Natural Coagulants for Pharmaceutical Removal from Wastewater

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Contributor: Motasem Alazaiza, Gomaa Ali

Pharmaceutical contamination threatens both humans and the environment, and several technologies have been adapted for the removal of pharmaceuticals. The coagulation-flocculation process demonstrates a feasible solution for pharmaceutical removal. However, the chemical coagulation process has its drawbacks, such as excessive and toxic sludge production and high production cost. To overcome these shortcomings, the feasibility of natural-based coagulants, due to their biodegradability, safety, and availability, has been investigated by several researchers.

Keywords: natural coagulation ; chemical coagulation ; pharmaceuticals ; Moringa oleifera ; green treatment technology

1. Introduction

The discharge of pharmaceutical waste into the environment poses a threat to both humans and environmental systems. The disposal of these contaminants without proper treatment has resulted in pharmaceuticals being widespread in ecosystems [1]. The presence and accumulation of these emerging compounds harm the ecosystem. Human drugs such as ibuprofen and acetaminophen are continuously accumulating in the environment, resulting in pollutants in water bodies and causing harmful effects [2]. In addition, the mineralization rate of pharmaceuticals such as diclofenac and ibuprofen through photocatalysis is low, resulting in the accumulation of these compounds in the environment [2]. The effluent of wastewater treatment plants is the typical source of pharmaceutical compounds, since the conventional wastewater treatment methods are not designed to remove these micropollutants [3]. Therefore, these harmful chemicals accumulate and contaminate soil, rivers, oceans, and groundwater [4].

Recently, several studies reported the efficiency of the coagulation-flocculation treatment method for pharmaceuticals' removal, especially in rich organic wastewater [1]. Coagulation-flocculation consists of two steps: (1) the tendency of colloidal particles to form large flocs by destabilization, and (2) settling these large flocs by precipitation. The removal of pharmaceuticals directly by means of the coagulation process is not reported in the literature. The mechanism of pharmaceuticals' removal by coagulation process is indirect by using colloidal particles as a vehicle for pharmaceuticals [3][5][6].

For many years, chemical-based coagulants such as aluminum sulfate (alum) and poly-aluminum [7][8] have had different environmental effects by producing highly toxic sludge. In addition, the consumption of water contaminated by the residual chemical coagulants may cause neurodegenerative diseases [9]. Thus, the transition towards natural-based coagulants for water and wastewater treatments has gained increasing attention in recent years [10].

Natural coagulants can be produced from natural sources such as plants and animals. Many studies reported several natural sources for extracting natural-based coagulants [11][12]. Natural resources that possess a higher molecular weight may contain a more extended polymer that increases these natural coagulants' efficiency [13][14][15]. These sources have been extensively studied to treat different types of wastewater, such as textile wastewater, dairy wastewater, and domestic wastewater [16][17]. In addition, coagulants can also be obtained from animal waste such as banes and shells [18]. The main challenge of using natural coagulants in general, especially animal-based coagulants, is their continuous availability for large-scale treatment [18].

Natural coagulants perform better at a wide pH range [19][20][21]. In addition, using natural coagulants does not change the pH of water compared to chemical coagulants. In addition, natural coagulants positively affect the ecosystem and the environment [10][22][23]

2. Application of Natural Coagulants for Pharmaceutical Removal

Recently, green water and wastewater treatment technologies have gained more attention. Among these technologies, natural coagulants are a promising method for wastewater treatment [24]. In this section, the recent advancements made in using natural-based coagulants are presented.

In a recent study, Nonfodji et al. [25] prepared a natural coagulant from *Moringa oleifera* seeds, and they studied its performance for hospital wastewater treatment. The results indicate that the removal efficacy of turbidity and chemical oxygen demand (COD) was 64 and 38%, respectively. In a subsequent study, Thirugnanasambandham and Karri [26] compared the COD, turbidity, and color removal by two types of coagulants; a natural coagulant (Azadirachta indica A. Juss) and a chemical coagulant (aluminum sulfate). Remarkably, the results indicate that natural-based coagulants may not only be effective for COD, turbidity, and color removal, but may also be economically competitive, as the operating costs were USD 0.56/m^3 and USD 1.73/m^3 for natural coagulants and the chemical ones.

In another study, Maharani et al. [27] investigated the removal of COD and BOD (Biochemical Oxygen Demand) from pharmaceutical waste using moringa seed coagulant and tapioca starch coagulant. The results point to high BOD (Biochemical Oxygen Demand) and COD removal for both natural coagulants. For moringa, the BOD and COD removals were 90 and 71%, respectively, whereas for tapioca, they were 95 and 94% for BOD and COD, respectively. These results indicate that natural coagulants might be a promising treatment technology for pharmaceutical waste treatment. Oliva et al. [28] studied the use of rice husk ash functionalized by *Moringa oleifera* protein for amoxicillin removal from water solutions. They also investigated the effect of operating parameters such as coagulant dosage, initial amoxicillin concentration, and contact time. The results indicate that the used biomaterials are feasible for pharmaceutical removal from water. Olivera [29] examined the potential of using biomaterial extracted from *Moringa oleifera* for the extraction of diclofenac and oxytetracycline from wastewater. The results show the high potential for pharmaceutical removal from wastewater using biomaterial.

