNF hydrogel containing trap showing the injection of fluorescent MPs and water. Reproduced under the CCBY-license.[124] Copyright 2022, Publisher Nature Portfolio. (c) Images, illustration of synthetic protocol of nanoCh hydrogels, and the syringe-based filtration device using dual crosslinked nanoCh hydrogel (Inset: SEM image of the top side and down side of the nanoCh hydrogel after filtration). Reproduced with permission.[125] Copyright 2025, Publisher Elsevier.

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Considering nature-inspired catechol-based chemistry, Han *et al.* fabricated polydopamine (PDA)-modified sodium alginate hydrogel (PMSAH) for eliminating MPs from drinking water.[90] Here, PDA-modified sodium alginate was crosslinked using calcium ions and tested for the elimination of different MPs including PE and PS of varying size and surface charges **(Fig. 6)**. For real life application, PMSAH was also utilized for the removal of MPs generated by boiling the commercially available tea bags. It was reported that PMSAH exhibited the highest removal efficiency of ~99.6% which could be attributed to various intermolecular interactions, including hydrogen bonding, electrostatic interactions, hydrophobic interactions, and π–π stacking interactions offered by PDA **(Fig. 6).**[90]

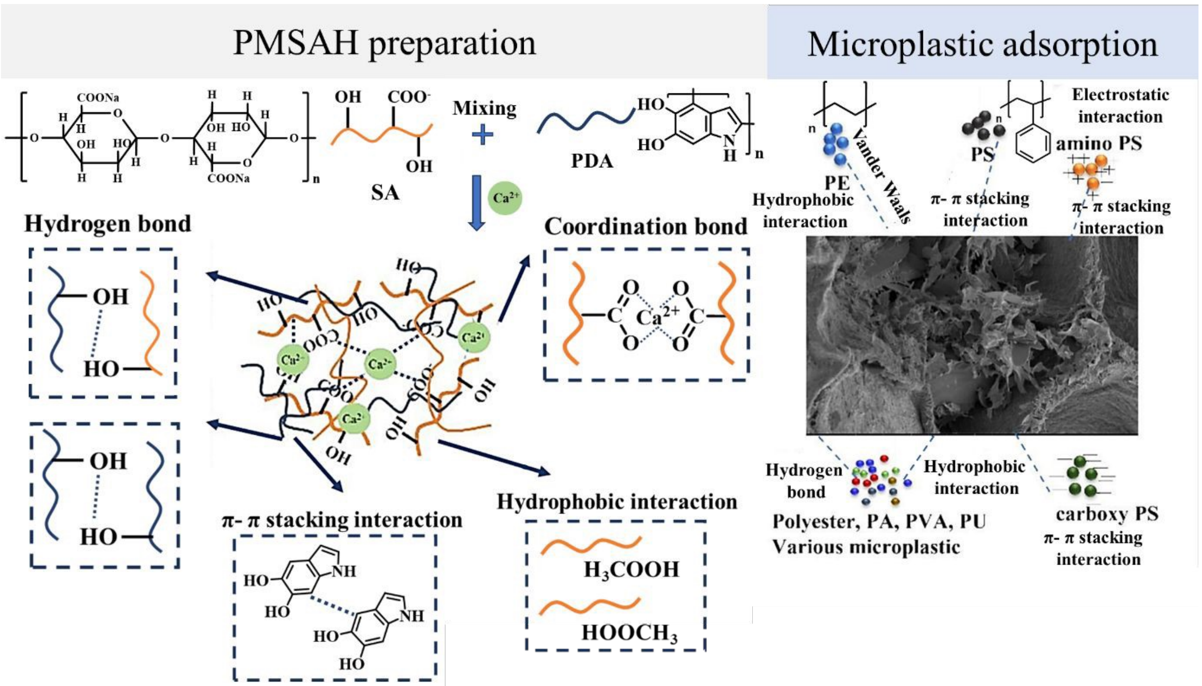

Fig. 6: Schematic representation for the fabrication of PMSAH hydrogel and interactions involved in MPs adsorption. Adapted with permission.[90] Copyright 2025, Publisher Elsevier.

Taking a step further in the direction of combined adsorption, detection, and actuation performance while still considering the catechol-based chemistry, Guo *et al.* imparted hydrogels

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with stimuli responsiveness and multiple functionality to improve their performance for heavy metal adsorbed MPs remediation.[126] In this case, a smart light-driven multifunctional hydrogel actuator was engineered for selective removal of iron adsorbed polystyrene microplastics (Fe^{3+}@PS) from water.[126] Here, covalently bonded acrylated polyethyleneimine and polydopamine copolymer (M-PEI@PDA), graphene oxide (GO) nanosheets, and poly(N-isopropyl acrylamide) (PNIPAM) formed hierarchical interpenetrating network of hydrogel (M-PEI@PDA/GO/PNIPAM) **(Fig, 7a)**. M-PEI@PDA copolymer was prepared utilizing Michael addition providing fluorescence responsiveness to the hydrogel. Subsequently, multifunctional crosslinked hydrogel was fabricated via *in situ* radical polymerization between thermo-responsive NIPAM monomer, M-PEI@PDA copolymer and *N*, *N'*-methylene bisacrylamide (MBA) crosslinker **(Fig. 7a)**. Notably, hydrogel showed strong interfacial interactions between catechol and amino groups of polymer and Fe^{3+}@PS, offering an excellent reversible adsorption property **(Fig. 7b)**. Furthermore, by leveraging temperature responsiveness of PNIPAM, hydrogel actuator demonstrated excellent actuation performance (bending speed: 2–4°/s, and swimming speed: 0.5 mm/s), thus facilitating the locomotive adsorption of MPs present in water **(Fig. 7b)**. The reported hydrogel displayed an ultralow detection for ferric ion (0.98 μM) and selective adsorption for Fe^{3+}@PS (~97%) with high adsorption (~95%) and desorption efficiency (~99%) for MPs, making it a suitable candidate to remove MPs from water.[126]

