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Review

Aquatic Microplastic Pollution Control Strategies: Sustainable Degradation Techniques, Resource Recovery and Recommendations for Bangladesh

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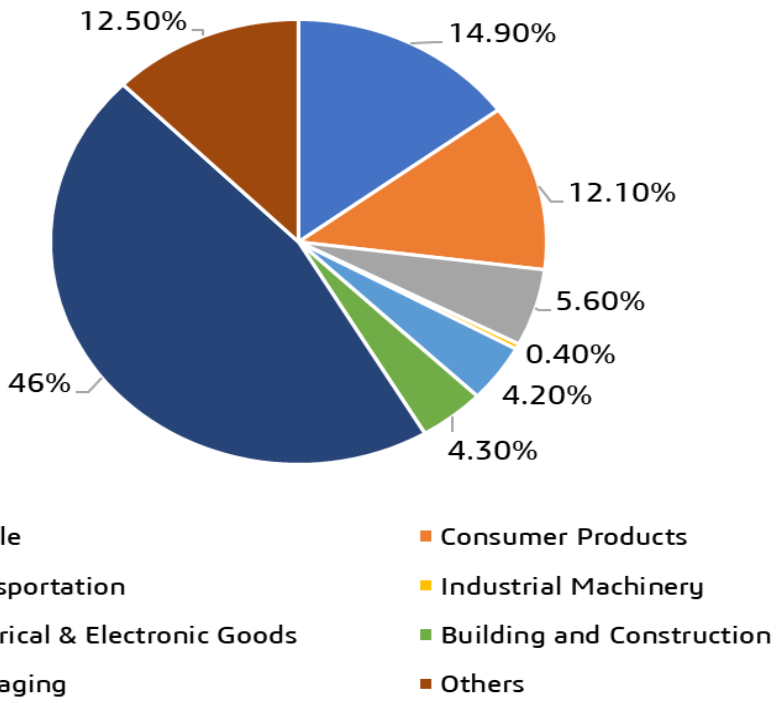

Figure S1. Global plastic waste generation by sector in 2018 [1].

Table S1. Summary of the biodegradation assay condition strain and rate.

Strain	Biodegradation Condition			Biodegradation Rate (%)	Ref
	Media	Duration	Temperature (°C)		
Bacillus cereus	A mineral salt medium containing 0.5g of polymer	1.25 month	Room Temperature	1.6; 6.6; 7.4	[2]
Bacillus gottheilli	A mineral salt medium containing 0.5g of polymer	1.25 month	Room Temperature	6.2; 3.0; 3.6; 5.8	[2]
Bacillus sp. YP1	Liquid carbon-free medium (also known as LCFBM), containing 1 g of polymer	2 months	30	10.7	[3]
Pseudomonas aeruginosa	Minimum salt media	0.94 month	Room Temperature	9.9	[4]

Table S2. Summary of the studies on periphytic biofilm degradation of microplastics.

Performed Study	Involved Microplastics	Experimentation Time/Incubation Period	Performance	Ref
Comparison and analysis of the capacity of native and bio-augmented microbial consortia to break down microplastic films under simulated marine conditions	Polystyrene (PS)	6 months	Bioaugmented consortia effectively reduced the mass of PS pieces by 4.7%, while indigenous consortia only managed to lose weight by 0.19% during that same period.	[5]
Use of periphytic biofilm in the background of various carbon sources to implement a novel strategy for the biological degradation of three structurally distinct	Polyethylene terephthalate (PET), Polyethylene (PE), and Polypropylene (PP)	60 days	Adding glucose as a carbon source boosted the biodegradation of microplastics by periphyton biofilm from 9.52% (for PP), 5.95% (for PE), and 13.24% (for PET), to 18.02% (for PP), 14.02% (for PE), and 19.72% (for PET).	[6]
Reassembling of a bacterial community on biofilm to achieve greater plastic degradation	Polyethylene terephthalate (PET), and Polyethylene (PE)	14 days	Bacteria-treated PET film decreased from 92.55% to 89.55%, while bacteria-treated PET film decreased from 49.10% to 29.50%.	[7]

Table S3. Different kinds of photocatalysts and their performance.

