Microplastic Removal from Wastewater by Natural Coagulation

Subjects: Engineering, Chemical

Contributor: Taskeen Reza, Zahratul Huda Mohamad Riza, Siti Rozaimah Sheikh Abdullah, Hassimi Abu Hasan, Nur ‘Izzati Ismail, Ahmad Razi Othman

Urban industrialization has caused a ubiquity of microplastics in the environment. A large percentage of plastic waste originated from Southeast Asian countries. Microplastics arising from the primary sources of personal care items and industrial uses and the fragmentation of larger plastics have recently garnered attention due to their ubiquity. Due to the rising level of plastic waste in the environment, the bioaccumulation and biomagnification of plastics threaten aquatic and human life. Wastewater treatment plant (WWTP) effluents are one of the major sources of these plastic fragments. Coagulation is a significant process in removing microplastics, and natural coagulants are far superior to their chemical equivalents due to their non-toxicity and cost-effectiveness.

Keywords: wastewater treatment plant ; coagulation ; microplastics

1. Introduction

It is crucial to mitigate the plastic pollution from wastewater treatment plants as they release a large percentage to the environment. An estimated 3.85 × 10^{16} microplastics per year are released from wastewater effluents [1]. Existing primary and secondary treatment processes can remove approximately 66% of the microplastics in the influent [2][3]. Coagulation is the process of removing contamination in suspended particle and colloidal forms by destabilizing and aggregating the particles into large flocs. The aggregates then settle and can be removed from water using a solid–liquid separation method [4]. Coagulation is a simple and cost-effective technology used in water treatment plants. In the wake of sustainable development, research on natural coagulants as replacements for chemical coagulants has increased. Natural coagulants are renewable, biodegradable, non-toxic and cheap, making them more attractive than chemical coagulants [5]. In recent studies, chemical and natural coagulants could effectively remove microplastics in wastewater streams. However, the research on natural coagulants for microplastic removal is limited, and most research focuses on turbidity and COD removal. Despite the limited research, natural coagulants have proven efficient and can help mitigate the microplastic problem. Coagulation using natural coagulants is a sustainable and suitable solution for the microplastic problem in wastewater effluent. The mechanism involved in natural coagulation is assumed to be a combination of two or more mechanisms. Charge neutralization and bridging are the most probable mechanisms of action of natural coagulants.

2. Microplastics from Wastewater Treatment Plants (WWTPs)

Almost 98% of the microplastics in the marine environment are generated from land activities, with road runoff being the primary source, followed by treated effluents from wastewater treatment plants (WWTPs) [6]. Primary microplastics are directly introduced in wastewater effluent streams, and the washing process of synthetic textiles is considered the major source of these primary microplastics in the oceans. The minute size of the plastic particles allows them to traverse wastewater treatment plants (WWTPs) and enter marine environments [7]. Wastewater treatment plants (WWTPs) are a significant source of microplastics in the environment. Microplastics can enter WWTPs in a variety of ways, such as sewage and stormwater runoff, and are discharged into the environment along with the treated wastewater. Primary microplastics from personal care items, the fibers from textiles during washing in domestic wastewater [8], and industrial effluents containing plastic fragments used in molding and other processes are major microplastics in plants. The wet sedimentation process washes off the tiny microplastic dust particles in the atmosphere resulting mainly from the wear and tear of tires, and road markings are carried to the treatment plants through stormwater runoff [9]. Plastic wastes undergo mechanical degradation, leading to fragmentation due to extreme environmental conditions in landfills. The leachate discharge carries the plastic debris to WWTPs [10].

Identifying the shapes of the microplastics present in the wastewater is necessary as this helps with the implementation of removal technology in a WWTP. The most common shapes in wastewater are fibers, pellets, fragments and films [11]. Fibers are the most dominant shape, accounting for nearly 52.7% of the microplastics present in wastewater. This can be due to the enormous quantity of fibers discharged in domestic washing discharges [12].