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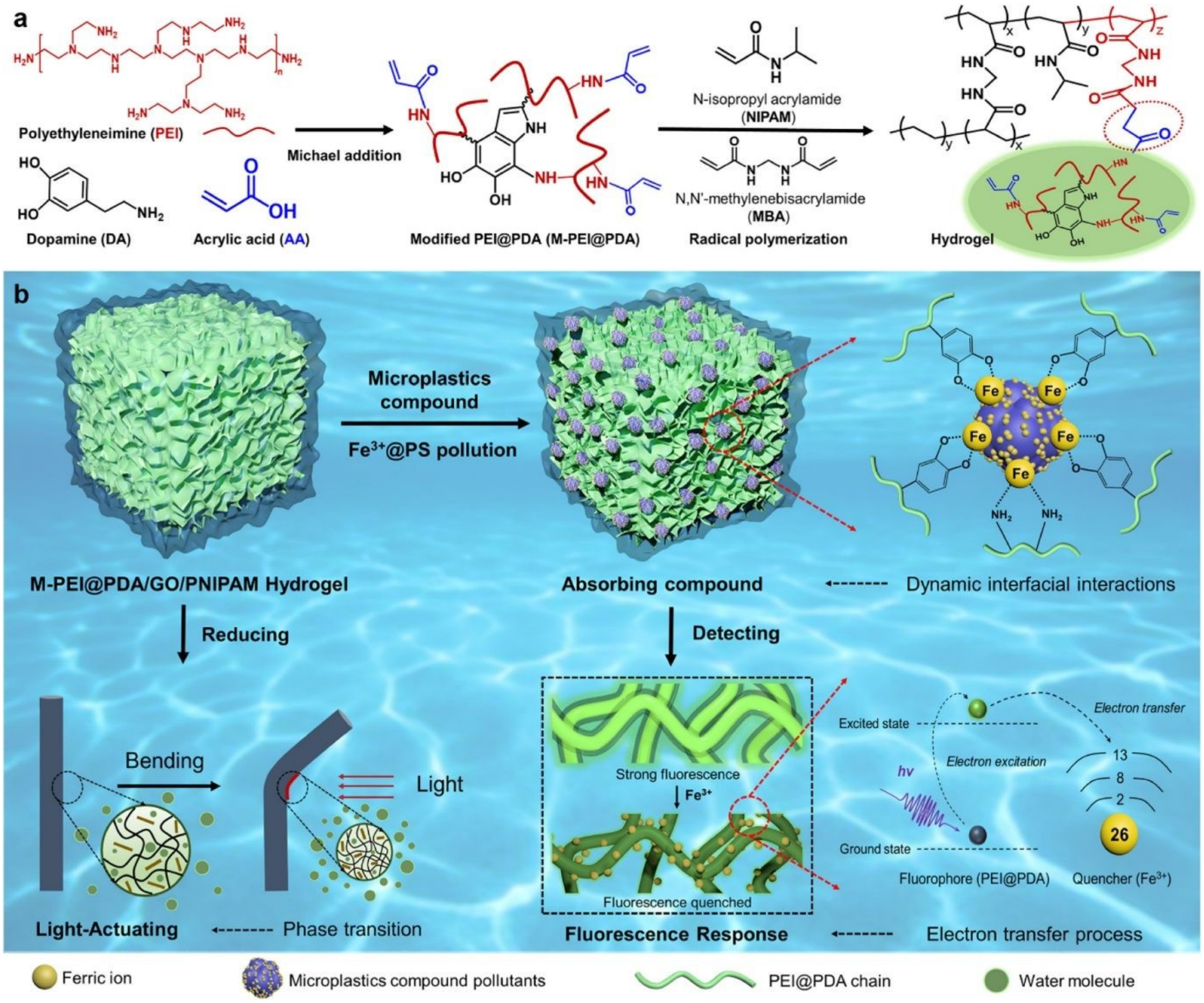

Fig. 7: (a) Schematic illustration for the fabrication of M-PEI@PDA/GO/PNIPAM-based multifunctional hydrogel. (b) Multifunctional hydrogel with combined adsorption, detection, and actuation performance for efficient removal of Fe^{3+}@PS. Reproduced with permission.[126] Copyright 2022, Publisher American Chemical Society.

6. Sponges for removal of MPs

In addition to hydrogels, hydrogel derived sponges have recently gained attention as a promising alternative for MPs removal owing to their highly porous structure, light weight, and good adsorption capacity. The sponges are typically fabricated by drying hydrogels using freeze drying

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method. For example, Sun *et al.* synthesized graphene oxide-incorporated chitin-based (ChGO) sponges by freeze-drying of corresponding hydrogel having epichlorohydrin-based crosslinking.[127] ChGO sponges could be reused and showed high adsorption efficacy of 89.8%, 72.4%, and 88.9% for PS, carboxylated polystyrene (PS-COOH), and aminated polystyrene (PS-NH_2) MPs, respectively, after 3 adsorption–desorption cycles. The pore size of the sponge was between 50-76 μm and it was capable of accommodating polystyrene MPs (1 μm in size) upto 3 cycles without any degradation. Here, the adsorption of MPs was facilitated by hydrogen bonding, electrostatic interactions, and π-π interactions between MPs and ChGO sponges. Additionally, biodegradation results also confirmed the degradability of ChGO sponges by microorganisms present in soil.[127] Taking a step further, Sun *et al.* developed various chitin-based sponges including pure chitin (Ch), chitin/O-C_3N_4 (ChCN), chitin/GO (ChGO), chitin/GO/carboxymethyl cellulose (ChGO-CL), and chitin/GO/chitosan (ChGO-CS) [128]. It was reported that the incorporation of additional material in chitin sponges further improved their mechanical strength and adsorption capacity. The pore size of the prepared sponges varied between 86 μm to 181 μm, and the sponges were efficient to remove different functionalized PS MPs (~1 μm) with removal efficiencies ranging from ~71% to ~92%. Moreover, these sponges could be reused up to three cycles for MPs removal.[128]

Next, Zhu *et al.* introduced polyaniline (PANI) into chitin matrix crosslinked with epichlorohydrin to develop chitin/polyaniline sponge (ChPANIs) having macroporous structure with uniform network.[129] It was reported that upon increasing the hydrophilicity of PANI in ChPANIs, the removal efficacy of polystyrene MPs (size = 1μm) was enhanced from 84% to 91%, which could be attributed to better dispersibility of PANI in chitin-based sponge matrix with porous structure ranging from 190 μm to 470 μm. More hydrophilic PANI in ChPANIs provided

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more contact area and adsorption sites, which enhanced the electrostatic interactions with MPs, providing excellent adsorption performance. Further, SEM images demonstrated that ChPANIs efficiently removed MPs without causing significant alterations in its sponge-like morphology, whereas higher magnification clearly confirmed numerous MPs adhered to the surface. The adsorption of MPs followed the Freundlich isotherm, suggesting multi-layer adsorption of MPs on ChPANIs. Moreover, ChPANIs demonstrated good mechanical strength, reusability up to five cycles, and ~89% degradation over 15 days in soil once their purpose is served.[129]

