## This is the preprint of the contribution published as:

**Mehmood, T.**, Mustafa, B., **Mackenzie, K.**, Ali, W., Sabir, R.I., Anum, W., Gaurav, G.K., Riaz, U., Xinghui, L., Peng, L. (2023):
Recent developments in microplastic contaminated water treatment: Progress and prospects of carbon-based two-dimensional materials for membranes separation
*Chemosphere* **316** , art. 137704

## The publisher's version is available at:

http://dx.doi.org/10.1016/j.chemosphere.2022.137704

# Recent developments in microplastic contaminated water treatment: Progress and prospects of carbon-based two-dimensional materials for membranes separation

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## Abstract

Micro(nano)plastics pollution is a noxious menace not only for mankind but also for marine life, as removing microplastics (MPs) is challenging due to their physiochemical properties, composition, and response toward salinity and pH. This review provides a detailed assessment of the MPs pollution in different water types, environmental implications, and corresponding treatment strategies. With the advancement in nanotechnology, mitigation strategies for aqueous pollution are seen, especially due to the fabrication of nanosheets/membranes mostly utilized as a filtration process. Two-dimensional (2D) materials are increasingly used for membranes due to their diverse structure, affinity, cost-effectiveness, and, most importantly, removal efficiency. The popular 2D materials used for membrane-based organic and inorganic pollutants from water mainly include graphene and

MXenes however their effectiveness for MPs removal is still in its infancy. Albeit, the available literature asserts a 70-99% success rate in micro/nano plastics removal achieved through membranes fabricated via graphene oxide (GO), reduced graphene oxide (rGO) and MXene membranes. This review examined existing membrane separation strategies for MPs removal, focusing on the structural properties of 2D materials, composite, and how they adsorb pollutants and underlying physicochemical mechanisms. Since MPs and other contaminants commonly coexist in the natural environment, a brief examination of the response of 2D membranes to MPs removal was also conducted. In addition, the influencing factors regulate MPs removal performance of membranes by impacting their two main operating routes (filtration and adsorption). Finally, significant limitations, research gaps, and future prospects of 2D material-based membranes for effectively removing MPs are also proposed. The conclusion is that the success of 2D material is strongly linked to the types, size of MPs, and characteristics of aqueous media. Future perspectives talk about the problems that need to be solved to get 2D material-based membranes out of the lab and onto the market.

**Keywords:** Microplastics, Membranes, 2D material, Graphene, MXene

# 1 Introduction

European plastics production analyses reported that global plastics output consumption and waste production in 2019 was estimated to be over 370 million tons (Mt) (Association of Plastic Manufacturers, 2020). More specifically, it is estimated that more than half of the yearly worldwide plastic produced, about 300 Mt (compared to 1.5 Mt produced in 1950), has ended up in landfills (Tiseo, 2020). A total of 12 billion tons of plastic garbage is expected to be dumped in landfills or released into the environment by 2050. Plastic waste accounts for much of the fuel used in trash incinerators and emits carbon dioxide. Worldwide, large amounts of plastic trash are discarded into the environment, contributing to the problem of white pollution (Editorial, 2021). According to an earlier study, over 8 million tons of plastic trash enter marine each year, which is projected to increase even more by 2030 (Zhang et al., 2020a). Furthermore, because plastics are difficult to breakdown organically, poorly discarded plastics end up in the ocean and build over time through land waste, sewage runoff, rivers, and wind.

Plastics collected in the environment can progressively degrade into MPs and even nanoplastics (NPs). Prior research on plastic trash focused on the ocean's surface but now includes deeper seas, sediments, freshwater, soil, air, and biological systems (Geyer et al., 2017). In the recent decade, concern has grown regarding MPs and Nps in the environment. MPs are fewer than 5 mm synthetic plastic particles originating from the breakdown of larger resins (Mehmood and Peng, 2022). MPs are released from primary (directly from the source) or secondary source form by UV radiation (photo-oxidation), crevice corrosion (e.g., wave impact), and microbial biodegradation (Chamas et al., 2020). Different sources of MPs actively contaminating water bodies are shown in Figure 1. Micro- and neoplastic particles can enter the food chain and damage humans (Smith et al., 2018). Thus, only reducing plastic emissions and expanding collection efforts will reduce plastic pollution in water bodies.

MPs have been removed from water in several ways. Although wastewater treatment facilities (WWTPs) can remove 95% of MPs (Malankowska et al., 2021), some can still permeate the aquatic environment. Due to the volume of wastewater discharged by WWTPs, 15,000 to 4.5 million MPs items are released to surface water daily, regardless of treatment efficacy (Yang et al., 2019). The common treatments in WWTPs, such as coagulation, did not prove effective for MPs removal (Ma et al., 2019a). Ozonation is a modern method for treating MPs; nevertheless, it is mostly ineffective in some situations since it primarily breaks down large MPs into smaller ones, increasing output concentration relative to input (Wang et al., 2020c).

Moreover, inadequate ozone management can lead to the formation of harmful intermediate compounds. Although wastewater treatment plants have implemented multi-stage water treatment, many MPs are still released into the water system. Previous research has found that primary treatment at WWTPs may eliminate 45 percent of MPs (Wu et al., 2016). Following treatment, 50% of MPs in wastewater may be removed (Pannetier et al., 2019).

From a practical standpoint, such advanced multi-stage water treatment (membrane filtration followed by primary and secondary treatment) severely limits its application on a broad scale. Membrane filtration methods are classified as nanofiltration (NF), microfiltration (MF), ultrafiltration (UF), and reverse osmosis (RO) based on separation membranes' distinct architectures and properties (Pauzan et al., 2022).

To date, many tactics or materials have been used to address this issue. MPs have been treated using a variety of approaches, including coagulation (Auta et al., 2017), extraction (Hu and Palić, 2020), and biological degradation (Revel et al., 2018; Mehmood et al., 2022). Furthermore, Wang et al. (2019b) used a $TiO_2$-based photocatalytic micromotor for MPs removal. Chen et al. (2020b) developed a novel acetone-assisted manufacturing technique for a variety of Zr-MOF foam materials, as well as their outstanding use in MPs removal simulation. Though these approaches have made significant progress toward efficient MPs removal, there is still a significant obstacle to overcome, as each method has its own limitations, such as the inability to handle small MPs or the inapplicability for large MPs, difficulty in large-scale processing, high energy consumption, or incapacity to operate in specific environments. As a result, there is an urgent need to develop novel technologies to effectively remove MPs (Talvitie et al., 2017a). Following tertiary treatment, around 2% of MPs with particle sizes smaller than 20 μm remain observable (Carr et al., 2016; Murphy et al., 2016), and this component of the MPs must be eliminated using membrane technology.

Depending on the size of the MPs, membrane type, and pore shape, membrane filtration can restrict the escape of MPs from wastewater. Researchers have proposed combined reverse osmosis (RO) and ultrafiltration (UF) membranes for virtually entirely remove MPs (Carr et al., 2016). Most membrane materials are organic polymers with low chemical resistance, restricted water permeability, and contamination risk (Murphy et al., 2016). Therefore, membranes with high water flow and MPs removal efficiency are needed. Inorganic two-dimensional (2D) multilayer membranes with excellent chemical and heat resistance, such as GO and MXene, provide significant separation and purification potential (Talvitie et al., 2015; Sun et al., 2019). In addition to assisting the membrane in the removal of MPs via adsorption, degradation, catalytic degradation, and filtration (Meng et al., 2022; Yang et al., 2022a), these materials are also used to reinforce the membrane material and prevent MPs release (Huang et al., 2021).

Nanofiltration membrane technology is widely used for desalination and water purification. Membrane filtration may limit the escape of MPs depending on their size, the membrane type, and pore structure, making it a very practical solution to the MPs in wastewater problems. Luogo et al. (2022) stated that under specific experimental conditions, two ceramic membranes composed of SiC and $ZrO_2$ have MPs removal rates from washing wastewater of 99.2 and 98.55 percent, respectively. Fryczkowska et al. (2021) found that the composite membranes made of polyacrylonitrile and rGO (rGO/PAN) had high rejection effectiveness for removing MPs from industrial effluent. Two-dimensional inorganic materials rely on interlayer gaps for aqueous separation, resulting in limited water flux.

Given MPs' direct and indirect hazards, searching for an efficient and environmentally friendly treatment method is imminent. Currently, the emission inventories (Bradney et al., 2019), distribution (Fu et al., 2020), transport (Guo et al., 2020), toxicity (Chen et al., 2020a), accumulation (Xu et al., 2020a), and risk (Ma et al., 2020b) are significant issues. However, removing MPs from aqueous media impedes water management, environmental sustainability, and health. Numerous factors, such as the type, composition, aging, coexistence with other pollutants, and membrane material, impact MPs removal. To the best of our knowledge, no complete evaluation of available literature on 2D material-based membranes and their efficiency for MPs removal has been conducted. This review addressed the recent literature on MPs contamination in different types of water, including groundwater, drinking water, marine water, wastewater, and stormwater, and the environmental implications of

MPs pollution. The current review focuses on perspectives and breakthroughs in synthesis, characterization, and variables influencing various 2D material-based membranes such as graphene, GO, rGO, and MXenes. A summary of factors impacting filtration and adsorption efficiencies of 2D material-based membranes is also included. This review presents both professionals and novices with broad principles and proposals for future study by combining and critically analyzing recent accomplishments in MPs removal technologies research on a variety of 2D material-based membranes.

# 2 MPs contamination of water

A recent review concluded that MPs (<5 mm) pollution had become a global environmental problem, and its accumulation in the environment is increasing, and the worldwide share of MPs in plastic pollutants will reach 13.2% by 2060 (Sharma et al., 2021). Polyamide (PA), polystyrene (PS), polyethylene terephthalate (PET), polypropylene (PP), polyvinyl chloride (PVC), and polyethylene (PE) are major plastics types found in water (Browne et al., 2008). Numerous studies summarized in table 1 have shown that MPs have polluted soils, water (including rivers, lakes, and oceans), and air (Fischer et al., 2016; Mehmood and Peng, 2022). The distribution of MPs is ubiquitous, and its distribution involves all latitudes of the Earth, even the Antarctic and the Arctic. Therefore, MPs contamination in water is particularly of major concern in the scientific community. In this section, the MPs contamination in different water types is discussed in detail.

## 2.1 MPs in wastewater and sewage

Wastewater treatment plants and combined sewer overflows are common pathways for MPs to enter the environment (McDougall et al., 2022), through treated effluent discharge, particularly after substantial rainfalls (Polanco et al., 2020). Wastewaters usually comprise different MPs forms including PE, PET, and PA (Sun et al., 2019). MPs quantities vary in treated and un-treated wastewater; for instance, the number of plastic particles was estimated as 447 particles $L^{-1}$ in treated effluents (Simon et al., 2018), while untreated wastewater had a huge number of > 10,000 particles $L^{-1}$ (particles retained on a 10 μm steel filter). In addition, cosmetic products, such as toothpaste and facial cleaners with added microbeads, directly add MPs into the wastewater. Along with synthetic clothing made of polyester and nylon, the washing process results in thousands of shed MPs fibers that accumulate in the sewage water. The water then enters wastewater treatment plants where they are filtered. Most water that comes through the sewage system this way is from households and other municipal services such as laundry and textile services. Other similar products like ropes can also shed plastic fibers into local sewage. Road paint particles can also be introduced into sewage as runoff (Coppock et al., 2017). Although 99.9% of the MPs can be filtered from wastewater, the MPs extracted can end up in sludge used as fertilizer, and MPs, are washed away after use and pollute the marine habitats (Lares et al., 2018).

Within the collected MPs found in the sewage system, it was found that MPs fibers were much more common than other MPs particles. One study found that polyester fibers made up 79.1% of all the MPs they had collected in their sample of multiple water plants (Lares et al., 2018). They were described to be equally thick and bent. Of the MPs polymers found in sewage plants, the most abundant was PE, which constituted 63.9% of the MPs polymers. The next most common type of plastic is PP. They make up most of the plastic particles found (Figure 1). They were the most present in sewage wastewater and came in different fragment shapes. Although the overall concentration of MPs particles is relatively low, the general discharge of MPs builds over time.

Wastewater also contains a high concentration of other elements like dissolved organic species. Such elements can influence the adsorption capability of MPs. For instance, humic acid can reduce or enhance the pharmaceutical pollutants' adsorption on MPs (Xu et al., 2021b; Upadhyay et al., 2022). However, pharmaceutical adsorption also depends on certain environmental factors like pH, salinity level, and temperature in wastewater (Puckowski et al., 2021). The excessive use and production of plastics threaten environmental sustainability and disturb aquatic life stability.

## 2.2 MPs in stormwater

Rainfall transports MPs into surface water from urban units such as garbage or litter, highways, soils, landfills, and biosolid-applied land (Koutnik et al., 2022). Urban road flush is commonly thought to be the principal source of MPs in stormwater control measures (SCM), which can catch these MPs and limit runoff, remove stormwater pollutants such as sediments, and minimize pollution downstream SCM (Mehmood et al., 2021; Österlund et al., 2022). Ziajaromi et al. (2020a) stated that 0-3500 MP items/kg were present in stormwater retention ponds in Australia; however, in river shore sediments, it was estimated as 4000 particles/kg and 11-3153 items/kg by Klein et al. Klein et al. (2015) and Liu et al. Liu et al. (2019c) respectively. In earlier studies, MPs concentrations in regions with no/less stormwater have also been reported (Free et al., 2014). According to recent studies, many researchers, however, focused on enhanced sampling techniques and advancements in detecting technologies and discovered a significant rise in MPs. In another study, Liu et al. (2019b) identified 1511-127, 986 plastic items per kg in stormwater retention ponds (in Denmark). Dikareva and Simon (2019) identified land use and urbanization as the reasons for the upsurge in MPs in urban tributaries. Despite evidence of MPs subsurface mobility after intermittent rainwater infiltration, the extent to which accumulated MPs can travel lower in SCM in field settings is unknown (Koutnik, 2022).

## 2.3 MPs in marine and seawater

Prior research has shown the sources of plastic litter and other toxins in the lagoon, lakes, rivers, and other bodies of water. MPs contribute to water pollution through various demodectic and economic activities, including residential rubbish dumping, aquaculture, fishing, river discharge, and commerce (Jiang et al., 2022). Plastic debris is everywhere, including the Indian Ocean and North Atlantic, Mediterranean Sea, even extending to the south of the world, i.e., the South Atlantic and South Pacific. Researchers estimate that over five trillion bits of plastic, weighing over 250,000 tons, have found their way into the ocean (Eriksen et al., 2014). Likewise, Peeken et al. (2018) and Chiba et al. (2018) reported that due to MPs transportation in the environment by water, dry and wet deposition, they'd reached extreme regions, including the Mariana Trench (the deepest part of the ocean), and Northern and Southern Polar Ice, respectively. Marine organisms suffer significant risks from land-based ocean garbage and lost or abandoned fishing gear due to being enclosed in plastic containers, trapped in fishing nets, and the intake of plastic particles. Seabirds, turtles, crabs, and fish are the most endangered species (Vegter et al., 2014). According to MacArthur, if current trends continue, more plastic will be in the oceans than fish by 2050 (MacArthur, 2017). Plastic trash accounts for eighty percent of marine contamination, with an estimated 8 to 10 million metric tons yearly introducing plastic into seas and oceans. Therefore, it is vibrant that people must reconsider their attitude toward plastic garbage and begin assisting in protecting aquatic ecosystems from pollution (Mehmood et al., 2023).

## 2.4 MPs in groundwater and drinking water

Although water treatment systems are increasingly updating for MPs removal, a substantial amount of MPs is an existing threat that has been reported to become groundwater and contaminated subsurface and drinking water (Peng et al., 2022b). The possibility of groundwater contamination is influenced by the total amount of MPs released in subsurface water and soil and the portion of those particles that are accessible for downward transport. Therefore, it is crucial to consider every process that adds to the MPs inventory and evaluate how likely these mechanisms may cause groundwater contamination. According to Koelmans et al. (2019), the abundance of MPs in some water bodies was lower than in tap water and bottled water, with groundwater having the lowest quantity of MPs (10-2 items $L^{-1}$). This finding suggests that other factors, such as the tap water supply or packaging process, may also impact the source of MPs in drinking water. Based on the sort of MPs found in bottled water, another research (Schymanski et al., 2018) identified bottle components as a leading potential source of MPs production (PET and polyester).

Zhang et al. (2020b) addressed that MPs larger than 50 μm could be removed during water treatment, with removal efficiencies ranging from 25% to 90% due to differences in water treatment methods. No MPs were detected in tap water in Italy and Denmark, and the highest concentration of MPs in tap water was 9.2 items $L^{-1}$ in the U.S. The maximum attention of MPs in bottled water was 5.4107 items $L^{-1}$, compared to water in reusable bottles, which contained substantially more MPs than water in single-use bottles. In comparison, water in reusable bottles has more MPs than water in single-use bottles.

A direct comparison of MPs abundance in different studies is difficult due to differences in the type of filter membrane and MPs identification methods in the water treatment process. Moreover, there is a phenomenon in which the concentration of MPs in tap water is more significant than that in bottled water, but the abundance is lower; this is likely owing to the lower detection limit of MPs detection methods in bottled water. In other words, the widely used FTIR method for tap water cannot identify MPs smaller than 10 μm, possibly resulting in an underestimation of MPs abundance.

# 3 Membranes based MPs removal technologies

In recent years substantial advancement in MPs treatment strategies has been achieved (Figure 2). A detailed description of these methods is given in the supplementary file, while a brief summary of these methods and their effectiveness in MPs removal is presented in table 2. Different strategies which are currently being used for MPs removal are presented. Membrane separation is also used to remove MPs.

Membrane filtration can restrict the escape of MPs based on MP size, membrane type, and porosities, which is an efficient solution to tackle the problem of MPs in wastewater . Evidence that emphasizes centralized processes like membrane bioreactors, coagulation, reverse osmosis, and dissolved flotation is more effective in removing MPs from wastewater than others (Shahi et al., 2020). Likewise, a combination of different approaches in membrane separation (generally MF or UF) has also been demonstrated (Mustafa et al., 2022b). One example is biodegradation in membrane bioreactors (rectors that use biological catalysts such as bacteria and enzymes) (Xiao et al., 2019). Complete MPs destruction was accomplished when MBR was combined with a preliminary anaerobic treatment and followed by a RO filter (Balabanič et al., 2012). PET was utilized as a carbon source by *Idonella sakaiensis*, a bacterial species, which degraded it into ethylene glycol and terephthalic acid (Yoshida et al., 2016). In a similar vein, *Euphasia superba* (Antarctic Krill) used an enzyme complex to degrade larger plastics from 31.5 μm to less than 1 μm (Dawson et al., 2018). Barth et al. (2015) also showed enzymatic PET degradation, demonstrating that enzymes may be easily included in the MBR . Hence, MPs degradation in the enzymatic membrane reactor is likely in the future. In Finland, MBR and UF combined (area: 8 $m^2$ and pore size: 0.4 μm) removed 99.9% of MPs, which was much more significant than average activated sludge (CAS) (Talvitie et al., 2017b).

A special membrane with controlled permeability is used in the membrane separation method. Small molecules preferentially penetrate the membrane under external pressure, preventing giant molecules from flowing through, resulting in the filtration of the multi-component mixture.

Because of its perks and key aspects such as minimal price, high efficiency, simple handling, high energy efficiency, and environmentally friendly nature, the UF membrane is believed to become a more significant role in the treatment of surface water (Zhang et al., 2018b; Lou et al., 2020). However, UF technologies can still not completely remove pollutants from the water. As a result, an efficient, cost-effective, and easy technique to remove micropollutants is still required to ensure the safety of drinking water. So far, only a few papers have investigated MPs removal through coagulation and UF for potable water production (Ma et al., 2019b; Xu et al., 2021a). Ma et al. (2019b) investigated the removal of different sizes of PE particles (0.5-5 mm) with UF and iron-based coagulation. Although PE is the most common plastic contamination detected in water, its density (0.92-0.97 g/cm3) is so close to water that removing it is challenging. After thickening, PE-MPs particle elimination was just below 15%, showing inadequate coagulation. Polyacrylamide (PAM) boosted coagulation performance and enhanced PE particle clearance from 13% to 91% Figure 3.

Likewise, in some cases capturing MPs with only membranes, on the other hand, is challenging. Any filter with a mesh size greater than 250 microns may enable some MPs to pass through (Lares et al., 2018). Another issue with mesh filters is clogging. Because the pores are so tiny, larger particles and any natural organic polymers present in the water sample with the MPs can easily block filters. It is difficult to strike a balance between filtering all plastics and cleaning the filter so that it may be reused. Given how important UF and coagulation operations are to making drinkable water, there needs to be a more in-depth look at how MPs are removed throughout complexation and UF operations (Shannon et al., 2008).

In urban and industrial WWTPs, reverse osmosis (RO) is used to remove salts. Micro organic pollutants and potentially toxic metals from water are removed using pore size > 2 nm non-porous or NF membranes. When a highly concentrated water solution is put under high pressure (10–100 bar), the water is forced through a semipermeable membrane, leaving the sludge in a highly concentrated liquid medium. Ziajahromi et al. (2017) reported on the effectiveness of the RO technique for MPs removal. However, the RO is less effective in the removal of MPs present in fibers form. Advanced polyester (alkyd resin) used in paints also produces MPs. Scientists ascribed MPs tracking to membrane deflections or minor pipe gaps, highlighting the necessity for ad hoc MPs removal technology. Combining RO with membrane bioreactor technology is the most effective for MPs cleanup. Membrane fouling affects reliability (Goh et al., 2018). A pretreatment process is also needed to keep flux rates steady, prevent fouling in large-scale RO desalination systems, reduce the number of times membranes need to be cleaned, and make RO equipment last longer. Fouling can also be reduced by mechanical and chemical cleaning, membrane backwashing, changing the surface, improving hydrophilicity, and using new membrane materials (Mustafa et al., 2022a).