The removal percentages were 88% for diclofenac and 50% for oxytetracycline. Santos et al. [30] examined tetracycline removal from river water by using *Moringa oleifera* seeds. The results show 50% tetracycline removal efficiency at 0.5 g/L *Moringa oleifera* dosage. Iloamaeke and Chizaram [31] examined the removal of pharmaceuticals by Phoenix dactylifera seeds-based coagulants. The results show that maximum removal efficiency of 99.86% was achieved at a 100 mg/L coagulant dosage, a 50 min settling time, and a pH of 2.

Sibartie and Ismail [32] studied the performance of H. Sabdariffa and J. Curcas as a neutral coagulant for pharmaceutical wastewater treatment. The results demonstrate that at a coagulant dosage of 190 mg/L and pH 4, the maximum removal efficiency was achieved for turbidity (5.8%) and COD (30%) by H. Sabdariffa, while J. Curcas works best at pH 3 and a coagulant dosage of 200 mg/L to remove 51% of turbidity and 32% of COD. **Table 1** presents the application of natural coagulants to remove different types of pharmaceuticals.

Table 1. Application of natural coagulant for the removal of different types of pharmaceuticals.

Coagulants	Properties	Contaminants	Conditions	Main Results	Reference
***Moringa oleifera* seeds**	**Plant-based**	**COD (hospital wastewater)**	**Initial COD 238 mg/L; pH 6, 8; Coagulant dosage 0–4000 mg/L; Rapid mixing: 200 rpm for 3 min; Gentle mixing: 45 rpm for 30 min; Settling time 60 min.**	***Moringa oleifera* seed polymers are promising bio-coagulants for hospital wastewater treatments.**	**[25]**
Azadirachta indica A. Juss.	**Plant-based**	**COD (urban sewage)**	**Initial COD 3030 mg/L; pH 4.5;Coagulant dosage 2000–6000 g/L; Rapid mixing: 100 rpm for 1 min; Gentle mixing: 40 rpm for 30 min; Settling time 60 min.**	**Natural coagulants effectively reduced COD, turbidity, and color at optimum conditions compared to chemical coagulants**	**[26]**

Coagulants	Properties	Contaminants	Conditions	Main Results	Reference
Moringa seed coagulant, tapioca starch coagulants	Plant-based	COD (Pharmaceutical waste)	pH 6–8; Coagulant dosage 3780 mg/L; Rapid mixing: 100 rpm for 10 min; Gentle mixing: 60 rpm for 15 min;	The high removal efficiency observed with the use of tapioca flour coagulant is due to an amide group that contains a high positive charge	[27]
Rice husk ash functionalized by *Moringa* oleifera protein	Plant-based	amoxicillin	Dosage 500, 1000, 1500 mg/L; Contact time 30, 60, 90 min; Mixing speed 150 rpm; Initial amoxicillin concentration (100, 200, 300) mg/L	Rice husk ash functionalized by *Moringa oleifera* protein can be an effective treatment method for an antibiotic from water	[28]
Moringa oleifera adsorpant	Plant-based	Diclofenac and Oxytetracycline	pH 3–10; Dosage 2000 mg/L; Initial diclofenac and oxytetracycline concentration 0.2–1 mg/L; Stirring speed 150 rpm.	The removal efficiency is highly pH-dependent; diclofenac removal efficiency was 4.8% at pH 8 and 87.3% at pH 2, while the removal efficiency of oxytetracycline at pH 3 and 10 was 31 and 50%, respectively	[29]
Moringa oleifera seed	Plant-based	Tetracycline antibiotic	Tetracycline initial concentration 5 mg/L; Coagulant dosage 250–2500 mg/L; pH 5–8; Rapid mixing: 120 rpm for 1 min; Gentle mixing: 30 rpm for 15 min; Settling time 30 min	*Moringa oleifera* seed is a natural, simple, and environmentally friendly technology for antibiotic removal from contaminated water	[30]
Phoenix dactylifera	Plant-based	Pharmaceutical effluent	pH 4–10; Coagulant dosage 200–400 mg/L; Rapid mixing: 100 rpm for 2 min; Gentle mixing: 40 rpm for 20 min; Settling time 50 min	SEM analysis indicated that phoenix dactylifera adsorbed pharmaceutical particles on the surface; thus, phoenix dactylifera can be an effective green coagulant for emergency pollutant removal	[31]
Hibiscus Sabdariffa and Jatropha Curcas	Plant-based	Pharmaceutical Wastewater	Contaminant initial concentration 660 mg/L; pH 2–12; Coagulant dosage 40–200 mg/L; Rapid mixing: 100 rpm for 10 min; Gentle mixing: 40 rpm for 25 min; Settling time 50 min	Compared to chemical coagulants (Alum), natural coagulants such as J. Curcas have better performance in terms of pharmaceutical wastewater treatments	[32]

3. The Transition from Chemical to Natural Coagulant: Comparative Evaluation on Performance

The transition from chemical coagulation to natural coagulation can be an important step towards increasing green water treatment technology, reducing health risks and environmental pollution [23]. Natural coagulants can be obtained from plant or animal sources. Natural coagulants were discovered years ago, before chemical coagulants; over the years, the application of natural coagulants decreased due to the development of chemical coagulants. Recently, the rise of green water treatment technology, besides the environmental problems related to chemical coagulants, has motivated the consideration of natural coagulants again. This section presents a comparative discussion of natural and chemical coagulants.