Nowadays, fibrous sponges have gained significant interest for the removal of MPs. In this direction, Wu *et al.* demonstrated that a fibrous framework sponge derived from exfoliated β-chitin nanofiber and suspended cellulose fiber can be derived simply through hydrogen bonding without the use of any crosslinking agents **(Fig. 8a)**. The fibrous sponge was prepared by interrupting original hydrogen bonds in the presence of an acid, stripping cotton into finer cellulose fibers, exfoliating the chitin into nanofibrous plane, and then interlacing these fibers to induce intermolecular hydrogen bonds. The fabricated sponge demonstrated a porous interconnected structure in addition to numerous activated functional groups such as OH^-, NH_3^+, and $NHCO^-$. The presence of these functional groups assured the removal of MPs through multilevel interactions, including electrostatic interactions, hydrogen bonding and Van der waals interactions **(Fig. 8b)**. The fabricated sponge demonstrated 98.0% to 99.9% MPs removal efficiency and was reusable upto five adsorption-desorption cycles without any effective loss in the degradation efficiency.[130]

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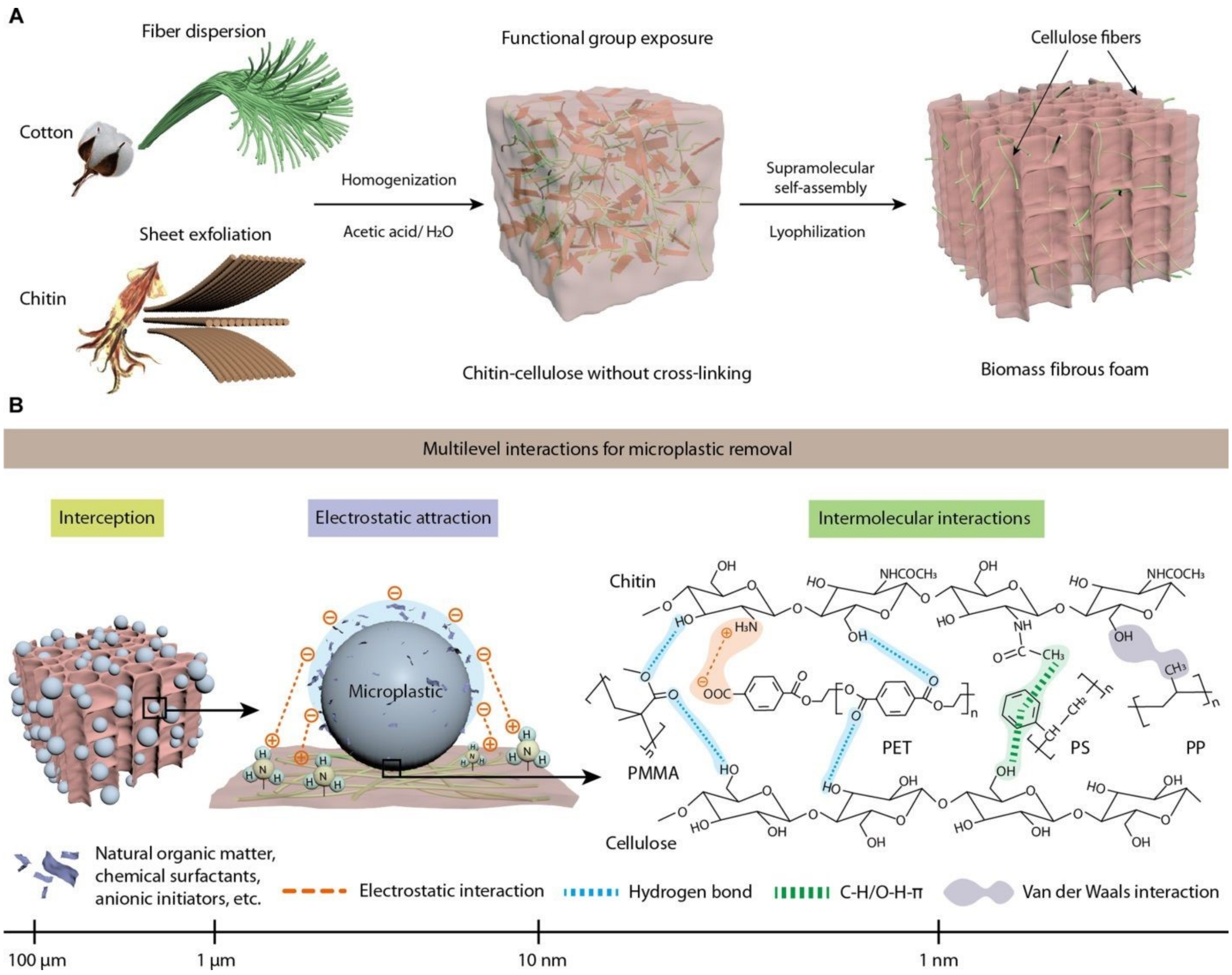

Fig. 8: (a) Schematic illustration of self-assembly of chitin/ cotton fibrous foam, and (b) Mechanistic pathway showing multilevel interactions for the removal of MPs. Reproduced with permission.[130] Copyright 2024, Publisher Science.

Further, Ko *et al.* utilized freeze-drying method coupled with genipin (GP)-based crosslinking to fabricate cost effective, and light-weight graphene oxide/chitosan/genipin sponges (GO_5/CS/GP).[131] These sponges were macroporous (porosity = 95%, pore size = 58.3 ± 47.8 μm, density = 32.6 mg/cm^3) and exhibited an excellent capturing rate (~73%) for PS NPs of diameter 0.026 μm as compared to PS MPs of diameter 1.1 μm (~41%). The adsorption of PS MPs on GO_5/CS/GP was attributed to hydrogen bonding, pore filling interaction, hydrophobic interaction,

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and π−π interaction. This work highlighted the need of developing systems that can be used for efficient removal of NPs.[131]

Similarly, Risch *et al.* performed freeze drying of chitosan electrospun nanofibers-based suspension followed by crosslinking with GA to develop chitosan nanofiber sponges (CSNFs) having good water stability.[132] CSNFs exhibited a bulk density of 5.77 mg cm^{-3}, pore diameter 163 ± 41 μm and a porosity of 99.59%. The hierarchical pore structure of CSNFs was utilized to remove PET MPs and Arizona test dust (ISO 12103-1) suspensions. These sponges exhibited high selectivity for the adsorption of PET MPs from water with ~99% removal efficiency, suggesting their application in tackling MPs based pollution.[132]