Nanofiltration is another membrane separation process known for its lower operating pressure, high molecular selectivity, and lesser salt rejection than the RO used in an advanced technique for organic micropollutants (OMPs) containing wastewater treatment (Shen et al., 2017). It is a pressure-driven membrane positioned between RO and UF liquid separation technology. Most commercially available NF membranes are synthesized using an interfacial polymerization approach (Guo et al., 2019). Luogo et al. successfully removed 98.55% and 99.2% of MPs $ZrO_2$ and used SiC in ceramic membranes (Luogo et al., 2022). Likewise, polyacrylonitrile and rGO (rGO/PAN) composite membranes also effectively removed MPs from industrial effluent (Fryczkowska and Przywara, 2021). Noticeably, NF and RO have some limitations, including partial removal of pollutants via size exclusion, adsorption, and charge interaction and work at high pressure (up to 10-60 bar), increasing their operating cost (Yang et al., 2016; Nguyen et al., 2021). A detailed comparison of the different characteristics of these membranes is given in Table 3. On the other hand, UF can be operated at lower pressure but is inefficient in removing some pollutants (Zhang et al., 2022) and MPs. Therefore, although UF technologies cannot altogether remove these contaminants from the water, however, it is still more efficient than NF membranes which have several technical issues, including low chemical resistance, a short lifespan, a complicated construction technique, and a high price (Shen et al., 2017; Ma et al., 2020a).

Time-consuming processes, high energy consumption, and significant investment hamper the application options. The membrane separation technology (MST), which is widely used in water purification and other industries, offers reliable operation, energy efficiency, and no secondary environmental impact (Wu et al., 2010). Noticeably the proposed dynamic membrane (DM) membranes are more energy efficient and showed 16 times less transmembrane pressure (80 to 180 mm) for water than conventional MF and UF. The use of DM technology to remove MPs has also been investigated (Li et al., 2018), as DM is good for eliminating low-density/poorly settling particles. Wastewater strained through an assisting membrane creates a cake layer as a supplementary barrier. Because: 1) it uses cheaper ingredients than traditional membranes, like the grid, non-woven textile, knitted filter thread, and stainless-steel wires; 2) it does not need additional chemicals, making any secondary pollutants; and 3) the experimental setup is usually smaller than in traditional membrane reactors (e.g., for UF and MBR). A lab-scale DM filtration device and a gravity-driven operation were employed to remove microparticles from synthetic wastewater using DM technology. The tap and synthetic water treatment with diatomite (D90 = 90.5 µm) demonstrated 90 percent of trapped MPs were of similar size (microparticles). In 20 minutes, experiment, the effluent turbidity was reduced to 1 NTU (Nephelometric Turbidity Unit), showing the DM's promising elimination of microparticles.

In addition to chemical precipitation, redox, electrolysis, and membrane permeation, there are various ways to heavy metal treatment (Heo et al., 2012). In practice, such sophisticated multi-stage water treatment (membrane filtration followed by primary and secondary treatment) significantly restricts its broad use. So far, many approaches or materials have been used to overcome this issue. MPs have been treated using a variety of techniques, including coagulation (Leiknes, 2009), extraction (Misra et al., 2020), and biological degradation (Paço et al., 2017). Also, Yifa Chen et al. (2020b) came up with a new way to make Zr-MOF foam with the help of acetone. These materials are great for simulating the withdrawal of MPs. In the same way, Juliane Simmchen et al. notably used a photocatalytic micromotor made of $TiO_2$ to remove MPs (Wang et al., 2019b). However, although these strategies have progressed significantly to efficient MPs separation, a substantial challenge still exists to overcome. Each technique has its own constraints, such as the inability to handle tiny MPs or massive MPs, complexity in large-scale handling, high energy requirement, or imitation to operate in specific environments. In order to effectively remove MPs in difficult environmental circumstances, novel ways must be developed.

Membranes can be constructed from polymers or metals, or ceramics. Ceramic membranes are constructed of alumina, glassy solids, silicon carbide, titania, and zirconia oxides. Synthetic organic polymers form polymeric membranes. When compared to polymeric membranes, ceramic membranes increased operational costs by 24-54%. Polymeric membranes have a higher total permeability than ceramic membranes. Furthermore, ceramic membranes are more mechanically fragile than polymeric membranes (Sagle and Freeman, 2004).

Before a decade, few articles on 2D carbon materials had been produced. These 2D materials showed excellent performance in removing diverse pollutants (Table S1). However, their effectiveness for MPs removal is in its early stages, describing the material's fascinating features, some of which showed superior performance including graphene and MXenes. The current state and future prospects of 2D carbon material materials for membrane-based MPs removal are presented here.

# 4 Progress and prospects of carbon-based two-dimensional materials for membrane

**separation**

On closer inspection, almost half of the carbon compounds belong to the graphene family. Graphene is a highly transparent, chemically stable material with improved carrier flexibility and outstanding electrical and thermal conductivity. It is ideally composed of a honeycomb-structured monolayer of $sp^2$-hybridized carbon atoms (Ferrari et al., 2015; Zhang et al., 2018a).

Membranes consisting of crystalline dense 2D materials are preferred for liquid phase filtration procedures such as NF, forward osmosis, and pervaporation since MPs vary in size and must be filtered (Kim et al., 2021; Mustafa et al., 2022b). Because the layers are close together on a sub-nanometer scale, dense 2DMs are commonly employed in membrane fabrication. The nano-channels are big enough to keep the solvent away from the larger organic molecules (Seo et al., 2021). A comparison and potential of different 2D materials, including graphene and MXene, in the removal of MPs, are summarized in Table 4. The following section examines the current state of 2D materials, such as graphene and MXene, as well as their future potential.

## 4.1 Application of graphene-based membranes in MPs removal

Graphene has lately sparked tremendous attention because of its higher chemical stability, hydrophilicity, and antifouling characteristics; graphene-based membranes outperform conventional membranes (Mustafa et al., 2022a). In comparison to other conventional adsorbents such as activated carbon, zeolite molecular sieve, and activated alumina (Altmann et al., 2016), graphene has gained the interest of researchers due to its unique properties. For instance, graphene has a high tolerance to harsh environmental conditions due to its unique hexagonal honeycomb structure, making it a strong candidate for pollutant remediation across a wide pH range. Likewise, $sp^2$-hybridized carbon atoms formation and intrinsic hydrophobicity, a large specific surface area, strong modifiability, high adsorption capacity, and abundant oxygen functional groups of graphene are considered desirable attributed for a material used to capture organic compounds such as polycyclic aromatic hydrocarbons (PAHs), methylene blue (MB), and neutral red. Therefore, other graphene-like carbon materials (GCs) with similar structures have been extensively researched for water purification (Peng et al., 2022a). Recently, oxygen-doped carbon nitride (O-$C_3N_4$), which has a similar design to GO, was found to be capable of removing PS attributed to the $\pi$-$\pi$ linkage in a pH range of 4-10 (Sun et al., 2021a). Although carbon nanotubes (CNTs) are another highly absorbent substance, graphene is less costly and easier to manufacture and poses less environmental concern. Graphene, a sort of remarkable 2D nanomaterial, is exceptionally effective in membrane manufacturing for the separation of salts, organic and inorganic pollutants (Li et al., 2022; Mustafa et al., 2022a), oil-water emulsion (Chen et al., 2022), and gas (Salahuddin et al., 2022), as well as in water treatment (Kamran et al., 2022).

Graphene-based membranes are commonly used at present water treatment plants (Yuan et al., 2020). Sun et al. (2020) found that chitin and graphene-made sponges had high reusability and MPs removal performance of 89.8%, even though graphene sponges are expensive if mass-produced for wastewater treatment. Other graphene materials include GO and rGO.

Graphene is the toughest, lightest, thinnest substance due to structural features such as interlayer crosslinks created by covalent bonding between side atoms of distinct sheets and Van der Waals forces caused by interactions between carbon atoms of various layers. Intralayer forces, such as sp2 carbon-carbon covalent bonds and crosslinks at graphene sheet boundaries, also enhance graphene and its derivatives' capabilities. Therefore, graphene-based materials are ideal catalyst support materials in catalysis because they increase catalyst durability and surface area, bringing contaminants closer to the catalyst surface, where catalytic reactions occur (Grigoriev et al., 2018). GO is the most extensively researched functionalized graphene, whereas rGO membranes are GO variants. Recent research by Olatunde and Onwudiwe (2021) identified the ability of graphene oxide (GO) to support organic pollutant degradation and is used in five distinct degradation processes: photocatalysis, chemical oxidation process, sonocatalytic/sono-photocatalytic processes, electrocatalytic processes, and direct catalytic degradation. Each process exhibited significant degradation efficiency, with the production of reactive radical species accounting for each phase's activity. It was observed that the integration of graphene/graphene derivatives enhanced the degrading efficiency by improving the formation of radical species due to the enhanced light absorption, surface area, and reduction in charge-carrier recombination. Determining the correct weight percentage of graphene required to enhance the catalytic activity of the composite is a crucial aspect of strengthening its activity. Comparative analysis of these techniques revealed that, despite having comparable degradation efficiencies, the photocatalytic process requires the least time to achieve 90% degradation of pharmaceuticals. In contrast, the electrocatalytic and sonocatalytic methods are the most desirable in terms of mineralization potential. However, very little is known about the ability of GOs to degrade MPs. According to Fadli et al. (2021), $TiO_2$ as a catalyst can generate significant amounts of oxidizing agents for the breakdown of MPs. However, it has a high rate of hole and electron recombination. To improve the photocatalytic capability of $TiO_2$ for the degradation of PE microplastics, $TiO_2$ was treated with Ag dopant utilizing rGO. Under UV light for 4 hours, 76% of MPs were degraded by 3%Ag/$TiO_2$–1%RGO, compared to 68% and 56% for Ag/$TiO_2$ and pure $TiO_2$, respectively.

#### 4.1.1 GO membranes

Researchers are interested in GO because it works so well as a biosensor (Cheraghi et al., 2022; Ilager et al., 2022), biomaterial (Liu et al., 2021), and adsorbent of organic and inorganic pollutants (Ma et al., 2022; Mustafa et al., 2022a) due to the numerous oxygen atoms in the form of epoxy, hydroxyl, and carboxyl groups on its surface. In terms of UF performance, it was discovered that membrane fouling decreased gradually after PE coagulation. Increasing the coagulant dose enhanced the floc cake layer's porosity due to PE particles, especially big PE particles. Compared to using flocs alone, membrane fouling was minimized. Larger PE particles decreased membrane blockage. Coagulation caused by two mmol/L dosages of each $FeCl_3 \cdot$ 6H2O and polyacrylamide decreased membrane flow by 10% when large PE particles (d > 5 mm) were present (Ma et al., 2019a). Furthermore, fouling is a continual challenge since it reduces permeate flow and membrane function, reducing the usable life of the membrane (Ding et al., 2021; Wu et al., 2021a). Several studies show that membrane design and modification strategies utilizing nanoparticles are viable ways to improve organic pollution removal by making membranes more resistant to fouling and, as a result, the issues connected with it (Heu et al., 2020; Wu et al., 2021a). As a result, hydrophilic alteration of membranes is required for use in pressure-driven filtration systems. GO has been found as a material with the potential to improve the effectiveness of the membrane in this water purification process due to its unique transport capabilities, hydrophilic and smooth nature. GO nanoparticles are also resistant to fouling and pollutant transmission, which sets them apart from other nanomaterials. On the other hand, GO may rapidly diffuse into water and is difficult to separate and reuse, potentially resulting in the re-pollution of treated water (Dey et al., 2019). Encapsulating GO in biodegradable porous materials like chitin-based sponges is suitable for MP-containing wastewater treatment.

Recently, Dey et al. (2022) fabricated a GO-PVA composite membrane and used it for removal of microplastics from synthetic wastewater. Briefly, the membrane was synthesized using vacuum filtering (Dave and Nath, 2016; Park et al., 2021). Various amounts of GO were dispersed in DI water (16 mL), and 3 mL PVA solution (10 g PVA/100 mL of DI) was mixed with the dispersed GO. Later, 1 mL of glutaraldehyde was added to the composite solution of GO-PVA to crosslink the two substances. The whole mixture was then sonicated for 15 minutes, filtered by vacuum filtration (filter area: 19.54 $cm^2$; pressure: 3 bar), and dried in an oven at 80 °C for 2 hours to increase the cross-linking between GO and PVA. The samples were then rinsed successively with DI water until the washing solution's pH reached neutral, and with ethanol to eliminate any remaining GA, followed by drying at room temperature. The obtained GO-PVA membrane (Figure 4) exhibited excellent permeability (179 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$) and eliminated 95% of HDPE in 15 seconds at pH 8 and 3.5 bar transmembrane pressure.

Following this, Sun et al. (2020) prepared a chitosan-functionalized graphene oxide (ChGO) sponge and oxygen-doped carbon nitride ($O-C_3N_4$) (Sun et al., 2021a). The prepared sponges were used to remove different functionalized 1µm size MPs including, PS, carboxylate-modified PS (PS-COOH), and amine-modified PS ($PS-NH_2$) in a water system at pH 6-8. The resultant sponges have mechanical strength, flexibility, and linked pores. These sponges, according to the authors, successfully eradicated functionalized MPs groups. For instance, chitin-based sponges have higher reusability due to their extraordinary compressibility (three cycles) i.e., adsorption efficiency for different MPs were as follows: PS (89.8%), PS-COOH (72.4%), and $PS-NH_2$ (88.9%) (Sun et al., 2020). Later on, Sun and coworker (2021b) synthesized a chitin-based sponge membrane with linked pores using GO and oxygen-doped carbon nitride ($O-C_3N_4$) and successfully removed 71.6-92.1% of PS, PS-COOH , and $PS-NH_2$ MPs in a water system at an environmentally relevant concentration of 1 mg $L^{-1}$. The ChGO is a compressive sponge with compressive stresses of 50 and 40 MPa, respectively, in dry and wet states. These sponges can be utilized to remove MPs from water due to their high reusability, biocompatibility, and biodegradability.

Meng et al. (2021) prepared a 3D graphene/cotton sponge with vertical gradient microchannels and used it in a photothermal platform. The sponge appeared as a highly efficient, low-cost, biomass-derived 3D sheet with a higher evaporation rate of 2.49 kg $m^{-2}$ $h^{-1}$ (top and side surfaces) and 8750-fold external stress withstanding rate. Approximately 90.6% of the water evaporated with the first attempt, removing all PE microfibers. It was done using 3D $MoS_2$/graphene/cotton, allowing for multilayer interception and reactive oxygen species attack.

GO is also considered a supporting unit for other membrane materials. Natural materials obtained from renewable resources (e.g., cellulose, chitin) are growing in popularity for MPs treatment due to their superior biocompatibility and biodegradability. However, the low mechanical strength of chitin materials due to molecular depolymerization during chitin modification has limited their practical applicability. The combination of GO with chitin significantly increases (1) the mechanical strength of the chitin-based sponge, (2) developed strong interfacial interactions with chitin chains via hydrogen bonding which GO works as an efficient crosslinker to reinforce the chitin materials, thus the mechanical property was distinctly improved (Chabbi et al., 2020; Liu et al., 2020a), and (3) oxygen-containing functional groups such as -COOH and -OH in GO also facilitate the removal of metal ions and organic pollutants (Sun et al., 2020), suggesting its usefulness in enhancing the performance of membranes for the co-existence of MPs with other pollutants.

In conclusion, developments in MST and innovative membrane technology, and simple manufacturing techniques are still required to simplify the membrane fabrication process and increase the 2D material membrane's removal efficiency. The potential of rGO in membrane fabrication and treatment of MPs from the aqueous environment was examined in the next section.

#### 4.1.2 rGO membranes

The rGO is a promising adsorbent with high reusability and biocompatibility that can mitigate the detrimental effects of PS (Dey and Jamal, 2021). Yuan et al. (2020) investigated the adsorption performance of 3-dimensional rGO for PS MPs; the membrane has a unique porous spatial structure. The experiment was carried out using a variety of variables such as temperature, pH, contact length, and ion concentration. According to the findings, PS MPs adsorption is an endothermic process. For example, a 3D RGO membrane could absorb PS MPs of 617.28 mg/g at 6 pH, $C_0$ = 600 mg/L, t = 120 min, and 26 °C. In another study, Fryczkowska and Przywara (2021) used composite membranes made of polyacrylonitrile and rGO (rGO/PAN) to remove MPs from industrial wastewater in an ultrafiltration system. When 0.11% to 0.83% w/w of rGO is added to the PAN matrix, more tiny holes (150 nm) were formed and enabled the separation of colloid formed in the aqueous $FeCl_3$ solution (rejection > 82%), particularly MPs. The proposed membrane showed excellent anti-fouling qualities that enable smooth clean of the cake layer, which makes them reusable. This study shows that a single membrane technique using rGO/PAN composite membranes can be used instead of a series of steps to treat wastewater that contains MPs.

The initial graphene-based membrane application for MPs removal is the fabrication of holey-reduced graphene oxide (h-rGO) by (Yang et al., 2022b). The rGO nanosheet was embraided with $Co_3O_4$ etching. Using a simple vacuum filtration approach, they integrated this membrane into the polymeric membrane for support (Figure 5). In brief, the GO dispersion (3.0 mg/ml, 30 mL) was produced by ultrasonication GO in DI water (1 h at 25 °C). Co NPs (0.12 g) was then loaded to the GO dispersion and agitated for 2 hours, then heated in autoclave heating (12 hours at 180 °C) followed by autoclave heating (12 hours at 180). After cooling to room temperature, Co/rGO was finally attained. Co/rGO was obtained by freeze-drying it for 12 hours. The Co/rGO samples were then annealed at 900 °C (5 °C/min) for 2 hours in a controlled situation. Co/CoO was removed with 10% HCl, neutralized with deionized water, and dried to get h-rGO. h-rGO nanosheets are uniformly deposited on a support membrane, and membranes based on h-rGO nanosheets are manufactured using the negative pressure filtration technique. The layout of the membrane is made up of the following parts: (1) The distinctive nanostructured (pore size 27.3 nm) form of h-rGO nanosheets captures MPs from nano to micro size and also has better water flux (37.19 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$) than GO and rGO alone (2) Because graphene is so physically and chemically stable, membranes made of h-rGO nanosheets can withstand the outside environment while removing MPs. The hrGO membrane was very good at removing FP microspheres and PS beads MPs (up to 99.9%), which makes it a promising candidate as a useable membrane for removing MPs in water, especially since it is easy to make and can be made on a large scale. h-rGO membranes may have a lot of potential for removing MPs from wastewater because of how stable they are physiochemically and how well they remove MPs.

In a baseline study, Liu et al. (2022) analyzed the tetracycline TC adsorption mechanism of multiple MPs (PP, PA, and PS) before and after UV radiation aging and the changes in TC adsorption trend when aged PP coexists with GO/rGO. In a coexisting system of aged MPs and GO/rGO, the TC adsorption of old PP-GO was risen by 336% and that of aged PP-rGO by 100%. Owing to the oxygen-containing functional groups, GO/rGO with a high oxidation level and level in an aged PP-GO/rGO coexisting system is better for TC adsorption. During the TC adsorption process, there was surface adsorption and partition adsorption. In the MPs-GO/rGO system, TC adsorption was strongly pH dependent, with acidic (pH = 3) or alkaline (pH = 11) conditions being preferable. Besides, microscopic fluorescence imaging of a 0.5 mg $mL^{-1}$ fluorescent polystyrene (FPS) microspheres (100 nm) solution before and after filtering (Figure 6a, c) indicates fewer FP microspheres after filtration. Under UV light (Figure 6b), the solution's diminished fluorescence after filtering is essentially unnoticeable. SEM and NTA studies were utilized to determine FP microsphere size, shape, and range in prefiltered and filtered samples (Figure 6d, e). Noticeably, 86% of the prefiltered samples contain particles bigger than 50 nm and some microscopic bits, showing that nanoscale FP microsphere particle sizes vary. The average h-rGO-1 filtration efficiency for 80 nm particles was 77.04%; for 200 nm, it was 94.35%; and for 1000 nm, it was 99.99%. Average h-rGO-2's filtration efficiency rose for FP microspheres with 80, 100, and 200 nm diameters but not for 500 and 1000 nm microspheres. Fluorescence intensity curves (Figure 6h) and removal efficiencies (Figure 6i) of FP microspheres with various concentrations before and after 200 nm filtration reveal that MPs content does not affect h-rGO nanosheet-based membrane filtration efficiency. The h-rGO membrane filters FP microspheres bigger than 80 nm 100% effectively. The h-rGO membrane can remove 99.9% of 12,500-mesh PS beads. The h-rGO membrane's porous nature allows for high water flow and MPs elimination.

Two-dimensional (2D) graphene can stay together in water due to strong stacking contact between sheets, making it less absorbent. Hydrothermal methods (Kazemeini et al., 2016) and freeze-drying (Zhu et al., 2013), and self-assembly may change 2D graphene into 3D (Payan et al., 2018). The 3D structure may help expedite the distribution and absorption of pollutants (Nardecchia et al.; Shen et al.). The 3D structure separates solids from liquids after adsorption. 3D graphene can absorb twice as much arsenic as 2D graphene, according to a report by Vadahanambi et al. 3D graphene has a greater possibility of cleaning water than 2D graphene.