Photocatalyst	Microplastics	Irradiation Sources	Removal Efficiency (%)	Degradation Time	Ref
TiO_2	Polystyrene	UV Light	4.65	50h	[8]
Protein Derived N-TiO_2	Polyethylene (HDPE)	Visible Light	6.4	18h	[9]
Sol-gel N-TiO_2	Polyethylene (HDPE)	Visible Light	2.86	8h	[9]
ZnO Nano Rods	Polyethylene (LDPE)	Visible Light	_	175h	[10]
ZnO-Pt	Polyethylene (LDPE)	Visible Light	_	175h	[10]
ZnO Nano Rods	Polypropylene	Visible Light	65	456h	[10]

BioCl-X	Polyethylene	Visible Light	5.38	5h	[11]
C, N-TiO_2	Polyethylene	Visible Light	71.77 ± 1.88	50h	[12]
Au@Ni@TiO_2	Polystyrene	UV Light	67	40s	[13]

***Section S1** (6.7.1. **Homogeneous AOPs** of the main manuscript)*

With their strong oxidation capabilities, AOP have been able to efficiently breakdown or mineralize a broad variety of contaminants, such as dyes, antibiotics, and persistent organic pollutants (POPs) [14-18]. When compared to other low molecular weight (MW) organic pollutants, microplastics are far more challenging to break down because of their much larger molecular weights (MWs). Pioneering efforts aiming to degrade microplastics via this protocol have recently been disclosed and publicized. AOPs like ultraviolet (UV) pyrolysis, ultraviolet (UV) combined with hydrogen peroxide, and ozone have been the subject of much research for microplastic degradation. UV light may affect the physicochemical characteristics of plastics including polyethylene (PE), polystyrene (PS), polyethylene terephthalate (PET), and polyvinyl chloride (PVC), according to research (PVC). In the presence of UV light, the surface of plastic samples cracked, making them feel significantly harsher to the touch. In the end, the samples disintegrated into fragments ranging in size from the nanometer to the micrometer. Plastics may be degraded by ultraviolet (UV) photolysis at wavelengths between 200 and 280 nm (UV-C), 280 and 315 nm (UV-B), and 315 and 400 nm (UV-A), according to other researches [19]. Since photolysis mainly targets the plastic's surface, it takes a long time for the material to disintegrate. And, microplastics, depending on their initial size, can break down into nanoparticles of plastic over time [20]. For the elimination of pollutants, UV/ H_2O_2 technology is employed with UV photolysis. The use of UV and H_2O_2 reactions to create •OH for environmental cleanup is a well-studied AOP. •OH's high redox potentials of 1.89-2.72 V have made it a popular choice for use in a wide range of experiments involving the breakdown of numerous substances. Because of its high redox potentials of 1.89-2.72 V, •OH has been widely used in a variety of studies to break down stubborn contaminants [21].

$$H_2O_2 + h\nu \rightarrow 2 \bullet OH$$
$$H_2O_2 + h\nu \rightarrow \bullet O + \bullet OH$$

Ozone is commonly employed in AOPs due to its strong reactivity and its potency as an oxidant. Once exposed to ozone, microplastics underwent a change in their physicochemical characteristics. In light of these shifting features, several types of research on microplastic disintegration and removal by surface oxidation have been done [21]. Studying how O_3 breaks down PE microplastics was a focus for Zafar et al. (2020) [22]. Carbonyl index (CI) and hydroxyl index (HI) measurements were taken at regular intervals during ozone breakdown of microplastics at 4-7 mg/min for 60, 120, and 180 minutes. Oxidation markers such as CI and HI values rose with O_3 dosing because PE oxidation was dose dependent. The CI and HI value went up as the response time went up. Longer reaction times resulted in more oxidation, as measured by higher CI and HI values. A further investigation by Chen et al. demonstrated that at temperatures between 35 and 40 degrees Celsius, O_3 efficiently degraded polymers in 60 minutes, with a degradation efficiency of more than 90% [23]. The oxidation of microplastics by O_3 and O_3/H_2O_2 was investigated. The results showed that different microplastics, including PE, PS, and polypropylene (PP) could be broken down effectively, however surface-level physicochemical property changes were the most prominent. Intriguingly, O_3 oxidation drastically affected the microplastics' adsorption behaviors, resulting in the largest sorption capacity ever recorded for the treated PE due to the increased adsorption potential [24]. Similarly, Fenton or Fenton-like processes for microplastic degradation would be a highly promising AOP technique. Liu et al. (2019) investigated the accelerated aging behaviors of PS and HDPE microplastics after being treated with Fenton and heat-activated PDS [25]. Size distributions of untreated and treated microplastics were studied using field emission scanning electron microscopy after being subjected to heat activated $K_2S_2O_8$ (PDS) and the Fenton treatment (Fe^{2+} /H_2O_2) (FESEM). Prior to AOPs treatment, PS, PEPS, and PE were mostly found in the 40-50 μm size range. After 30 days of PDS treatments, almost all of the microplastics were smaller than 30 μm, and 80.1% and 97.4% of the PS and PE, respectively, were smaller than 20 μm. Degradation of microplastics by SO4•- based AOPs (SR-AOPs) was pioneered by Kang et al. (2019) [26]. Based on their findings, they developed a helical-shaped N-doped carbon nanotube catalyst (Mn@NCNTs) for the SR-AOPs reaction to degrade PE beads. Because of this method, after an 8-hour reaction at 160 °C, microplastics lost a considerable 54 wt% of their weight. To explain how the microplastic was oxidized and completely mineralized to H_2O and CO_2, the