In another direction, taking inspiration from anisotropic vessels in hardwood, Xu *et al.* developed biomimetic, double crosslinked CS-based sponges (BGCS) by combining directional freezing with freeze drying and GA-based crosslinking.[133] Here, sponges were reinforced with flexible bacterial cellulose fibrils, which served as physical crosslinkers by forming supramolecular assemblies with chitosan (soft phase). Additionally, treatment with GA vapor further resulted in covalently crosslinked network (hard phase) in sponges **(Figure 9a)**. Moreover, BGCS sponge displayed excellent wet compressive resilience with retention rate of ~95% even after 100 compression cycles. It was reported that the anisotropic structure of sponges served as a rapid water treatment channel and removed ~78% of PS MPs (~1μm in size) within 420 min at room temperature. Owing to its higher wet compressibility, the developed sponge demonstrated good removal efficiency for PS MPs (~47%) even after 20 cycles **(Figure 9b)**. Here, electrostatic interactions, conjugation, and intraparticle diffusion were identified as primary forces for adsorption of PS MPs on sponges.[133]

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Building on the concept of enhancing mechanical strength and improving water stability via crosslinking, Ma *et al.* synthesized Ca^{2+} crosslinked sodium alginate-based sponges via secondary freeze-drying method.[134] These sponges exhibited excellent water absorption (~1193–5232%), high porosity (~89%), good mechanical properties, and remarkable PS MPs (≤5 µm) removal efficiency (~92%). The adsorption of MPs on sponges was attributed to intra-particle diffusion, hydrogen bonding, and π-π interactions. Overall, the above-mentioned examples clearly signify the potential of polymeric sponges in addressing the issue of MPs pollution for moving towards sustainable development.

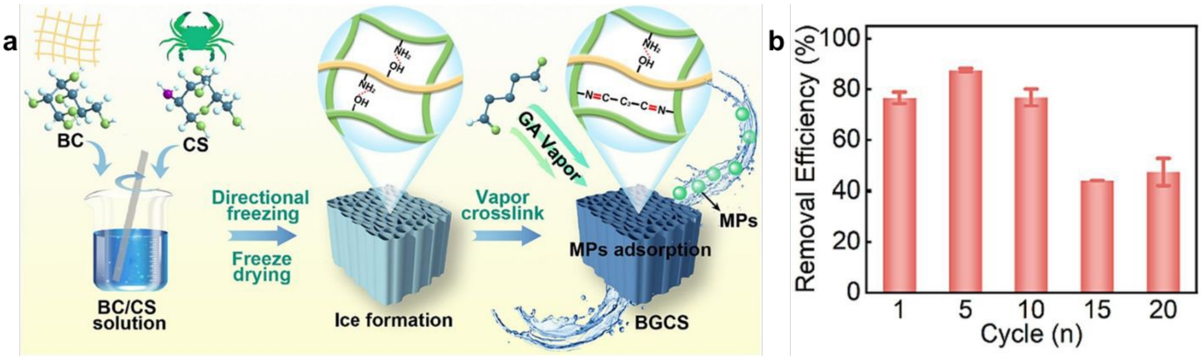

Fig. 9: (a) Synthetic route for the fabrication of biomimetic, double crosslinked chitosan-based sponges (BGCS). (b) Histogram showing the efficiency of BGCS in removing PS MPs up to 20 cycles. Reproduced with permission.[133] Copyright 2023, Publisher Elsevier.

7. Electrospun fibers-based membranes for removal of MPs

In recent years, electrospun polymeric nanofiber-based membranes have emerged as a potential candidate for water purification. These membranes display high surface area to volume ratio, highly interconnected pore structure, and adjustable physicochemical properties (e.g., wettability) that helps in increasing the fluid flow rate while reducing the fouling.[135-138] In the literature, nanofibrous membranes have been developed through electrospinning of both natural and

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synthetic polymer-based solutions.[139-140] Morphology and architecture of these electrospun fibers can be tuned by adjusting several parameters including polymer solution related parameters (concentration, viscosity, and conductivity), electrospinning process parameters (flow rate, voltage, and distance between spinneret and collector), and environmental parameters (temperature or humidity).[139,141]. The resulting fibrous mat can also be processed to form dispersion of nanofibers which in turn can be used for the preparation of sponges or wet-laid membranes.[142-143] So far, electrospun fibers-based membranes have been widely explored for removing organic and inorganic pollutants including heavy metal ions, dyes, and oil from aqueous environment due to their large surface area and adjustable wettability.[144-145] The main driving force behind the removal of pollutants from aqueous environment is adsorption. Recently, scientists have started to explore electrospun nanofibers-based membranes for the removal of MPs.

In this direction, Wang *et al.* fabricated polyacrylonitrile (PAN)-based nanofiber membranes by electrospinning followed by hot-pressing method.[146] Fiber diameter, membrane thickness, and membrane porosity were optimized here by adjusting various processing parameters. These membranes were utilized for the removal of PS beads (0.2 μm and 0.1 μm) from water. It was reported that the rate of rejection of membranes for 0.2 μm PS beads approached ~100% upon reducing the porosity of membrane. Moreover, membranes with relatively small nanofiber diameters were able to completely reject all the 0.1 μm PS beads while maintaining high flux and low degree of fouling. This study demonstrated that the fiber diameter and membrane porosity play crucial role in determining the equivalent/apparent pore size, which in turn directly relates to microfiltration performance.[146]

With the aim of developing self-standing membrane without any supporting substrates, Juraij *et al.* developed polyurethane/graphene oxide-montmorillonite-based electrospun composite

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(PGT) membrane **(Figure 10a).**[147] Here, different amounts of graphene oxide-montmorillonite (GOMt) ranging from 5 to 20 wt% were loaded into PGT membrane. Based on high tensile strength (6.6 MPa), porosity (~61%), superhydrophilicity, pressure-driven water flux (8163 L m^{-2} h^{-1}), and higher gravity-driven water flux (793 L m^{-2} h^{-1}), PGT membrane containing 20 wt% GOMt was chosen for MPs and NPs separation from water. For separation studies, three individual dispersions of MPs and NPs were prepared including acrylonitrile butadiene styrene MPs (ABS MPs), PS NPs, and poly(methyl methacrylate) NPs (PMMA NPs). Experimental results showed that ABS MPs, PMMA NPs, and PS NPs followed cake formation (R^2 = 0.9997), intermediate blocking (R^2 = 0.9998), and standard blocking (R^2 = 0.9907) fouling mechanism, respectively. Here, removal efficiencies for ABS MPs, PMMA NPs, and PS NPs were 98%, 97%, and 95%, respectively. In the case of ABS MPs, constant flux was observed due to easy removal of cake layer from membrane. However, flux reduction was reported in the case of PMMA NPs (385.87 L m^{-2} h^{-1}) and PS NPs (395 L m^{-2} h^{-1}) filtration owing to the adherence of NPs on the membrane **(Figure 10b)**. Overall, this electrospun fibers-based membrane showed good performance for the separation of both MPs and NPs upto 10 cycles, further confirming its reusability. In addition to MPs and NPs removal, the membrane also exhibited selective adsorption capacity of methylene blue (MB) (417 mg g^{-1}) from methylene blue/methyl orange (MB/MO) mixture, thus rendering itself as a promising candidate for water purification.[147] ABS MPs followed the cake formation mechanism with R^2 = 0.9997, while the intermediate blocking mechanism was the predominant mechanism in case of PMMA NPs (R^2 = 0.9998) and PS NPs (R^2 = 0.9907). This may be due to the reason that the size of ABS MPs (624 nm) is greater than the pores of the electrospun composite, which favours the formation of a cake layer and causes extensive relative flux decay (85%) during the filtration of ABS MPs. Here, cake formation was responsible for the efficient