Yuan et al. (2020) employed 3D RGO to remove PS MPs. However, 3D RGO can't eliminate PS MPs from tap water (56.0%, 448.60 mg/g) and micropollutant water (53.85%, 430.78 mg/g) as well as distilled water (66.63%). This discovery may be produced by metal ions, organic debris, algal secretions, ammonia nitrogen, and other contaminants in tap water and micropollutant water. Environmental factors charge humic acids and metal ions. A few tiny particles might impede the adsorbent's porous structure. 3D RGO removes more than 50% of PS MPs in actual water samples, proving effective.

## 4.2 MXene based membranes

MXenes are promising 2D materials for making new microrobots with many different functions (Thirumal et al., 2021). The formula for them is $M_{n+1}X_nT_x$ (n = 1, 2, 3), where M is an early transition metal (Ti, Mo, V), X is carbon and/or nitrogen, and T is the surface-terminating functionality (O, F, OH, H etc.) (Ghidiu et al., 2014). MXenes are made by etching only the A-element layers of a MAX phase, where A is an element from group IIIA or IVA (Al, Si) (Ghidiu et al., 2014). This method makes "exfoliated" MXene microparticles with a large surface area and many layers, like an accordion. These materials are also highly conductive, chemically stable, thermally conductive, hydrophilic, functional on the surface, and compatible with the environment (Bao et al., 2018). Because of this, they have been used quickly in many different ways, such as to clean water (Ding et al., 2018). $Ti_3C_2T_x$ is the MXene that has been studied the most.

A novel membrane constructed of holey $Ti_3C_2T_x$ nanosheets (h-$Ti_3C_2T_x$) removes MPs effectively from wastewater. Etching $Co_3O_4$ nanoparticles on $Ti_3C_2T_x$ nanosheets was followed by vacuum filtering using a polymer membrane. The design includes (see Figure 7A): (1) Nanoporous $Ti_3C_2T_x$ nanosheets can filter and collect nano- to micro-scale polymers. h-$Ti_3C_2T_x$ membranes had quicker mass transit and better water reflux than graphene or $Ti_3C_2T_x$ membranes. (2) The physicochemical stability of $Ti_3C_2T_x$ renders h-$Ti_3C_2T_x$ membranes MPs-resistant. They are more beneficial in tough situations. The h-$Ti_3C_2T_x$ membrane comprising Co3O4 nanoparticles embedded in $Ti_3C_2T_x$ nanosheets demonstrated experimental performance of up to 99.3%. The distinct nanoporous holey MXene structures of h-$Ti_3C_2T_x$ nanosheets can additionally screen and trap polymers from the nano- to the micro-scale. According to the study, these membranes feature a planar porous structure with nano-holes with an average hole size of 25 nm. This allowed for membranes with high MPs removal efficiency, physiochemical stability, and 196.7 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$ water flux (Yang et al., 2022a).

MXenes have been found to be good at killing bacteria, to have a surface that attracts water, and to be stable enough to use in membranes that clean water. Also, MXenes' improved mechanical flexibility and excellent ability to form films are promising for making highly selective water desalination membranes with high throughput.

However, MXenes can't be used in many situations because ultra-thin 2D materials have problems, such as a high tendency to overlap and no way to control their porousness. In the past few years, much work has gone into making porous MXenes that fit these criteria: (i) MXene assembly; (ii) MXene deposition or insertion into porous substrates; (iii) Loading or coating functional porous materials on the surface of MXenes; and (iv) Formation of in-plane pores inside MXenes (Bu et al., 2020). Even though more research needs to be done before MXene can be used to get rid of MPs through membranes, it has already shown that it is stable and does a good job of getting rid of organic pollutants.

Urso et al. (2022) used 3D multifunctional Mxene-derived oxide microrobots for an NPs cleanup study. $Ti_3C_2T_x$ MXene was converted into photocatalytic multilayer $TiO_2$ through a heating method. These microrobots have zeta potential and self-propulsion abilities, attracting and entrapping nano polymers on their surfaces. The researchers propose this membrane as a portable, low-cost technique for dealing with MPs pollution.

Recent research used MXenes as catalyst promoters to expedite charge-hole separation, enhancing the photocatalytic performance of $Zn_xCd_{1-x}S$ (Cao et al., 2022). A variety of molar ratios of Zn and Cd were used to prepare $Zn_xCd_{1-x}S$ (here, x = 0, 0.2, 0.4, 0.5, 0.6, 0.8, 1.0). They discovered that 2% MXenes combined with $Z_{0.6}C_{0.4}S$ was the optimal ratio for photodegrading plastic and producing $H_2$. Due to the extremely low Fermi level of MXene compared to that of $Zn_xCd_{1-x}S$, the $Zn_xCd_{1-x}S$ treatment ($Z_{0.6}C_{0.4}S$) incorporated with MXene exhibits a lower photoluminescence (PL) peak intensity, which promotes the electron transfer from $Z_{0.6}C_{0.4}S$ to MXene through the interface of the heterogeneous junction, thereby enhancing the separation efficiency of charge carriers and achieving highly efficient photocatalytic activity (Li et al., 2020a). MXene loading exhibited a higher photocurrent density than $Z_{0.6}C_{0.4}S$ alone, demonstrating its superior charge transfer capability. The smaller radius also displayed a lower charge transfer resistance and a more efficient surface charge migration, indicating a higher charge carrier efficiency (Dong et al., 2020). There was no decline in $H_2$ generation up to four cycles, and it was feasible to prevent the photocorrosion effect. However, a high amount of MXene may effectively prevent $Z_{0.6}C_{0.4}S$ from absorbing visible light. Therefore, $Z_{0.6}C_{0.4}S$ with an adequate MXene loading is significant for enhancing photocatalytic $H_2$ evolution activity. The synergistic effect of MXene/ $Zn_xCd_{1-x}S$ and enhanced charge-hole separation efficiency, visible light absorption capacity, and oxidation capability. The PET bottle is simultaneously oxidized into organic macromolecule components, such as methanol, glycolate, and acetate. By modifying the band topologies of photocatalysts, this finding provides a method for producing $H_2$ while degrading plastic contaminants.

# 5 Mechanisms of removal of micro/nano plastics from membranes

Based on kinetic adsorption studies, a strategy for removing MPs was developed. In a GO-assisted chitin-based sponge membrane doped with carbon nitride, the removal of MPs by (i) electrostatic attraction, (ii) hydrogen boning, and (iii) the main force behind MPs absorption was $\pi$-$\pi$- contacts and intra-particle diffusion were very important parts of the whole adsorption process (Sun et al., 2020; Sun et al., 2021a). MPs were incorporated into the sheet's pore structure and folded and uniformly and thickly spread throughout the 3D rGO sheet, according to SEM and XRD analyses (Yuan et al., 2020). 3D rGO offers many active sides capable of firmly adsorbing PS MPs even at the sheet's borders because of the -effect provided by a thick and smooth graphite layer. Removing organic contaminants from the sheet is simpler because they are readily absorbed and form a stable complex on the sheet. Because PS MPs are organic macromolecules with conjugated systems of uniformly charged electrons, 3D rGO functions by way of a - contact created by the carbon rings of PS and 3D rGO, respectively (Pei et al., 2013).

Yuan and associates (2020) demonstrate the surface morphology of 3D rGO following PS MPs adsorption using SEM (Figures 8(a)-8(d)) and XRD (Figure 8(e)). Figures 8(a) and 8(b) illustrate how certain PS MPs were distributed equally and thickly throughout the sheets of the 3D rGO after adsorption, while others were stuck in the pore structure and folds (b). Figure 8(c) and Figure 8(b) show that 3D rGO has multiple active sites that may securely adsorb PS MPs on its surface Figure 8(d). A thick, smooth graphite layer creates the - effect in 3D rGO. It rapidly and effectively removes organic contaminants by adsorbing organic impurities containing electrons, generating a stable compound. MPs are constructed of organic macromolecules with electrons. Charges in the conjugated system are equally distributed, and its characteristics are stable. It was assumed that PS MPs clinging to 3D rGO generated strong contact between their carbon rings (Chen et al., 2011). Figure 8(e) indicates PS MPs had amorphous diffraction peaks of about 20, but 3D rGO had a broad peak at 25. Both peaks in the 3D rGO's XRD pattern widened following adsorption, showing that PS MPs had been absorbed. Because of electrostatic attraction, PS MPs traveled fast from the 3D rGO's edge to its surface. The huge specific surface area and loose pore structure of the 3D rGO offered PS MPs a variety of active locations. Due to the strong $\pi$-$\pi$ force and the porous structure's physical retentive effect, the MPs dispersed slowly inside the 3D rGO. Consequently, the strong force via (i) electrostatic attraction and (ii) physical retention was primarily responsible for the adsorption of PS MPs on 3D rGO.

To further understand how 3D rGO adheres to PS MPs, the adsorption kinetic models were assessed using a varied adsorption time. The mixture was shaken for 5 to 480 minutes in a vibrator with constant temperature and water bathing. The removal efficiency of MPs significantly increased in the first 30 minutes, going from 28.71%

to 54.35%. The clearance effectiveness of MPs rose significantly between 30 and 120 minutes, from 54.35% to 66.10%. MPs removal effectiveness and 3D rGO adsorption capacity both remained effective after 120 minutes. These findings show that after 120 minutes, the adsorption of MPs on 3D rGO reached equilibrium. Moreover, two kinetic model pseudo-first-order and pseudo-second-order kinetic plots, show how PS MPs stick to 3D rGO. The main thing that changed the pseudo-second-order kinetic model (Freundlich) was the formation of chemical bonds, which was strongly linked to the adsorption of PS MPs on 3D rGO. So, the main way that MPs stick to 3D rGO is through a chemical process. The intraparticle diffusion model was also used to look at the movement of PS MPs that had stuck to the surface of 3D rGO.

Given that it is a three-stage curve, it suggests that intraparticle diffusion is not the only factor influencing how the adsorption process is regulated. The high slope of the line in the initial phase shows that MPs swiftly penetrate the 3D rGO's surface and edge. The migration of MPs into the interior structure of 3D rGO via internal diffusion is a gradual adsorption process; the line's slope is less steep in the second stage than it was in the first. In the third step, the slope of the line tends to be horizontal, demonstrating the achievement of the 3D rGO and PS MPs adsorption equilibrium. As a result, membrane diffusion and internal particle diffusion are the main steps of MPs adsorption on 3D rGO. According to some prior investigations, the concentration of PS MPs in the solution and the distribution and size of the 3D rGO pore may affect the mass transfer limitation (Barghi et al., 2014; Fattahi et al., 2014). This discovery may be looked into independently. To understand the energy changes in the adsorption process, several temperatures of adsorption thermodynamics were investigated. The efficacy of MPs removal improved from 66.83% to 72.63% as the temperature rose, and the 3D rGO's adsorption capacity increased from 534.60 mg/g to 598.98 mg/g. This study demonstrates that raising the temperature is advantageous for MPs adsorption on 3D rGO. This is most likely because MPs quickly diffuse into 3D rGO. However, the temperature was fixed at 26 degrees Celsius to conserve energy and since the removal efficiency did not vary significantly. MPs adsorption is an endothermic process because the enthalpy change H is larger than zero.

Indicating that the interface between 3D rGO and MPs becomes more disordered during adsorption, and the entropy shift S is larger than zero (Tan et al., 2009). Since the Gibbs free energy change is always negative, adsorption must occur spontaneously (Çolak et al., 2009). As a result, the endothermic process of MPs adsorption on 3D rGO has the potential to enhance adsorption. For most adsorptions, the Langmuir and Freundlich models are applicable. The Langmuir adsorption isotherm states that monolayer adsorption can only occur on uniform surfaces. The Freundlich isotherm may also determine how much adsorption occurs on different surface qualities. The Langmuir and Freundlich isotherm plots revealed that the adsorption of PS MPs on 3D rGO corresponded to the Langmuir adsorption isotherm model with a good correlation coefficient ($R^2$ = 0.992). Based on these findings, it appears that the active sites on the surface of 3D rGO are identical and that 3D rGO adsorbs MPs in a single layer.

## 5.1 Effect of Co-existence of different pollutants

The interaction between MPs and graphene may significantly impact adsorption processes and capacity in the presence of other contaminants. Moreover, the actions of MPs concerning the absorption of other contaminants may alter. The addition of new contaminants may result in significant modifications to processes. The behavior of contaminants will change when inorganic nanoparticles (INPs) like $Al_2O_3$ nanoparticles are added. This shows that heteroadsorption of INPs is much more difficult than homoadsorption (Liu et al., 2020b). Researchers have looked at the ability of graphene-like magnetic biochar (GLMB) to remove 17β-estradiol (E2). GLMB was an adsorbent for removing E2 from water that was cheap, very effective, safe for the environment, and could be reused. $Al_2O_3$/MPs made it take much less time for E2 to reach adsorption equilibrium on GLMB. $Al_2O_3$/MPs sped up the time it took for adsorption to reach equilibrium. Nonetheless, at equilibrium, increased $Al_2O_3$/GLMBB or MPs/GLMBB ratios did not significantly enhance or restrict E2 removal. E2 may adsorb onto GLMB by many simultaneous processes, such as -interactions, micropore-filling effects, electrostatic interaction, etc.

When GO/rGO and aged PP were in the same solution, they made it easier for TC to stick to the surface. This effect was stronger in acidic conditions than in alkaline conditions. This had to do with the form of the TC ion in the solution and the charge on the surface of GO/rGO. When compared to rGO, the aged PP and GO coexisting system may make it much easier for TC to stick on the surface. Previous research showed that the interactions and van der Waals forces between rGO and TC might have been caused by their huge ring shapes (He et al., 2018).

On the other hand, hydrogen bonds may have been formed between TC molecules and oxygen-containing functional groups in GO. These hydrogen bonds may have been important for adsorption (Ai et al., 2019). TC likes hydrogen bonds more than van der Waals forces and interactions with negative charges. The process of antibiotic adsorption is shown in Figure 9.

Due to the loss of oxygen functional groups on the surface, like carboxyl groups, the reduction of GO to rGO would modify the way graphite stacks up from a sheet-like shape to a stacked shape (Qi et al., 2019). This would make it much harder for pollutants to stick to the rGO surfaces. As a result, the removal of MPs by graphene material is complicated in the presence of other pollutants, which may also vary depending on the different kinds of pollutants (i.e., organic, inorganics, ionic, etc.). With the aging of MPs in the environment, the overall adsorption may change drastically. According to a recent study, the chlortetracycline (CTC) and amoxicillin

(AMX) adsorption capacities of PE rose 1.08-14.24-fold following aging (Fan et al., 2021). Furthermore, research has shown that functionalized GO and rGO display nucleating capabilities when used as additives in isotactic PP crystallization (Broda et al., 2020). Adding graphene to plastics allows graphene and MPs to cohabit. Graphene is also vulnerable to environmental degradation (Yu et al., 2021). It has been demonstrated that sunlight, natural reductants, and bacterial decomposition rapidly convert GO to rGO (Matsumoto et al., 2011). Because the reduction procedure lowers the number of oxygen-containing groups, the adsorption capacity of GO will be altered (Zhang et al., 2019b). More study is needed to enhance membrane filtering of MPs and graphene-based membranes for use in water treatment systems. The first study to look at the ciprofloxacin adsorption behavior of multiple MPs (PP, PA, and PS) before and after UV radiation aging, as well as the alterations in TC adsorption behavior when aged PP coexists with GO/rGO recently, was performed by Liu et al. (2022). In a coexisting system of aged MPs and GO/rGO, the TC adsorption capacity of old PP-GO was increased by 336% and that of aged PP-rGO by 100%. GO/rGO with a high degree of oxidation and concentration in an aged PP-GO/rGO coexisting system are more advantageous for TC adsorption due to the involvement of oxygen-containing functional groups. Surface and partition adsorption happened during the TC adsorption process. In the MPs-GO/rGO coexisting system, TC adsorption was strongly pH-dependent, with acidic (pH = 3) or alkaline (pH = 11) conditions preferable.

In the case of the co-existence of heavy metals and MPs, the literature suggests surface morphological and chemical changes brought on by environmental weathering and plastic degradation may regulate heavy metal mobility on MPs surface by changing the physical attachment of heavy metals on plastic surfaces, accelerating the electrostatic interaction and chemical interaction between functional groups of MPs and heavy metal species (Huang et al., 2020; Aghilinasrollahabadi et al., 2021). For instance, compared to fresh low density polyethylene (LDPE) samples, surface oxidized LDPE films and pellets showed greater hydrophilicity and surface area, which led to enhanced uptake of $Pb^{2+}$, $Cu^{2+}$, $Mn^{2+}$, and $Zn^{2+}$ from synthetic drinking water (Ahamed et al., 2020; Huang et al., 2020). Likewise, a marine study found that beached plastic pellets acquire more heavy metals than virgin plastic samples (Holmes et al., 2012). Aghilinasrollahabadi et al. (2021) reported that $Pb^{2+}$ and $Zn^{2+}$ uptake by weathered MPs appeared to be influenced by the heterogeneous surface of the MPs, the partitioning of heavy metals between MPs, the accumulation of silt particles on the MPs, and the detachment of silt particles from the surface of weathered MPs during the heavy metal exposure process. It was found that $Zn^{2+}$ adsorption on PET films was below the detection threshold. Future research will be needed to quantify how much silt separates from weathered MPs and to evaluate how certain heavy metals are distributed among MPs, aqueous solution, and silt particles. The MPs adsorption of heavy metals may also be influenced by the electronegativity of the metals, with larger electronegativity ions [ $Pb^{2+}$ (2.33) and $Zn^{2+}$ (1.65), Pauling unit] adhering more firmly to the surface (Allen and Brown, 1995). The $Pb^{2+}$ with higher electronegativity may induce a stronger electrostatic attraction on the plastic surface, enabling its preferential sorption on the plastic surface (Huang et al., 2020). These factors alter MPs charecterisitcs and influence the physicochemical reaction impading membrane surface and consequently can changed their removal performance.

The hydrophobicity profoundly affected the interaction of organic pollutants (e.g., PAHs) with MPs since the researchers found that PE could adsorb more high molecular weight PAHs (HMW-PAHs, 4-6 rings) than low molecular weight PAHs (LMW-PAHs, 2-3 rings) from the aquatic environment as HMW-PAHs are more hydrophobic (Wang et al., 2018). Yu et al. (2020) studied the sorption behavior of NAP and their derivatives on MPs and found that charged functional groups in NAP derivatives can lower the hydrophobicity and greatly restrict their sorption onto MP-COOH and MPs as compared to NAP and NAP derivatives with uncharged functional groups.

The sorption isotherms of NAP and NAP derivatives ($R^2$ = 0.937–0.981) exhibited higher $K_d$ (L/g) values for NAP and NAP-$CH_3$ (9.4–9.9 L $g^{-1}$) for MP-COOH and (11.6–11.9 L $g^{-1}$) for (MP) compared to those for NAP-$NH_2$, NAP-COOH and NAP-OH (4.5–6.3 L $g^{-1}$) ) for MP-COOH and (6.1–8.4 L $g^{-1}$) for MP. Moreover, Log $K_{OW}$ values NAP-$CH_3$ (3.9) and NAP (3.3) were likewise significantly greater than those for NAP-OH (2.9), NAP-COOH (3.1), and NAP-$NH_2$ (2.3) demonstrating that the hydrophobicity of NAP and NAP derivatives is a key component in determining their sorption onto MPs. This is consistent with the fact that perfluorooctanoic acid, which frequently transports ionic charges in aqueous media, has comparably poorer MPs binding capabilities (Wang et al., 2018). Yu et al. (2020) studied since the Log $K_{OW}$ values of NAP and NAP derivatives were positively and linearly correlated with the $K$d values of their sorption on MPs ($R^2$ = 0.725), which further confirmed the hydrophobicity is the determinant for MP-COOH. The sorption capacities of the NAP and NAP derivatives on MP-COOH were generally lower than that on MPs (except for NAP-$NH_2$), which was coincident with a previous study reporting perfluoroalkyl substances preferred to sorb on PS than PS-COOH (Llorca et al., 2018). It was reported that MPs with oxidized functional groups formed on the surface would have higher hydrophilicity (i.e., a lower hydrophobicity) than virgin ones (Shams et al., 2020), which might, therefore, induce their lower affinities with NAP and NAP derivatives. Therefore, changing the surface properties of MPs impacts their hydrophobicity corresponding interaction with organic pollutants. Rius-Ayra et al.(2021) applied superhydrophobicity and superoleophilicity to the surface of MPs to change its wettability in order to leverage hydrophobic influence for MPs recovery. These two approaches have the potential to generate long-term MPs extraction solutions with greater process optimization. These techniques enhance the selectivity of MPs extraction, hence enhancing their potential utility and reducing trace amounts in WWTPs. In reality, MPs surface functionalisation using $ClO^-$ or $Fe_3O_4$ provides hydrophilic characteristics to solid contaminants, hence enhancing separation process selectivity. This should be further studied to optimize MPs membrane separation technologies and for enhancing the efficiencies of carbon-based two-dimensional materials membranes in MPs contaminated water treatment systems.

# 6 Factors, challenges, and limitations in 2D material-based membranes in MPs removal

The development of effective MST for eliminating MPs is sought, as seen by the preceding debate. Filtration, adsorption, and degradation are major working routes by which membranes can eliminate MPs from water. Likewise, membranes for removing MPs may be divided into two categories. First, membrane filtration uses a screening process to exclude pollutants (Cetinkaya and Ozdemir, 2018; Heu et al., 2020; Mirzaei et al., 2021), and the second one is affinity (AF) membranes which use an adsorption process to eliminate pollutants (Yang et al., 2016; Ivanković et al., 2021). However, the incorporation of 2D materials showed promising prospects for many types of pollutants (Ivanković et al., 2021) and MPs removal. Albeit, there are some factors that impacted the MPs removal performance by such membranes. The factors that directly impact the MPs removal performance of 2D material-based membranes are further categorized into factors impacting filtration and those which intervene adoption of MPs. A detailed description is provided in section 7.