authors of this research postulated a reaction mechanism in which they claimed that the continual creation of •OH and SO4•- free radicals was responsible for the oxidation. In spite of the encouraging results of the SR-AOPs reaction, it is not currently possible to recreate the required high pressure and high temperature conditions in a wastewater treatment facility. The odds of success for other methods, such as Electro Fenton-like processes, are higher in a real-world setting. Degradation efficiency is strongly influenced by the cathode material used here. Hydrogen peroxide (H_2O_2) is generated locally via a two-electron oxygen reduction reaction at the cathodes, and is then converted to the •OH free radical [27]. To degrade PVC-based microplastics in a Na_2SO_4 electrolyte, Miao et al. (2020) designed an electro Fenton-like system with a heterogeneous TiO_2/C cathode [28]. The counter electrode was graphite, while the reference electrode was an Ag/AgCl electrode. The system was evaluated based on its ability to reduce body weight and improve dichlorination. Under optical circumstances, 75% dichlorination efficiency and 56% weight loss were obtained after 6 hours of electrolysis at 100 °C at -0.7 V vs. Ag/AgCl. Prior to being attacked by •OH, the mechanical strength, flexural strength, and tensile modulus of microplastics were lowered due to the high temperature experienced throughout the process. Using a Boron doped diamond (BDD) electrode and a PS sample, Kiendrebeogo et al. (2021) degraded 89 ± 8 % microplastics in 6 hours of electrolysis time using Na_2SO_4 (0.03M) as a supporting electrolyte at a current intensity of 9 A [29]. The study's authors argue that, with further study, this procedure would be economically viable and suitable for use on an industrial scale.

*Section S2: (**6.8 Coagulation and Flocculation** of the main manuscript)*

For wastewater treatment of microplastics, one of the most feasible techniques could be coagulation and flocculation. To avoid exaggeration of information in the manuscript, this section is represented in Section S1 (Supplementary File). Coagulation is the process of using different coagulants to change dissolved and suspended solids' physical states. After the flocs of suspended solids are formed, they can be easily separated via sedimentation. This technique is utilized the most in the globe for wastewater treatment [30]. The hydrogen bonding and/or electrostatic interactions between the coagulants and suspended particles are essential for this procedure to achieve optimal separation efficiency. The principle of any coagulation process is to separate microplastic from wastewater using coagulants. Charge neutralization, adsorption, and sweep flocculation serve as the cornerstones of microplastic removal by coagulation [31]. By eliminating unstable colloid particles such as organic matter and suspended solids, flocculants are utilized further to improve the quality of water [32]. Al-based coagulants, Fe-based coagulants, synthetic organic coagulants, natural organic coagulants, etc. are coagulants used to remove microplastics. These hydrolysates of metal coagulants are applied to the surface of negatively charged microplastics to neutralize their initial charge, making them unstable and less repulsive. The positively charged microplastics then form flocs with the negatively charged microplastics and are separated via sedimentation.

Ma et al. used a Fe-based salt ($FeCl_3.6H_2O$) as a coagulant to remove polyethylene microplastics at pH 7.0 in a lab-scale simulated drinking water system to explore the effectiveness of microplastic removal by coagulation [33]. Under the diameter of 0.5 mm, the microplastic removal rate was just 13%. Zhou et al. noticed a similar phenomenon. Al-based coagulants, such as $AlCl_3.6H_2O$, have a greater microplastic coagulation rate than $FeCl_3.6H_2O$, according to studies [31]. Compared to $FeCl_3.6H_2O$'s 13 % removal efficiency, the removal efficiency improved to 36.89 % ± 3.24 %. The study also suggested that poly aluminum chloride be used instead of Fe-based coagulants [31]. This might be because Al-based flocs have a larger zeta potential than Fe-based flocs, which can better neutralize negatively charged polyethylene microplastics and facilitate subsequent sweep flocculation [34].