2.1. Microplastic Removal in WWTPs

Current wastewater treatment plants are not intended to remove the microplastics that appear with the waste. As microplastics are an emerging pollutant, specific treatment plants have yet to be created to eliminate them. A removal efficiency of more than 88% could be reached with secondary treatment [12] with the efficiency increasing to 99.9% with tertiary treatment [9]. The fundamental design of municipal WWTPs around the world is relatively the same, with **Figure 1** showing the standard processes included in the primary, secondary and tertiary treatment steps.

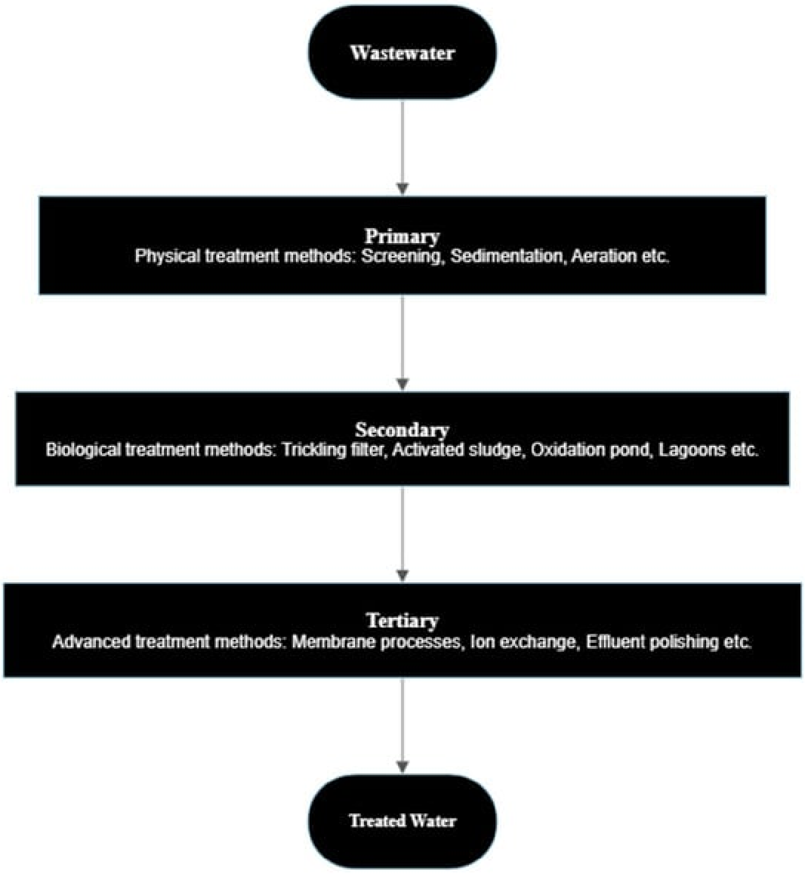

Figure 1. Classification and technologies in different steps of a wastewater treatment plant.

In the primary treatment, microplastics larger than 1000 µm can be removed [13]. Primary treatment can also remove fiber microplastics as the fibers are trapped due to flocculation and settling [12]. Conventional primary treatment was able to remove approximately 65% of the microplastics in the influent [2]. In a study conducted by Bayo et al., a removal efficiency of 74% was achieved in the primary stage of the WWTP [14]. In one study conducted, primary treatment including coagulation was able to remove 98% of microplastics, and in another study, the removal efficiency reached up to 95.3% [15][16]. In a WWTP without the coagulation process, the mean removal efficiency after treatment was found to be 72% [17]. Microplastics can become entrapped within the aeration tank during the secondary treatment of microbial polymers or sludge flocs. The microplastic removal efficiency was 67% in the activated sludge process [3]. Microplastics with a particle size of more than 500 µm were found to be absent after secondary treatment [18]. Tertiary treatment can reduce the amount of microplastics in the influents to 0.2 to 2%. Talvitie et al. conducted a study to compare the removal efficiency of different tertiary treatment technologies and found the membrane bioreactor to be the most efficient, with an efficiency of 99.9% [19].