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removal efficiencies of ABS MPs. On the contrary, in the cases of PS NPs and PMMA NPs, electrostatic interactions between negatively charged electrospun composite fibers (ζ potential =−42 mV) and positively charged NPs (ζ potential = +38 mV for PS NPs and ζ potential = +41.7 mV for PMMA NPs) were responsible for the removal of NPs.[147]

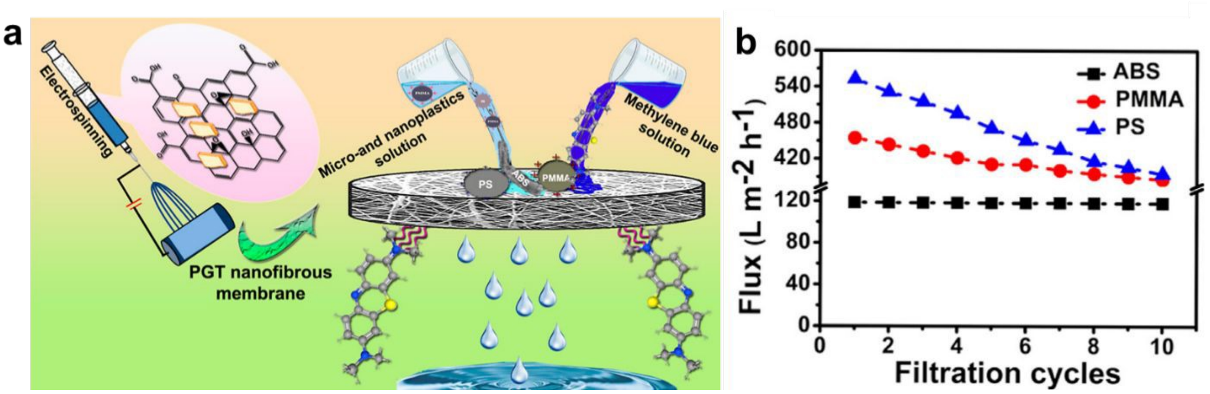

Fig. 10: (a) Schematic representation showing PGT-based electrospun composite membrane for the removal of MPs, NPs, and MB from water. (b) Graph showing variation in flux associated with the reusability study of the PGT electrospun membrane upto 10 filtration cycles. Reproduced with permission.[147] Copyright 2023, Publisher American Chemical Society.

Taking a step further, Rist *et al.* developed self-standing electrospun fiber membranes from bio-based polyamide 6.9 (PA@EFMs).[142] Here, polyamide 6.9 was prepared using hexamethylene diamine and azelaic acid derived from plant oil. PA@EFMs exhibited good mechanical strength and high resistance to solvents. Highly porous architecture of PA@EFMs (pore diameter = 0.55 µm to 1.14 µm) showed remarkable efficacy of >99.7% for the filtration of PS MPs (~679 nm) from water. Additionally, the high hydrophobicity of PA@EFMs facilitated efficient adsorption of oil from water, achieving separation efficacy of 99.9%, while retaining a high permeate flux of 5345 L m^{-2} h^{-1}. Furthermore, the recyclability of PA@EFMs was demonstrated through back flushing, highlighting sustainable potential of these membranes.[142]

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Next, to enhance the structural integrity of electrospun membranes, Gopakumar *et al.* fabricated electrospun fibers of PVA which were crosslinked with GA to form stable nanofibrous membrane.[148] MPs removal efficacy of the membrane was evaluated using PE MPs (5 µm $\leq d \leq$ 25 µm) (~99% removal efficiency) and PS MPs ($d \leq$ 1 µm) (~77% removal efficiency). Furthermore, excellent recovery in performance was observed by backwashing even after five cycles, thus showcasing the durability and reusability of PVA membrane. Additionally, the membrane displayed ~69% removal efficiency for lead at pH 6, demonstrating its capability to handle water pollution from different fronts.[148]

In another direction, Wang *et al.* utilized a layer-by-layer assembly (LbL) technique to fabricate three different membranes (M^0, M^-, and M^+), exhibiting neutral, negative, and positive charges at physiological pH 7.4.[149] These membranes were fabricated by alternatively assembling three layers of PEI and PAA onto hydrolysed electrospun PAN membrane The hydrolysis step was carried out to incorporate negatively charged carboxylic groups, promoting the LbL assembly process. Furthermore, three different sizes of PS NPs (50 nm, 100 nm and 500 nm) were chosen to evaluate the removal efficacy of membranes under very low pressure. Among these modified membranes, M^+ membranes showed excellent performance in removing PS NPs of different sizes, with rejection efficiencies of ~99.4% for 500 nm, ~99.3% for 100 nm, and ~89.9% for 50 nm, while maintaining high flux. It was reported that the removal of PS NPs was primarily governed by electrostatic interactions rather than pore-size exclusion. However, M^- and M^0 membranes had limited rejection efficiency due to their similar or neutral surface charge in connection to PS NPs. Additionally, these membranes successfully retained bacterial contaminants like *E. coli* and *S. aureus* during filtration, which was attributed to smaller pore size of membranes. Thus, the

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multifunctional nature of M^+ membrane presents them as a promising alternative for addressing water pollution by NPs.[149]