Many studies evaluated the performance of natural coagulants for removing pollutants from water and wastewater; they concluded that natural coagulants can be competitive in terms of removal efficiency [33]. **Table 2** presents the comparison performance of natural and chemical coagulants. The combination of chemical and natural coagulants may increase the performance of the coagulation process. In a study, the combination of alum and banana peels removed 94% of turbidity, whereas the use of alum and banana peels alone resulted in turbidity removal efficiency of 73.1 and 65.6%, respectively [34].

Table 2. Comparison performance of natural and chemical coagulants.

Type of Wastewater	Chemical Coagulant	Removal Performance	Natural Coagulant	Removal Performance	Reference
Arsenic-contaminated surface water	**Ferric chloride**	**Maximum arsenic removal of 69.3% at 40 mg/L coagulant dosage**	**Cellulose and chitosan**	**Maximum arsenic removal of 84.62% at a 1 mg/L cellulose dosage and 75.87% at a 25 mg/L chitosan dosage.**	[35].
Turbidity in Surface water	**Alum**	**Turbidity removal of 78.72% at a dosage of 100 mg/L**	**Sago and chitin**	**Turbidity removal of 69.15% at a sago dosage of 300 mg/L, and 67.73% at a chitin dosage of 300 mg/L**	[36]
Paper mill industry	**Alum**	**Turbidity removal of 97.1%, COD removal of 92.7%**	***Moringa oleifera* seed**	**Turbidity removal of 96%, COD removal of 97.3%**	[37]
Paint industry	**Ferric chloride**	**Color removal of 89.4%, COD removal of 83.4%**	**Cactus**	**Color removal of 88.4%, COD removal of 78.2%**	[38]
Concreate plant	**Ferric chloride and Alum**	**Turbidity removal of 99.9%**	***Moringa oleifera* seed**	**Turbidity removal of 99.9%**	[39]
Confectionary	**PAM**	**TSS removal of 93.5% COD removal of 95.9%**	**Cactus**	**TSS removal of 92.2% COD removal of 95.6%**	[40]
Paper and mill	**Alum**	**Color removal of 80% TOC removal of 40%**	**Chitosan**	**Color removal of 90% TOC removal of 70%**	[41]
Dam water	**Alum**	**Turbidity removal of 98.5%, color removal of 98.5%**	**Watermelon seed**	**Turbidity removal of 89.3%, color removal of 93.9%**	[42]

The advantages of using natural-based coagulants over chemical ones are: (1) natural coagulants may produce less sludge than chemical coagulant; thus, the environmental sustainability increases, while the sludge handling cost decreases; (2) the natural coagulant dosage is less than that of chemical coagulants; thus, the cost and sludge production is lower; (3) the toxicity of natural coagulants is lower than that of chemical coagulants [43]; and (4) the use of natural coagulants does not require skilled workers, as they have a low health impact and do not represent such as potential environmental hazard [23].

However, natural coagulants have some disadvantages that hinder their widespread use: (1) rapid mixing during the coagulation process induces cell rupture; thus, the organic matter load may increase and react with disinfectants in the following treatment process, resulting in disinfectant by-products [44][45]; (2) the vast majority of natural coagulants are extracted from plants, so the supply of these coagulants may be affected by seasonal production [46]; (3) natural coagulants are bio-based materials; thus, this material can decompose during long-term storage [9]; and (4) some natural coagulants are used as medicines; the high consumption of these materials in water treatment could affect their supply to the medicine sector [10].

4. Conclusions

The removal of pharmaceuticals from water and wastewater is challenging due to their low concentration and their resistance to biodegradation. Several studies reported the feasibility of using natural-based coagulants for water and wastewater treatments. The main mechanisms that natural coagulants use for pharmaceutical removal from water and wastewater are charge neutralization and polymer bridging. Plant-based natural coagulants are more affordable than animal-based ones. Although the application of natural coagulants for emergency pollutants, especially pharmaceuticals, is limited in the literature, the available data demonstrate a promising future for these bio-coagulants in this domain. A natural coagulant has advantages over a chemical coagulant as a low dosage is required, less sludge is produced, and low/no toxicity is presented. For the complete transition from chemical coagulants to natural coagulants, further research is required in areas such as developing reliable extraction methods, searching for new natural sources, determining the optimal conditions for pharmaceutical removal, and evaluating the effect of environmental parameters on the process' performance.

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