2.2. Advanced Removal Technologies

Although microplastics in wastewater can be removed during the primary, secondary, and tertiary segments, none of the processes involved is specifically devised to remove microplastics. This causes a significant amount of microplastics to remain in the WWTP effluent, which releases these microplastics into the environment. Most microplastics are contained in sewage sludge and can be distributed through sludge land application. Advanced technologies, such as rapid sand filtration, the sol–gel method, electrocoagulation and photocatalytic degradation, are some approaches proposed for removing microplastics in WWTPs [20]. Most technologies are designed as add-on technologies for the existing secondary and tertiary treatment facilities. Rapid sand filtration is proposed as a tertiary treatment substitute to eliminate microplastics. Thus, a pre-treatment with techniques such as sedimentation and coagulation is necessary before the application of this process [21]. The electrocoagulation process causes the separation of microplastics through flotation by dissolving sacrificial anodes to free the coagulant precursors, which causes electrolysis to occur at the cathode and is depicted in **Figure 2** [22].

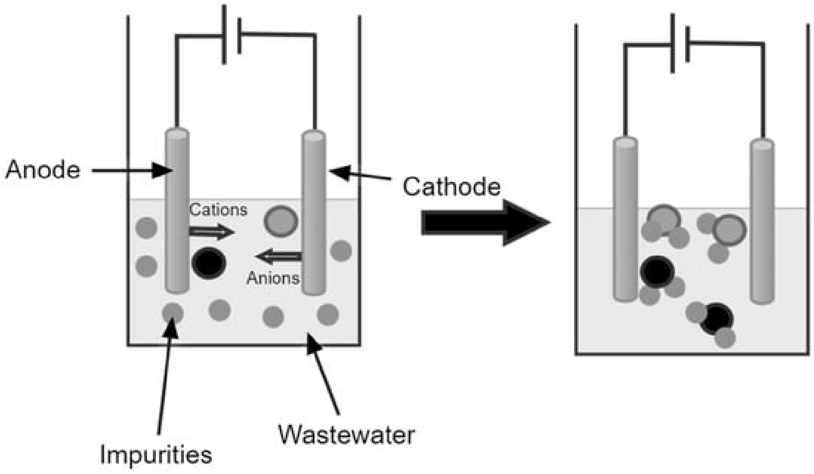

Figure 2. Electrocoagulation process.

2.3. Wastewater Management in Southeast Asia

Most WWTPs in Asia cannot remove the microplastics in wastewater influents. This is a major issue as it has been noted that Asia is the primary source of plastic waste pollution. The wastewater facilities in Southeast Asia are currently dominated by decentralized wastewater technologies (DWTs), with cluster wastewater technologies (CCTs) in the urban areas. Only a few centralized wastewater technologies (CWTs) were observed [23]. The different systems can be defined based on the treatment capacity and the proximity to the wastewater source. DWT systems' approach is treating the wastewater at or near the source. DWTs have a lower capacity of approximately 5000 person equivalents, whereas the capacity of CWTs is twofold that of DWTs [24]. CCTs are usually classified under CWTs as both systems treat wastewater from multiple households, whereas DWTs can only treat a single household. The major difference between CCTs and CWTs is the size of the facility, and CWT systems are considered large-scale CCT systems. CWTs collect wastewater from multiple households, and the wastewater is then carried to an end-of-pipe treatment facility. DWTs, on the other hand, treat the wastewater within the building with minimal collection. The facilities are minimal, and the standard technologies used are aerobic and anaerobic digestion, composting, sand/soil filtration and wetlands [25]. Both CCTs and CWTs are formed by a vast sewer network that carries the wastewater from sources to the treatment plant. With the growing population and urbanization in the countries considered, CWTs are considered the preferred treatment facility. However, most CWT facilities have deteriorated over time and cannot perform at their full capacity. A major issue with DWT systems is that they are unable to comply with the limits of environmental discharges [26].