## 6.1 Effect of different factors on membered filtration

Several membrane-based filtering processes are used in water treatment to remove particles, notably MPs (Poerio et al., 2019; Rocha-Santos et al., 2020). However, when particle size drops, the filtering system's removal capacity gets more challenging. Membrane filtering approaches become less effective in this regard as their complexity and expense rise. Furthermore, membrane filtration is costly, necessitating substantial energy inputs as well as regular maintenance due to membrane fouling and scaling.

Coagulation reduced the fouling of the UF membrane caused by PE. Increasing the coagulant dosage increased the permeability of the floc surface due to PE particles, particularly large PE particles. Membrane fouling was reduced as compared to employing flocs alone. Membrane fouling was reduced when the PE particles were larger. In the presence of large-sized PE particles (d > 5 mm), the membrane flow fell by just 10% after coagulation with 0.2 mmol/L PAM and 2 mmol/L $FeCl_36H_2O$, respectively (Ma et al., 2019a). However, this behavior may not be a general norm because it depends on various membrane-process-related variables and plastic characteristics (chemical composition, size, and shape). UF might be used to remove all PE particles as a general rule, but additional research is needed to know how the presence of plastic particles influences the development of the cake layer and subsequent fouling. The structure of MPs can affect their removal in some water treatment techniques. According to Talvitie et al. (2017a), WWTPs do not retain a portion of "fiber-shaped" plastic. As a result, in order to enhance plastic removal efficiency, the final stage treatments must be adequately formulated for fiber removal. Dissolved air flotation (DAF) is proposed as an alternative to membrane filtrate to avoid membrane fouling concerns (Rocha-Santos et al., 2020). Several studies have shown that DAF's capacity to eliminate MPs is unsuccessful (Talvitie et al., 2017a). Wang et al. (2021c) improved its performance by using a range of flocculants and surface modifiers, attaining a removal efficiency of 68.9-43.8%. Because MPs exist in various polymer forms and differ in their properties, particularly their surface chemistry, they significantly influence flocculants, making it even more challenging to select the most matched flocculant (Ma et al., 2019b). Common flocculants are iron or aluminum-based and have limited flexibility (Lapointe et al., 2020). Other polyelectrolyte-based flocculants, despite their greater flexibility in removing MPs, are detrimental to aquatic life because they stay in the water (Harford et al., 2011; Pereira et al., 2018).

MPs and NPs removal is critically dependent on their sizes; for example, a study found that the removal efficiency of MPs was 99.5% in membrane bioreactors systems and 97% in oxidation ditch systems, but 40% of MPs were >500 µm in size and 29% MPs fell in the 62.5-125 µm range (Lv et al., 2019). Moreover, MPs and NPs with a diameter of 1 µm are more dangerous because their increased surface area allows toxic substances to be absorbed. However, the efficiency of these MP/NP elimination is little understood (Tufenkji and Elimelech, 2004; Wang et al., 2021c). Future research must thus prioritize the elimination of MPs from aquatic systems.

Although integrating different strategies could enhance the MPs removal performance in the membrane filtration system, when two MPs removal methods, MBRs, and conventional active sludge, were compared, MBRs had a little higher removal efficacy (99.4%) than CAS (98.3%) (Lares et al., 2018). However, membrane filtering can be challenging due to pore blockage and biofilm thickness (Joo et al., 2021). According to Xiong and colleagues, MPs evoked the manufacture of proteins and polysaccharides, which can accumulate on membrane surfaces and cause massive membrane fouling. The components of extracellular polymeric substances that were accelerated by the availability of MPs were the primary contributors to membrane fouling. In contrast, microbial community distribution analysis revealed that *Alphaproteobacteria*, *Flavobacterium*, and *Pseudomonas* were more prevalent in samples containing MPs. Therefore, it can be speculated that the presence of MPs may promote the growth of specific microorganisms capable of biodegrading plastic and plastic-related chemicals. In membrane bioreactors (MBRs), quicksand filtration was proposed by Ngo et al. (2019) as a low-cost and affordable maintenance approach with a greater removal capacity. Nonetheless, a comparison of MBR and quicksand filtration found that membrane filtration removed 99.9% of MPs, whereas rapid sand filtering removed just 97% (Talvitie et al., 2017a). Ngo et al. (2019) ascribed the decline in fast sand filtering efficacy to the high porosity of the filter material after the working period. The more porous material is created by combining anthracite coal with sand.

Likewise, the activated sludge approach is used for wastewater treatment; however, according to Carr et al. (2016) and Rummel et al. (2017), characteristics such as nutrient content and retention duration influence the MPs removal rate. Increasing the retention duration and nutrient content in wastewater in such cases may result in cross-contamination, affecting removal effectiveness. Increasing reagent dosage, membrane fouling, and nitrogen conversion rate inhibition, among other variables, diminish MPs removal efficiency in wastewater treatment (Wu et al., 2021b). Dong et al. (2021b) observed that humic acid accelerated the mobility of PET microplastics with a mass recovery rate of up to 49.8%, with the impact being more noticeable at increasing electrolyte concentrations. HA in solutions may coat the surfaces of both micro/nanoparticles and porous media, which increases electrical repulsion by increasing electronegativity and creates a steric hindrance to impede the deposition of PET microplastics onto the media surface (Dong et al., 2017). Furthermore, the HA adsorbed on the surface of fragmental PET microplastics might modify their irregular shape, thus increasing their mobility in porous settings (Dong et al., 2021a).

Membranes with the potential to degrade MPs would also be demanded in the future. In a previous study, Tofa et al. (2019) used ZnO nanorods (a nano-coating approach), and a 30% breakdown rate of MPs (PE films and fragments) was found. The degradation process was accelerated by using heterogeneous Zinc oxide photocatalysts activated by visible light. Photocatalyst degradation is presented as a cost-effective option for water treatment due to fewer byproducts. In their study, Li et al. (2018) revealed an effective technique for eliminating MPs. They offered a polymer-coated elongated mesh screen as a modification strategy, which was strong and easily produced from a range of materials; moreover, this modified screen was advantageous since it did not require mechanical devices or electrical power.

## 6.2 Effect of different factors on the adsorption

The potential of PS to adsorb more material was enhanced by higher GO concentrations (Sun et al., 2020). The results showed that 12 h elapsed following a gradual increase in PS adsorption on ChGO sponges. The positive correlation between removal efficiency and sponge concentration (r = 0.989) demonstrated that the removal of PS-COOH and PS-$NH_2$ by Ch, ChGO-50, and ChGO-100 was dose-dependent (Fig. 9B-C). The - interactions between organic molecules and the graphene surface are crucial for adsorption, according to recent research (Song et al., 2018). The ChGO-100 treatment's accessible sites were sufficient for MPs adsorption at 1 mg $L^{-1}$, the maximum concentration recorded in actual water systems (Lu et al., 2016b). Similar to GO concentration, MPs concentration demonstrated some beneficial adoption impact (Yuan et al., 2020). In a preliminary investigation on MPs adsorption on 3D rGO, the standard solution concentration is linearly related to its absorbance at 720 nm. The linear fitting has a correlation coefficient $R^2$ of 0.999, suggesting that it is faultless. The initial concertation of MPs and 2D materials could be a limiting factor in adsorption performance. For instance, the adsorption efficiency of 3D rGO increased from 118.72 mg/g to 533.83 mg/g with an increase in PS initial concentration from 100 to 600 mg/L. Whereas the initial rise in pH from 2 to 6 improved adsorption and reached maximum (66.63 % and 522.06 mg/g), further increase in pH reduced the MPs adsorption on 3D rGO. Despite electrostatic repulsion between PS MPs and 3D rGO under acid and alkaline environments, PS MPs remove more than 55% of 3D rGO, with less than a 10% discrepancy between peak and weakest adsorption rates. This reveals that pH has little influence on PS MPs adsorption onto 3D rGO. Another study (Sun et al., 2020) used ChGO to evaluate PS adsorption as a function of pH because of its high adsorption capacity and found that pH affects the surface charge density and ionic strength of MPs and ChGO sponges. Because $OH^-$ may compete with negatively charged PS for exchange sites at higher pH (Liu et al., 2014; Liu et al., 2016a), the maximum adsorption impact (92.9% removal efficiency) was reported at pH 6, while the lowest was at pH 10. pH showed no effect on adsorption performance within wastewater discharge standards (pH = 6-9; GB18918-2002) (Lu et al., 2016a). PS-COOH and PS-$NH_2$ elimination were similar at pH 6 and 10 (Figure 10E-F). PS-removal carboxylic group's effectiveness decreased when pH neared 8 compared to pH 4 and 6. $H^+$ and PS-$NH_2$ in the acidic solution made adsorption problematic. This is why PS-$NH_2$ elimination was less successful at pH 4. It is generally acknowledged that many functional groups, such as -$NH_2$ and -COOH, are imparted to the surface of PS by solar radiation or weathering (Kim et al., 2017). As shown in Figure S3, the FTIR spectra of ChGO-300 and MPs suggested a combination of ChGO and MPs features. After adsorption, the peak at 1663 $cm^{-1}$ in the spectra of ChGO-300 and PS-$NH_2$ moved to 1608 $cm^{-1}$, indicating an increase in the amide I. A signal corresponding to PS-COOH carboxyl C = O is detected at 1737 $cm^{-1}$.

Furthermore, the SEM image supported the adsorption of MPs by ChGO-300 and indicated the following adsorption capacity order: PS-$NH_2$ ≈ PS > PS-COOH (Fig. 9a–c, Fig.S2). Because of the electrostatic interaction between positively charged MPs (Table 1) and GO, whose surface was covered with different functional groups such as OH- and -COOH, PS, and PS-$NH_2$ were removed more quickly (Song et al., 2018). As the temperature climbed, the removal efficiency of MPs increased from 66.83% to 72.63%, while the adsorption capacity of 3D rGO increased from 534.60 mg $g^{-1}$ to 598.98 mg $g^{-1}$. This study shows that increasing the temperature is beneficial for MPs adsorption on 3D rGO, most likely due to the fast diffusion of MPs inside 3D rGO. The temperature was reduced to 26 degrees Celsius to save electricity, and the removal efficiency did not differ considerably. Ion's concentration is another factor as PS MPs' removal efficacy fell from 66.71 to 53.23% when NaCl concentration rose, whereas 3D rGO's adsorption capacity fell from 533.68 to 432 mg $g^{-1}$. 3D rGO was positively charged, while PS MPs were negatively charged at pH 6. $Na^+$ adsorbs onto PS MPs, neutralizing negative charges and reducing electrostatic attraction between 3D rGO and PS MPs (Wu and Yu, 2019).

## 6.3 Membrane fouling

The size range of pollutants largely dictates membrane fouling (for example, MPs), the size and surface features of the membrane pore, the support layer architecture, and the module design (e.g., number and thickness of spacer sheets, number of fibers and membrane sheets in modules, water flow rate, etc.). Smaller particles than the pore size of the membrane can cause permanent internal fouling, whereas larger particles can plug holes or create cake layers by physical adsorption (Abdelrasoul et al., 2013; Enfrin et al., 2019).

Enfrin et al. (2020) examined the fouling produced by nano- and microplastics (size range: from 13 to 690 nm) on a commercial UF poly(sulfone) membrane with 31 nm pores. Cross-flow filtration of plastic particles at 1 bar pressure for 48 hours reduced permeate water flux by 38% owing to NPs/MPs interactions with the membrane pores and surface. Within 48 hours of filtering, more than 25% of the feed's nano- and microplastics were absorbed onto the membrane surface. Concentration polarization increases solute content on the membrane surface and promotes surface fouling, which can create a boundary layer near the membrane walls (Guha et al., 2017; Zargar et al., 2020). Li et al. (2020b) observed that PVC-MPs at 10 MP/L enhanced MBR membrane fouling. MBR eradicated practically all MPs due to the membrane pore size (0.1 μm) and PVC particle size (<5 μm). Membrane fouling induced by MPs increased both reversibly and permanently due to tiny MPs entering membrane pores. In contrast, Maliwan et al. (2021) found MPs decreased MBR system fouling. The scouring by the presence of polyester (fiber), PA (fiber), PP (fragment), and PE (fragment) (particle sizes ranging: from 0.3 to 5.6 mm) and concentrations (from 7 MP/L 10 75 MP/L) reduced the MBR fouling. There is a disagreement between two recent studies due to MPs with various size ranges. The first study utilized MPs smaller than 5 μm that might clog MBR filters, whereas the second used MPs larger than 300 μm that could function as scouring agents. This suggests membrane pore size for MPs removal should be changed to account for MPs size ranges in WWTP effluent. Because there have been few studies on MPs smaller than 1 μm and NPs in WWTP effluents, more research is needed to identify the most typical MPs and NPs size ranges to design the tertiary membrane processes.

# 7 Conclusion and future perspective

Although MPs are ubiquitous and all living bodies are facing similar risks, marine life is significantly deteriorating with MPs. The number of MPs in the environment and the inefficiency of current equipment make cleaning marine regions challenging. Another difficulty with MPs filtering is the potential exclusion of other marine animals. MPs can be as small as or smaller than plankton and other microorganisms on the ocean's surface. Because of their comparable size and density, selectively removing MPs from sea water is not possible. Therefore, further introduction in the environment needs to be prevented and the methods discussed be used to treat effluents before entering the water cycle.

MPs contain a large specific surface area and hydrophobicity which are conducive to adsorbing other toxic substances from the environment, including polychlorinated biphenyls, polycyclic aromatic hydrocarbons, and dichlorodiphenyltrichloroethane which enter into the human body via inhalation, ingestion and produces hazardous impacts.

Plastic must be removed from the sludge before it may leave the sewage treatment facility. Sludge combustion is being used in certain WWTPs. MPs are removed alongside the residual sludge in this case. However, not all wastewater treatment plants do this, therefore if water is reused, MPs should be filtered here to prevent environmental pollution.

For applications for which biodegradable polymers cannot be used, synthesis of more stable plastic polymers, particularly for the marine environment, is also recommended, which ultimately releases fewer MPs. Scientists have used nanofillers such as graphene, carbon black, carbon nanotubes, and other carbonaceous components to reinforce plastic polymers to improve their thermal, mechanical, and electrical properties (Tarfaoui et al., 2019). Nanofillers have a high elastic modulus and the ability to prevent fracture formation. In addition, 2D material like graphene can restrict MPs' creation, easing pollution control and lowering management costs. Recently, melt-processing was used to incorporate graphene nanosheets (GNs) into thermoplastic polyurethane (TPU) to create high-performance composites (Huang et al., 2021). MPs release of the generated composites is significantly inhibited during oxidative degradation due to the barrier effect of GNs and the strong interfacial connection, while mechanical characteristics are simultaneously enhanced. In contrast to those application where a really long and stable life is needed, a strategy should also be to use biodegradable polymers for all the purposes where no lang life time is needed. This also removes MPs on the long term.

The membrane filtration alone and with other strategies such as coagulation, flotation, and degradation are reasonable advantages over conventual MPs removal treatments. Yuan et al. (2020) demonstrated that porous 3D rGO has good adsorption affinity for PS-MPs limited scale. This technique has also shown outstanding success in eliminating PS MPs from tap water and lake water. In the future, this method has the potential to be an effective MPs treatment industrial level for the removal of other types of MPs from natural water.

Other 2D materials, particularly the graphene family and MXenes, showed complete MPs removal depending on operating conditions. However, more studies are required to optimize MPs' more economical and energy-efficient removal via 2D material-based membrane filtration systems, which may include pre-treatment, incorporation of coagulants, flotation, and other mechanical and biological strategies.

One challenge in MPs filtration is the presence of a higher amount of organic matter and other contamination in water; a pre-treated stream enters the bioreactor, where organic contaminants biodegrade for this semi-crossflow filtration is promising to separate the mixture. In addition, this technique may have further numerous fouling-control methods. Current plans include scouring aeration, in-situ chemical cleaning, enzymatic and bacterial foulant breakdown, and nanomaterial-based membranes (Meng et al., 2017). This might also be accomplished by altering the system's hydrophobicity.

Noticeably membranes themselves are also potential sources of MPs in water. Membrane technology is widely used in water and wastewater treatment today and has a thriving industry. According to a new GIR (Global Info Research) analysis (Market, 2020), the global market for Membrane Filtration is expected to grow from 4710 million US dollars in 2019 to 7030 million US dollars in 2024. The rapid proliferation of membrane processes has necessitated the development of methods to reuse and recycle these materials (Lejarazu-Larrañaga et al., 2020).

The removal of MPs from wastewater has received a lot of interest recently; however, the collection/removal of MPs from stormwater has received less attention (Liu et al., 2022). The gathering of MPs is also supported for more effective and long-term control of MPs. Integration of membrane filtration systems with other strategies, such as constructed wetlands, which can remove 28% of MPs, might be beneficial. On the other hand, Alam et al. (2018) claim that decentralized stormwater treatment methods such as catch basins, filter strips, gross pollutant traps, and grass swales are unsuccessful at eliminating MPs.

Competitive development is being pursued in MPs extraction to design standard extraction methods with the maximum output. The cutting-edge model includes plasma electrolytic oxidation, graphene-based adsorption, electrocoagulation, nanocomposite membrane filtering, and magnetic separation. Sun and coworkers (2020) proposed a nano-adsorbent made with GO and chitin that has a high efficiency for MPs recovery after separation. With the predominance of PS and its effects in mind, research on rGO is undertaken in order to acquire the optimum reusability and biocompatibility as an adsorbent. Similarly, the polymeric membrane more effectively traps polystyrene-based MPs. Perren et al. (2018) stated 99% removal of polyethylene-based MPs can be achieved using a low-cost electrochemical pathway, and magnetic extraction yielded 95% removal of polyvinyl chloride-based MPs (Rhein et al., 2019). Integration of different sorbents could also be promising in MPs removal. For instance, corn straw and hardwood biochar removed 95% PS microspheres (Wang et al., 2020d), and magnetic nano-$Fe_3O_4$ demonstrated >80% removal of PET and PE MPs (Shi et al., 2022). Furthermore, photocatalytic micromotors made of Au@Ni@$TiO_2$ have only recently been used for MPs separation (Rius-Ayra et al., 2021). Likewise, novel strategies for extracting polymeric MPs from aqueous media are being developed.

Some strategies, particularly MPs degradation, can completely disintegrate the MPs in the solution; therefore, no MPs residue is available for further treatment. For instance, Meng et al. (2022) demonstrated 100% PE removal. Real-time photodegradation assisted by m-$TiO_2$/rGO oxygen gas (16.1%) or trace of $H_2O_2$ (0.008 mM) (27.7%). This strategy holds high promise in alleviating the “white pollution” and generating no secondary pollution for sustainable development.

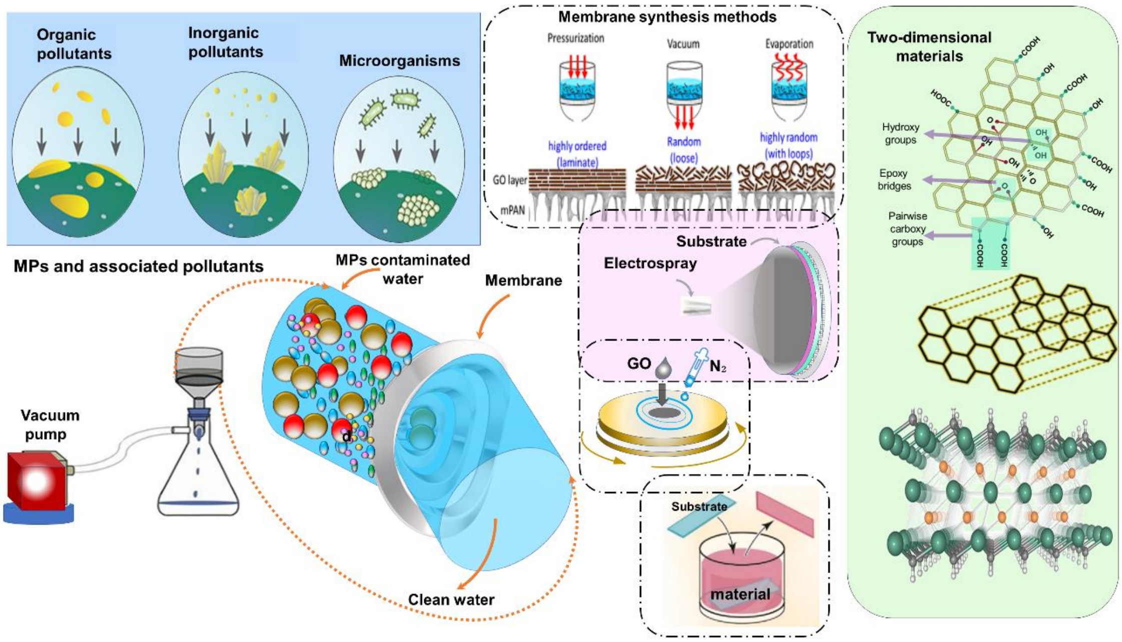
Several routes exist for the recycling of MPs residue obtained from membrane separation: (1) heating process of MPs to transform them into fibers, followed by spinning into thread for use in garments (Sarioglu and Kaynak, 2017), (2) degradation of collected MPs into $CO_2$ and $H_2O$. Then, carbon dioxide can e.g., be electrochemically transformed into CO for use as a gasoline precursor (Zhou et al., 2019). (3) Carbon dioxide can also be used to extract carbon for the synthesis of graphene and other very valued compounds (Chakrabarti et al., 2011). (4) The catalytic oxidation method used during PE elimination can transform CO2 products into high-value hydrocarbon fuels (Meng et al., 2022).