Recently, creating eco-friendly materials with advanced 3D printing technology has paved the way for developing tailored water filtration systems, notably beneficial in regions facing clean water scarcity. A persistent challenge in sustainable water treatment is the need of multifunctional, durable, reusable, and high-flux filtration solutions. In this regard, 3D printing has emerged as an efficient technique which has revolutionized design, prototyping, and manufacturing.[150] Flexibility of 3D printing facilitates the designing of complex shapes and customized structures, permitting the development of membranes that are tailored to meet specific requirements of wastewater treatment.[151] Considering the advantages of 3D printing, Fijoł *et al.* constructed fully bio-based and biodegradable polylactic acid (PLA)-based composites water filters via fused deposition modelling (FDM) 3D printing.[152] PLA filters reinforced with TEMPO-oxidized cellulose nanofibers (TCNF) and chitin nanofibers (ChNF) resulted in improved water throughput and mechanical strength as compared to pristine PLA. The biocomposite filters were 3D printed in cylindrical as well as hourglass geometries with varying, multiscale pore architectures. In addition to their structural benefits, the filters exhibited significant adsorption capacity for Cu^{2+} ions (234 and 208 mg/g_{NF} for TCNF and ChNF-reinforced filters, respectively), and PS MPs (50-100 μm) removal from laundry water (54% for TCNF, and 35% for ChNF-reinforced filters). Moreover, metal ion adsorption was mainly governed by electrostatic interactions, while MPs removal was owing to size exclusion and physical bonding to the filter surface. Other than that, PLA-based composite water filters demonstrated recyclability upto multiple times.[152] To the best of our knowledge, only one literature report is available on 3D printed filters for MPs removal,

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emphasizing the need for further research and development to unlock their full potential in real-world wastewater treatment applications.

8. Comparative performance metrics

An extensive literature survey demonstrated that several materials, including hydrogels, electrospun fibers and sponges, have already been reported in literature for the removal of MPs. Here, we have compared and summarized the recent examples of hydrogels, sponges and electrospun fibers based upon their maximum removal efficiency and maximum adsorption capacity **(Table 2)**. In addition to MPs removal, some polymer-based materials have shown good potential to remove NPs based on size exclusion mechanism and electrostatic interactions among the oppositely charged surfaces. These examples have also been included in **Table 2** as degradation of MPs in surrounding medium over time may lead to the formation of NPs, which are also hazardous like MPs due to their small size.

It should be noted that although the electrospun fibers have shown significant potential to remove MPs from the contaminated wastewater, still, they are rarely employed on industrial-scale wastewater treatment plants due to their relatively higher cost in comparison to the hydrogels and sponges. Moreover, despite the growing interest of utilizing polymeric systems in biological and chemical approaches for MPs removal, this area remains less explored than the conventional physical adsorption and filtration systems. This might be due to the difficulty of integrating catalytic and biological functionalities into polymeric matrices while maintaining sufficient permeability, mechanical stability, and long-term operational performance. The biological treatment process using enzyme-functionalized hydrogels usually exhibit limited environmental

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stability, slow degradation kinetics, pH sensitivity, temperature variations, and biofouling-related performance decline, and hence they are rarely explored.

Table 2: Comparative analysis of different materials used for the removal of MPs

Material Used	Type of matrix	Synthesis method	Type of microplastic removed	Size range of targeted MPs	Surface charges on the adsorbent	Pore/ mesh size of the adsorbent	Maximum removal efficiency (%)	Maximum adsorption capacity (mg/g)	Removal mechanism and remarks	Ref.
Lignin/poly(vinyl alcohol) hydrogel	Hydrogel	Functionalized of lignin through phenolization and ammoniation	Polystyrene	5 µm	12 mV to 25 mV	23.77 nm	97.9	288.6	Adsorption	108
			Polyamide				99.7			
ChNFs/ lignin composite hydrogel	Hydrogel	Chemical crosslinking of epichlorohydrin onto the chitin nanofibrils and quaternized Kraft lignin	Polystyrene	166.5 nm	~ + 12.1 mV	> 100 µm	93.7	1790.8	Electrostatic interactions and π-π interactions	153
Polydopamine-modified sodium alginate hydrogel	Hydrogel	Ionic gelation in combination with in situ polydopamine coating/polymerization	Polystyrene	0.1 to 1.5 µm	−19.63 mV	200 nm to 10 µm	99.6	154.57	Chemical adsorption, π-π interactions and electrostatic interactions	90
Gelatin–sodium alginate aerogel	Hydrogel	Chemical crosslinking of gelatin and sodium alginate with glutaraldehyde	Polystyrene	50 µm	-	1–10 µm	90	-	Adsorption, hydrogen bonding, ionic interactions and physical	154

									interception within the aerogel's 3D-network	
Taro stem-sourced cellulose/polypyrrole hybrid aerogel	Hydrogel	Chemical crosslinking and freeze-drying	Polystyrene	200 nm to 2000 nm	-	6.45 nm	91.55	818.06	Physical entrapment, hydrogen bonding, electrostatic interactions, and π-π interactions	[155]
Melamine sponges modified with mussel-inspired polydopamine	Sponge	Surface functionalization of melamine sponge with PDA-PEI coating	Micro and nanoplastics	10 µm	-	160.85 µm	90	302	electrostatic attractions, hydrophobic interactions, and π–π stacking with aromatic polymers	[156]
MOF-based superhydrophobic sponge	Sponge	auto-polymerization of polydopamine followed by coprecipitation of Ni-MOF and PDA-modified sponge	Polystyrene, acrylonitrile-butadiene-styrene, Polyvinyl chloride, polyethylene,	6.5 µm to 150 µm	65 mV	-	95.1	67.4	Electrostatic interactions, Capillary force of inner pores, hydrogen bonding, hydrophobic	[157]

			and polypropylene						interactions, and p–π conjugation	
Chitin-cellulose nanofibers nanosponges	Electrospun nanofiber Sponge	controlled chemical modifications of starch and pectin followed by freeze-thaw technique to design ChCNFs nanosponge	Polystyrene	1 µm	-	-	93.07	116.34	Intraparticle diffusion mechanism	158
Cu/Co-LDHs-based superhydrophobic sponge	Sponge	PDA coating, in situ growth of CuCo-LDHs, and HDTMS surface modification on a pretreated melamine sponge.	Polyethylene and polypropylene	6.5 µm to 150 µm	62 mV	-	100	56.2	Electrostatic attractions, and hydrogen bonding through Intraparticle diffusion mechanisms	159
Wood derived cellulose sponges	Sponge	Delignification of Balsa wood and sulfonation modification of wood derived cellulose sponges	Amine-modified polystyrene microspheres	500 nm	−8.5 mV to -94.4 mV	-	88.8	586.95	electrostatic attraction, hydrogen bonding and *Van der Waals* forces	160