3. Coagulation

There has been recent research on advanced technological methods to mitigate the number of microplastics in wastewater effluent. However, the specific treatment processes still need to be applied on a full scale to any wastewater treatment plant. Furthermore, implementing the technologies in the existing wastewater treatment plants could increase the plants' capital and operational costs. An economical solution to the cost problem would be to tune the operational parameters of the existing treatment processes to increase the efficiency of removing microplastics. Improving the flocculation and coagulation process could be essential in removing microplastics [12]. In a study conducted by Ma et al., it was observed that an aluminum-based coagulant showed improved efficiency in the removal of microplastics, which implies the possibility of improving the process of coagulation in wastewater treatment plants [27].

The coagulation process, as shown in **Figure 3**, consists of merging small particles into larger aggregates or flocs, followed by the adsorption of dissolved organic matter into the flocs. The flocs are removed as impurities in subsequent solid–liquid separation processes [4]. Coagulation is an important operation in wastewater treatment plants and for sludge dewatering in industries such as the pharmaceutical, pulp and paper processing and metalworking industries [28]. Coagulation is a commonly used treatment method due to it being cost-friendly and easy to operate [29].

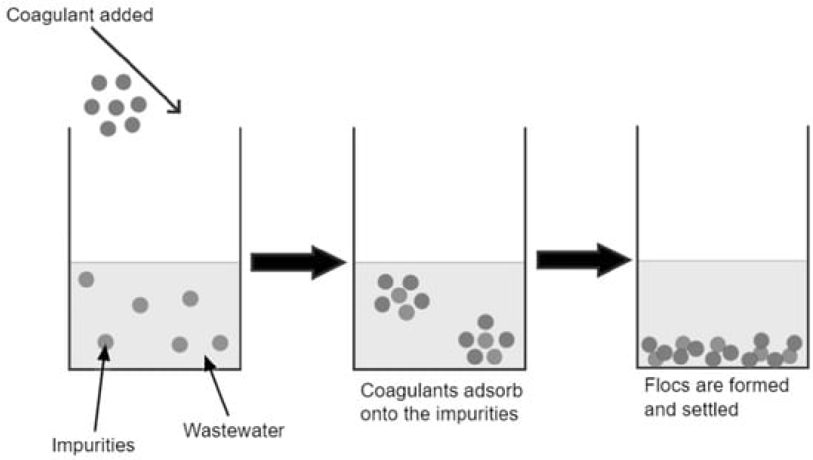

Figure 3. Coagulation process.

3.1. Mechanism

The interaction of different coagulants is varied via a broad range of mechanisms and kinetic processes. In general, the kinetics of coagulation is described as contact between the coagulant and suspended colloids by absorption through electrostatic interactions. The conformation of adsorbed polymers is rearranged, resulting in aggregation in the suspended particles, which causes them to form large flocs [29]. The mechanism involved in the coagulation process largely depends on the type of coagulant used as well as the properties of suspended particles.

In general, the coagulation mechanisms can be sorted into four types: (1) Simple charge neutralization is the neutralization of the charges present in the colloidal surface. A decline in the electrostatic repulsion to a minimum value causes the particles to aggregate and form large flocs. (2) In charge patching, heterogeneous charges on the colloids are unevenly distributed, which generates electrostatic attraction in the particles. The non-zero value of the zeta potential at the optimal dose forms a flocculation window. The electrostatic attraction leads to the eventual aggregation of the particles, forming large flocs. (3) Bridging usually occurs when the molecular weight of the coagulants is high. The long-chain coagulants connect the finer flocs to accumulate into a large one. (4) The sweeping mechanism is used by inorganic coagulants. Hydroxide precipitates are formed as a fine colloidal dispersion. Further aggregation produces hydroxide flocs [29][30][31][32]. The detailed mechanism method is shown in **Figure 4**.