**Acknowledgement**

Tariq Mehmood is thankful for Postdoctoral Research Fellowship awarded by the Chinese government at Hainan University, Hainan, Haikou, China. This study was supported, in part, by Hainan Province Science and Technology Special Fund, the Key Project of Natural Science Foundation of Hainan Province, China (ZDYF2022SHFZ278), the High-level Talents Project of Natural Science Foundation of Hainan Province, China (Grant No. 2019RC043), Start-up funding from Hainan University (kyqd(zr)1719), Alexander von Humboldt Foundation of Germany.

## Highlights

- Excessive MPs pollution has severely damaged marine ecology.
- Surface adsorption of toxic pollutants on MPs intensifies their environmental implications.
- 2D materials are effective membrane materials for MPs removal from water.
- The coexistence of MPs with other pollutants affected their removal mechanism.
- Present status and future-oriented assessment of 2D material-based membranes are discussed




## Graphical Abstract

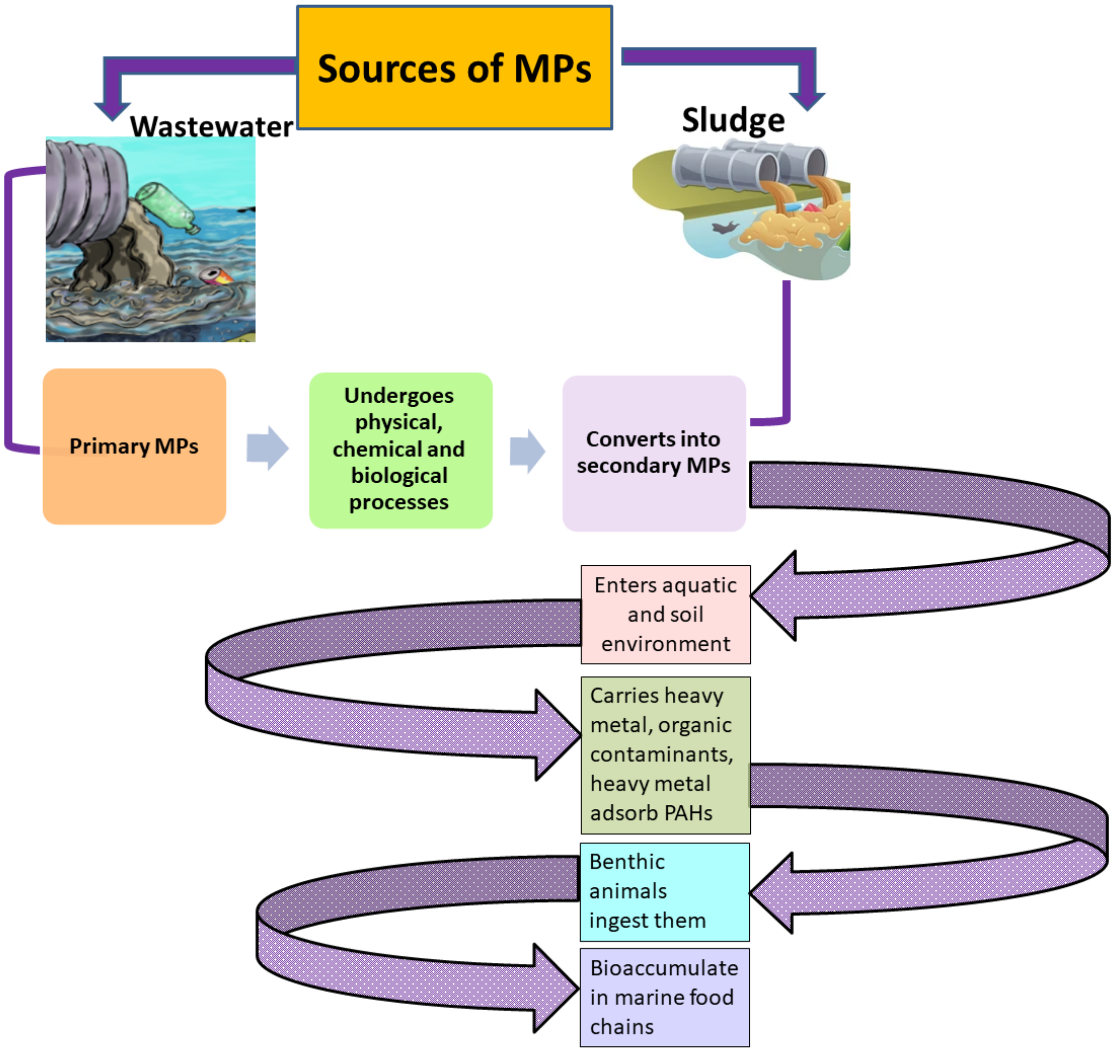



Figure 1: Cycle of MPs from water to accumulation in human food

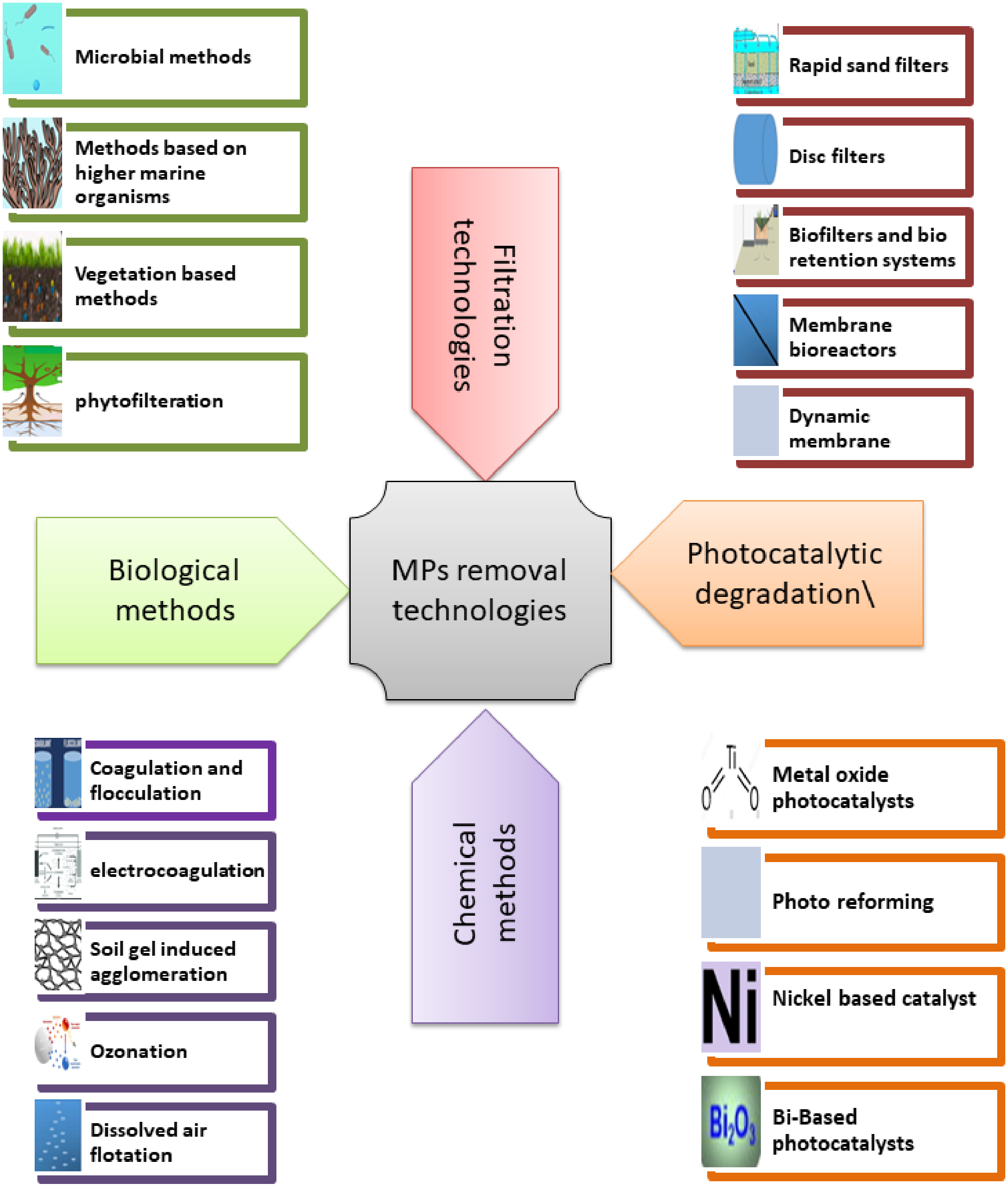



Figure 2: Current technologies used for removing MPs

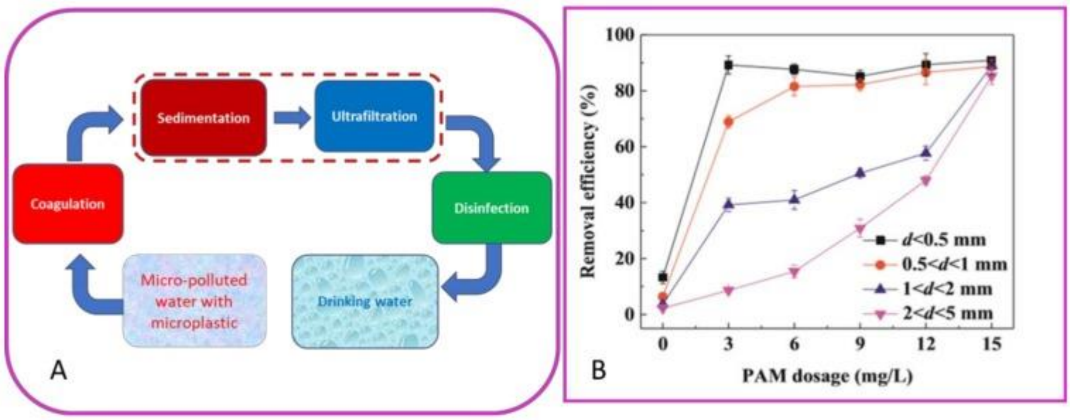



Figure 3: Scheme of the process for removal (A) and removal efficiency (B) of polyethylene (PE) using $FeCl_3{\cdot}6H_2O$ and anionic polyacrylamide (PAM) (elaborated from Ma et al. (2019b)).

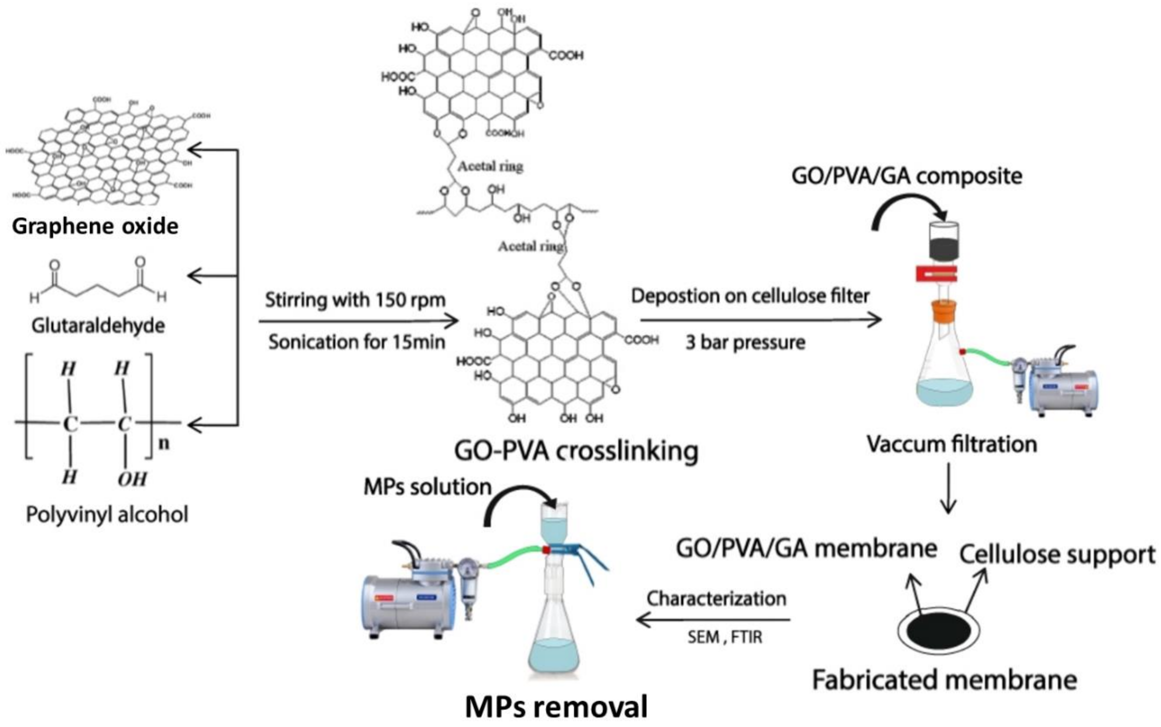



Figure 4: Synthesis of GO/PVA/GA membrane. Reproduced from (Dey et al., 2022).

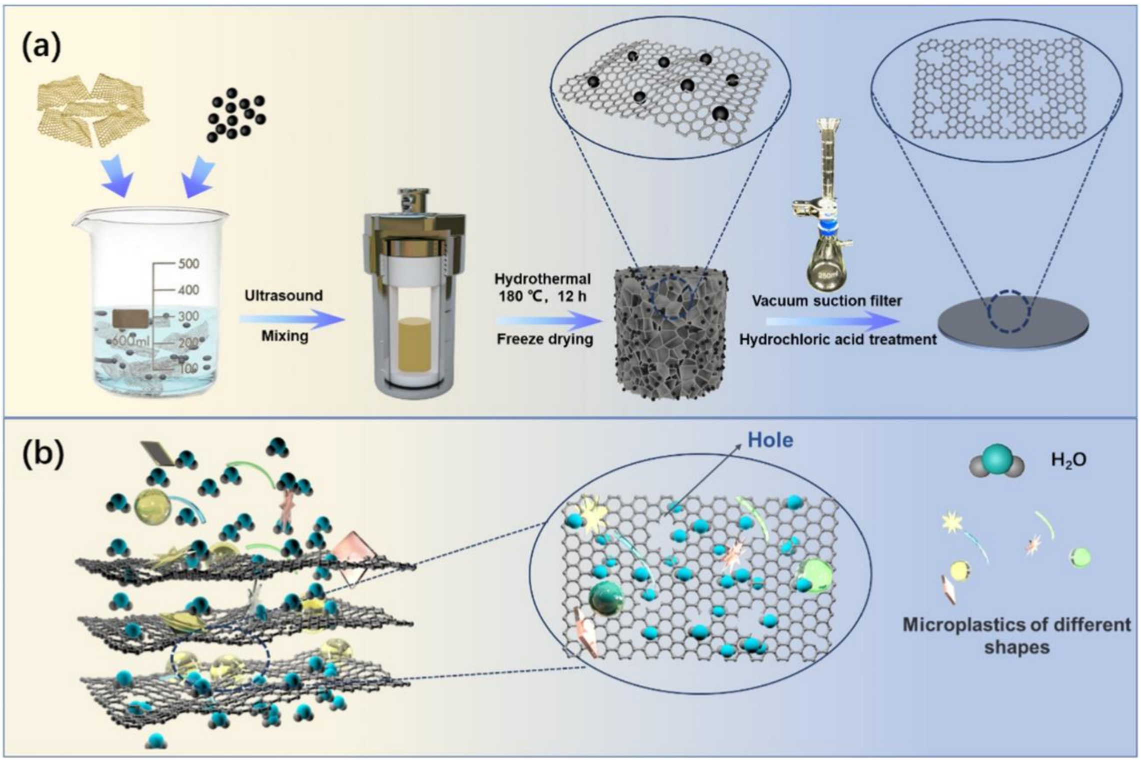



Figure 5: (a) Schematic diagram of the morphological formation of h-rGO and membranes based on h-rGO nanosheets formation, (b) schematic diagram of the filtration of MPs. (Reproduced from Yang et al. (2022b) with permission under creative commons CC-BY license, 2022).

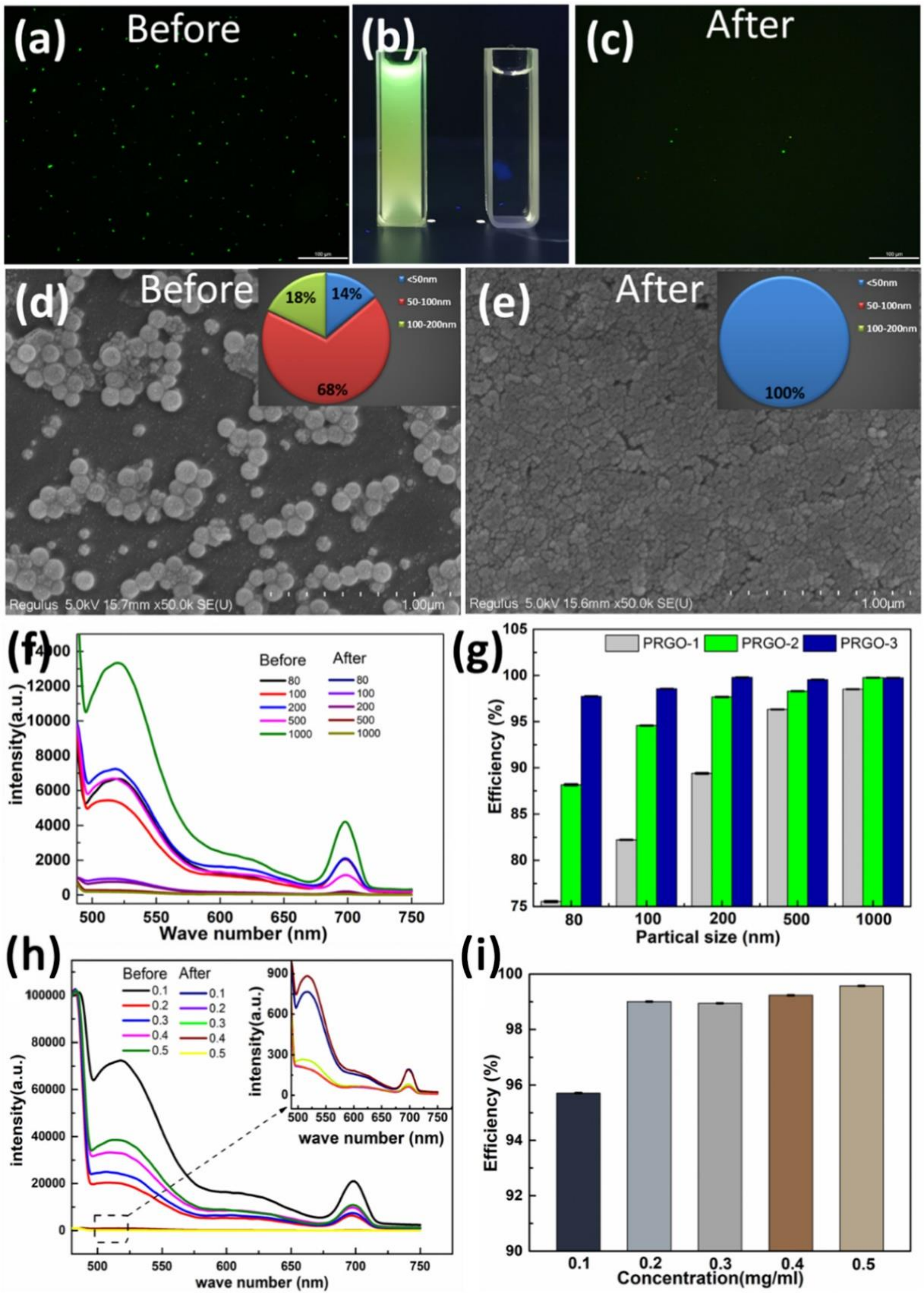



Figure 6. Removal of FP microspheres using membranes based on h-RGO nanosheets. (a), (c) Fluorescence microscopy view of pre-filtered and filtered sample, (b) FP microspheres suspension before and after filtration under UV lamp, (d) and (e) SEM images of FP microspheres before and after filtering, the inset plot is particle size distribution based on nano particle size and Zeta potential analysis. (f) Fluorescence intensity curves of FP microspheres with different particle sizes before and after filtration by h-RGO-1. (g) The removal efficiency of FP Microspheres with different particle sizes through three kinds of membranes. (h) Fluorescence intensity curves of different concentrations of 200nm FP microspheres before and after filtration by h-RGO-2. (i) The removal efficiency of different concentrations of 200nm FP microspheres filtration by h-RGO-2. (Reproduced from Yang et al. (2022b) with permission under creative commons CC-BY license, 2022).