Loofah plant—derived biodegradable superhydrophobic sponge	Sponge	Dip-coating method	Polystyrene microplastics	5 µm	-	-	99.9	569	Monolayer and multilayer adsorption mechanisms	161
Hardwood vessel-inspired chitosan-based sponge	Sponge	Directional freeze-casting of a chitosan solution followed by glutaraldehyde vapor crosslinking, neutralization, and freeze-drying	Polystyrene	1 µm	-	-	94.9	0.26	Adsorption mechanism through electrostatic bonding and p-π interactions	133
Bio-based electrospun polyamide membrane	Electrospun fiber	Melt polycondensation of hexamethylenediamine–azelaic acid, followed by electrospinning and emulsion polymerization	Polystyrene	0.3 µm	-	0.55 µm to 1.14 µm	99.8	-	Surface filtration mechanism with 123% high initial permeability, 99.9% separation efficiency for oil-water emulsion with a high flux of 5345 L m^{-2} h^{-1}, significant antifouling property of the	142

									membrane upto 10 cycles	
Electrospun polyurethane nanofiber membrane	Electrospun fiber	surfactant-free emulsion polymerization followed by electrospinning	Acrylonitrile butadiene styrene, polystyrene and polymethylmethacrylate	90 nm to 825 nm	−42 mV	1309 nm	93%	-	Cake formation, intermediate blocking, and standard blocking fouling mechanisms, 8163 L m^{-2} h^{-1} pressure-driven water flow and 793 L m^{-2} h^{-1} gravity-driven water flux, reusability up to 10 filtration cycles	147
Polyvinyl alcohol nanofibrous membrane	Electrospun fiber	Electrospinning 7% and 10 wt% aqueous PVA solutions at 20 kV using a rotary drum collector under optimized flow rate and spinning conditions	Polyethylene and polystyrene	<50 µm	-	169 nm and 113 nm	77.3	-	Size exclusion mechanism 109 ± 1.67 L m^{-2} min^{-1} membrane flux at 5 psi, reduced fouling time	148

									with 75% ± 3.5% efficiency after 5 round of filtration cycles	
Electrospun polyacrylonitrile membrane	Electrospun fiber	Electrospinning fabrication of PAN nanofibrous membrane followed by layer-by-layer (LbL) surface modification using PEI/PAA polyelectrolyte assembly	Polystyrene	50 nm to 500 nm	+ 18 mV to - 60 mV	-	89.9	-	Electrostatic attraction-driven adsorption and low pressure driven by low pressure, 1452.4 Lm^{-2}·h^{-1} highest flux of the membrane in pure water, promising fouling resistances to bacteria, recyclability of the membrane up to two filtration cycles	[149]

Chitosan/ polyethylene oxide Nanofiber Sponge	Electrospun fiber sponge	Electrospinning, followed by Neutralization of Chitosan-polyethylene oxide Nanofiber and preparation of Chitosan NF Sponges	Poly(ethylene terephthalate)	48.7 µm	-	130 µm	99.46	335.3	-	132
Electrospun polyacrylonitrile membrane	Electrospun fiber	Electrospinning and subsequently surface-modified via layer-by-layer assembly	Polystyrene	50 nm to 500 nm	-	-	89.9	-	Electrostatic attractions	149

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9. Management strategies

The management of MPs contamination for safe environment requires multidimensional approach including the involvement of public, industries, government, and international organizations. In this direction, the first step is to reduce overall plastic consumption. Furthermore, the public awareness programs should be conducted to educate people regarding negative impacts of MPs on our environment and the significance of reducing plastic usage. This can be done by conducting outreach activities in schools, colleges, and community organizations. It has been found that the main source of MPs production is single-use plastic (SUP).[162] To eliminate SUP from environment, people should be encouraged to utilize reusable containers, bottles, and bags.

Further, government, industries, scientists, and individuals must work together in a team to reduce plastic trash, improve plastic waste management, and create new alternatives. Government may consider monitoring and limiting the usage of MPs in cosmetics, cleansing agents, and other personal care products. Further, modern filtration technologies may be implemented to upgrade existing wastewater treatment facilities to eliminate MPs prior to their discharge into waterbodies.[163-164] Additionally, the enhanced producer responsibility can be implemented.[165] Furthermore, government should encourage industry to work on sustainable practices such as minimizing plastic-based packaging and increasing their efforts for plastic waste management. This may be done by rewarding organizations that take significant steps in this direction and invest in eco-friendly alternatives.

Waste management follows five Rs hierarchy: reduce, reuse, recycle, redesign, and recover.[166] Generation of MPs can be reduced by minimizing the production of plastic. Reusing and recycling plastic products further helps in reducing the amount of plastic debris.[167] Further, sustainable management of plastic wastes can also be achieved via upcycling which provides other

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value added materials.[168] A portion of the energy can be recovered from plastic by incineration and approaches like co-fuelling of kilns, offering a practical route to achieving reasonable energy efficiency.[169] These strategies are advantageous as compared to landfill disposal, as they allow energy recovery from plastic to some extent.[166]

Moreover, the molecular redesign of plastics has emerged as a new approach in green chemistry, and should be integrated into the design and life cycle analysis of plastics. In line with this, some alternatives can be used to minimize the usage of excessive chemicals in plastic manufacturing, for example, citrates can be used as a substitute for synthetic plasticizers.[166] Similarly, zeolites can be employed to produce sustainable plastics from bio-based feedstock.[165] Zeolites can turn lactic acid into lactide, which is a key ingredient for making biodegradable plastics like PLA.[165] These innovations not only lower the environmental footprint of plastic production but also support the transition towards a circular and more sustainable materials economy.

10. Challenges and future perspectives

MPs have been globally recognized as a major pollutant leading to a surge in scientific efforts for developing strategies to handle MPs pollution. In this regard, hydrogels have recently emerged as a potential candidate due to their unique swelling properties allowing them to adsorb MPs on their surface and within the porous network, thus removing MPs from water. However, for effective application, hydrogels must be optimized to enhance their adsorption efficiency and mechanical robustness under diverse environmental conditions. It is also crucial to investigate how these hydrogels interact with MPs, specifically under different pH, temperature, and saline conditions. In future, studies should be performed to understand molecular-level interactions focusing on the

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effects of charge, hydrophobicity, surface chemistry, and other physicochemical properties of hydrogels on the adsorption process.

Further, attention should be given on tuning the chemical structure of polymeric hydrogels, sponges, and electrospun fibers for selectivity towards specific type of MPs. Incorporation of stimuli responsive feature in these adsorbent materials may further help in easy capture and removal of MPs.[126] In addition to adsorption of MPs, the overall composition of these materials should be chosen in such a way that degradation of MPs can also be achieved. This direction can be explored by embedding photocatalytic nanomaterials inside the polymeric matrix.[122] Moreover, sustainability, biodegradability, reusability, processability, scalability, and reduced secondary pollution are important criteria for analyzing the practical applicability of these materials. Hence, a thorough analysis of the existing materials in these aspects is the need of the hour.