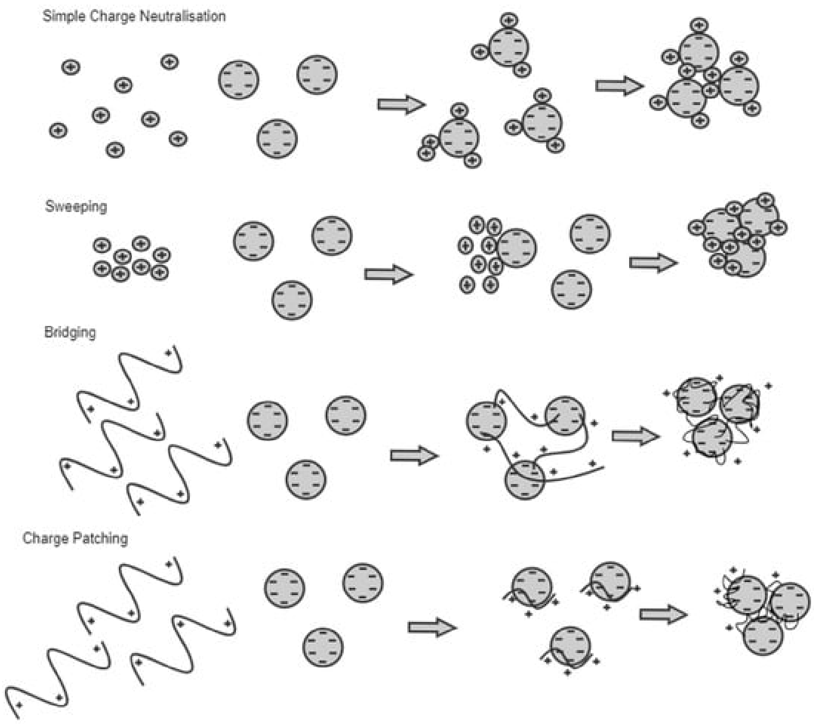

Figure 4. Mechanism of the coagulation process.

3.2. Factors Affecting Coagulation

Several factors and operational conditions can affect the process of coagulation. It is important to understand the general and specific factors affecting the process to help with the optimization. The principal operating conditions that affect coagulation are pH and temperature. Different coagulants tend to have different pH dependencies. The initial pH level can affect the surface charge on microplastics, the hydrolysis mode of a coagulant and other factors [33]. The pH value determines the type of hydrolysis taking place when using inorganic coagulants [32]. An increase in the pH value causes the negative charge on the particles' surface to become greater [34]. pH also affects the particle size of the flocs, with larger floc sizes for alkaline conditions than acidic conditions [35].

The types and dosages of coagulants also substantially influence the efficiency of coagulation. The dose of the coagulant used has a substantial effect on microplastic removal. The relation between the efficiency and dosage of coagulation depends on the primary mechanism of coagulation. If the primary mechanism is simple charge neutralization, the removal efficiency will generally increase with the coagulant dosage. This is because the absolute zeta potential value of the microplastics will gradually decrease with the addition of a coagulant. Maximum removal is achieved when the zeta potential of the microplastic is 0 [29]. If the coagulation process occurs due to several coexisting mechanisms, then microplastic's relative stabilization phenomenon does not occur if the coagulant dose is too high [36]. For the sweeping mechanism, it is assumed that a large dose of a coagulant will cause the density and structure of the flocks to be greater with stronger adsorption and sweeping effects [37]. The types of coagulants typically used are categorized into two: chemical coagulants, which include inorganic coagulants, organic synthetic coagulants and polymeric coagulants, and natural coagulants [32].

3.3. Chemical Coagulants

Chemical coagulants, which include inorganic coagulants and organic synthetic polymer coagulants, are efficient in removing microplastics in wastewater systems. Inorganic coagulants have a strong reaction with the negatively charged microplastics through the cations produced by hydrolysis [38]. In general, aluminum-based coagulants have better efficiency in removing microplastics than iron-based coagulants.