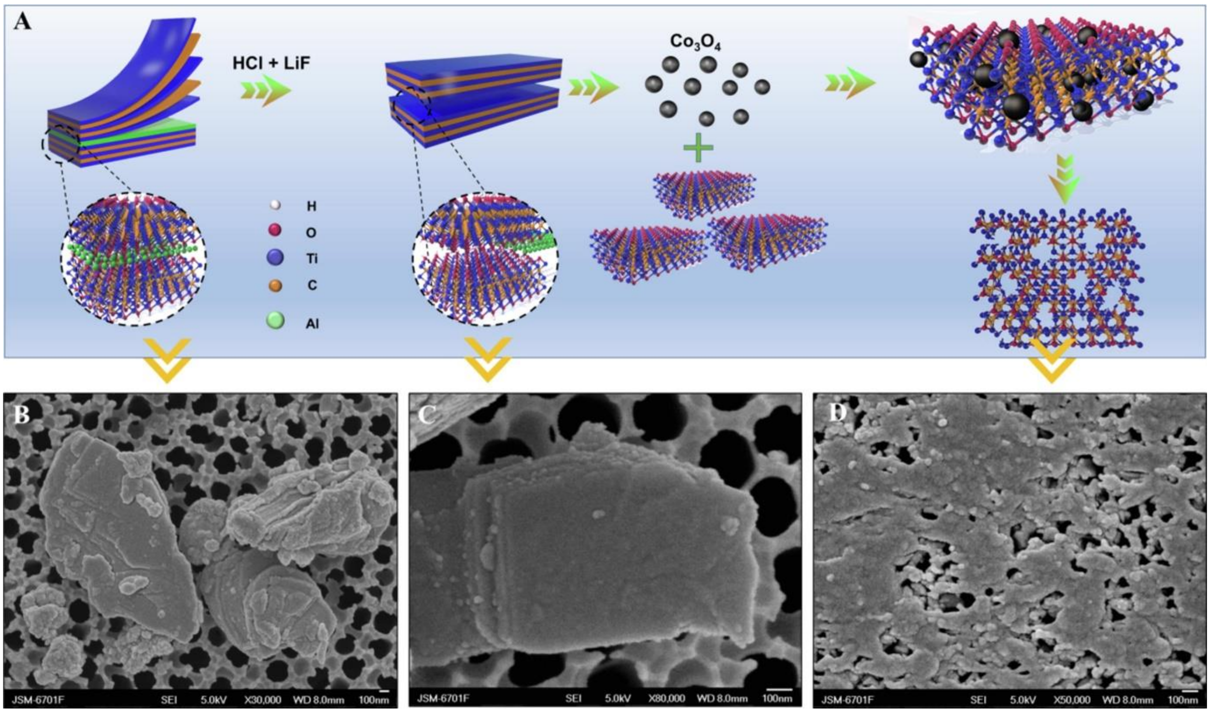



Figure 7. A) Schematic diagram of the morphological formation of h-$Ti_3C_2Tx$ formation, SEM image of $Ti_3AlC_2$(B), $Ti_3C_2Tx$(C) and h-$Ti_3C_2Tx$(D). Reproduced from Yang et al. (2022a) with permission under creative commons CC-BY license, 2022).

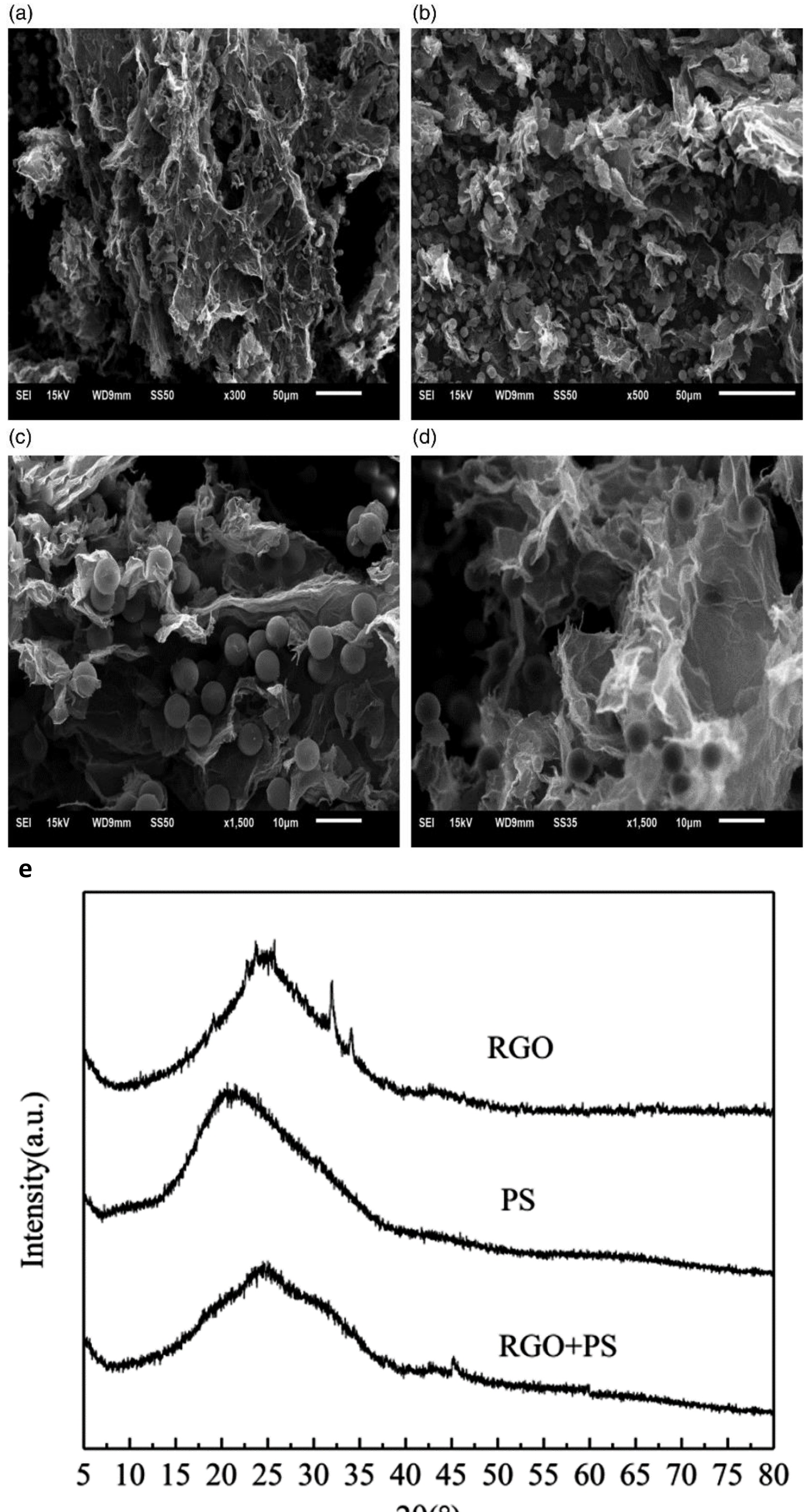
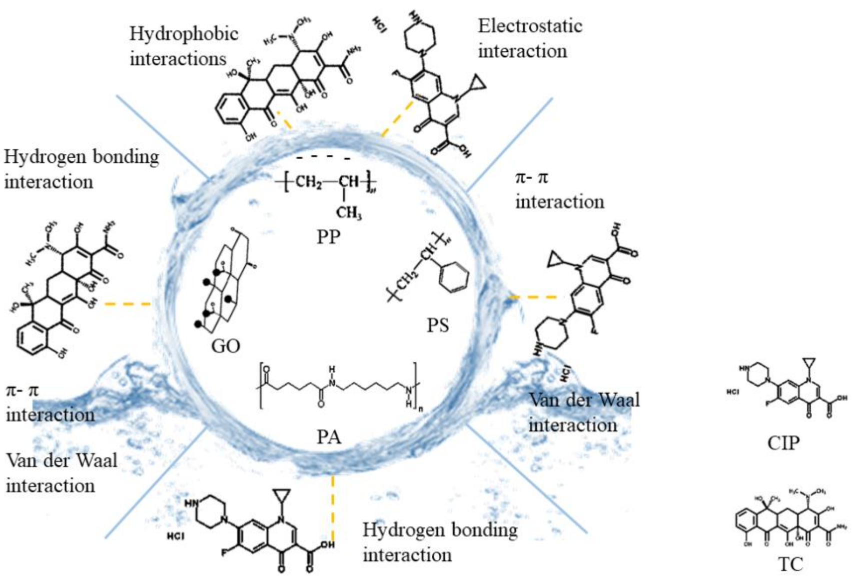



Figure 8: SEM images of 3D RGO after adsorption, magnified (a) 300 times, (b) 500 times, (c) 1,500 times, (d) 1,500 times, and (e) XRD patterns of 3D RGO, PS microplastics, and 3D RGO after adsorption. Reproduced from Yuan et al. (2020) with permission under creative commons CC-BY license, 2022).




1025

1026 Figure 9 Schematic diagram of the CIP and TC adsorption mechanism on MPs and GO. Reproduced from Liu
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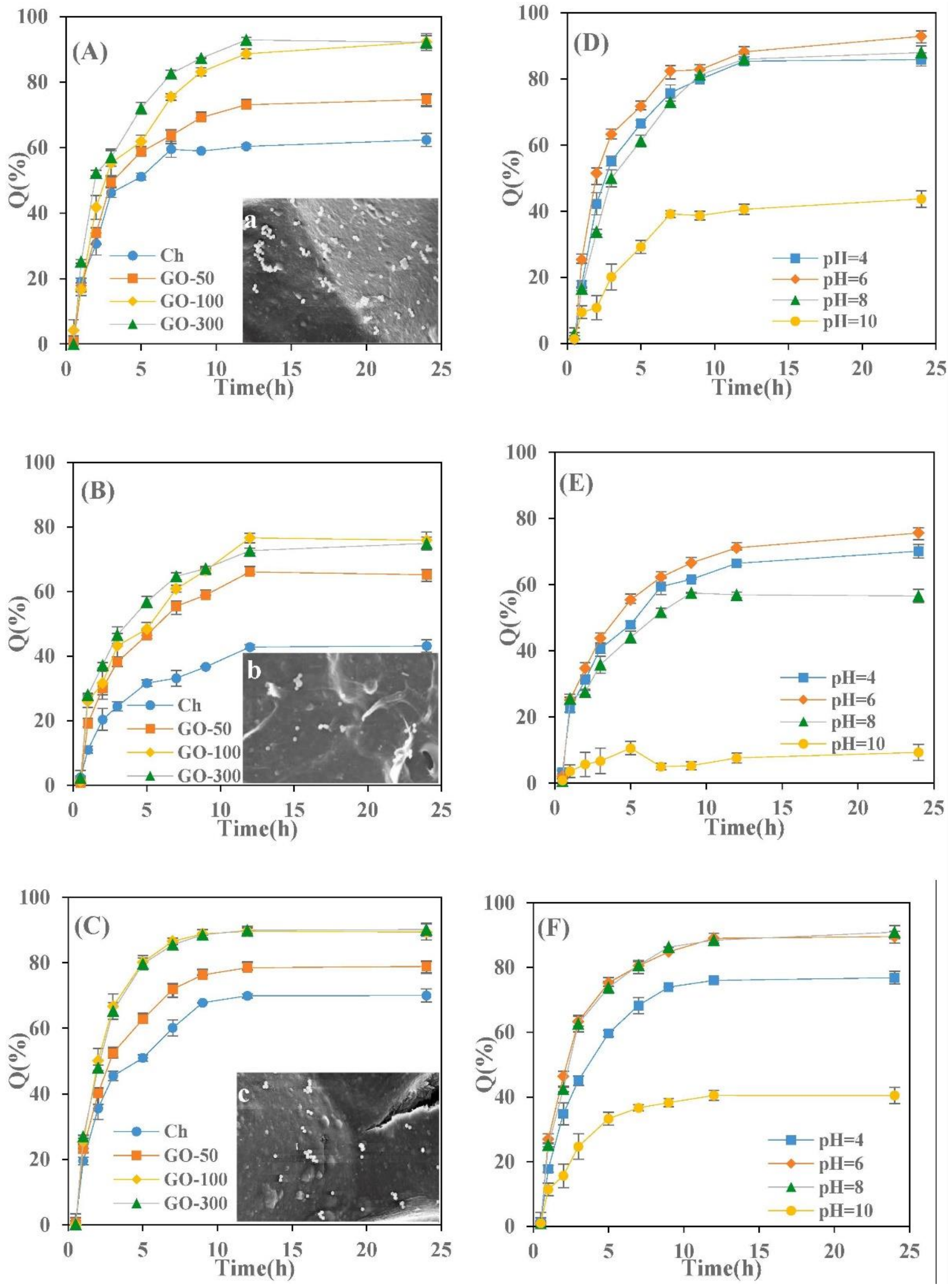


Figure 10. Adsorption capacity of PS (A), PS-COOH (B), and PS-$NH_2$ (C) by Ch, ChGO-50, ChGO-100, and ChGO-300 sponges at 25 °C when the pH is 7, the SEM image of the adsorption of PS (a), PS-COOH (b), and PS-$NH_2$ (c) on ChGO-300 sponges at 25 °C when the pH is 7, and the adsorption capacity of PS (a), PS-COOH (b), and PS-$NH_2$ (c) by ChGO-300 sponges at 25 °C when the pH is 4, 6, 8, and 10. The error bars represents standard deviation ($n$ = 3). Reproduced from (Sun et al., 2020), Copyright 2020, Elsevier.

**Table 1: Occurrence of various types of MPs found in various aqueous media**

| Sample type | Location | MPs concentration | MPs types | References |
|---|---|---|---|---|
| Sewage (locally treated) stormwater | Victoria Harbor (Hong Kong) | Effluents: 10,816 pieces $m^{-3}$ | PE and PP | (Mak et al., 2019) |
| Stormwater (retention ponds) | Denmark | 127,986-1511 items $kg^{-1}$ | PP, PS, PE, Polyurethane, PVC. | (Liu et al., 2019b) |
| Stormwater drainage system (sediments) | Perth region (Western Australia) | 0-3500 MPs $kg^{-1}$ of dry sediment<br>Mean concentration: 664 particles $kg^{-1}$ | Fibres, PE, PP, PET Polyamide (Nylon) | (Lutz et al., 2021) |
| Surface water | Yellow river (China) | 1760 ± 710 to 10,120 ± 4090 MPs $m^{-3}$ | Colored particles (fibres), PE, PS, Polybutylene terephthalate | (Wang et al., 2019d) |
| Sea water | Mediterranean Sea (NW) | 0.23 ± 0.20 MPs $m^{-3}$ | PET | (Lefebvre et al., 2019) |
| Mollusc species | Tunisia | 1031.1 items $kg^{-1}$ (ww) | PP and PE | (Abidli et al., 2019) |
| Surface water | Western Lake Superior | Mean: 1200 mg $km^{-2}$<br>Range: 91–3538 mg $km^{-2}$ (mass per unit area). | PVC, PP, PE | (Hendrickson et al., 2018) |
| Drinking water | Germany | Mean: 0.7 MPs $m^{-3}$<br>Range: 0–7 MPs $m^{-3}$ | PE, PA, Polyester PVC or Epoxy resin | (Mintenig et al., 2019) |
| Mineral water (bottled) | Bavarian (Germany) | From 2649 ± 2857 to 6292 ± 10521 MPs $L^{-1}$ | PE, Styrene-butadiene-copolymer | (Oßmann et al., 2018) |
| Surface water | Qinghai Lake (China) | 0.03 × 105 -0.31 × 105 MPs $km^{-2}$ | PE, PP | (Xiong et al., 2018) |
| Surface water | Snake and lower Columbia Rivers | 483-967 MPs $m^{-3}$ | PET, PE, PP, PS, PA | (Kapp and Yeatman, 2018) |
| Sea surface | Southeast Spain | 0.10 ± 0.09 MPs $m^{-2}$ | PE(54.5%), PP (16.5%), PS (9.7%) | (de Haan et al., 2019) |
| Water | Danjiangkou Reservoir (China) | 467–15,017 MPs $m^{-3}$ | Fibres, small-sized items, PP | (Di et al., 2019) |
| Waste water | Wastewater was collected at the Seine-Centre wastewater treatment plant | 260–320 × $10^3$ MPs $m^{-3}$ | Fibres | (Dris et al., 2015) |
| Surface water | Lake Winnipeg (Canada) | 4.74x $10^{943}$ | Fibres, Films and foams (less common) | (Anderson et al., 2017) |
| Surface water | Ottawa River | 0.05- 0.24 fragments $L^{-1}$ | Microfibers (70%-100%), plastic microbeads secondary plastic fragments | (Vermaire et al., 2017) |
| Plastic water bottles | Germany | Average: 118 ± 88 particles $L^{-1}$ in returnable bottles, 14 ± 14 particles $L^{-1}$ (single-use plastic bottles) | Most of the particles in PET ( 84%), PP (7%). | (Schymanski et al., 2018) |
| Surface water | Lake Bolsena, Lake Chiusi (Central Italy) | 2.68- 3.36 particles $m^{-3}$ (Lake Chiusi)<br>0.82- 4.42 particlesm$^{-3}$ (Lake Bolsena) | Fibres | (Fischer et al., 2016) |
| Highly urbanized river | Chicago (Illinois, USA) | 1.94 (0.81) $m^{-3}$ (upstream) 17.93 | Fibres, Fragments Pellets, Styrofoam | (McCormick et al., 2014) |

| | | | | |
|---|---|---|---|---|
| | | (11.05) $m^{-3}$ (downstream) | | |
| Surface water | Hudson River (USA) | Average= 0.985 microfibers $L^{-1}$ | Microfibers | (Miller et al., 2017) |
| Raw sewage sludge | Netherlands | Mean particle concentrations= 68–910 $L^{-1}$, 51–81 $L^{-1}$ and 510–760 $kg^{-1}$ wet weight (ww) (particle sizes between 10 and 5000 μm). | Fibres | (Leslie et al., 2017b) |
| Saigon River | Vietnam | 172,000-519,000 items $m^{-3}$ (fibres) 10-223 items $m^{-3}$ (fragments) | PE, PP, Fibers mainly made of polyester. | (Lahens et al., 2018) |
| Fresh water | Lake Hovsgol (Mongolia) | 20,264 particleskm$^{-2}$ | Fragments, Films | (Free et al., 2014) |
| Fresh water | Lake Maggiore Lake Iseo Lake Garda. | Fragments (73.7%) | PE (45%), PS (18%), PP (15%) | (Sighicelli et al., 2018) |
| Reservoir ecosystems | Xiangxi River (backwater area) | Concentrations ranging from 0.55 × $10^5$ to 342 × $10^5$ itemskm$^{-2}$ | PE, PP, and PS | (Zhang et al., 2017) |
| Freshwater | Taihu Lake ( China) | 3.4–25.8 items $L^{-1}$ | Fiber (100–1000 μm size, cellophane composition) | (Su et al., 2016) |
| Fresh water | Antuã River (Portugal) | 58–193 items $m^{-3}$ | PE, PP | (Rodrigues et al., 2018) |
| springs and wells (<65m) | karst aquifers, Illinois (USA) | Average= 6.4 particles$L^{-1}$ maximum concentration= 15.2 particles $L^{-1}$. | Fibres (>0.45 μm) | (Panno et al., 2019) |
| Groundwater (30 m deep wells) | Holdorf (Germany) | minimal concentrations= 0–0.007 particles $L^{-1}$ average concentration of =0.0007 particles $L^{-1}$ | Fragments 50–150 μm in size. | (Mintenig et al., 2019) |
| Groundwater boreholes | North West of South Africa | 0.0417 fragments $L^{-1}$ 0.1250 fibres $L^{-1}$ | Fragments | (Bouwman et al., 2018) |
| Marine water | Coastal waters (India) | 1.25 particles $L^{-1}$ | Fragments, Fibre/line, foam, PE, PP | (Robin et al., 2020) |
| Marine | - | - | Fragments, Fibre/line,foam, PE, PP, PS | (Karthik et al., 2018) |
| Marine | - | 0.004–4,137 particles$L^{-1}$ | - | (Avio et al., 2017) |
| River | - | 10–520 particles $L^{-1}$ | Film, Fragments, fibres,PE, PA, PP,PET | (He et al., 2020) |
| River | - | 50.3–1,600 particles $L^{-1}$ | PP, Polyester, Rayon cotton, viscose phenoxy resin, Poly (vinyl stearate) | (Peng et al., 2018) |