It may be noted that the MPs are comparably easier to be detected, monitored, and removed using the existing analytical and separation techniques due to their micron-scale dimensions. However, the NPs pose a significant challenge even with the existing techniques due to their smaller size. Additionally, NPs exhibit more ecotoxicological impacts and can easily penetrate through the cell membranes as compared to MPs. Due to large surface to volume ratio, NPs can exhibit higher uptake of toxic chemicals as compared to the same mass of MPs, and may produce Trojan horse effect.[170] Although, the advanced characterization methods such as electron microscopy, Raman spectroscopy, dynamic light scattering analysis, Fourier-transform infrared spectroscopy and thermal analysis have been explored for quantification and detection of NPs, however, their precise identification and monitoring remain a major challenge, and requires further technological advancements.

Effective utilization of the aforementioned adsorbent materials on large scale is another hurdle. Although lab-scale results are promising, scaling up to larger operations demands cost effectiveness, easy material accessibility, and broad applicability of a single system. Moreover, strategies for incorporating these materials into filters, membranes, or reactors utilized in municipal and industrial water treatment should be investigated to facilitate their large-scale utilization. Further, exploration of AI in the context of wastewater treatment still remains in nascent stage. Application of AI models in this context may help in automation of such water treatment facilities resulting in easy and low-cost operations.[171-172] This can also help in predicting the removal efficiency of hydrogels, sponges, and electrospun fibers-based materials under complex and continuously varying waste water conditions.[173]

Finally, the researchers should concentrate on fabricating biodegradable hydrogels, sponges, and electrospun fibers-based materials that can be readily disposed of or repurposed at the end of their lifecycle, thereby minimizing secondary environmental pollution. Considering *waste-to-wealth* approach, different waste resources or biomass can be employed for fabrication of these materials. Additionally, new approaches must be developed to reprocess used materials into other useful value-added materials that can be further utilized for the benefit of the society. Moreover, long-term ecological and biological impact of employing these materials for MPs and NPs extraction should also be evaluated along with their life cycle analysis.

11. Conclusion

MPs pollution has become an alarming environmental concern due to the widespread applicability of plastic products and adversely impact both terrestrial and aquatic ecosystems. This review highlights the importance of different removal methods and recently explored materials. Distinct

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studies represented the effectiveness of the adsorption method for MPs removal. In this regard, polymeric hydrogels, sponges, and electrospun fibers have attracted a significant interest as a potential adsorbent material owing to their highly porous architecture which provides more surface area to capture MPs and NPs. Recent laboratory studies have shown > 99% MPs removal efficiencies using polydopamine-modified sodium alginate hydrogel. Similarly, Loofah plant—derived biodegradable superhydrophobic sponge have shown > 99.9% MPs removal potential. Further, in some cases, the incorporation of different additives in these materials improves their mechanical strength and helps in shortening the adsorption time, while also offering the capability of MPs degradation. However, the research on polymeric hydrogels, sponges, and electrospun fibers for MPs and NPs removal still remains in its nascent stage. Continued research and innovation in these materials are essential for optimizing their usage and ensuring their economic feasibility to take a step further for moving towards global sustainable development goals.

Abbreviations

AA: Acrylic acid; ABS: Acrylonitrile butadiene styrene; AM: Acrylamide; AOPs: Advanced oxidation processes; BC: Bacterial cellulose; BGCS: Double crosslinked chitosan-based sponges; Ch: Chitin; ChCN: Chitin/O-C_3N_4; ChGO: Graphene oxide-incorporated chitin; ChGO-CL: Chitin/GO/carboxymethyl cellulose; ChGO-CS: Chitin/GO/chitosan; ChNF: Chitin nanofibers; ChPANIs: Chitin-PANIs sponge; CNF: Cellulose nanofiber; CS: Chitosan; CSNF: Chitosan nanofiber sponges; Cu-POM: Copper substituted polyoxometalate; FDM: Fused deposition modelling; Fe^{3+}@PS: Iron doped polystyrene microplastics; GA: Glutaraldehyde; GO: Graphene oxide; GO_5/CS/GP: Graphene oxide/chitin/genipin sponges; GOMt: Graphene oxide-montmorillonite; GP: Genipin; IPNs: Interpenetrating networks; LbL assembly: Layer-by-layer

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assembly; LG-GH-PVA: lignin/epichlorohydrin/polyvinyl alcohol; MA: Methacrylic acid; MB: Methylene blue; MBA: *N, N'*-methylene bisacrylamide; MO: Methylene orange; MOFs: Metal-organic frameworks; MPs: Microplastics; NIPAM: N-isopropylacrylamide; NPs: Nanoplastics; NVCL: N-vinyl caprolactam; NVP: N-vinylpyrrolidone; PA: Polyamide; PAA: Polyacrylic acid; PAHs: Polycyclic aromatic hydrocarbons; PAN: Polyacrylonitrile; PANI: Polyaniline; PC: Polycarbonate; PCBs: Polychlorinated biphenyls; PDA: Polydopamine; PE: Polyethylene; PEG: Polyethylene glycol; PEI: Polyethylene imine; PA@EFMs: Polyamide electrospun fibers-based membranes; PEI@PDA: Polyethyleneimine and polydopamine copolymer; PET: Polyethylene terephthalate; pGel@IPN: Cu-POM infused triple interpenetrating network hydrogel; PGT membrane: Polyurethane/graphene oxide-montmorillonite electrospun composite membrane; PLA: Polylactic acid; PMMA: Polymethyl methacrylate nanoplastics; PNIPAM: Poly(N-isopropyl acrylamide); PP: Polypropylene; PS: Polystyrene; PS-COOH: Carboxylate-functionalized polystyrene; PS-NH_2: Amine-functionalized polystyrene; PU: Polyurethane; PVA: Polyvinyl alcohol; PVC: Polyvinyl chloride; PVDF: Polyvinylidene fluoride; ROS: Reactive oxygen species; SEM: Scanning Electron Microscopy; SUP: Single-use plastic; TCNF: TEMPO-oxidized cellulose nanofibers.

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Notes

The authors declare no conflicts of interest.

Acknowledgement

GA acknowledges the financial support from the Anusandhan National Research Foundation ANRF-SERB, India (Grant No. WEA/2023/000001). PB thanks Council of Scientific and Industrial Research (CSIR) for fellowship.

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Data Availability Statement

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.