Al-based and Fe-based inorganic coagulants are the most commonly used coagulants in the research conducted on microplastic removal in wastewater. In recent years, Mg-based coagulants have been used in certain studies and have shown promising potential. Factors such as pH, the dosage of water and the presence of other substances in the sample water affect the efficiency of these coagulants. In summary, Fe-based coagulants are more effective in removing microplastics than others. However, the size and type of microplastics present in the water also play a major role in the coagulation process, and further research needs to be conducted. An increase in the coagulant dosage does not necessitate higher removal efficiency. It can be observed that increasing the coagulant dosage past the optimum value tends to decrease the removal efficiency. It is difficult to determine the effect of pH on the efficiency as it depends on the microplastic present and the coagulant used.

3.4. Natural Coagulants

Natural coagulants have a cost and environmental benefit over chemical coagulants. In recent years, natural coagulants have gained much importance in scientific communities as chemical coagulants are found to be toxic to the environment. Chemical coagulants are not biodegradable and tend to persist in water unless treated specifically [39]. The presence of aluminum, one of the most commonly used chemical coagulants, in drinking water has been linked to contributing to Alzheimer's and related diseases in humans [40]. Natural coagulants derived from plant sources can overcome these health concerns. In addition to the health issues, chemical coagulants pose a threat to the environment as they produce hazardous sludge. Natural coagulants, on the other hand, do not increase the metal load, and they produce minimal waste sludge, making them a sustainable alternative [39]. The dosage of natural coagulants required is also lower than that of their chemical counterparts, making them cost-effective. Considering all these advantages, natural coagulants are a far superior and sustainable alternative to chemical coagulants. Limited research is found on the efficiency of natural coagulants, with most research focusing on turbidity and COD removal.

Natural coagulants play a substantial role in removal efficiency when used in water treatment. Most natural coagulants used and summarized in the table above exhibit high removal efficiency. The studies summarized in the table above show that a higher coagulant dosage was directly related to a higher removal efficiency. However, this varies with the coagulant used, and further research needs to be conducted before a conclusion can be made. The active coagulant agents, polysaccharides, proteins and polypeptides also need to be studied as they play an essential role in the efficiency of these coagulants. It is observed from most studies that natural coagulants work best at an optimum pH of 7 with slight variations.

3.5. Use of Natural Coagulants in Southeast Asia

Coagulation is a common process involved in most wastewater treatment plants. Most plants use chemical coagulants, which have toxic effects on the environment and are also expensive. As cost is one of the major barriers to wastewater management in Southeast Asia, the coagulation process with the use of natural coagulants is a suitable alternative.

Diascora hispida is a plant found in the tropic and subtropic regions of the world, especially in West Africa, the Caribbean, and Southeast Asia [41]. *Diascora hispida* was used as a natural coagulant to treat textile wastewater effluent and could achieve an efficiency of 28%, 94% and 64% at an optimum pH [42]. Banana has the highest production amount in Southeast Asia [43], and a study conducted on banana peels as a natural coagulant showed the removal of 88% of turbidity under optimum conditions [44].

The removal efficiency of natural coagulants shows promising effects for removing microplastics from WWTPs. Even though the efficiency of chemical coagulants is also high, the substantial impact of chemical coagulants on the environment and living beings is a major drawback. Natural coagulants are cheap and non-toxic, making them a suitable alternative for mitigating microplastics released into the environment from WWTPs.

4. Conclusions

(i) Billions of tons of microplastic are present in the marine environment, with the majority coming from land sources. (ii) Wastewater treatment plants can remove a certain amount of microplastics during treatment. However, the plants need to be equipped with specific treatment technologies. Implementing new technologies is costly; thus, the optimization of current processes is a better alternative. (iii) The optimization of the coagulation process could help mitigate the microplastic problem in WWTPs. Natural coagulants are cheaper and more sustainable than chemical coagulants. (iv) The abundance of natural materials in Southeast Asia represents a potential for the region to implement natural coagulation in WWTPs to diminish the microplastic problem.

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