The addition of flocculants improves the microplastic removal efficiency drastically. Adding only a small amount of polyacrylamide (PAM) to the $FeCl_3.6H_2O$, excellent microplastic removal efficiency of 90.91% ± 1.01% was achieved [33]. Stronger adsorption bridging effects in the presence of PAM might explain the much-improved removal of polyethylene microplastics. PAM might improve the particle size and density of the flocs, resulting in a better sweep flocculation effect. Further analysis of different flocculants indicates its immense effect on microplastic removal efficiency. PAM-induced increased elimination was also seen in Al-based coagulants and polystyrene microplastics. The removal efficiency of polyethylene microplastics ($d < 0.5$ mm) rose to 45.34% ± 3.93% in the presence of $AlCl_3.6H_2O$ and PAM, according to the study. It is also reported in some studies that, along with PAM, polyamine-coated (PC) sand also increases the removal efficiency. Shahi et al. discovered that treating polyethylene microplastics ($d < 0.5$ mm) with a combination of 20 mg/L alum and 500 mg/L PC sand resulted in removal effectiveness of 92.7 %, which was greater

than that of solitary alum (65.9%) [35]. The addition of cationic PC sand boosts the bridging action between flocs and microplastics and therefore improves the settling of suspended flocs, which is responsible for the better removal performance [36]. Lapointe et al. also looked at the effectiveness of coagulation in removing pristine and aged microplastics and came up with some impressive microplastic removal figures. With an aluminum salt-based coagulant, the removal effectiveness of pristine polyester, polyethylene, and polystyrene microfibers was found to be 99%, 82%, and 84%, respectively. Weathered polyethylene removal was dramatically boosted to 99 %, compared to 82 % for pristine polyethylene [36].

Table S4. Performance of different coagulants and coagulant aids for microplastic removal.

Coagulant	Coagulant Aid	Microplastic	Removal Efficiency (%)	Ref
Fe based salts ($FeCl_3.6H_2O$)	-	Polyethylene (PE)	Below 15	[33]
Fe based salts ($FeCl_3.6H_2O$)	Anionic Polyacrylamide (PAM)	PE	90.9 ± 1.01	[33]
Poly Aluminum Chloride (PAC) (180 mg/L)	-	PE	29.70	[31]
Poly Aluminum Chloride (PAC) (180 mg/L)	-	Polystyrene (PS)	77.83	[31]
$FeCl_3$ (180 mg/L)	-	PE	Below 20	[31]
$FeCl_3$ (180 mg/L)	-	PS	Below 70	[31]
$AlCl_3.6H_2O$ (405 mg/L)	-	PE	36.89 % ± 3.24 (d < 0.5 mm)	[34]
$AlCl_3.6H_2O$ (5 mM)	-	PE	25.83% ± 2.91 (d < 0.5 mm)	[34]
$AlCl_3.6H_2O$ (5 mM)	PAM (15mg/L)	PE	45.34% ± 3.93 (d < 0.5 mm)	[34]
Alum (20 mg/L)	-	PE	(65.9) (d < 0.5 mm)	[35]
Alum (20 mg/L)	Polyamine coated sand (PC sand)	PE	(65.9) (d < 0.5 mm)	[35]
Aluminum based salt (2.73 mg Al/L)	PAM (0.3 mg/L)	Polyester Fiber	99	[36]
Aluminum based salt (2.73 mg Al/L)	PAM (0.3 mg/L)	PS	84	[36]
Aluminum based salt (2.73 mg Al/L)	PAM (0.3 mg/L)	PE	82	[36]

Finally, it can be concluded that coagulation and sedimentation are one of the most researched and sophisticated methods for removing microplastics. However, the studies also point out that chemical dosages might exceed the authorized limit, resulting in detrimental consequences. However, with some more refinement of the technique, coagulation might be our option for controlling aquatic microplastic, as the technology has already demonstrated acceptable removal of microplastic.

Table S5. Wastes collected from four sea-beaches of Cox's Bazar and Chittagong.

Type of Waste	Proportion of Total Amount (Approximate %)
Plastic	63
Foamed Plastic	13
Paper and Cardboard	9
Rubber	3
Cloth	2
Glass and Ceramic	1
Metal	1
Wood	1
Others	7

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