1043 **Table 2: Different strategies and outcomes of MPs removal from water**

| Strategies to remove MPs | Water type | Study nature/location | Removal rate of MPs/removal efficiency | References |
|---|---|---|---|---|
| Mechanical and chemical pretreatment | wastewater | Finland | 97.4-98.4% | (Talvitie et al., 2017a) |
| Screening, grit removal, sedimentation | wastewater | Australia | 31.8% | (Ziajahromi et al., 2021) |
| Screening, grit separation | wastewater | Finland | 90% | (Lares et al., 2018) |
| Primary settling tank | wastewater | South Korea | 62.7% | (Hidayaturrahman and Lee, 2019) |
| Aerated grit chamber Oxidation ditch | wastewater | China | 16.5% | (Lv et al., 2019) |
| Aerated grit chamber Sedimentation tank | sewage water | China | 58.84 ± 8.05% | (Yang et al., 2021) |
| Coagulation sedimentation | DWTP | China | 40.5 to 54.5% | (Wang et al., 2020c) |
| Activated sludge Rapid sand filtration | wastewater | Denmark | 99% | (Simon et al., 2018) |
| Membrane bioreactor | WWTP | Finland | 99.9% | (Talvitie et al., 2017a) |
| Secondary setting tank | WWTP | China | 97% | (Lv et al., 2019) |
| $A^2O$ treatments | sewage treatment plant | China | 54.47 ± 14.73% | (Han et al., 2019) |
| Disc filter | wastewater | Finland | 40% to 98.5% | (Talvitie et al., 2017a) |
| Rapid sand filter | wastewater | Finland | 97.1% | (Talvitie et al., 2017a) |
| Dissolved air flotation | wastewater | Finland | 95% | (Talvitie et al., 2017a) |
| RO | wastewater | Australia | 90.5% | (Ziajahromi et al., 2017) |
| Ozone | wastewater | South Korea | 99.2% | (Hidayaturrahman and Lee, 2019) |
| Membrane disc filter | wastewater | South Korea | 99.1% | (Hidayaturrahman and Lee, 2019) |
| Coagulation (ferric chloride and aluminum chloride treatment) | In a synthetic drinking water matrix. | | Al-based coagulant= 36% Fe-based coagulant=17% | (Ma et al., 2019a) |
| Sedimentation biofilter | WWTP | Paris France | 88.1% | (Xu et al., 2021c) |
| Grit separation | WWTP | Mikkeli (Finland) | 99.3% | (Xu et al., 2021c) |
| Reverse osmosis | WWTP | Sydney (Australia) | 98.25% | (Xu et al., 2021c) |
| Activated sludge process | WWTP | Finland | 99.9% | (Xu et al., 2021c) |
| Aeration, activated sludge | WWTP | Los Angeles (United States) | 99.9% | (Carr et al., 2016) |
| Grit removal, aeration, sedimentation | WWTP | Glasgow, Scotland (United Kingdom | 98.41% | (Murphy et al., 2016) |
| Activated sludge, grit removal, screening | WWTP | Detroit (United States) | 95.6% | (Michielssen et al., 2016) |
| Membrane bioreactor | WWTP | Netherlands | 25% | (Leslie et al., 2017a) |
| $A^2O$, sedimentation | WWTP | Wuhan (China) | 40.7% | (Liu et al., 2019d) |
| $A^2O$, aerated grit chamber, sedimentation | WWTP | Beijing (China) | 58.8% | (Yang et al., 2019) |
| Aeration | WWTP | Seyhan, Adana (Turkey) | 73% | (Gundogdu et al., 2018) |

| | | | | |
|---|---|---|---|---|
| Sand filter treatment, disinfection, sedimentation | WWTP | Northern Italy | 84% | (Magni et al., 2019) |
| MBR (Ultrafiltration) | MWWTP | - | 99.9-100% | (Talvitie et al., 2017a) |
| RO (reverse osmosis) | MWWTP | - | 99-100% | (Sun et al., 2019) |
| Powdered activated carbon and ultrafiltration | MWWTP | - | 99-100% | (Baresel and Olshammar, 2019) |
| Dynamic membrane of non-woven fabric, woven filter, stainless steel mesh | MWWTP | - | 99.5% | (Zhang et al., 2019a) |
| RSF (sand filtration) | MWWTP | - | 97% | (Talvitie et al., 2017a) |
| Dissolved air flotation | MWWTP | - | 95% | (Talvitie et al., 2017a) |
| Hybrid sand and biochar filtration | - | - | >95% | (Wang et al., 2020d) |
| Iron-modified biochar pyrolyzed at 550 °C & 850 °C | - | - | ∼100% | (Singh et al., 2021) |
| Steam activated pine and spruce bark biochar pyrolyzed at 475 °C & steam-activated at 800 °C | - | - | ∼100% for cylindrical PE pieces and fleece fibers. | (Siipola et al., 2020) |
| Mg/Zn modified magnetic biochar | - | - | 98.75% for Mg-MBC 99.46% for Zn-MBC. | (Wang et al., 2021b) |

**Table 3: Comparison of contrasting characteristics of different membranes (Shon et al., 2002; Sagle and Freeman, 2004)**

| Properties | RO | NF | UF | MF |
|---|---|---|---|---|
| Material | Cellulose acetate or polysulfone coated with aromatic PAs | Cellulose acetate blends or PA composites like the RO membranes, or they could be modified forms of UF membranes such as sulfonated polysulfone | Poly(vinylidene fluoride), polysulfone, poly(acrylonitrile) and poly(acrylonitrile)-poly(vinyl chloride) copolymers. Poly (ether sulfone) | Poly(vinylidene fluoride), polysulfone, poly(acrylonitrile) and poly(acrylonitrile)-poly(vinyl chloride) copolymers |
| Effective in removal | All contaminants | Can remove ~50% hardness, >90% color | Colloids and macromolecules such as protein, dyes and polymeric substance | Bacteria, suspended solids, organics, |
| Common use | For salts and LMW pollutants phenolic wastewater from paper mill, oily wastewater, dumpsite leachate | Textile | Industrial WWTPs, vegetable oil factory, metal finishing industry, oily wastewater, phenolic wastewater from paper mill | Membrane bioreactors, municipal wastewater (disinfection and phosphorus removal), synthetic emulsified oily wastewater |
| Advantages | More efficient for drinking water treatment | Higher removal of organic than RO, Economic due to low operating pressure | High recovery for paints, lignin, black liquor | Permeate can removed by backwash |
| Limitations | More vulnerable to organic fouling | Both charge and size of pollutants impacts more vulnerable to organic fouling | Unable to remove soluble pollutants, pH sensitive, more frequent backwash require | High membrane fouling, high cost |
| Composition | Composite/asymmetric Nonporous | Composite/asymmetric Finely porous | asymmetric Porous | Isotropic Porous |
| Filtration routes | Diffusion | Ions and small molecule: Electrostatic hydration diffusion; Macromolecules and colloids: sieving | Sieving/preferential adsorption | Sieving and adsorption |
| Pure water flux | 10-100 | 20-200 | 100-2,000 | 500-10,000 |
| Pressure | 20-100 atm | 7-30 atm | 1-10 atm | 0.5-5 atm |
| Molecular weight | <200 Da | Tight: 200-300; Loose: 300-1000 | Tight: 1000-10000; Loose: 1000-100000 | Tight: 100000-0.01µm; Loose: 0.01 µm -0.05µm |

**Table 4: Efficiency of graphene, GO, rGO and MXenes in treatment of MPs**

| Types of Materials | MP types | Experimental conditions | Removal | Observation | Reference |
|---|---|---|---|---|---|
| Holey $Ti_3C_2$ membrane | fluorescent PS MPs | 0.4 mg $ml^{-1}$ (M1) to 1.2 $mgml^{-1}$ (M5); 0.8 mg $ml^{-1}$ of h-$Ti_3C_2Tx$ | 99.3% | The removal mechanism was size exclusion. Archived high flux rate: 196.7 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$ to 68.9 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$ | (Yang et al., 2022a) |
| Chitin and graphene oxide sponges | PS, PS-COOH- and PS-$NH_2$ | 0, 0.2, 0.5, 1, and 3 mL of GO | 89.8%, 72.4%, and 88.9% for PS, PS-COOH- and PS-$NH_2$ | Electrostatic interactions, hydrogen bond interactions, and $\pi - \pi$ interactions, the adsorption efficiency of PS:5.89. 7.52, and 8.46 mg $g^{-1}$ at 25, 35, and 45 ℃ | (Sun et al., 2020) |
| Chitin-GO and Chitin-O-$C_3N_4$ sponges | PS, PS-COOH- and PS-$NH_2$ | GO=0.3 g; O-$C_3N_4$=0.3 g; Chitin=6 g | Removal rate of all three types of MPs=71.6–92.1% | Chitin provides robust sponge structure and support embedded GO and O-$C_3N_4$ to have $\pi - \pi$ for MPs adsorption. | (Sun et al., 2021a) |
| Holey reduced graphene oxide (h-rGO) nanosheets | PS | 0.25 mg $ml^{-1}$, 0.5 mg $ml^{-1}$, 0.75 mg $ml^{-1}$ h-rGO; 30% $H_2O_2$; 48 h; | 99.9% | Size exclusion and adsorption 37.19 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$ | (Yang et al., 2022b) |
| rGO/PAN | PET | 0.11% to 0.83% w/w; alkaline solution 300 W Xe lamp (>420 nm); Catalyst dose/mg 10 | > 82% | 150 nm to be effectively separated. | (Fryczkowska and Przywara, 2021) |
| MXene/$Zn_xCd_{1-xS}$ photocatalysts | PETMPs | 50 mL PET solution | 100% | The photocatalytic $H_2$ evolution rate was 14.17 mmol $g^{-1}$ $h^{-1}$, glycolate, acetate and methanol were generated | (Cao et al., 2022) |
| Ag/$TiO_2$ modified using reduced graphene oxide | $TiO_2$ and UV | 3%Ag/$TiO_2$–1% RGO; Ag/$TiO_2$ and pure $TiO_2$ | 56-76% | 3%Ag/$TiO_2$–1% RGO catalyst 76% degradation compared with 68 and 56% for Ag/$TiO_2$ and pure $TiO_2$, respectively | (Fadli et al., 2021) |
| Reduced graphene oxide 3D fibrous aerogels (*m*-$TiO_2$/RGO aerogel) | mixed suspension including dominant *m*-TiO2 nanofibers (with the average diameter of 167 nm | PE microfibers with a length of 3 mm | MPs removal efficiency (~100%) electricity-free system | pH influenced interactions among organic pollutants, MPs and *m*-$TiO_2$/RGO fibrous surfaces abundant $O_2^{\cdot}$– and $OH^{\cdot}$ free radicals were generated from *m*-$TiO_2$/RGO aerogel during solar evaporation, indicating the ROS accelerated photodegradation in PE removing | (Meng et al., 2022) |
| 3D rGO adsorbent | PS | 3D rGO concentration: 1.5 mg; reaction time:2 h | 0.6 g $L^{-1}$ | Spontaneous endothermic process. The strong π–π interaction between the carbon ring of 3D RGO and the benzene ring of PS MPs. Structural specification of rGO facilitates PS removal | (Yuan et al., 2020) |
| Three-dimensional reduced graphene oxide | pH=6, $C_0$ = 600 mg/L, t = 120 min, and T =26 °C | PS | 617.28 mg $g^{-1}$ | Spontaneous endothermic process; adsorption mechanism was mainly attributed to the strong π–π interaction between the carbon ring of 3D RGO and the benzene ring of PS MPs | (Yuan et al., 2020) |

| | | | | | |
|---|---|---|---|---|---|
| $TiO_2$/graphite | polyvinyl chloride (PVC) | at 100 °C for 6 h; potentiostatic electrolysis at -0.7 V vs. Ag/AgCl | 56 wt % removal and 75 % dechlorination | PVC degradation at a high temp. PVC dechlorination was mainly via direct reduction by the applied cathode potential, meanwhile, oxidization of OH resulted in the oxidation and breakage of PVC backbone, Organics shedding promoted further dechlorination of PVC MPs indirectly. | (Miao et al., 2020) |
| $Fe_3O_4$/laser-induced graphene | MPs particle size: Melamine (2 and 10 µm), PS (10 µm), PA (50 µm) | $Fe_3O_4$-LIGPs dosage: 5 g $L^{-1}$ | MPs were 1400 (PA), 1250 (PS), 1050 (melamin:10 µm), and 775 (melamine:2 µm) mg $g^{-1}$ | Adsorbes all types of MPs which are easily desorbed by magnetic force. Reusable over six cycles. possesses great potential for industrial applications, such as domestic sewage or slow-flow water treatment plants | (Jeong et al., 2022) |
| Graphene oxide (GO) - polyvinyl alcohol (PVA) based composite membrane | HDPE | GO: 0.5-5.5 ratio to PVA pH 8, with 3.5 bar of transmembrane pressure, and 15s time | 95% HDPE rejection was noticed | Electrostatic repulsion between membrane surface and MPs along with sieving capacity; highest permeability of fabricated membrane was 179 L $m^{-2}$ $h^{-1}$ k $Pa^{-1}$ | (Dey et al., 2022) |

**Authorship contributions**

**Conceptualization**: Mehmood and B.Mustafa; Data curation: T. Mehmood; B.Mustafa and W. Anum; **Formal analysis**: T.Mehmood, K. Mackenzie, and B. Mustafa; **Funding acquisition**: L.Peng; **Investigation**: B. Mustafa, T. Mehmood and W. Anum; Methodology: B. Mustafa, W. Ali, R. I. Sabir and W. Anum; **Project administration**: L. Peng, K. Mackenzie; **Resources**: L. Peng; K. Mackenzie; **Software**: T. Mehmood and W.Anum and B.Mustafa; **Supervision**: L. Peng; K. Mackenzie; **Validation**: B. Mustafa, L. Xinghui, and G.K. Gaurav; **Visualization**: B. Mustafa, W. Ali, R. I. Sabir, L. Xinghui, W. Anum and G.K. Gaurav; **Writing – original draft**: T. Mehmood, W. Anum and B. Mustafa; **Writing – review & editing**: K. Mackenzie, T. Mehmood, and L. Peng.

All authors have approved the manuscript and agree with its submission to Chemosphere.

**Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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# Supplementary information

## Recent developments in microplastic contaminated water treatment: Progress and prospects of carbon-based two-dimensional materials for membranes separation

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## 1. Environmental implication of MPs in water

As synthetic organic polymers, MPs can exist stably in aquatic environments for a long time due to their special properties (small particle size, large specific surface area, lightweight, strong hydrophobicity, and variable density). In seawater, plastic particles of high-density sink towards sediments, accumulate there, and become more accessible to benthic organisms. The less-dense particles endure at the sea surface until bio-fouling occurs, sunlight exposure enables breakdown, or when they entrain in aggregates like fecal pellets. After gaining density, they sink into the sediment (Andrady, 2011). However, due to atrophy, the size of nanoparticles increased. Characteristics of aqueous media, like pH, salinity, etc., may influence nanoparticles' physiochemical structure (coating, surface charge). Resultantly, aggregation/agglomeration and hydrophobicity processes fluctuate, eliciting varying distribution in the water column. Marine sediments are thus considered long-term sinks for nano plastic particles (Oliveira and Almeida, 2019; Javeed et al., 2021). Zoo planktonic, for example, marine copepod *Tigriopus japonicus* mortality, has occurred due to a high level of polystyrene plastic particles (Lee et al., 2013).

MPs are self-toxic, and their ingestion by animals has negative effects on growth, intestinal tissues, etc. (Rodriguez-Seijo et al., 2017; Wang et al., 2019a), and a variety of additives often accompanies the manufacturing process of plastics (e.g., antioxidants, pigments, plasticizers, etc.), and their exposure to external factors (e.g., shear, UV irradiation, weathering, etc.) are easily released into the environment (Lambert et al., 2014; Paluselli et al., 2018; Wang et al., 2020b), thus endangering the health of organisms. In addition, MPs are also loaded with toxic, and their smaller particle size and larger surface area lead to their strong adsorption properties, which can readily adsorb various toxic substances in the environment (e.g., polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), antibiotics, etc.), thus indirectly exerting toxic effects on biological processes (Wang et al., 2020a; Wang et al., 2021a).

The range of problems resulting from inadvertent uptake of MPs by organisms (including reduced foraging ability, digestive tract blockage, and nutrient loss) is a major severe environmental challenge (Graham and Thompson, 2009; Bakir et al., 2012), and it has been shown that MPs have a wide range of biological effects, with filter-feeding and deposit-feeding invertebrates (Xu et al., 2020b), seabirds (van Franeker, 1985), crustaceans (Murray and Cowie, 2011) and commercially (Lusher et al., 2013) have been shown to ingest MPs that will eventually enter humans body through the food chain (Habib et al., 2020). MPs pollute the land, lakes, and waterways, as well as coastal areas, estuaries, and oceans. On land, plastic particles have been found in human consumables (Kosuth et al., 2018), and accumulation has been observed in human embryos (Ragusa et al., 2021; Wang et al., 2021a), detected in the blood (Leslie et al., 2022), in human stools (Schwabl et al., 2019), suggesting widespread contamination of the human food chain, and thus, representing a potential threat to human health.

Aquatic and terrestrial ecosystems are threatened by MPs pollution since MPs are absorbed by aquatic microbiota (e.g., microalgae) and fishes (Galloway et al., 2020). Humans consume a lot of fisheries since it's a high-quality protein source. MP-contaminated fish and other seafood can cause human illnesses (Gündogdu et al., 2022). MPs, associated metals, and organic compounds can affect human health by interfering with metabolism. MPs polymerization process absorbs metal contaminants. In this way, it can carry organic contaminants and aid bioaccumulation in exposed organisms (Mehmood and Peng, 2022).

MPs can disrupt the food chain by causing physiological stress in living bodies, altering the balance and health of the ecosystem (Gündogdu et al., 2022). MPs combined with harmful metals and organic species can disturb low trophic species, especially microalgae like *Chlorella vulgaris* and *Chlorella pyrenoidosa*. It can slow algae development procedure by boosting oxidative stress and affecting superoxide dismutase, catalase, etc. (Yu et al., 2022). Due to oxidative stress, reactive oxygen species develop and accumulate in algal cells. Increased ROS and stress oxidation drive lipid peroxidation by creating more malondialdehyde (MDA), a peroxidation byproduct. Higher MPs concentrations and metal-absorbed MPs affect algae growth and chlorophyll production (Wu et al., 2019; Yu et al., 2022). Additionally, MPs lessen sunlight penetration in the water column and travail photosynthetic organisms.

Fish are also particularly sensitive to MPs exposure and ingestion. Pathogenic microorganisms, plastic additives, and organic and metal pollutants on bonds on MPs surface can concentrate in exposed fish. Several investigations found MPs polymers in fish digestive tracts. Because of their dimensions and shape, microscopic MPs particles are able to penetrate the epidermis, lymphatic systems, and gills of sea species (Calderon et al., 2019). Ingesting MPs particles can cause fish digestive tract damage and obstruction. It affects fish eating, growth, and nutritional absorption (Jabeen et al., 2018). MPs can cause allergic reactions and affect fish's natural immunity (Wen et al., 2018). MPs can pass through fish's circulatory system and injure organs, i.e., the liver (Barboza et al., 2018). The study reveals that MPs chemical additives, including PAH, PCB, and PBD, can be kept in fish intestines and transported between trophic levels. Biofilms adhering to MPs can cause bacterial infections in fish (Wang et al., 2019c). MPs can affect fish's metabolic, oxidative stress, enzyme activity, and reproductive and endocrine systems (Law, 2017). MPs are more likely to affect young fish (Duran and Beiras, 2017). MPs can impair fish larval development, motility, head-to-body length, and hatching time.

Along with these, MPs and chemical additives cause cardiovascular irregularities, DNA breakdown, and larval death (Kogel et al., 2020). Water bodies contain visible amounts of wrappers, containers, and plastic bags. They not only pose an unaesthetic look, and cause an interruption in natural river and stream flows but also marine animals intertwined in them, which cause suffocation.

Up to 60-80% of marine life is affected by plastic pollution, which originates from Land bases (Barboza et al., 2018; Abalansa et al., 2020)  Tsang et al. (2017) reported ample MPs in Hong Kong surface waters sediments and in edible fish like *Mugil cephalus*. Lin et al. (2018) identified the origin of microplastics as upstream cities from where water flows to Pearl River Plume in Hong Kong waters, while Prata (2018) stated MPs discharge from local stormwater outfalls and sewage treatment works. Ordinarily, Organic matter (OM), suspended solids (SS), and nutrients present in sewage are removed in sewage treatment works. However, illicit drugs, trace metals, pharmaceuticals, and smaller-sized  MPs (less than 1mm) are still retained there  (Binelli et al., 2015). Fish and other aquatic animals face diverse and life-threatening conditions after MPs are discharged into the recipient's water from STWs or SWOs (Güven et al., 2017).  MPs enter food chains via biomagnification and bioaccumulation  (Peng et al., 2022b). Other sources of MPs are frequently used personal care products, which result in the accumulation of MP/NPs in microbeads and microfibers (Napper and Thompson, 2016).

Although there are various regulations for marine litter pollution, there are fewer regulations for MPs. Since 2014, when the Netherlands prohibited microbeads in cosmetics, regulations have been enacted in many countries, including Sweden, Australia, New Zealand, Italy, Canada, the United Kingdom, and the United States (OECD, 2021).

MPs are exogenous particles with hydrophobic surfaces and aid in carrying numerous pesticides and organic compounds with hydrophobic properties preexisting in soils (Jiang et al., 2020). The recurring exertion of such fungicides leads to their accretion in soil, and after coming in contact, pesticides stick to MPs surfaces. Diverse interactions occur, which leads to ecosystem disturbances. Therefore, with time, a replacement of polymers with biodegradable materials is a prerequisite (Jiang et al., 2020).

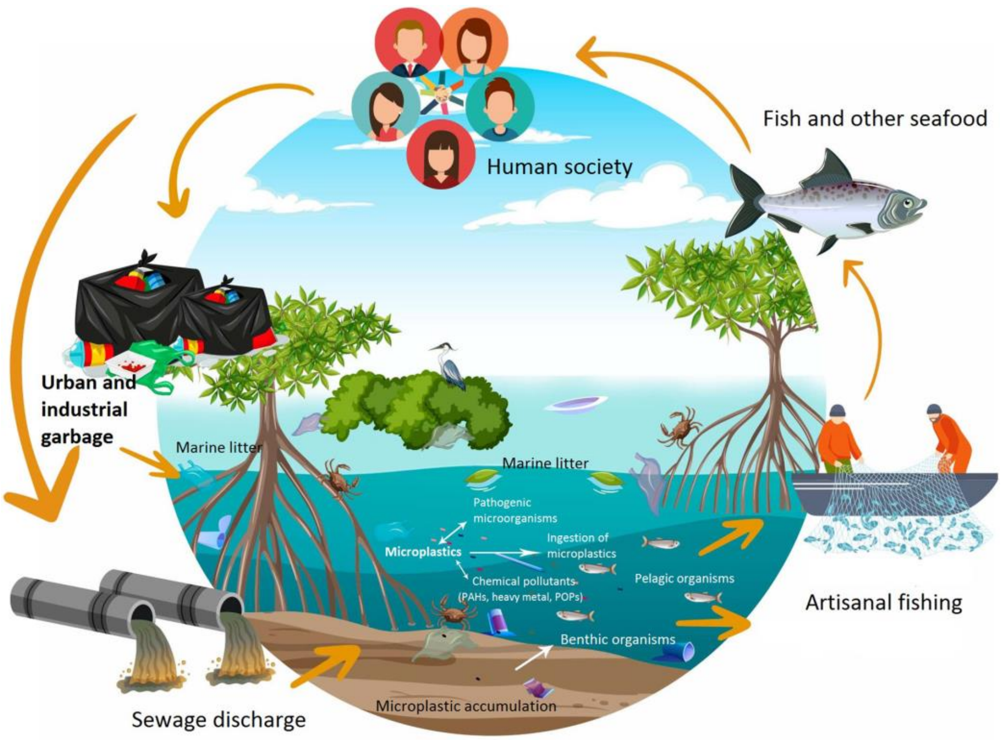


Figure S1 Sources, transportation and consumers of MPs pollution in water. Reproduced with permission from ref. (Garcés-Ordóñez et al., 2022) Copyright 2022, Elsevier.

## 2. Current strategies for removal of Micro/nano plastics from aquas media

Various ways have been explored to date to eradicate MPs from water. Although wastewater treatment facilities (WWTPs) can remove 95% of MPs (Talvitie et al., 2017a), some MPs can still circumvent WWTPs and reach the aquatic environment. Due to the high volume of wastewater discharged by WWTPs, it is predicted that between 15,000 and 4.5 million microplastic particles are released daily into surface water, independent of treatment effectiveness (Shen et al., 2021). In addition, the coagulation procedures described in prior investigations do not eliminate MPs (Ma et al., 2019a). Ozonation is a novel approach for treating MPs; however, in some circumstances, it is nearly ineffective since it mainly reduces big MPs to smaller ones, resulting in a slight increase in the output concentration of MPs relative to the input (Heinrich et al., 2020). Inadequate ozonation treatment may also result in the development of chemicals that are hazardous to human health and ecosystems. Membrane filtration may limit MPs' escape based on the MPs' size and membrane type, and pore structure, making it a very effective solution to the MPs in wastewater problem(Li et al., 2018; Poerio et al., 2019). Several removal processes exist for MPs, but sedimentation and deposition are the most prevalent (Liu et al., 2019a). MPs' form, size, and density dictate their eventual destination and mobility in water. Biofilm formation on the surface of MPs aids in their deposition and sedimentation, particularly for polymers with a low density (Carr et al., 2016).

Some researchers have advocated mixing RO and UF membranes to completely remove MPs (Goh et al., 2018; Tang and Hadibarata, 2021), improving the microplastic removal rate. The majority of conventional membrane materials are organic polymers with limited water permeability and vulnerability to contamination(Guha et al., 2017; Maliwan et al., 2021). Recent research has shown that inorganic two-dimensional (2D) layered membranes with high chemical and thermal resistance, such as graphene oxide (GO) and MXene, offer considerable separation and purification potential (Zazoum et al., 2021; Mustafa et al., 2022a). In aqueous separation, these two-dimensional inorganic materials rely on interlayer gaps for water flow, resulting in low water flux. Consequently, it is essential to continue searching for membranes with high water flow and MP removal effectiveness. Some major strategies which are used for MPs removal include bioretention(Lange et al., 2021b; Smyth et al., 2021; Stang et al., 2022), coagulation(Ma et al., 2019b; Lapointe et al., 2020; Skaf et al., 2020), floatation (Egger et al., 2020; Ziajahromi et al., 2020b), chemical degradation (Rodríguez-Narvaez et al., 2021).

## 2.1. Bioretention

Bioretention systems have been proven to be effective in removing emerging pollutants in earlier studies (Mehmood et al., 2021), as well as the elimination of particulate pollutants such SS, particulate materials, and PAH-associated particles (Liu et al., 2016b). Meanwhile, only two studies have demonstrated that bioretention effectively removes microplastic. A rain garden bioretention system was able to remove MPs of particle size effectively>125 m; however, the data was based on just three rainfall events. A parking lot bioretention cell was successful in removing MPs particles (>106 m) (Smyth et al., 2021). Pre-treatments like oven drying, pre-sieving (for coarse and small particles), and chemical oxidation have an impact on the effectiveness of sedimentation. The effectiveness of the technique depends on the sediment's composition. The mineralogy, the amount of organic matter, and the particle size all impact the final findings and the method's complexity. Optically separating and identifying materials is always challenging, especially now that challenges in distinguishing plastic from non-plastic materials have been recognized (Tamminga et al., 2019; Li et al., 2020c). Understanding particle removal and movement in SCM reveal microplastic removal and transport pathways (Tirpak et al., 2021). MPs are removed from stormwater through settling, adsorption, and filtering (Stang et al., 2022). Even though most MPs will be filtered out in topsoil due to their size (Han et al., 2022), smaller MPs may travel downstream with runoff. Research shows microspheres larger than 2 m have limited transport capacities (Gao et al., 2021). SCMs are prone to precipitation penetration or dry-wet cycles, which can accelerate microplastic downward migration (O'Connor et al., 2019). Due to a lack of depth distribution data from SCM, we cannot understand the vertical movement of MPs. A membrane filtering facility before and within the bioretention system reduces pollutants in stormwater (reference); however, this needs to be thoroughly researched for MPs treatment.

## 2.2. Floatation

One method currently being used to filter MPs is to separate them based on densities. By using density and floatation, the denser particles sink to the bottom of the sample, and the lighter particles will float to the top. Most plastics, such as polypropylene and polyethylene, are lighter than seawater (Mai et al., 2018). Common chemicals used in this type of separation are zinc chloride, sodium chloride, and sodium iodine due to their inexpensiveness (Coppock et al., 2017). Any sediments found in a sample can sink while other plastic particles floating in the remaining water. This method helps isolate the microplastic by removing the denser sediments.

MPs are also separated from sediments (marine, estuarine) through density separation methods. As stated by Kononov et al. (2022), agitation of sediments samples with salt solution results in the floatation of MPs. Yet, floatation methods are governed by the density of their corresponding salt solutions. Hidalgo-Ruz et al. (2012)specified those common plastics are in the range of 0.8-2.35 g/cm3 density, further along with Coppock et al. (2017) indicated that low-density salt solutions (sodium chloride) are not appropriate for eradicating high-density plastics. Contrariwise, Na Br, $ZnCl_2$, and NaI as high-density salts do not discern among plastic particles and other elements, thus making the separation process challenging. Furthermore, prodigious variation in toxicity, price, reactivity, and waste disposal of salt makes the density separation method tough and hinders or prohibits laboratories from endeavoring high-density salts (Cashman et al., 2020).

## 2.3. Chemical Degradation

Few studies have employed chemical digestion, and even fewer have used specialized removal procedures such as wet oxidation and advanced oxidation for MPs removal (Rodríguez-Narvaez et al., 2021). Kang et al. (2019) demonstrated high temperature ((>100 °C) and acidic conditions (pH=3) in combined system of peroxymonosulfate (PMS) and manganese modified carbon nanotubes (Mn@CNTs) and acidic conditions (pH = 3) resulted in 50% weight of MPs in 8 hours reaction time. Fenton-like system, can substantially change physicochemical properties and reduced MPs weight. Nonetheless, despite the positive findings, significant knowledge gaps persist. There is no information, for example, on the impact of the primary variables studied for MPs degradation (e.g., PMS concentration, Mn@CNTs load). MPs could possibly be degraded using other sophisticated oxidation techniques (for example, the Co/PMS reaction), which have been found to be extremely effective at degrading organic pollutants (Ghanbari and Moradi, 2017). Miao et al. (2020) confirmed that the electro-Fenton method with a $TiO_2$/graphite cathode was extremely successful for the degrading of polyvinylchloride (PVC) MPs. The researchers revealed that using a $TiO_2$/graphite cathode boosted the synthesis of $H_2O_2$, creating a higher number of hydroxyl radicals than the typical Fenton reaction. The dechlorination of PVC MPs was found to be 80% under these conditions. However, a fundamental obstacle to the general application of chemical degradation techniques is secondary MPs produced by chemical or biological mechanisms. Meanwhile, MPs have been effectively removed by chemical and electrochemical coagulation-flocculation techniques (Rodríguez-Narvaez et al., 2021).

## 2.4. Coagulation

Coagulation is a widely used, affordable method of wastewater treatment (Monira et al., 2021). It is a chemical process that destabilizes the colloidal fraction, leading to sedimentation because of flocculation (Pramanik et al., 2016). The efficacy of coagulation for removing MPs is estimated at 95% in various studies (Ahmed et al., 2021). Rajala et al. (2020) and Ma et al. (2019a) along with others, identified coagulation as effective means for confiscating hydrophobic pollutants. However, a research gap is relevant to coagulation mechanisms working under different environments (Monira et al., 2021). Lange et al. (2021a) recently highlighted the role of gross pollutant trap/forebay, a sedimentation step often used in stormwater bioretention systems, and concluded it as an appropriate alternate for removing MPs as well. Combined coagulation with UF coupling can improve the quality of effluent of discharged pollutants. Ma et al. (2019a) have stated that polyethylene is among the most abundant plastic pollutants, and their removal was estimated through Fe-based coagulation through ultrafiltration; however, the efficacy of PE particles (<0.5mm) was observed (13 to 90.9%). They attributed the result to dense floc formation in consort with the high adsorption ability of Fe-based flocks, which were positively charged under neutral conditions. PE particles are MPs' main constituents and can easily float/suspend in water. Their effect was studied with Al and Fe-based salts and the result revealed that Al-based salts were more efficacious (40%) in removing PE, which was observed at a high (15 mM) Al-based salt dosage.

Time-consuming methods, high energy consumption, and significant investment restrict such treatment possibilities. The MST, which is widely used in water purification and other sectors, is known for its dependability, energy efficiency, and lack of secondary environmental damage (Wu et al., 2010). Additional heavy metal treatment processes exist in addition to chemical precipitation, redox, electrolysis, and membrane penetration (Heo et al., 2012). In practice, such sophisticated multi-stage water treatment (membrane filtration followed by primary and secondary treatment) significantly restricts its broad use. So far, many approaches or materials have been used to overcome this issue.

Furthermore, Wang et al. recently used a $TiO_2$-based photocatalytic micromotor for microplastic removal (Wang et al., 2019b). Yifa Chen et al. (2020b) proposed a unique acetone-assisted procedure for the production of numerous Zr-MOF foam materials, as well as their excellent application in microplastic removal simulation. Though these approaches have made significant progress toward efficient MP removal, there is still a significant obstacle to overcome, as each method has its own limitations, such as the inability to handle small MPs or the inapplicability for large MPs, difficulty in large-scale processing, high energy consumption, or inability to operate in specific environments. As a result, there is an urgent need to investigate innovative methods for the efficient removal of MPs in a realistic manner that can be utilized under harsh environmental conditions.

**Table S1. Graphene and Mxene-based membranes for removing pollutants**

| **Membrane type** | **Major components of membrane** | **Membrane surface characteristics** | **Experimental conditions** | **Removal rate/performance** | **Reference** |
|---|---|---|---|---|---|
| Graphene based | graphene nanofiltration membranes CCG sheets | ≈22–53 nm thick T well packed layer structure formed by CCG sheets, 34 mg of CCG was sufficient for making square meter membrane | Evaluated on a dead-end filtration device and the pure water flux of uGNMs were high (21.8 L $m^{-2}$ $h^{-1}$ $bar^{-1}$). | High retention for organic dyes= (>99%) Moderate retention for ion salts= (≈20–60%) | (Han et al., 2013) |
| Graphene based | - | supported on a ceramic hollow fiber prepared by a vacuum suction method | At 25 °C and 2.6 wt % feed water content, | excellent water permeation for dimethyl carbonate/water mixtures High permeation flux (1702 g $m^{-2}$ $h^{-1}$). | (Huang et al., 2013) |
| Graphene based | Mixing GO with Ti3C2Tx. | lattice period= 14.28 Å interlayer spacing=f around 5 Å | pressure-driven filtration at 5 bars | Rejected dye molecules (with hydrated radii above 5 Å) Rejection rates: methyl red= 68% methylene blue= 99.5% Bengal= 93.5%, | (Kang et al., 2017) |

| | | | | | |
|---|---|---|---|---|---|
| | | | | brilliant blue =100% | |
| Graphene and MXene based | GO and MXene. | two-dimensional (2D) interlayer channels hydrophilicity (~550 nm) GO/MXene mass ratio= 1/4 water flux= (71.9 L $m^{-2}$ $h^{-1}$ $bar^{-1}$) | | Rejection of common small molecule organic dyes (NR, MB, CV, BB) = exceeds 99.5%, | (Liu et al., 2020c) |
| Graphene and MXene based | a two-dimensional (2D) GO/MXene (GM) composite lamellar membrane | lamellar structure increased interlayer spacing Excellent surface wettability to water and organic solvents. | | ultrahigh flux for pure solvents= (21.02, 48.32, 25.03, 10.76, 6.18 L/$m^2$h for water, acetone, methanol, ethanol and IPA) Outstanding dyes molecular separation performance =(over 90%) | (Wei et al., 2019) |
| Graphene based | Laminated graphene oxide (GO) Branched polyethylene-imine (BPEI). | | sonication (>1 h duration, 40 kHz frequency) pH range (2–12). | Rejection rate of methylene blue, rose bengal, and brilliant blue = (>90%) | (Nam et al., 2016) |
| Graphene based | dopamine-functionalized graphene oxide (DGO) intercalated into the MXene (Ti3C2Tx) nanosheets, | increased mechanical stability reduced interlayer spacing M4 (MXene: DGO = 1:2) 2 µm thickness of functional layer | | rejection ratio 98.1% (for Direct Red 28) and 96.1% (for Direct Black 38) high value of water flux (63.5 Lm−2 h−1) | (Zeng et al., 2021) |
| Mxene based | covalently cross-linked $Ti_3C_2T_X$ (MXene)/cellulose acetate (MXene@CA) composite | 10%MXene@CA (10:90 wt % of MXene:CA) High pure water flux= ~256.85 L $m^{-2}$ $h^{-1}$ $bar^{-1}$, 123.28% water uptake, and 69.7% porosity. | | 92% and 98% rejection of rhodamine B (RhB) and methyl green (MG), respectively. | (Pandey et al., 2020) |
| MXene based | $Ti_3C_2T_x$ by etching and ultrasonicating $Ti_3AlC_2$. | loose lamellar structure efficient permselectivity in the separation of dyes | | excellent flux (115 L $m^{-2}$ $h^{-1}$) Rejection to Congo red dye (92.3% at 0.1 MPa). | (Han et al., 2017) |

| | | | | | |
|---|---|---|---|---|---|
| MXene based | Two-dimensional (2D) MXene ($Ti_3C_2T_x$) modified with Ag nanoparticles (Ag@MXene | AgNP loadings=between 0–35%<br>21% Ag@MXene have 470 nm thickness<br>2.1 nm average pore size, | | high rejection efficiency for organic molecules excellent flux recovery | Pandey et al. (2018) |
| Graphene based | Graphene | Nano channels with a narrow size distribution (3–5 nm)<br>porous structure and significantly reduced channel length | | - | Huang et al. (2013) |
| Graphene based | Graphene oxide | highly permeability $MoS_2$ nano-supporting spacer among the graphene oxide (GO) layers | | water permeability =10.2 ± 1.68 L/(m $^2$·h·bar)<br>High rejection to different charged dyes (≥ 95%) | Zhang et al. (2019) |

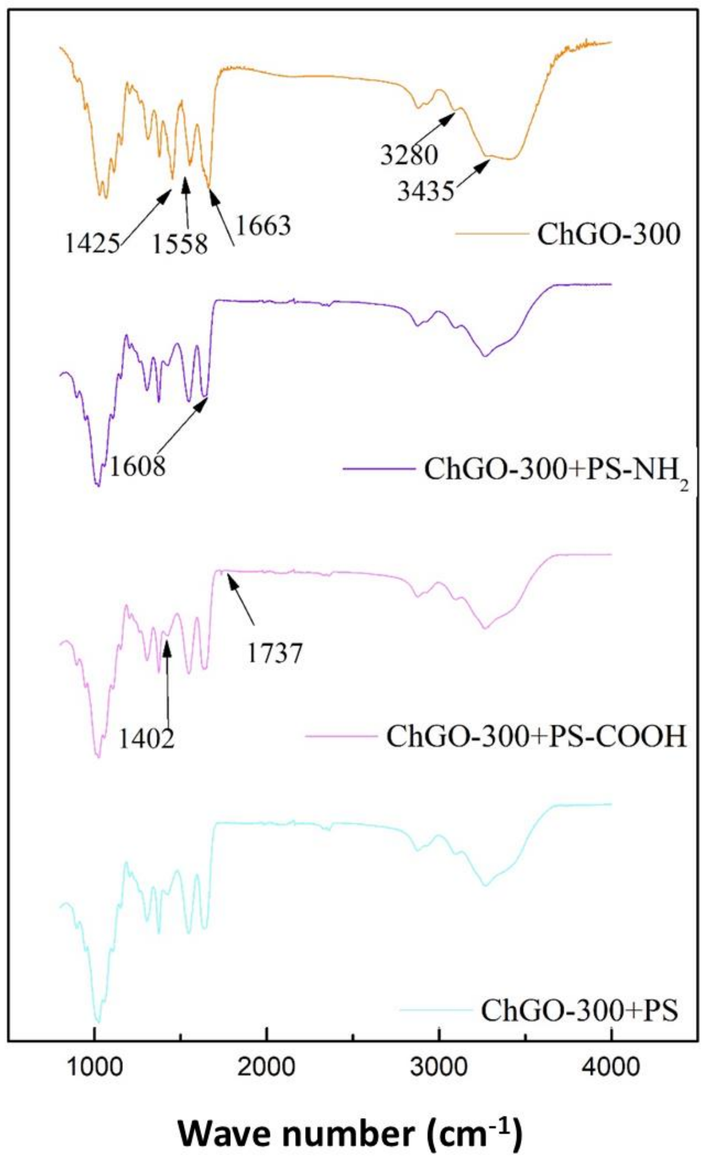


Figure S2. FTIR spectra of the original ChGO-300 sponges and the ChGO-300 sponges after adsorbing PS-$NH_2$, PS-COOH, and PS. Reproduced from Sun et al. (2020), Copyright 2018, Elsevier.

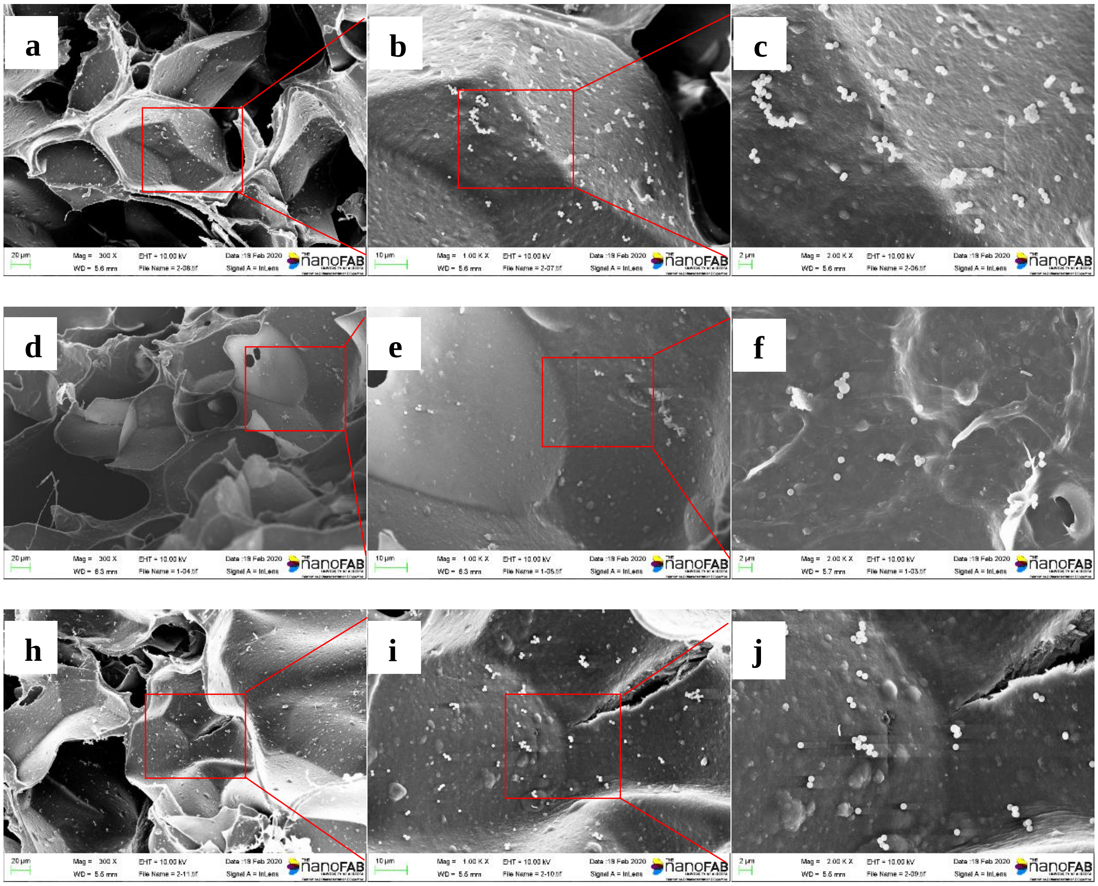


Figure S3. SEM image of the adsorption of PS-$NH_2$ (a, b, c), PS-COOH (d, e, f), PS (h, i, j) on ChGO-300 sponges Reproduced from Sun et al. (2020), Copyright 2018, Elsevier.

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